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GASEOUS DIFFUSION COEFFICIENTS.
A COMPREHENSIVE CRITICAL EVALUATION
OF EXPERIMENTAL STUDIES AND CORRELATIONS
OF RESULTS

by

CASE FILE
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ABSTRACT

Title of Thesis: Gaseous Diffusion Coefficients. A Comprehensive Critical Evaluation of Experimental Studies and Correlations of Results

Thomas R. Marrero, Doctor of Philosophy, 1970

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Diffusion coefficients of binary mixtures of dilute gases are comprehensively compiled, critically evaluated, and correlated by new semiempirical expressions. Data for seventy-four systems are sufficiently extensive and accurate to be recommended for the National Standard Reference Data System of the U. S. National Bureau of Standards. Deviation plots are given for most of these systems. Almost every gaseous diffusion coefficient which was experimentally determined and reported prior to 1969 can be obtained from the annotated bibliography and table of gas pairs.

A detailed analysis of experimental methods is given, and intercomparison of their results helps establish reliability limits for the data, which depend strongly on temperature. Direct measurements are supplemented by calculations based on knowledge of intermolecular forces derived from independent sources—molecular beam scattering for high temperatures, and London dispersion constants for low temperatures. In addition, diffusion coefficients for several mixtures are obtained from experimental data on mixture viscosities and thermal diffusion factors. Combination of all these results gives diffusion coefficients over a very extensive temperature range, from very low temperatures to 10,000°K.

All data are corrected for composition dependence and for quantum effects. New semi-empirical equations are derived for making such corrections easily.

The correlated diffusion data are used to develop a corresponding-states procedure for predicting diffusion coefficients on the basis of fragmentary information. This procedure involves only two parameters, and appears to be more accurate than any prediction scheme heretofore proposed.

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Figure

Group III

LIST OF FIGURES. (Continued)

Figure

1. INTRODUCTION

The purpose of this report is to recommend gaseous diffusion coefficients as Standard Reference Data for the National Bureau of Standards. This survey of gaseous diffusion coefficients was prepared under a program for the critical evaluation of physical science data sponsored by the National Bureau of Standards through the National Standard Reference Data System.[1,2] Short catalogues of measured diffusion coefficients have previously appeared in the International Critical Tables,[3] the Landolt-Börnstein Tables,[4] and the Thermophysical Properties Research Literature Retrieval Guide.[5] Additional limited reviews are also available.[6-10] In the present survey gaseous diffusion coefficients are comprehensively compiled and critically evaluated, including analysis for consistency with theory and with other measured molecular properties. Such detailed evaluation is possible because of the existence of a highly developed theory for dilute gases.

The scope of this survey is therefore limited to the dilute gas region. In this region the density is such that the rates of transport of mass, momentum, and energy are entirely controlled by binary molecular collisions. In practice this means gases with densities approximately corresponding to standard conditions (i.e., of the order of 10^{19} - 10^{20} molecules per cm^3). The emphasis here is on two-component (binary) mixtures. Multicomponent diffusion can be accurately described in terms of the binary diffusion coefficients for all possible pairs of gases in the mixture. All the atoms or molecules considered are neutral species. The mass transport processes outside the scope of this survey are as follows: (1) diffusion of ionized particles (ion mobility), (2) mixture separations in a temperature gradient (thermal diffusion), (3) mixing due to convection or turbulence, and (4) the permeation of gases through liquids or solids. The sole interest is the mixing of gases caused by composition gradients.

The gaseous diffusion coefficients recommended here as Standard Reference Data are succinctly reported by means of semi-empirical functions; temperature limits range up to 10,000°K and to a lower temperature of the order of 100°K (Tables 12 and 13). There is a small composition dependence in the diffusion coefficients which may be estimated from parameters listed in Table 15. Figures 5 to 81 are deviation plots, for 62 gas pairs, of experimental measurements from the semi-empirical equations; these graphs illustrate discrepancies in the data.

The procedure used in this report is as follows. At all possible temperatures published diffusion coefficient values were critically evaluated on an individual gas-pair basis. The data assessment was determined without any additional experimental measurements. From the rigorous kinetic theory of gases an approximation was developed to make corrections for small composition effects. Coefficients could then be normalized to a specific mixture concentration for comparison and subsequent correlation. Diffusion coefficients derived from other transport property measurements, particularly mixture viscosities, were useful for the extension of values to intermediate temperatures and for consistency checks. In the absence of direct measurements, intermolecular forces from theory and from beam experiments served to determine diffusion coefficients at very low and elevated temperatures, respectively. Semi-empirical functions were constructed to precisely correlate the data over three decades of temperature within the experimental uncertainty.

This report is divided into five major sections. It begins with a chapter - Theoretical Background - which includes the diffusion coefficient definition and its theoretical expression according to the rigorous kinetic theory of gases. The kinetic-theory foundations are necessary for the understanding of temperature and composition dependences, and quantum effects. This chapter closes with equations for the determination of diffusion coefficients from intermolecular forces and from other transport properties. The principal experimental techniques are described next. Methods of measurement are classified by the geometry of the apparatus and their reliability is estimated. Procedures used to critically evaluate the entire body of experimental data for accuracy, composition and temperature dependencies are outlined under Treatment of Data. The analyses and results related to the small composition dependence of the diffusion coefficient are entirely in this section. The semi-empirical correlation equation was chosen on the basis of knowledge of how intermolecular forces affect the temperature dependence of the diffusion coefficients. Previously uncalculated values of low-temperature asymptotes of diffusion coefficients are tabulated.

The fourth major section - Results - can be subdivided into four areas. First, diffusion-coefficient uncertainty limits are classified according to temperature and gas pair. Second, the tabulation of correlation parameters for recommended Standard Reference Data are listed. Then a series of graphs shows the relative deviations between the recommended coefficients and the data. An inspection of these graphs will readily indicate that the unqualified selection of a diffusion coefficient from the literature may be uncertain by at least several percent. The last part of the Results section contains detailed remarks about data appraisals for specific systems. The final major part of this report is the Bibliography; two annotated bibliographies are given, one is all the experimental sources, complete through 1968, according to author (gas pair and method are noted), and the second is a supplementary listing of citations according to gas pair. Practically all diffusion coefficients ever measured can be traced through these bibliographies; however, for many systems the results are too fragmentary or too uncertain to be recommended as Standard Reference Data.

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2. THEORETICAL BACKGROUND

In this chapter the presentation of the theoretical background is preceded by the phenomenological definition of diffusion coefficients. Then in outline form expressions for diffusion coefficients are derived by the Chapman-Enskog procedure for a solution of the Boltzmann equation. Most mathematical details of the derivation are omitted, and the discussion accentuates the application limits of these rigorous kinetic-theory formulas. More complete information is available in three monographs,[1-3] and recent developments appear in several articles.[4-13] The emphasis here is on molecular physics as a prerequisite to the understanding of accepted theoretical results which are necessary in order to achieve the following:

(1) suggest the mathematical form for the correlation of diffusion coefficients as a function of temperature,

(2) correct diffusion coefficient measurements for composition dependence,

(3) estimate quantum effects for low-temperature diffusion coefficients,

(4) calculate diffusion coefficients directly from knowledge of intermolecular forces, and

(5) calculate diffusion coefficients from other transport properties.

2.1 Phenomenological Definition of the Gaseous Diffusion Coefficient

In a nonuniform mixture the diffusion coefficient is a proportionality constant between the molecular flux and the composition gradient of a species. Diffusion coefficients are defined by phenomenological equations for two-component and multicomponent mixtures.

a. Two-component Mixtures

In two-component mixtures, in the absence of temperature and pressure gradients, external forces, and chemical reactions, the flux equations are

$$\mathbf{J}_1 = -n\mathcal{D}_{12}\boldsymbol{\nabla}x_1 \quad , \tag{2.1-1}$$

$$\mathbf{J}_2 = -n\mathcal{D}_{21}\boldsymbol{\nabla}x_2 \quad . \tag{2.1-2}$$

Each species (or component) is labelled by subscripts 1 or 2. The flux densities are $\mathbf{J}_1$ and $\mathbf{J}_2$ (molecules/cm^2-sec), the total number density is n (molecules/cm^3), and the composition gradients are in terms of mole fractions x_1 and x_2. These equations hold only in the case of zero net flux, $\mathbf{J}_1 + \mathbf{J}_2 = 0$. If the net flux is not zero, Eqs. (2.1-1) and (2.1-2) can be considered to hold in a coordinate system moving with the net flux, that is, at a velocity equal to $(\mathbf{J}_1 + \mathbf{J}_2)/n$.

The diffusion coefficients $\mathcal{D}_{12}$ and $\mathcal{D}_{21}$ are positive constants with units of cm^2/sec. It is easy to show from Eqs. (2.1-1) and (2.1-2) that $\mathcal{D}_{12} = \mathcal{D}_{21}$, because $\mathbf{J}_1 + \mathbf{J}_2 = 0$ and $x_1 + x_2 = 1$ for a binary mixture. Thus diffusion in a binary mixture is described by a single diffusion coefficient.

Molecular diffusion, strictly speaking, cannot occur under conditions in which both the net flux and the pressure gradient are simultaneously zero. If the pressure is uniform, then in general fluxes are different for different species, and the net flux is not zero. If the net flux is zero, a small pressure gradient must exist in order to counter the tendency for the different species fluxes to be different.[14-16] For instance, in a closed system the difference in the species fluxes causes the number density and hence the pressure to increase on one side of the system and decrease on the other side until the resulting pressure gradient forces the net flux to be zero. If the fluxes were to remain unequal in a closed system, then the pressure would continue to increase on one side and decrease on the other side.

The pressure gradients in diffusing gas mixtures turn out to be very small in magnitude, however; in fact, they are almost immeasurably small except in capillary tubes, where they have been measured.[17-26] Because of this, it is unnecessary to include in the flux equations any term directly proportional to a pressure gradient. The whole effect of any pressure gradient is simply to modify the net flux, and this is the only term that needs to be directly included.

The generalization of Eqs. (2.1-1) and (2.1-2) for nonzero net flux is therefore simply

$$\underset{\sim}{J}_1 = -n\mathcal{D}_{12}\underset{\sim}{\nabla}x_1 + x_1\underset{\sim}{J} \quad , \qquad (2.1\text{-}3)$$

$$\underset{\sim}{J}_2 = -n\mathcal{D}_{21}\underset{\sim}{\nabla}x_2 + x_2\underset{\sim}{J} \quad , \qquad (2.1\text{-}4)$$

where the net flux is $\underset{\sim}{J} = \underset{\sim}{J}_1 + \underset{\sim}{J}_2$. Equations (2.1-3) and (2.1-4) define the diffusion coefficient in a stationary coordinate system. It can be easily shown, as before, that $\mathcal{D}_{12} = \mathcal{D}_{21}$.

The preceding equations, which define binary diffusion coefficients, are applicable to any fluid, and hold regardless of any dependence of the diffusion coefficient on composition, pressure, or temperature. For dilute gases the pressure and composition dependences are especially simple-the diffusion coefficient is inversely proportional to pressure and is only slightly dependent on mixture composition. The temperature dependence is more complicated. All these factors are treated in more detail in subsequent sections.

b. Multicomponent Mixtures

The flux of a species in a multicomponent mixture is not conveniently expressed in terms of composition gradients as in the foregoing equations for binary mixtures. The reason is that the multicomponent diffusion coefficients thereby defined have an excessively complicated composition dependence which makes the calculation of a flux a formidable task. A simpler set of equations for multicomponent diffusion is obtained by a different arrangement of terms - the composition gradient of a species is related to differences in fluxes of gas pairs.[2,3,8,27] The outstanding advantage of such a relationship is a description of multicomponent diffusion coefficients for binary mixtures. These equations, credited to Stefan and Maxwell, are

$$\underset{\sim}{\nabla x_i} = \sum_{j=1}^{\nu} \frac{n_i n_j}{n^2 \mathcal{D}'_{ij}} \left(\frac{\underset{\sim}{J_j}}{n_j} - \frac{\underset{\sim}{J_i}}{n_i} \right) , \qquad (2.1\text{-}5)$$

where i and j denote the species. For a mixture of ν species there are ν equations, but only $\nu-1$ are independent. The diffusion coefficients $\mathcal{D}'_{ij}$ depend primarily only on the nature of the species i and j, but are not quite the same as the corresponding binary diffusion coefficients $\mathcal{D}_{ij}$ (hence the prime). However, the difference between the $\mathcal{D}'_{ij}$ and the $\mathcal{D}_{ij}$ lies only in their weak composition dependence, the exact value of $\mathcal{D}'_{ij}$ depending slightly on the composition of the whole multicomponent mixture and not on just the relative amounts of i and j. The variation of the binary $\mathcal{D}_{ij}$ with composition is empirically a few percent at most, and is of the same magnitude as the experimental uncertainties in the few available multicomponent $\mathcal{D}'_{ij}$, or even in most measured binary $\mathcal{D}_{ij}$ for that matter. Therefore it is reasonable on an empirical basis to take $\mathcal{D}'_{ij} \approx \mathcal{D}_{ij}$ for multicomponent diffusion. This is also justified theoretically, for in the first Chapman-Enskog approximation $\mathcal{D}'_{ij}$ and $\mathcal{D}_{ij}$ are identical and independent of composition.[2,3,7]

The special case of a trace species diffusing through a uniform multicomponent mixture is of interest for two reasons. It provides a simple test of $\mathcal{D}'_{ij} \approx \mathcal{D}_{ij}$, and makes possible the calculation of diffusion coefficients of various species in air. First, denote the trace species by 1 and assume the absence of a net flux ($\underset{\sim}{J} = 0$), then Eqs. (2.1-5) reduce to a single equation,

$$\underset{\sim}{\nabla} x_1 = -\underset{\sim}{J}_1 \sum_{j=2}^{\nu} \frac{x_j}{n\mathcal{D}'_{ij}} \quad . \tag{2.1-6}$$

If the trace diffusion coefficient $\mathcal{D}_1$ is defined to be the constant of proportionality between $\underset{\sim}{J}_1$ and $\underset{\sim}{\nabla} x_1$, then

$$\frac{1}{\mathcal{D}_1} = \sum_{j=2}^{\nu} \frac{x_j}{\mathcal{D}'_{ij}} \quad , \tag{2.1-7}$$

where $\mathcal{D}_1$ specifies the diffusion coefficient of the tracer in the multicomponent mixture. If $\mathcal{D}_{ij}$ replaces $\mathcal{D}'_{ij}$ then Eq. (2.1-7) becomes an expression of Blanc's law.[28] Detailed calculations[29] of $\mathcal{D}'_{ij}$ and $\mathcal{D}_{ij}$ for this special case show that the deviations from Blanc's law are small for ordinary gases. This further justifies the application of binary diffusion coefficients to Eq. (2.1-5) for multicomponent diffusion. The second case of interest, the calculation of diffusion coefficients of a species in air (when direct measurements are unavailable or inadequate), is especially convenient by the application of Blanc's law with available binary diffusion coefficients of the species in nitrogen and in oxygen.

2.2 Molecular Theory of Diffusion

a. General Background

This presentation of the molecular theory of diffusion briefly outlines some major points of the rigorous kinetic theory of gases. Kinetic theory postulates transport due entirely to molecules in motion. In diffusion the individual molecules themselves carry mass through the gas. Since there are immense numbers of molecules moving about in a gas it is to be expected that molecular encounters (or collisions) are of cardinal importance in controlling the overall rate at which transport occurs. The collisions in turn are controlled by the forces of interaction between the molecules. By the formulas of kinetic theory, knowledge of these fundamental intermolecular forces can lead to gaseous diffusion coefficients.

The importance of molecular collisions in diffusion can be illustrated by some typical numerical values. At ordinary conditions of temperature and pressure molecules in gases have molecular speeds of the order of 10^4 cm/sec, which is about the speed of sound. In contrast, actual diffusion velocities $(\underset{\sim}{J}_i/n_i)$ are much less - about 1 cm/sec. This great decrease in apparent molecular speed occurs because diffusion is dominated by collisions which cause the molecular paths to be twisted into tortuous shapes. The actual path of a molecule is approximately 10^4 times the net distance travelled during diffusion. For gases at ordinary conditions only binary collisions are important; ternary and higher-order collisions are very unlikely. Binary collisions, two-particle encounters, are characteristic of gases with the ratio of mean free path to molecular diameter of the order of 100.

Transport phenomena - diffusion, viscosity, thermal conductivity, and thermal diffusion - arise by deviations, however slight, from the equilibrium molecular velocity distribution function known as the Maxwell distribution. At equilibrium conditions an isolated gas mixture has no gradients in composition, pressure, or temperature; thus no fluxes. Therefore to obtain transport coefficients on a theoretical basis knowledge of a nonequilibrium velocity distribution function is a necessary requirement.

b. Theoretical Methods

Diffusion coefficients can be calculated from a flux derived from a molecular concept - the integral of molecular velocity over the nonequilibrium velocity distribution function. The velocity distribution function represents the probability for a molecule to have a specific velocity and location at some instant. The changes in the velocity distribution due to molecular interactions must satisfy the nonlinear Boltzmann integrodifferential equation. The basic problem of rigorous kinetic theory is to solve the Boltzmann equation.

A solution of the Boltzmann equation was independently obtained by Chapman and by Enskog. Both used a method of successive approximation, and even though procedures by Chapman and Enskog differ in detail the results are identical. The transport properties appear finally in the Chapman-Enskog theory as solutions of infinite sets of simultaneous algebraic equations, and the transport properties can be expressed formally as ratios of infinite determinants whose elements are the coefficients of the algebraic equations. The coefficients of the equations are complicated functions which depend on the species and the composition of the mixture, and on integrals related to binary molecular interactions. These sets of equations can be solved, fortunately, by rapidly converging approximation schemes.

An outline of the Chapman-Enskog procedure is as follows. First the velocity distribution function is expanded in terms of a perturbation function added to the Maxwell (equilibrium) distribution. By the assumption of a small perturbation, the expansion substituted back into the Boltzmann equation leads to a linearized integrodifferential equation for the perturbation.

The perturbation term is assumed proportional to gradients, and expanded in a series; the series expansion coefficients are functions of molecular velocities. The assumption of linearity in the gradient of composition is precisely consistent with the preceding phenomenological definition of diffusion coefficients; other transport coefficients may be accounted for by additional appropriate gradients. The diffusion coefficient now appears as an integral of the expansion coefficient over the molecular velocities. The expansion coefficient satisfies a linear integrodifferential equation obtained from the Boltzmann equation. This equation is solved by a second series expansion in terms of squares of molecular velocities. For the second expansion it is convenient, but not necessary, to use orthogonal functions because orthogonal properties lead to subsequent simplification of the calculations. The orthogonal functions usually used are Sonine polynomials. When this second expansion is substituted back into the integral expression for the diffusion coefficient, it turns out (because of the orthogonality) that the diffusion coefficient is exactly equal to just one of the coefficients in the second expansion. The problem now is to find the coefficients of the second expansion. To do this, the expansion is substituted back into the linear integrodifferential equation, which is then solved by a moment method. The result is an infinite set of algebraic equations in which the unknowns are the coefficients of the second expansion, and the coefficients of these unknowns are complicated multiple integrals over molecular velocties. These integrals result from the moment formation; most of the integrations can be carried out explicitly, but not all, until the law of intermolecular force is specified.

The diffusion coefficient is thus equal to a single unknown in an infinite set of algebraic equations. This set cannot be solved exactly except in very special cases, and some successive approximation procedure must be used. The set is systematically truncated in some plausible way (two ways are commonly used, one due to Chapman and Cowling, and the other to Kihara); the simplest truncation gives the first approximation to the diffusion coefficient, the next step gives the second approximation, and so on. In the first approximation the diffusion coefficient is independent of composition; the second and higher approximations introduce composition dependence. Since the approximation procedure converges rapidly, the third approximation for the diffusion coefficient is almost identical with the second approximation.

The solution of the Boltzmann equation by the Chapman-Enskog procedure depends on the following assumptions:

Binary collisions. The Boltzmann equation itself has a fundamental assumption - binary collisions. This assumption - that only two-molecule interactions are important - limits the application of theoretical results to transport properties of dilute gases.

Small mean free path. The Chapman-Enskog solution assumes that the dimensions of the gas container are large compared to the molecular mean free path. In gases at extremely low densities molecules collide more frequently with the walls of the container than with each other. When molecular collisions with a container surface are significant, the theory fails.

Small perturbation. In the Chapman-Enskog theory the assumption of a small perturbation function describes small departures from the equilibrium velocity distribution function; in other words, at conditions slightly away from equilibrium the transport property fluxes are linear in the gradients.

Classical mechanics. Historically, classical mechanics was necessarily used by Boltzmann, Chapman, and Enskog; however, their theory can be reformulated to account for quantum-mechanical effects. The modification needed is merely to replace an integration over classical impact parameters for molecular interactions by an integration over deflection angles involving the quantal differential cross section.

Elastic collisions. The original Boltzmann equation and its solution by Chapman and Enskog were limited to elastic collisions between molecules interacting with central forces. Inelastic collisions occur between molecules with internal degrees of freedom, and kinetic energy is no longer conserved, although mass and momentum are conserved. Thus diffusion and viscosity are not strongly affected by the presence of internal degrees of freedom, but thermal conductivity is. The theory may be reformulated to account for inelastic collisions.

c. Theoretical Results

In this section the Chapman-Enskog theoretical expressions for diffusion coefficients are given, as well as the definition of collision integrals, and a number of complementary definitions related to binary molecular collisions. The extension of the collision integral formulas to include inelastic collision effects is also given.

Approximation Scheme for Diffusion Coefficients. The higher approximations for diffusion coefficients in a dilute gas binary mixture with species of type 1 and 2 are written

$$[\mathcal{D}_{12}]_M = [\mathcal{D}_{12}]_1 \, f^{(M)} \quad , \qquad (2.2\text{-}1)$$

where $[\mathcal{D}_{12}]_1$ is the first approximation, $f^{(M)}$ accounts for the effects of higher approximations, and M indicates the order of approximation. In the first approximation for diffusion coefficients, $f^{(1)} = 1$; the effect of higher approximations is described by

$$f^{(M)} = 1/(1 - \Delta_{12} - \ldots), \qquad (2.2\text{-}2a)$$

$$f^{(M)} = (1 + \Delta_{12} + \ldots) \quad , \qquad (2.2\text{-}2b)$$

where Δ_{12} is the first correction term to $[\mathcal{D}_{12}]_1$.

First approximation for the Diffusion Coefficient. The expression for $[\mathcal{D}_{12}]_1$ is

$$[\mathcal{D}_{12}]_1 = \frac{3}{16} \left(\frac{2\pi kT}{\mu_{12}}\right)^{1/2} \left(\frac{1}{n\bar{\Omega}_{12}^{(1,1)}}\right) \quad , \qquad (2.2\text{-}3)$$

where $\mu_{12} = m_1 m_2/(m_1 + m_2)$ is the reduced mass of a pair of molecules, m is the molecular mass of a species, k is the Boltzmann constant, and T is the absolute temperature. The diffusion collision integral $\bar{\Omega}_{12}^{(1,1)}$ has units of area and is dependent on the temperature and the forces of molecular interaction of the gas. The collision integral for diffusion is

$$\bar{\Omega}^{(1,1)}(T) = \frac{1}{2}(kT)^{-3} \int_0^\infty e^{-E/kT} E^2 S^{(1)}(E)dE, \qquad (2.2\text{-}4)$$

where E is the initial relative translational energy of two molecules in a binary collision, $E = \frac{1}{2}\mu_{12}v^2$, v being the initial relative speed of the molecular pair, and the diffusion transport cross section is

$$S^{(1)}(E) = 2\pi \int_0^\pi (1 - \cos \chi) I(\chi,E) \sin \chi d\chi \quad , \qquad (2.2\text{-}5)$$

where $I(\chi,E)$ is the differential scattering cross section. For classical systems $I(\chi,E)\sin\chi d\chi = bdb$, where b is the impact parameter - the perpendicular distance between one molecule and the initial line of relative approach of the other molecule. The classical scattering angle for a pair of colliding molecules is

$$\chi = \pi - 2b \int_{r_c}^{\infty} \frac{dr}{r^2} \left[1 - \left(\frac{b}{r}\right)^2 - \frac{\varphi(r)}{E}\right]^{-1/2} \quad , \qquad (2.2\text{-}6)$$

where r_c, the distance of closest approach, is given by

$$1 - \left(\frac{b}{r_c}\right)^2 - \frac{\varphi(r_c)}{E} = 0 \quad . \qquad (2.2\text{-}7)$$

In Eq. (2.2-6) r is the internuclear separation distance, and $\varphi(r)$ is the spherically symmetric intermolecular potential.

The expression for $[\mathcal{D}_{12}]_1$ in practical units is

$$[\mathcal{D}_{12}]_1 = 0.008258 \left(\frac{M_1+M_2}{2M_1M_2}\right)^{1/2} \frac{T^{3/2}}{p\bar{\Omega}_{12}^{(1,1)}} \quad , \qquad (2.2\text{-}8)$$

where T is in degrees Kelvin, p is the pressure in atmospheres, M_1 and and M_2 are the molecular weights in grams per mole, and $\bar{\Omega}_{12}^{(1,1)}$ is in angstroms squared.

The first approximation for the diffusion coefficient is independent of mixture composition.

Second Approximation for the Diffusion Coefficient. The second approximation for the diffusion coefficient is

$$[\mathcal{D}_{12}]_2 \equiv [\mathcal{D}_{12}]_1(1 + \Delta_{12}) \quad , \qquad (2.2\text{-}9)$$

where

$$\Delta_{12} = \frac{1}{10}\,(6C_{12}^{*}-5)^2 \left(\frac{x_1^{\,2}P_1 + x_2^{\,2}P_2 + x_1x_2P_{12}}{x_1^{\,2}Q_1 + x_2^{\,2}Q_2 + x_1x_2Q_{12}}\right). \qquad (2.2\text{-}10)$$

The P's and Q's are complicated algebraic expressions which contain various collision integrals and are defined in Section 2.4; C_{12}^{*} is a collision integral ratio given by Eq. (2.2-16). The first correction term

Δ_{12} is temperature dependent, and contains the small composition dependence of the diffusion coefficient.

Accuracy of Formulas for the Diffusion Coefficient. How close $[\mathscr{D}_{12}]_1$ is to $\lim_{M\to\infty}[\mathscr{D}_{12}]_M$ depends on composition, molecular masses, and the intermolecular forces of the gas mixture. By numerical comparison of $[\mathscr{D}_{12}]_1$, $[\mathscr{D}_{12}]_2$, $[\mathscr{D}_{12}]_3$, etc. for a variety of special cases, the accuracy of $[\mathscr{D}_{12}]_1$ may be assessed.[30] For the case of nearly equal molecular masses $[\mathscr{D}_{12}]_1$ is probably accurate to within 2% regardless of the composition or intermolecular forces. If the molecular masses are very unequal and the heavy component is the trace species then $[\mathscr{D}_{12}]_1$ is accurate to within 1%. If the light component is the trace then $[\mathscr{D}_{12}]_1$ may be quite inaccurate; the worst case known is a mixture of rigid spheres for which $[\mathscr{D}_{12}]_1$ is low by about 13%. In practical cases it is probably safe to regard $[\mathscr{D}_{12}]_1$ as accurate within about 5% for all gas pairs, and $[\mathscr{D}_{12}]_2$ as accurate within 2%.

Pressure Dependence of Diffusion Coefficients. All theoretical approximations for dilute-gas diffusion coefficients are inversely proportional to density, or pressure. It can be shown by elementary kinetic theory arguments that the molecular flux is independent of pressure for binary collisions. The reason is that the number of flux carriers (i.e., the molecules) is directly proportional to their number density n, but the number of particles that impede the flux by collisions is also proportional to n. The two effects exactly compensate. If the associated gradient is chosen so as not to involve n, then the constant of proportionality must also be independent of n. Thus the coefficients of viscosity and thermal conductivity are independent of density. But the proportionality constant for diffusion is arbitrarily chosen to be $n\mathscr{D}_{12}$ (for historical reasons), so that $\mathscr{D}_{12}$ itself must be inversely proportional to n.

Collision Integrals for Elastic Collisions. The general equation for collision integrals is

$$\bar{\Omega}^{(\ell,s)}(T) = [(s+1)!(kT)^{s+2}]^{-1} \int_0^\infty e^{-E/kT} E^{s+1} S^{(\ell)}(E)\, dE, \quad (2.2\text{-}11)$$

with

$$S^{(\ell)}(E) = \left[1 - \frac{1 + (-1)^\ell}{2(1 + \ell)}\right]^{-1} \int_0^{2\pi} d\phi \int_0^\pi (1 - \cos^\ell\chi) I(\chi,\phi,E) \sin\chi\, d\chi, \quad (2.2\text{-}12)$$

where ℓ and s specify weighting factors related to the mechanism of transport by molecular collisions; χ and ϕ are the polar azimuth angles which describe the orientation of the final relative molecular velocity to the initial relative velocity in a collision. From Eq. (2.2-3) it is evident that for diffusion $\ell = 1$ and $s = 1$; the viscosity and thermal conductivity collision integrals have $\ell = 2$ and $s = 2$. Other values of ℓ and s occur only in the expressions for higher approximations. Collision integrals are calculated for realistic intermolecular force models only by difficult numerical integrations.

The definition of collision integrals as dimensionless reduced quantities, that is, collision integrals divided by the analogous quantities for rigid-sphere molecules, makes calculations of transport coefficients more convenient. The reduced collision integral is defined as

$$\Omega^{(\ell,s)*} \equiv \frac{\bar{\Omega}^{(\ell,s)}}{\pi\sigma^2}, \quad (2.2\text{-}13)$$

where σ is an arbitrary molecular size or range-of-force parameter. Numerical values of reduced collision integrals are usually about unity if σ is chosen in a reasonable way, and differences from unity reflect molecular "softness" for the selected intermolecular force model in comparison to an ideal rigid-sphere model.

In the higher approximations for diffusion coefficients, and in other transport properties as well, several recurring ratios of collision integrals, or reduced collision integrals, are defined for calculation convenience, namely

$$A^* = \Omega^{(2,2)*}/\Omega^{(1,1)*} \quad , \qquad (2.2\text{-}14)$$

$$B^* = [5\Omega^{(1,2)*} - 4\Omega^{(1,3)*}]/\Omega^{(1,1)*} \quad , \qquad (2.2\text{-}15)$$

$$C^* = \Omega^{(1,2)*}/\Omega^{(1,1)*} \quad , \qquad (2.2\text{-}16)$$

$$E^* = \Omega^{(2,3)*}/\Omega^{(2,2)*} \quad . \qquad (2.2\text{-}17)$$

The magnitude of each of these ratios is approximately unity, and exactly unity for rigid spheres.

Collision integrals and collision integral ratios are functions of temperature and the parameters of the selected model for intermolecular forces. Since such models usually have at least two parameters, one with dimensions of distance and one with dimensions of energy, it is economical to tabulate collision integrals in dimensionless form, in which the reduced collision integral is given as a function of a reduced temperature. Reduced collision integrals have already been defined; reduced temperature is usually defined as

$$T^* \equiv kT/\varepsilon \quad , \qquad (2.2\text{-}18)$$

where ε is the energy parameter of the potential (usually the depth of the minimum).

Collision Integrals for Inelastic Collisions. As previously mentioned, the kinetic theory of gas transport properties by the Chapman-Enskog procedure applies strictly to molecules that have no internal degrees of freedom. To extend the preceding equations to polyatomic and polar molecules the theory of transport properties must account for inelastic collisions. This can be achieved only by a reformulation of the Boltzmann equation in which the nonequilibrium velocity distribution function must be specified for all the internal energy states of molecules. A semiclassical treatment is used in which the translational molecular motion is described classically, as before, but the internal motions are described quantum-mechanically. The formal kinetic theory of transport properties that includes inelastic collisions in the Chapman-Enskog scheme was originally developed for pure gases by Wang Chang, Uhlenbeck, and deBoer,[31] and by Taxman.[32] Additional theoretical work[7-9,13] has extended the theory to mixtures; the derived collision integrals correspond to the first approximations of the Chapman-Enskog theory.

The available results for inelastic collision integrals are for the most part formal in the sense that the integrations are too difficult to carry out for realistic models, even with the fastest available computers. But useful conclusions can be drawn from them without going through elaborate calculations. These conclusions are stated at the end of this subsection.

The general equations for the diffusion and viscosity collision integrals are as follows:

$$\bar{\Omega}_{qq'}^{(\ell',s)}(T) = 2[(s+1)!Z_q Z_{q'}]^{-1} \sum_{ijk\ell} e^{-\varepsilon_{qi}-\varepsilon_{q'j}} \int_0^\infty \gamma^{2s+3} e^{-\gamma^2} S_{ij}^{(\ell')k\ell}(E)\,d\gamma \quad , \tag{2.2-19}$$

where

$$\gamma^2 S_{ij}^{(1)k\ell}(E) = \int_0^{2\pi} d\phi \int_0^\pi (\gamma^2-\gamma\gamma'\cos\chi) I_{ij}^{k\ell}(\chi,\phi,E)\sin\chi\,d\chi \quad , \tag{2.2-20}$$

$$\gamma^4 S_{ij}^{(2)k\ell}(E) = \frac{3}{2}\int_0^{2\pi} d\phi \int_0^\pi I_{ij}^{k\ell}(\chi,\phi,E)\sin\chi\,d\chi[\gamma^2(\gamma^2-\gamma'^2\cos^2\chi)-\frac{1}{6}(\gamma^2-\gamma'^2)^2] , \tag{2.2-21}$$

$$\gamma^2-\gamma'^2 = \varepsilon_{qk}+\varepsilon_{q'\ell}-(\varepsilon_{qi}+\varepsilon_{q'j}) \quad , \tag{2.2-22}$$

$$\gamma^2 = E/kT \quad , \tag{2.2-23a}$$

$$\gamma'^2 = E'/kT \quad , \tag{2.2-23b}$$

in which the prime on γ refers to the relative kinetic energy after a collision and the species are denoted by q and q'. The various ε's are the energies of the internal quantum states of the species, divided by kT. Z_q and $Z_{q'}$ are the internal partition functions for the q and q' species: $Z_q = \sum \exp(-\varepsilon_{qi})$ and $Z_{q'} = \sum \exp(-\varepsilon_{q'j})$. They appear only as normalization factors in Eq. (2.2-18). The indices i and j denote the i th and j th internal quantum states of the q th and q' th species before a collision, and k and ℓ the corresponding states after a collision. The differential scattering cross section $I_{ij}^{k\ell}(\chi,\phi,E)$ describes collisions between two molecules initially in internal states i and j which undergo a collision and finally are in states k and ℓ. In the collision integral of Eq. (2.2-18) the superscript ℓ is primed so as not to be confused with the ℓ th quantum state.

The collision integrals for inelastic processes reduce exactly to collision integrals for elastic collisions when E' = E and the differential scattering cross section is the same as the elastic cross section, $I_{ij}^{ij} = I_{el}$ for all i and j.

Inelastic collisions enter $\bar{\Omega}^{(1,1)}$ only through the term $\gamma\gamma'\cos\chi$; to a first approximation $\gamma \sim \gamma'$ and the inelastic collisions have no effect. For a second approximation γ' can be written as γ plus some terms in $\Delta\varepsilon_{qq'}$, where $\Delta\varepsilon_{qq'} = \gamma^2 - \gamma'^2$; the inelastic contributions are then of the form $\gamma(\Delta\varepsilon_{qq'})\cos\chi$. For isotropic molecular scattering the correction term vanishes, and even for nonisotropic scattering the inelastic contribution is probably small unless there is some special correlation between $\Delta\varepsilon_{qq'}$ and χ. The $\bar{\Omega}^{(2,2)}$ may also reduce to a manageable form, in the first approximation $\Delta\varepsilon_{qq'} << \gamma^2$ and the terms in $\Delta\varepsilon_{qq'}$ may be dropped. For a second approximation, the terms in $\Delta\varepsilon_{qq'}$ vanish for isotropic scattering.

The determination of $\mathcal{D}_{12}$ from mixture viscosity measurements is especially related to A^*_{12}. In these calculations the algebraic expressions (see section 2.7) appear mathematically the same whether the molecular collisions are elastic or inelastic. This is important because the only effect depends on what value is substituted for A^*_{12}. A first-order expansion for A^*_{12} indicates only a small correction for inelastic collisions, but good approximations are not yet available.

2.3 Temperature Dependence of Diffusion Coefficients

The temperature dependence of $\mathcal{D}_{12}$ according to the preceding expressions must be investigated in order to develop a general equation useful for the correlation of diffusion coefficients. Almost the entire temperature dependence is given by the factor $[T^{3/2}/\bar{\Omega}^{(1,1)}(T)]$ appearing in $[\mathcal{D}_{12}]_1$; that is, the higher approximations have only a slight effect. Accordingly the temperature dependence of Δ_{12} is disregarded in the following discussion. The temperature dependence of $[\mathcal{D}_{12}]_1$ can be calculated if the law of force between two molecules is known. Details about intermolecular forces will follow later in this section. Calculations for plausible molecular force laws have shown that the derivative $d \ln \bar{\Omega}^{(1,1)}(T)/d \ln T$ usually lies between 0 and -1/2, so that the derivative $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ lies between 3/2 and 2. Thus $\mathcal{D}_{12}$ should vary as $T^{3/2}$ to T^2, and this is usually found to be the case experimentally. These general features are depicted in Fig. 1.

This figure shows the derivative $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$, obtained from experimental data and intermolecular force models. The simple molecular model of ideal rigid spheres, sets a lower bound of 3/2 to the derivative, independent of temperature. Actual gas pairs, however, have appreciably greater values of the derivative than 3/2.

The general characteristics of $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ are as follows. At extremely low temperatures the dominant interaction is the long-range r^{-6} London dispersion energy, which causes $\bar{\Omega}^{(1,1)}(T)$ to vary as $T^{-1/3}$. At extremely high temperatures the dominant interaction is the (roughly) exponential short-range repulsion energy, which causes $\bar{\Omega}^{(1,1)}(T)$ to have a weaker temperature dependence than at low temperatures. Thus $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ is equal to 11/6 at low temperatures, and equal to a smaller value, ∿ 1.7, at high temperatures, the high-temperature value being slightly dependent on temperature. In the intermediate temperature region $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ is not monotonic, and exhibits a maximum where both short-range and long-range forces are significant.

In Fig. 1 the insert shows $\ln(p\mathcal{D}_{12}/T^{3/2})$ vs. $\ln T$. This curve illustrates the behavior to be expected from very low temperatures up to about 10^4°K, and indicates the form of relationship needed to fit $\mathcal{D}_{12}$ data as a function of temperature. Possible quantum effects at very low temperatures have been ignored in this illustration since they are important only for a few very light gases.

More quantitative information about the temperature dependence of $\mathcal{D}_{12}$ requires additional details about intermolecular potentials. As is well known, molecules attract each other at large separation distances and repel each other at small separations. In principle, quantum theory provides a method for calculating the interaction between a pair of molecules.[33] The long-range interactions are dominated by London dispersion forces, and can be calculated fairly accurately,[34] but the short-range interactions are too complicated to be calculated in any simple way. The various interactions and their effect on $\mathcal{D}_{12}$ are considered below.

a. Long-Range Interactions

These interactions behave asymptotically as (neglecting retardation effects)

$$\varphi(r) = -C/r^6 \quad , \qquad (2.3\text{-}1)$$

where C is the London constant. According to classical mechanics the collision integral has the form

$$\bar{\Omega}^{(1,1)} \propto (C/T)^{1/3} . \qquad (2.3\text{-}2)$$

Thus as T → 0, $\mathcal{D}_{12} \propto T^{11/6}$ classically, but at sufficiently low temperatures quantum corrections become important. A general expression for the quantum-mechanical $\bar{\Omega}^{(1,1)}$ as $T \to 0$ is not presently available.

b. Short-Range Interactions

Short-range interactions can be approximated by an exponential function, and over a more limited range by an inverse power. These single-term potentials have a simple algebraic form which permits the collision integral to be calculated numerically; such results lead to values of $\mathcal{D}_{12}$ at high temperatures, $T \gtrsim 1000$ °K.

The expression for the exponential potential is

$$\varphi(r) = \varphi_o \exp(-r/\rho) \quad , \qquad (2.3\text{-}3)$$

in which φ_o and ρ are empirical parameters. For this potential the $\bar{\Omega}^{(1,1)}(T)$ has been evaluated[35] over a wide temperature range by numerical methods, and its temperature dependence found to be approximately

$$\bar{\Omega}^{(1,1)}(T) \propto [\ell n(\varphi_o/kT)]^2 \quad . \qquad (2.3\text{-}4)$$

Thus at high temperatures diffusion coefficients are expected to be proportional to $T^{3/2}/[\ell n(\varphi_o/kT)]^2$.

The inverse-power repulsive potential can be written as

$$\varphi(r) = K/r^s \quad , \qquad (2.3\text{-}5)$$

where K and s are empirical parameters. For this potential the temperature dependence of the collision integral is exactly[1,2]

$$\bar{\Omega}^{(1,1)}(T) \propto (sK/kT)^{2/s} \quad . \qquad (2.3\text{-}6)$$

For this model the diffusion coefficients are proportional to $T^{3/2 + 2/s}$.

c. Intermediate-Range Interactions

At intermediate internuclear separation distances the potential is not dominated by either attractive or repulsive forces. The potential has a "well" whose detailed shape is not precisely known; descriptive approximations are frequently given by semi-empirical expressions which interpolate between functions derived for solely attractive or repulsive interactions. For spherical nonpolar molecules two such well-known approximations are,

Lennard-Jones (n-6)

$$\varphi(r) = \left(\frac{n\varepsilon}{n-6}\right) \left[\frac{6}{n} \left(\frac{r_m}{r}\right)^n - \left(\frac{r_m}{r}\right)^6 \right] , \qquad (2.3\text{-}7)$$

and

Exp-6

$$\varphi(r) = \left(\frac{\alpha\varepsilon}{\alpha-6}\right) \left\{\frac{6}{\alpha} \exp\left[\alpha\left(1 - \frac{r}{r_m}\right)\right] - \left(\frac{r_m}{r}\right)^6 \right\}, \qquad (2.3\text{-}8)$$

where ε is the depth of the potential energy well, r_m is the location of the potential energy minimum, and n and α are parameters which reflect the steepness of the repulsive forces. Such potentials give a complicated relationship for the temperature dependence of the collision integral, and no analytic expression can be given corresponding to intermediate temperatures (about 200 °K to 1000 °K for most gas pairs). However, Sutherland[36] developed a simple relationship for rigid-sphere molecules with weak attractive interactions, and showed that

$$\Omega^{(1,1)*} = 1 + S/T \quad , \qquad (2.3\text{-}9)$$

where S is a positive constant. The temperature dependence of $\mathscr{D}_{12}$ is then

$$\mathscr{D}_{12} \propto T^{3/2}/(1 + S/T) \quad , \qquad (2.3\text{-}10)$$

which correlates experimental results well over moderate temperature ranges. This form can also accurately represent collision integrals for the Lennard-Jones (12-6) potential (within 0.2% for $1.4 < kT/\varepsilon < 3.5$).[37] Another relationship, suggested by Reinganum,[38] is

$$\Omega^{(1,1)*} = e^{S/T} \quad , \qquad (2.3\text{-}11)$$

or

$$\mathscr{D}_{12} \propto T^{3/2}\, e^{-S/T} \quad , \qquad (2.3\text{-}12)$$

which reduces to the Sutherland form for small values of S/T.

2.4 Composition Dependence of Diffusion Coefficients

In this section the theoretical results are given for the small composition dependence of gaseous diffusion coefficients. The composition correction, less than 5% for most gas pairs, is needed to eliminate systematic discrepancies in the evaluation and correlation of $\mathscr{D}_{12}$ measurements. The composition correction term, Δ_{12}, is repeated here for convenience, and the P and Q terms are expressed as follows:

$$\Delta_{12} = \frac{(6C_{12}^{*} - 5)^2}{10} \left[\frac{X_1^{\,2}P_1 + X_2^{\,2}P_2 + X_1X_2P_{12}}{X_1^{\,2}Q_1 + X_2^{\,2}Q_2 + X_1X_2Q_{12}} \right] \quad , \qquad (2.2\text{-}10)$$

where

$$P_1 = \frac{2M_1^{\,2}}{M_2(M_1+M_2)} \left(\frac{2M_2}{M_1+M_2} \right)^{1/2} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{22}} \right)^{2} \quad , \qquad (2.4\text{-}1)$$

$$P_{12} = 15\left(\frac{M_1-M_2}{M_1+M_2}\right)^2 + \frac{8M_1M_2A^*_{12}}{(M_1+M_2)^2} \quad , \qquad (2.4\text{-}2)$$

$$Q_1 = \frac{2}{M_2(M_1+M_2)}\left(\frac{2M_2}{M_1+M_2}\right)^{1/2} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{12}}\right)^2$$

$$x[(5/2-6/5\ B^*_{12})M_1{}^2 + 3M_2{}^2 + 8/5M_1M_2A^*_{12}] \quad , \qquad (2.4\text{-}3)$$

$$Q_{12} = 15\left(\frac{M_1-M_2}{M_1+M_2}\right)^2\left(\frac{5}{2}-\frac{6}{5}B^*_{12}\right) + \frac{4M_1M_2A^*_{12}}{(M_1+M_2)^2}\left(11-\frac{12}{5}B^*_{12}\right)$$

$$+\frac{8}{5}\frac{(M_1+M_2)}{(M_1M_2)^{1/2}} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \frac{\Omega_{22}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{12}}\right)^2 \left(\frac{\sigma_{22}}{\sigma_{12}}\right)^2 \ . \qquad (2.4\text{-}4)$$

The relations for P_2 and Q_2 are obtained from those for P_1 and Q_1 by an interchange of subscripts. The subscript "11" denotes molecular interactions between two type 1 molecules, and so on. The Chapman and Cowling relations for the Q's have been presented, not Kihara's.[39]

The above complicated formulas for Δ_{12} are tedious to use, and attempts have been made to simplify the expressions.[40-42] The results are semi-empirical approximations, one of which[42] takes a form that determines the most sensitive parts of Δ_{12} from experiment and the remainder from theoretical calculations. An improved semi-empirical approximation for Δ_{12} is developed in this report; details appear in Section 4.1.

2.5 Quantum Effects on Diffusion Coefficients

Quantum effects become significant when the de Broglie wavelength, $\lambda = h/\mu v$, approaches the size parameter σ. Thus the ratio λ/σ is a measure of quantum effects, and gases behave classically for $\lambda/\sigma << 1$. In kinetic theory it is common practice to use the deBoer parameter Λ^*,

$$\Lambda^* \equiv h/[\sigma(2\mu\varepsilon)^{1/2}] \quad , \qquad (2.5\text{-}1)$$

which is simply λ/σ for a colliding pair of reduced mass μ and kinetic energy equal to the depth, ε, of the potential well. The larger the value of Λ^*, the more important are the quantum effects at a given reduced temperature, $T^* \equiv kT/\varepsilon$. This is illustrated in Table 1, which is based on calculations for the Lennard-Jones (12-6) potential.[43] A gas behaves classically at all temperatures for $\Lambda^* = 0$; typical values of Λ^* are as follows: 0.35 for Ne-Ar, 1.3 for He-Ne, 1.5 for H_2-D_2, and 2.9 for ^{3}He-^{4}He. From Table 1 it is evident that quantum deviations in $\mathcal{D}_{12}$ can be quite large for light gases at low temperatures. However, the collision integral ratio A^*_{12} has deviations of only a few percent, so that reliable values of $\mathcal{D}_{12}$ can be computed from accurate viscosity measurements even when quantum effects are important, as explained in more detail in Section 2.7.

The only modification necessary for quantum effects is the replacement of the integration over classical impact parameters by one over the quantum mechanical differential cross section. The quantum transport cross section as given by a scattering phase-shift analysis,

$$I(\chi) = |f(\chi)|^2 \quad , \qquad (2.5\text{-}2)$$

$$f(\chi) = \frac{1}{2i\kappa}\sum_{\ell=0}^{\infty}(2\ell+1)[\exp(2i\delta_\ell)-1]P_\ell(\cos\chi) \quad , \qquad (2.5\text{-}3)$$

in which δ_ℓ is the phase shift, ℓ denotes the angular momentum quantum number, and κ is the wave number of relative motion, equal to $2\pi\mu v/h = 2\pi/\lambda$. The phase shifts are obtained by the solution of the radial wave equation. $P_\ell(\cos\chi)$ is a Legendre polynomial in $\cos\chi$, and $f(\chi)$ is the scattering amplitude. When Eqs. (2.5-2) and (2.5-3) are substituted into Eq. (2.2-12), the integrations can be carried out to yield the following expressions for the diffusion and viscosity (or thermal conductivity) transport cross sections:

$$S^{(1)}(E) = \frac{4\pi}{\kappa 2} \sum_{\ell=0}^{\infty} (\ell+1) \sin^2(\delta_{\ell+1}-\delta_\ell) \quad , \qquad (2.5\text{-}4)$$

and

$$S^{(2)}(E) = \frac{4\pi}{\kappa 2} \sum_{\ell} \frac{(\ell+1)(\ell+2)}{(2\ell+3)} \sin^2 (\delta_{\ell+2}-\delta_\ell) \quad . \qquad (2.5\text{-}5)$$

The summations are over all integral values of ℓ from 0 to ∞ for distinguishable particles, but only over the even or odd integral values for indistinguishable particles (in which case the summation is multiplied by a normalization factor of 2). In order to describe observable processes $S^{(1)}$ must always refer to distinguishable particles, but $S^{(2)}$ can refer to either. These formulas apply only to the case of elastic collisions; corresponding formulas for inelastic collisions have never been derived.

It is often desired to adjust measurements of $\mathcal{D}_{12}$ for a set of isotopes to a common molecular weight basis. This is especially important for hydrogen isotopes (H_2, D_2, T_2, HD, etc.) for which there are many measurements for different isotope pairs. The diffusion coefficient has a mass dependence which may involve three factors. First, the principal dependence of $\mathcal{D}_{12}$ on mass is the proportionality to the inverse square root of the reduced mass of the gas pair. A second mass dependence factor is in the composition correction term Δ_{12}, but this is almost always negligible. The third dependence is in the diffusion collision integral, which in the quantum case depends on mass through the deBoer parameter Λ^*. In order to make the necessary computations a potential model is assumed, and the diffusion collision integral is then obtained for both isotopic mixtures; for the Lennard-Jones (12-6) potential quantal collision integrals have been published[43,44] as a function of the deBoer parameter and reduced temperature in convenient tabular form.

For any two mixtures a simple ratio of $\Omega^{(1,1)*}(\Lambda^*, T^*)$ is taken to adjust the data according to Eq. (2.2-8). Since this procedure is model-dependent, it is reliable only when the adjustment is small.

It is sometimes useful to express the exact formulas of Eqs.(2.5-4) and (2.5-5) as semiclassical expansions, in which the leading term is the classical formula and the quantum corrections appear as a series in powers of Planck's constant (or Λ^*). Explicit expressions have been obtained for the first two quantum corrections,[45] but little use has yet been made of these results. Most numerical calculations to date have used the exact formulas in terms of phase shifts.

2.6 Determination of Diffusion Coefficients from Intermolecular Forces

In this section expressions for diffusion collision integrals are presented which lead to $\mathcal{D}_{12}$ at conditions unavailable by direct experiment. The expressions for $\bar{\Omega}^{(1,1)}$ are given only for long-range and short-range interactions; for intermediate-range interactions, the $\bar{\Omega}^{(1,1)}$ are not given because the corresponding values of $\mathcal{D}_{12}$ are available by direct experiment. Information about long-range interactions is obtained from molecular polarizabilities, oscillator strengths, and other optical data; a summary of the various results has been published.[34] The short-range interactions are based on molecular beam scattering experiments.[46-48] For both ranges of interaction the specific data sources used in this report are listed in the Bibliography, Sections II and III.

a. Diffusion Collision Integrals for Long-Range Interactions

The collision integral for the London r^{-6} attractive potential is

$$\bar{\Omega}^{(1,1)} = 71.1\ (C/T)^{1/3} \quad , \qquad (2.6\text{-}1)$$

in which C is the London constant in atomic units ($e^2 a_0^5$)and $\bar{\Omega}^{(1,1)}$ has units of angstroms squared. Equation (2.6-1) gives the classical-mechanical low-temperature asymptote for the first approximation of the diffusion coefficient, that is, $[\mathcal{D}_{12}]_1$ as $T \to 0$.

The accuracy of the available London constants is within 5% for most gases, and at worst 10% for gas pairs containing xenon.[34] By Eq. (2.6-1) the first approximation for the diffusion coefficient is inversely proportional to the 1/3 power of the London constant; thus the errors in $[\mathcal{D}_{12}]_1$ due to errors in C are less than 4% for all gases considered. Numerical values are given in Section 5.2.

The valid range of temperature for the low-temperature asymptote is difficult to estimate accurately, but this range may be approximated as follows. First, the upper limit is given by the condition at which the London dispersion energy ceases to dominate interactions. From Fig. 1 this is estimated to occur at reduced temperatures $\lesssim 0.2$. Second, the lower limit is determined by the magnitude of quantum effects. These effects depend strongly on the deBoer parameter Λ^* and reduced temperature in a complex manner, and no simple estimate seems possible for the lower limit of temperature for Eq. (2.6-1). For gas pairs with large values of the deBoer parameter, quantum effects are quite significant at $T^* < 0.2$, as shown in Table 1. This suggests that Eq. (2.6-1) is of only qualitative value for $\Lambda^* > 1$ and $T^* < 0.2$. At $\Lambda^* = 1$ and $T^* \geq 0.1$, Eq. (2.6-1) is useful only to a 10% level of uncertainty. For $\Lambda^* = 0.5$ and $T^* \geq 0.02$ the low-temperature asymptote is accurate to within 3%, and for $\Lambda^* < 0.5$ it is even better.[43]

b. Diffusion Collision Integrals for Short-range Interactions

The diffusion collision integral expressions for short-range interactions in terms of the exponential and inverse power models are as follows. The exponential potential, Eq. (2.3-3), gives

$$\bar{\Omega}^{(1,1)} = 4\pi\alpha^2 \rho^2 I_{(1,1)} \qquad (2.6\text{-}2)$$

in which $\alpha = \ln(\varphi_o/kT)$ and $I_{(1,1)}$ is an integral available from tables[35] as a function of α. The inverse power potential, Eq. (2.3-5), gives

$$\bar{\Omega}^{(1,1)} = \pi\left(\frac{sK}{kT}\right)^{2/s} \Gamma\,(3-2/s)\; A^{(1)}(s) \quad , \qquad (2.6\text{-}3)$$

in which $\Gamma(3-2/s)$ is the gamma function of argument $(3-2/s)$ and $A^{(1)}(s)$ is an integral, independent of temperature and available in tables[49,50] for different values of $\underline{s}$.

The reliability of diffusion coefficients calculated from molecular beam experiments is estimated as follows. First, the consistency of $\mathscr{D}_{12}$ by molecular beam results and by direct diffusion experiments can be checked at about 1000°K, a temperature at which these results overlap. The agreement is within a few percent for the gas pairs He-Ar, He-N_2, and H_2-Ar. Other gas pairs do not have sufficient data for such a comparison. Second, the uncertainties of the potentials can be evaluated by comparison (1) with reliable theoretical calculations, (2) with potentials obtained from different apparatus in the same laboratory and from different laboratories, and (3) with potentials derived from other transport property measurements at elevated temperatures.[46-48,51] The potentials are determined from molecular beam scattering experiments which have been done only at two independent laboratories: Amdur et al. at the Massachusetts Institute of Technology, and Leonas et al. at the Moscow State University. This information has a level of reliability that varies with the type of gas. The noble gas pairs have uncertainties in the potentials that range from about 10 to 30%. Gas pairs with diatomic molecules have higher uncertainties, about 20 to 45%, and for polyatomic molecules even higher uncertainties, 30 to 60%.

The diatomic and polyatomic molecules have less reliability than the noble gases because nonspherical characteristics of molecules are not completely taken into account in the derivation of the potential from the experimental scattering observations. In addition, for the dissociated gases H, N, and O, there are only a few molecular beam measurements, which are relatively difficult to obtain; for these mixtures uncertainties in the potentials range from about 30 to 60%. However, these rather large uncertainties in the potentials appear only as much smaller uncertainties in the calculated diffusion coefficients. This is clearly evident from Eq. (2.6-3) for the inverse power potential, since its collision integral is proportional to a fractional power of the potential parameters.

The valid temperature range for diffusion coefficients calculated for short-range interactions can be predicted as follows. The potentials derived from molecular beam scattering experiments are reported with an applicable internuclear separation range. These are obtained directly from the minimum and maximum values of the measured scattering cross sections.[46] In order to calculate the upper and lower limits of the temperature range, the minimum and maximum values of the separation range, respectively, are assumed to be approximately related to the collision integral as $\bar{\Omega}^{(1,1)} \approx \pi r^2$. Since the collision integral is also given in terms of the potential parameters and temperature by Eqs.(2.6-2) or (2.6-3), a temperature range can easily be computed. The accuracy of the predicted temperature limits has two significant figures at most.

c. Combination Rules

Often no direct determinations are available for the intermolecular potential of a particular gas pair, but the potentials for the individual species may be known. Various semi-empirical combination rules are available for the prediction of potential parameters for a 1-2 interaction from those for the 1-1 and 2-2 interactions. Such rules work well enough to allow the prediction of $\mathcal{D}_{12}$ to a level of uncertainty in the order of 10%.

The combination rules for the long-range and short-range interactions are as follows. For long-range interactions, theory indicates a geometric-mean rule for the London dispersion coefficient,

$$C_{12} = (C_{11}\ C_{22})^{1/2} \quad . \qquad (2.6\text{-}4)$$

This rule has been tested[52] and found to be quite accurate. Theory also suggests, but more weakly, a geometric-mean combination rule for the short-range interactions;[53]

<u>Exponential Potential</u>

$$(\varphi_o)_{12} = [(\varphi_o)_{11}(\varphi_o)_{22}]^{1/2} \quad , \qquad (2.6\text{-}5a)$$

$$\rho_{12}^{-1} = \frac{1}{2}(\rho_{11}^{-1} + \rho_{22}^{-1}) \quad , \qquad (2.6\text{-}5b)$$

and

<u>Inverse-Power Potential</u>

$$K_{12} = (K_{11}\ K_{22})^{1/2} \quad , \qquad (2.6\text{-}62)$$

$$s_{12} = \frac{1}{2}(s_{11} + s_{22}) \quad . \qquad (2.6\text{-}6b)$$

These rules have been directly tested by means of the molecular beam scattering experiments, and the results are quite satisfactory.[54-56]

2.7 Determination of Diffusion Coefficients from Other Transport Property Measurements

In this section procedures are described for the determination of

$\mathcal{D}_{12}$ from other transport property measurements according to results of the Chapman-Enskog theory.[1,2] These procedures are virtually independent of knowledge of the molecular interactions, and are an alternate route to the reliable prediction of $\mathcal{D}_{12}$.

a. Viscosity

The Chapman-Enskog first approximation for the viscosity of a binary mixture can be expressed[57] as a quadratic equation in the diffusion coefficient:

$$(p\mathcal{D}_{12})^2 a + (p\mathcal{D}_{12}) b + (p\mathcal{D}_{12}) c\, A^*_{12} + A^*_{12} d = 0, \qquad (2.7\text{-}1)$$

in which

$$a = (x_1 x_2)^2 (\eta_{mix} - \eta_1 - \eta_2)/\eta_1 \eta_2 \quad , \qquad (2.7\text{-}2)$$

$$b = 2x_1 x_2 (M_1 + M_2)^{-1} RT[\eta_{mix}(x_1{}^2 \eta_2 + x_1{}^2 \eta_1) - \eta_1 \eta_2]/\eta_1 \eta_2 \quad , \qquad (2.7\text{-}3)$$

$$\begin{aligned} c = \tfrac{6}{5} x_1 x_2 (M_1 + M_2)^{-1} RT[\eta_{mix}(x_1{}^2 M^2 \eta_2 + x_2{}^2 M_2{}^2 \eta_1) \\ - (x_1 M_1 - x_2 M_2)^2 \eta_1 \eta_2]/M_1 M_2 \eta_1 \eta_2 \quad , \end{aligned} \qquad (2.7\text{-}4)$$

$$d = \tfrac{3}{5}(2x_1 x_2 RT)^2 \eta_{mix}/M_1 M_2 \quad , \qquad (2.7\text{-}5)$$

where R is the gas constant (82.0567 cm^3-atm/mole °K), η is the viscosity in g/cm-sec, η_{mix} denotes the mixture viscosity, and the subscripts have their usual meaning. The determination of $\mathcal{D}_{12}$ requires experimental data for mixture composition, the molecular weights and viscosities of the pure components, and the mixture viscosity of the gas pair. The only non-experimental quantity required is the collision integral ratio A^*_{12}. The variation of A^*_{12} with temperature is only a few percent in the intermediate temperature region, is relatively independent of the choice of a realistic intermolecular potential model, and is insensitive to inelastic collisions (Section 2.2, part c) and quantum effects (Section 2.5). Thus the determination of $\mathcal{D}_{12}$ from viscosity measurements essentially eliminates the need for accurate information about molecular interactions.

For a mixture of a gas with itself the binary mixture expression, Eq. (2.7-1), reduces to

$$p\mathcal{D}_{11} = \frac{6}{5} A^*_{11} (RT/M_1)\eta_1 \quad , \tag{2.7-6}$$

in which $\mathcal{D}_{11}$ is known as the self-diffusion coefficient.

The determination of $\mathcal{D}_{12}$ from viscosity measurements has been derived from first approximation formulas. On this basis the diffusion coefficients calculated cannot be the true values of $\mathcal{D}_{12}$, which have a small composition dependence. The diffusion coefficients calculated cannot be exact $[\mathcal{D}_{12}]_1$ because experimental viscosity data are used.[58] However, the diffusion coefficients calculated from experimental binary mixture viscosity data are nearly equal to $\mathcal{D}_{12}$ at a mixture composition corresponding to the heavy component in trace amounts, as shown by numerical computations of the higher Chapman-Enskog approximations.[10] The uncertainty in this conclusion was found to be less than any error in available diffusion coefficient measurements.

The reliability of $\mathcal{D}_{12}$ calculated from mixture viscosity measurements is almost the same as obtainable by $\mathcal{D}_{12}$ measurements with the best modern techniques, as shown by the following analysis. First, assume that A^*_{12} is known exactly. On the basis of an error propagation analysis of Eq. (2.7-1), the calculation procedure for $\mathcal{D}_{12}$ can introduce a loss in accuracy by as much as a factor of five.[57] However, reliable viscosity measurements are obtained with inaccuracies of 1/10% at about room temperature and about 1/2% at 1000°K. These uncertainties are approximately 10 times less than in direct $\mathcal{D}_{12}$ measurements at the corresponding temperatures. Second, remove the restriction of a perfectly known A^*_{12} in order to obtain the total uncertainty of calculated $\mathcal{D}_{12}$. For spherical or homonuclear diatomic molecules at intermediate temperatures A^*_{12} is reliable to about 1%; non-spherical or polar gases have slightly larger uncertainties in A^*_{12}. Uncertainties in values of A^*_{12} will be directly reflected in $_{12}$, that is, a 1% error in A^*_{12} corresponds to an error of approximately 1% in $\mathcal{D}_{12}$. Thus the total uncertainty in diffusion coefficients calculated from accurate viscosity measurements is about 2% at room temperature, an uncertainty comparable to the available direct $\mathcal{D}_{12}$ measurements.

b. Thermal Conductivity

The first approximation of the Chapman-Enskog theory for the thermal conductivity of binary mixtures can be used to compute values of $\mathcal{D}_{12}$.[59] The procedure is similar to that used for diffusion coefficients calculated from viscosity data, but the values calculated from thermal conductivity measurements are not as reliable as available $\mathcal{D}_{12}$ measurements for two reasons. First, the relationship between thermal conductivity and $\mathcal{D}_{12}$ is slightly more sensitive to temperature and molecular interactions; that is, the applicable relationship has the collision integral ratio B^*_{12}, as well as A^*_{12}. Second, the accuracy of thermal conductivity data is only equal to, and often less than, that of $\mathcal{D}_{12}$ measurements, and the experimental errors propagate by a factor of as much as five through these calculations. Thus thermal conductivity is a transport property from which only mediocre estimates of $\mathcal{D}_{12}$ are possible at present.

c. Thermal Diffusion Factor

The Chapman-Enskog theoretical first approximation for the thermal diffusion factor of binary mixtures may give reliable values of $\mathcal{D}_{12}$. The thermal diffusion factor describes how a gas mixture separates under the influence of a temperature gradient. Diffusion coefficients can be calculated from the strong composition dependence of the thermal diffusion factor, α_T.[60] But, the available measurements of the composition dependence of α_T have rather large uncertainties, which lead to mediocre values of $\mathcal{D}_{12}$ at present. Another procedure relates the temperature dependence of $\mathcal{D}_{12}$ to that of α_T, and the derived relationship is combined with a single measurement of $\mathcal{D}_{12}$ to produce diffusion coefficients over a wide temperature range.[61] From this procedure the accuracy of $\mathcal{D}_{12}$ is good, because uncertainties in the measurements appear only as much smaller uncertainties in the calculated diffusion coefficients. In principle the calculations are applicable generally, but have been limited to gas pairs with $M_2/M_1 << 1$ and a trace concentration of the heavy component. The procedure has involved iterative type calculations which are described next.

An "experimental" value of $(6\ C^*_{12}-5)$ is compared to the auxiliary theoretical expression

$$(6C^*_{12}-5) = 2[2-(\partial\ \ell n[\mathcal{D}_{12}]_1/\partial\ \ell n\ T)_p] \quad , \qquad (2.7\text{-}7)$$

in which the "experimental" $(6\ C^*_{12}-5)$ is derived as follows:

$$(6\ C^*_{12}-5) = \alpha_T[(1+\kappa_2)(-S_2/Q_2)]^{-1} \quad , \qquad (2.7\text{-}8)$$

$$-S_2 = \frac{15}{2}\,\frac{M_1(M_1-M_2)}{(M_1+M_2)^2} + 4\,\frac{M_1M_2}{(M_1+M_2)^2}\,A^*_{12} - \frac{5}{3}\,\frac{M_2{}^2}{(M_1+M_2)}\,\frac{p[\mathcal{D}_{12}]_1}{[\eta_2]_1RT} \quad ,$$

$$(2.7\text{-}9)$$

$$Q_2 = \frac{10}{3} \frac{M_2}{(M_1+M_2)^2} \frac{p[\mathscr{D}_{12}]_1}{[\eta_2]_1 RT} (3M_1^{\,2} + M_2^{\,2} + \frac{8}{5} M_1 M_2 A^*_{12}) \quad , \tag{2.7-10}$$

$$\kappa_2 = \frac{1}{42} (8E^*_{22}-7)^2 + \frac{2}{21} (1 - \frac{M_2}{M_1}) (8E^*_{22}-7) [1 - \frac{3}{4}(5-4B^*_{12})(6C^*_{12}-5)^{-1}] \quad , \tag{2.7-11}$$

$$8E^*_{22}-7 \approx 2[1 - (\partial \ln \eta_2/\partial \ln T)_p]. \tag{2.7-12}$$

In these equations the subscript 2 denotes the light component and 1 the heavy, κ_2 is a small correction term, and for Q_2 the Kihara expression is used in this case. As previously discussed, A^*_{12} is virtually independent of temperature and the potential model. The values of $[\mathscr{D}_{12}]_1$ and $[\eta_2]_1$ can be interpreted as "experimental first approximations". In Eq. (2.7-8) the denominator is weakly dependent on temperature, but the major temperature dependence is in α_T, and this is obtained from experiments. The substitution of Eqs. (2.7-9) to (2.7-12) into the right-hand side of Eq. (2.7-8) gives the "experimental" (6 C^*_{12}-5) value principally in terms of the temperature dependence of α_T. For the first iteration step ($-S_2/Q_2$) and κ_2 are assumed independent of temperature, the temperature at which they are evaluated is conveniently taken to be the same as for the experimental $\mathscr{D}_{12}$. This value of (6 C^*_{12}-5) is substituted into the differential equation Eq. (2.7-7), and the subsequent integration completes the first iteration cycle. The constant of integration is evaluated from one isothermal measurement of $\mathscr{D}_{12}$. The result is a relationship for the temperature dependence of $\mathscr{D}_{12}$ over the range for which measurements of α_T are available. The second iteration step uses values of $[\mathscr{D}_{12}]_1$ from the first cycle together with experimental values of $[\eta_2]_1$ to evaluate the temperature variation of ($-S_2/Q_2$); κ_2 can be assumed independent of temperature. The second set of ($-S_2/Q_2$) gives new values for (6 C^*_{12}-5), and new values of $[\mathscr{D}_{12}]_1$ by the integration of Eq. (2.7-7). The $[\mathscr{D}_{12}]_1$ of the second set are usually almost identical with the first set, but a third iteration step can be used as a check, if desired. The diffusion coefficients calculated are as reliable as most direct measurements of $\mathscr{D}_{12}$; at present this means about a few percent.

In some cases the calculation procedure can be simplified, and made to involve the thermal-diffusion factor in a more direct way, but this method was not used for any results in this report.

The determination of $\mathscr{D}_{12}$ from thermal diffusion data is, strictly speaking, limited to noble gas pairs. The equations are based on monatomic molecules which are free of internal energy. The theoretical expressions can be used for polyatomic gases when the translational energy contribution is much greater than that of internal energy factors which contribute to α_T.

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3. EXPERIMENTAL METHODS FOR DIFFUSION COEFFICIENTS

The purpose of this chapter is to assess reliabilities and limitations of the various experimental methods used for determinations of gaseous diffusion coefficients. The critical evaluation of $\mathcal{D}_{12}$ necessarily requires a comprehensive appraisal of experimental methods, which has not been prepared before. The various methods are outlined in Section 3.1 in order to give an overall perspective of the types of apparatus and the reliabilities of results. In Section 3.2 five major methods are described and their systematic errors considered. A major method means one that has been frequently used by different investigators and has well-known experimental uncertainties. The results of four of these five methods have generally contributed to Standard Reference Data, but one technique - the evaporation-tube - has had disappointing results and is included as a major method only because it has been used more often than any other. In Section 3.3 brief descriptions are given for six methods which have not been used very often, but which have sufficient results available so that their reliability may be estimated. These are called minor methods and the results have made small contributions to Standard Reference Data. The final section of this chapter contains remarks about seven miscellaneous methods which have not contributed to Standard Reference Data, but which are of general applicability or of unusual inventiveness. Every experimental method ever used is not included in these groups, but those omitted are considered unimportant.

The discussion of each method includes a comprehensive list of references to specific studies; these listings contain the reference information for the discussion of each method unless special footnotes are given in the text.

Several of the experimental methods have been previously described in specialized surveys.[1-6]

The first significant measurements of diffusion in gases were made by Thomas Graham, starting in 1829. His ingenious experimentation included observations of gaseous diffusion in closed-tube and two-bulb apparatuses.[7,8] These techniques were later developed into the most reliable methods, by modern standards, for the determination of diffusion coefficients. In addition Graham used what is now called the capillary-leak method and an equivalent to the diffusion bridge. However, Graham never calculated a diffusion coefficient, and actually most of his work preceded the mathematical statement of the law of diffusion by Fick in 1855.[9] From some of Graham's later observations, reported in 1863, the first accurate $\mathcal{D}_{12}$ were calculated by Maxwell in 1867.[10,11] Until recent times most of Graham's work in diffusion had been overlooked.[11]

In the 1870's two experimental methods, the closed tube and the evaporation tube, were developed; their results include almost all values of $\mathcal{D}_{12}$ up until World War II. Then several other methods (two-bulb apparatus, point source, diffusion bridge, dissociated gases, and gas chromatography) were developed because of interests in isotope separations, combustion processes, and theoretical studies of intermolecular forces which were in need of values of $\mathcal{D}_{12}$ over an extensive range of temperatures. The availability of radioisotopes made measurements for many gas pairs easier. In addition a number of other techniques have been occasionally used over the last half century. The experimental methods are classified in Table 2, and the reference sources can be found in Bibliography I.

3.1 Outline of Experimental Methods

Table 2 serves as an outline of the assessment of methods that follows. The reliabilities given are based on reproducibilities and on intercomparisons of $\mathcal{D}_{12}$ by various methods. At present the reliability[12] is not exactly known for each method; these measurement techniques are amenable to possible refinements. The determinations of $\mathcal{D}_{12}$ are considered good when uncertainties are within about 2%, although for a given apparatus the reproducibility of results may be better than 1%. A vast majority of available data does not have this level of either reproducibility or reliability. Determinations of $\mathcal{D}_{12}$ are considered of average quality when uncertainties are within about 5%. These magnitudes indicate that accurate determinations of diffusion coefficients are rather difficult, even with the best of modern instrumentation.

The major and a few minor methods are schematically illustrated in Fig. 2, classified according to overall geometry of apparatus and time behavior of the diffusion process. The apparatuses listed under the first two columns have no carrier gas flow in the zone where diffusion takes place. The two apparatuses in the third column have diffusion occurring within a flowing gas stream.

3.2 Major Experimental Methods

a. Closed Tube

In 1870 the closed-tube method was developed by Loschmidt, who carefully determined $\mathcal{D}_{12}$ for 10 gas pairs at temperatures of 252° to

293°K. The essential characteristic of this method is a variation of mixture composition with time and position throughout a long tube closed at both ends. The gases of the mixture are initially separate in the closed tube, then interdiffuse at constant temperature and pressure. The diffusion time is controlled by an opening mechanism at the middle of the tube. The composition changes are measured as a function of time, either continuously or after a definite period of diffusion.

Determinations of $\mathcal{D}_{12}$ by the closed-tube method are usually quite reliable. The results have been obtained at temperatures from 195° to 478°K. This range indicates an indirect disadvantage - determinations at more extreme temperatures have not been made because of difficulties that arise from the construction and the operation of a thermostat around a long tube (about 1 meter) with moving parts.

The reported determinations are listed in Table 3 in chronological order. There are various versions of the closed-tube apparatus, but details of these refinements are omitted here.

The basis of all closed-tube determinations is a solution of the one-dimensional time-dependent diffusion equation,

$$\partial x_1/\partial t = \mathcal{D}_{12} \, (\partial^2 x_1/\partial z^2), \qquad (3.2\text{-}1)$$

where $\mathcal{D}_{12}$ has been assumed independent of mixture composition and position. In Eq. (3.2-1) x_1 is the mole fraction of component 1 in the binary mixture, and t and z are time and axial distance, respectively. For the initial and boundary conditions,

$$x_1 = x_1^{\ell} \qquad 0 \le z < L/2, \; t = 0$$

$$x_1 = x_1^{u} \qquad L/2 < z < L, \; t = 0$$

$$\partial x_1/\partial z = 0 \qquad z = 0 \text{ and } z = L, \; t \ge 0,$$

the solution of Eq. (3.2-1) is

$$x_1(z,t) = \frac{1}{2}\ (x_1{}^{\ell} + x_1{}^{u}) + \frac{2(x_1{}^{\ell} - x_1{}^{u})}{\pi} \sum_{n=0}^{\infty} \frac{e^{-(2n+1)^2 t/\tau}}{(2n+1)} \sin \frac{(2n+1)\pi z}{L} , \tag{3.2-2}$$

and the relaxation time is

$$\tau = L^2/\pi^2 \mathcal{D}_{12} , \tag{3.2-3}$$

where L is the total length of the closed tube. A few additional assumptions were taken to obtain Eq. (3.2-2), namely a uniform cross section and symmetry about the midplane at L/2. Equation (3.2-2) may be simplified in accordance with the technique used for composition analysis; the simplified expressions are readily available elsewhere,[3,6,13,14] as well as from the original articles of Table 3.

The determination of $\mathcal{D}_{12}$ requires measurements of composition, temperature, pressure, geometrical factors, and time. The uncertainties of these measurements are usually much less than the reliability of the results; thus the reliability is apparently dependent on other factors. Occasionally, however, poor methods of composition analyses have led to inaccurate results.

Other possible uncertainties of the closed-tube method are as follows. Errors due to convective mass flux are possible. To avoid convection from buoyancy effects, the lighter gas should always be placed in the top half of vertically mounted apparatus. If the closed tube is in a horizontal orientation, a "spillage" convective flux may occur; that is, the higher density component in one half may spill across the diffusion "interface" into the lower portion of the other half, and the low density component would then flow into the upper portion of the opposite half of the closed tube. Spillage is not significant if the diffusing component is a tracer. At the start of diffusion, convection effects are also possible because of the movements of the opening mechanism; this has been investigated,[15-17] and the reproducibility of results from run to run indicates that the effect is small. Convection effects are also possible because of nonuniform temperatures axially along the tube. The design and operation of closed-tube apparatus should eliminate all possible convection effects.

A significant uncertainty, even though not limited to the closed-tube method, is the small dependence of $\mathcal{D}_{12}$ on the mixture composition. The diffusion coefficient was assumed independent of mixture composition. Equation (3.2-1) rewritten to express the composition dependence of $\mathcal{D}_{12}$ is

$$\partial x_1/\partial t = \mathcal{D}_{12}(\partial^2 x_1/\partial z^2) + (\partial x_1/\partial z)^2(\partial \mathcal{D}_{12}/\partial x_1). \qquad (3.2\text{-}4)$$

If one of the components is a tracer then the composition gradient, $\partial x_1/\partial z$, is very small, and the uncertainty essentially zero. If two pure gases fill each half of the closed tube, then both the gradient or the composition dependence of $\mathcal{D}_{12}$ may be significant. However, for the case of the closed tube, the exact integration of the diffusion equation with a composition-dependent $\mathcal{D}_{12}$ has not been performed. The uncertainty of results caused by the composition dependence of $\mathcal{D}_{12}$ depends on the duration of the experimental run, the gas pair investigated, and the initial composition of the mixture. Calculated values of $\mathcal{D}_{12}$ would apparently depend on the length of the experimental run. This has been investigated,[15,18] and the variation of $\mathcal{D}_{12}$ is small for diffusion times between 10 minutes and about 2 hours. To a first approximation the composition of the mixture may be taken equal to a uniform mixture of the components, or the arithmetic mean of the initial conditions.[17]

The uncertainty in determinations of $\mathcal{D}_{12}$ caused by the assumption of a one-dimensional diffusion equation has not been estimated.

Additional uncertainties may be caused by the Dufour effect. The Dufour effect is a small temperature transient that occurs when two gases interdiffuse, and may occur even with ideal gases. The uncertainty in $\mathcal{D}_{12}$ caused by the Dufour effect can be made small by suitable choice of apparatus geometry,[17] but most experimenters have apparently simply ignored the problem. If the mixture were nonideal then diffusion would be accompanied by heats of mixing or pressure changes.

The closed-tube determinations of $\mathcal{D}_{12}$ often have reproducibilities better than 1%, and the measurements have been reported accurate to 1 to 3%. However, independent determinations of $\mathcal{D}_{12}$ for the same gas pair indicate that this method has a reliability no better than 2%. These comparisons are presented in deviation plots given in Section 5.3. To achieve greater reliability a major effort involving careful variation of many experimental parameters would probably be necessary.

b. Two-Bulb Apparatus

The two-bulb method was developed by Ney and Armistead in order to determine the self-diffusion coefficient of UF_6; their results were published in 1947. Two bulbs, or chambers, are connected by a narrow tube through which the diffusion occurs. After an initial transient, the composition in the bulbs varies exponentially with time, and $\mathcal{D}_{12}$ can be found from the relaxation time.

The determinations of $\mathcal{D}_{12}$ by the two-bulb method have been made over a temperature range of 65° to 400°K, with one datum available at 473°K. This range of temperatures is ∿50°K larger than results obtained by the closed-tube method. In general, measurements at different temperatures are easier to make with the two-bulb apparatus because its relative compactness facilitates thermostating, and its opening mechanism can be designed without moving parts. These conveniences, however, are only operational advantages; the ultimate accuracies of $\mathcal{D}_{12}$ are probably the same as determined by both the closed-tube and two-bulb methods.

Studies by two-bulb apparatus are listed in chronological order in Table 4. This listing shows that the method has been widely used in recent years. The meticulous studies by van Heijningen _et_ _al_. are especially noteworthy because the results attained are probably the most reliable measurements of $\mathcal{D}_{12}$ to date, within 1%, over a temperature range of 65° to 400°K.

As for the closed tube, the simple theory for the two-bulb apparatus involves the assumptions of constant pressure and temperature, constant $\mathcal{D}_{12}$, and one-dimensional diffusion. It also involves the following additional assumptions:

(1) Quasi-stationary state - the flux of a component is constant along the connecting tube. Since $\mathcal{D}_{12}$ is assumed constant, this implies a linear variation in composition in the tube.

(2) The connecting tube volume is much smaller than either bulb volume. This is related to the quasi-stationary-state assumption.

(3) The composition gradient is entirely contained in the connecting tube.

With all these assumptions introduced into the diffusion equation for one component, Eq. (2.1-1), a simple solution can be obtained of the form

$$\Delta x(t) = \Delta x(0) \exp(-t/\tau), \qquad (3.2\text{-}5)$$

where $\Delta x(t)$ is the composition difference at $t = \infty$ and at time t in one bulb, $\Delta x(0)$ is the composition difference between $t = \infty$ and $t = 0$, and τ is the relaxation time. The component subscript 1 has been dropped from the notation. The relaxation time is

$$\tau = \frac{1}{\mathscr{D}_{12}} \left(\frac{L}{A}\right) \left(\frac{V_1 V_2}{V_1+V_2}\right) \quad , \qquad (3.2\text{-}6)$$

where A is the cross-sectional area, L the length of the tube connecting the bulbs, and V_1 and V_2 denote the bulb volumes. From measurements as a function of time of the composition in one bulb, or alternately the composition difference between the two bulbs, the relaxation time is obtained from Eq. (3.2-5). Corrections to the relaxation time for the above assumptions are as follows.

The quasi-stationary state assumption is unnecessarily severe;[19] it is sufficient to assume only that the mean flux in the tube is proportional to the effective mean flux at the two ends of the tubes. On this basis a correction factor K for the relaxation time may be derived,

$$\tau = \frac{K}{\mathscr{D}_{12}} \left(\frac{L}{A}\right) \left(\frac{V_1 V_2}{V_1+V_2}\right) \quad , \qquad (3.2\text{-}7)$$

where

$$K = 1 + \frac{AL}{3V_1} \left(\frac{1-\beta + \beta^2}{1 + \beta}\right) \quad , \qquad (3.2\text{-}8)$$

with $\beta = V_1/V_2$. This approximate solution assumes that the composition analysis is performed in bulb V_1 and that AL/V_1 is small. Deviations from a quasi-stationary state show up as values of K unequal to unity. The deviations from the quasi-stationary state are due to the fact that the bulbs are not infinitely large compared to the connecting tube. To minimize these deviations the apparatus should be constructed such that the volume of the tube is very much less than the volume of either bulb. For bulbs of equal size $K = 1 + AL/6V$, where V is the volume of a bulb.

Transient effects arise from the finite time required to establish a constant gradient across the entire length of the tube.[20] These transients can be avoided by waiting some time to elapse after the start of mixing. Normally, the transients decay rapidly, and they completely disappear within a few minutes.

The assumption that the composition gradient is all in the connecting tube requires an end correction. This correction is required because the gradient does not truncate immediately at either outlet of the tube. The extension of the gradient into the bulbs is corrected for by a small increase in the actual length of the tube. The end correction is given by

$$L_{eff} = L + 2\alpha R \quad , \qquad (3.2\text{-}9)$$

where L_{eff} is the effective length of the tube, R is its radius, α is a numerical constant whose value depends on the geometrical configuration of the end of the tube, and the factor 2 accounts for both ends of the tube. The value of α is obtained from an analogous case for sound passage in a tube.[21,22] Typical values of α are as follows: $\alpha = 0.58$ when the connecting tube end is in free space, $\alpha = 0.82$ when the end is flush with a flat surface, and $\alpha \gtrsim 0.82$ when the end is flush with the inside surface of a spherical bulb. The value of $\alpha = 0.82$ has been invariably used in the analysis of two-bulb apparatus experiments, but this is not always correct. The choice of an incorrect α value has led, in a few studies, to systematic errors of the order of 1%.

Most two-bulb apparatus are constructed with the connecting tube of uniform bore; if not so, then the ratio L/A is taken to mean $\Sigma_j(L_j/A_j)$ for each element of length L_j and cross section A_j.[23]

In addition to the above corrections, observations may require connections for Knudsen flow,[24,25] which can occur during diffusion in narrow capillaries at low pressures, where the mean free path is not negligible compared to the diameter of the connecting tube.

The determinations of $\mathcal{D}_{12}$ by the two-bulb method have uncertainties similar to those of the closed tube, but with less chance of convective effects because of the narrowness of the connecting tube. In several two-bulb investigations, errors from non-negligible sample volumes are possible because samples of the mixture were removed from the apparatus during the diffusion run. The inaccuracies of two-bulb measurements have been reported to be between 1 and 3.5%. Except for results by van Heijningen _et_ _al_. the reliability of data by this method is considered no better than 2%. This is the same as for the closed tube. The two-bulb method is capable of yielding reliable $\mathcal{D}_{12}$ provided care is taken to optimize the geometry of the apparatus and to make corrections.

c. Point Source

The point-source method was developed especially for the determination of diffusion coefficients at high temperatures. In 1958 Walker and Westenberg fully reported the first results by this method, in which a trace amount of gas is steadily introduced through a fine hypodermic tube into a carrier gas flowing in the same direction. The tracer spreads by diffusion through the carrier gas, which has characteristics of steady-state laminar flow with a flat velocity profile. The mixture composition is measured by means of a sample probe located at various distances downstream of the tracer inlet.

Point source determinations of $\mathcal{D}_{12}$ are available from room temperature up to 1944 °K.

Studies by this method are listed in chronological order in Table 5. A few special remarks are as follows. Walker and Westenberg used electrical heat and attained temperatures up to ∿ 1200 °K; Ferron _et al._ used combustion heat (mixtures containing H_2O or CO_2 were studied) and reached higher temperatures, up to 1944 °K.

The basic equation for the point-source method is

$$\mathcal{D}_{12} \left[\frac{\partial^2 x}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\,\frac{\partial x}{\partial r}\right)\right] - U(r)\,\frac{\partial x}{\partial z} = 0 \quad , \qquad (3.2\text{-}10)$$

where x denotes the tracer component, U is the carrier velocity, z is the axial distance from the point of injection, and r is the radial coordinate measured from the axis. The appropriate boundary conditions are:

$$\lim_{R \to \infty} x = 0 \quad ,$$

$$(\partial x/\partial r)_{r=0} = 0 \quad , \quad \text{and}$$

the normalization condition

$$Q = \lim_{R \to 0} 4\pi R^2 \mathcal{D}_{12} (\partial x/\partial R) \quad ,$$

where $R^2 = r^2 + z^2$ and Q is the volumetric flow rate of the tracer. Equation (3.2-10) involves the usual assumptions of constant temperature and pressure, and the composition-independence of $\mathcal{D}_{12}$. Additional assumptions are as follows:

(1) Steady-state conditions of flow.

(2) Axial symmetry of the tracer concentration in the carrier gas.

(3) One-dimensional flow.

(4) Flat velocity profiles across a cross section normal to the direction of flow, i.e., U(r) = constant.

(5) Equal velocities for the carrier and tracer gases.

(6) Absence of convection effects.

(7) Absence of chemical reactions. This assumption is particularly

noted, even though applicable to all major methods, because point-source measurements at high temperatures may be accompanied by reactions between the components or the components and surfaces of the diffusion apparatus.

The solution of Eq. (3.2-10) is

$$x = (Q/4\pi R\mathcal{D}_{12})\ \exp\ [-(R-z)U/2\mathcal{D}_{12}]. \qquad (3.2\text{-}11)$$

A concentration profile can be used to determine $\mathcal{D}_{12}$, that is, at a fixed axial distance z samples of the stream are taken as a function of R. A plot of $\ell n(xR)$ versus $(R-z)$ should be linear with slope $-U/2\mathcal{D}_{12}$, and measurements of the carrier velocity U lead to values of $\mathcal{D}_{12}$. Alternately, the stream can be sampled at points on the axis, $z = R$, and values of $\mathcal{D}_{12}$ calculated from

$$\mathcal{D}_{12} = Q/4\pi(x)_{\max} z \quad , \qquad (3.2\text{-}12)$$

where $(x)_{\max}$ is the concentration of the tracer at points on the z axis. This axial decay method has the advantage of not requiring knowledge of U, and of requiring significantly fewer composition measurements. The more difficult concentration profile method can serve as a check on the consistency of both the theory of the experiment and the results.

The instrumentation errors of the point-source method have been estimated by Ferron <u>et al</u>. to be about 5%. This estimate was based on an approximate analysis of errors arising from measurements of flow, sample probe position, composition, and temperature. These experiments are probably not as reproducible as those by Walker and Westenberg, who performed experiments at lower temperatures and with a more precise technique for composition analysis.

In addition to the instrumentation errors, the possible causes of uncertainty for the point-source method are:

(1) Unequal flow rates between the tracer and the carrier gases.

(2) Difference in density between the tracer and the carrier.

(3) Variations in the steady-state flow rate of the tracer, or carrier.

(4) Skewness of the mixture velocity profile.

(5) Temperature gradients in the stream.

A priori estimates of uncertainties caused by such effects are difficult to make; but these effects have been empirically investigated.[26,27] The reliability of point-source measurements of $\mathcal{D}_{12}$ is best estimated by comparisons with the results by other methods. Such comparisons show deviations of up to 4% for 10 gas pairs at about 300 °K. The deviations are slightly greater at 1000 °K by comparison of point-source results and those calculated from short-range interaction forces obtained by molecular-beam scattering experiments (Section 2.6, part b). The general reliability of $\mathcal{D}_{12}$ by the point-source method is considered to be better than 5%, or average.

d. Gas Chromatography

The gas-chromatography method is a flow method in which a trace amount of gas is injected as a pulse into a carrier gas flowing through a long hollow tube. The dispersion of the pulse is caused by the combined action of molecular diffusion and the parabolic velocity profile of the carrier gas. As the pulse emerges from the tube outlet, measurements of the dispersion - characterized by a Gaussian distribution function - lead to values of $\mathcal{D}_{12}$.

The advantages of the gas-chromatography method are as follows. Determinations of $\mathcal{D}_{12}$ can be completed in a matter of minutes and vapor-gas mixtures can be studied. Once the carrier gas is at temperature and pressure, the injection of a number of sample pulses into the gas is possible, with the result that several samples may be simultaneously dispersing in the tube. The dispersion characteristics of the pulse can be obtained by one simple measurement of its variance. A vapor-gas determination of $\mathcal{D}_{12}$ is practical because of the small amount of sample required to make a pulse. These advantages are operational,only.

Determinations of $\mathcal{D}_{12}$ by gas chromatography are available between temperatures of 77° and 523°K. These studies are listed in chronological order in Table 6, all of which are based on the instrumentation and certain aspects of the convential theory of gas chromatography. In 1960 four independent manuscripts, which described the method, were submitted for publication: Bohemen and Purnell (23 June); Fejes and Czárán (20 July); Giddings and Seager (3 August), and Bournia, Coull, and Houghton (8 November). Of these authors, only Giddings et al. have continued to publish new determinations of $\mathcal{D}_{12}$.

Packed chromatography columns have been used, on occasion, to determine $\mathcal{D}_{12}$. A packed column has a complex geometry because of the interstitial flow volume. Since the geometry and the pulse velocity profile are not well defined in packed columns, their use for determinations of absolute values of $\mathcal{D}_{12}$ is rather uncertain.[28]

Outside the scope of this report, but worthy of mention, is the following. Gas chromatography is well suited for high-pressure determinations of $\mathcal{D}_{12}$ because the column actually consists of small-bore tubing which is easily pressurized. High-pressure studies in other apparatuses ordinarily require the fabrication of relatively expensive pressure vessels.

Before gas-chromatography apparatus was applied to the determinations of $\mathscr{D}_{12}$, the theory had been developed for diffusion phenomena in the flow of fluids.[29-34] The basic equation for the gas-chromatography method is

$$\mathscr{D}_{12}\left[\frac{\partial^2 x}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial x}{\partial r}\right)\right] - U(r)\,\frac{\partial x}{\partial z} = \frac{\partial x}{\partial t} \quad , \qquad (3.2\text{-}13)$$

where x is the mole fraction of the pulse component, U is the velocity of the carrier gas, z is the axial distance, r is the radial coordinate of the tube, R_o is the constant radius of the tube, and t denotes time. The boundary conditions are:

$$(\partial x/\partial r)_{r\,=\,0} = 0 \quad , \text{ and}$$

$$(\partial x/\partial r)_{r\,=\,R_o} = 0 \quad .$$

The initial condition depends on the pulse shape at the injection point. As for the point-source method, gas-chromatography involves the assumptions of constant pressure and temperature, constant $\mathscr{D}_{12}$, one-dimensional flow, and axial symmetry. Additional assumptions are as follows:

(1) The carrier flow velocity is laminar with a parabolic profile; i.e., $U(r) = 2\bar{U}[1-(r/R_o)^2]$, where $\bar{U}$ is the average velocity.

(2) Convection effects are absent.

(3) The initial pulse of sample may be well approximated by a delta-function.

Subject to these conditions and assumptions, the solution of Eq. (3.2-13) is given by

$$\bar{x} = \frac{N}{2\pi R_o^2 n}\,(\pi\mathscr{D}_{eff}\,t)^{-\frac{1}{2}} \exp\left[\frac{-(z-\bar{U}t)^2}{4\mathscr{D}_{eff}\,t}\right] \quad , \qquad (3.2\text{-}14)$$

where $\bar{x}$ denotes the mean mole fraction of sample in a cross section, n is the total number density, N is the number of molecules of the pulse injected into the carrier gas at $z = 0$, $t = 0$, and $\mathcal{D}_{eff}$ denotes the effective diffusion coefficient,

$$\mathcal{D}_{eff} = \mathcal{D}_{12} + R_o^{\,2}\,\bar{U}^2 / 48\,\mathcal{D}_{12} \quad . \qquad (3.2\text{-}15)$$

The first term on the right-hand side of Eq. (3.2-15) accounts for the dispersion of the pulse caused by diffusion in the axial direction. The second term is known as the Taylor diffusion coefficient, and accounts for the dispersion of the pulse caused by the parabolic velocity profile, as modified by diffusion in the radial direction.

The experimental procedure is to observe the concentration $\bar{x}$ as a function of time at the end of the tube ($z = L$); Eq. (3.2-14) for this case is

$$\bar{x}(t) = \frac{N}{2nV}\left[\pi\left(\frac{\mathcal{D}_{eff}}{\bar{U}L}\right)\left(\frac{\bar{U}t}{L}\right)\right]^{-\frac{1}{2}} \exp\left[\frac{-(1-\bar{U}t/L)^2}{4\left(\frac{\mathcal{D}_{eff}}{\bar{U}L}\right)\left(\frac{\bar{U}t}{L}\right)}\right] \quad , \qquad (3.2\text{-}16)$$

where $V = \pi R_o^{\,2} L$ is the tube volume. This is a skewed, not Gaussian, distribution, but if $\mathcal{D}_{eff}/\bar{U}L \leq 0.01$, the distribution becomes nearly Gaussian.[34] The reason is that $\bar{U}t/L$ must be nearly unity when $\mathcal{D}_{eff}/\bar{U}L$ is small, or else $\bar{x}(t)$ becomes too small to measure accurately. In the approximation that $\bar{U}t/L \sim 1$, Eq. (3.2-16) becomes a Gaussian, with variance τ given by

$$\tau^2 = \frac{2\mathcal{D}_{eff}}{\bar{U}L} = \frac{2\mathcal{D}_{12}}{\bar{U}L} + \frac{R_o^{\,2}\bar{U}}{24\mathcal{D}_{12}} \quad . \qquad (3.2\text{-}17)$$

A simple way to determine the variance is to measure the peak width at half height, $w_{1/2}$, related to τ as

$$w_{1/2} = 2(2\ell n 2)^{1/2}\,\tau \quad . \qquad (3.2\text{-}18)$$

The calculation of $\mathcal{D}_{12}$ from a measured value of $w_{1/2}$ requires solution of a quadratic equation, which has two roots; one root corresponds to the physical value of $\mathcal{D}_{12}$ when $\bar{U} \leq (48)^{1/2}\mathcal{D}_{12}/R_o$, and the other root when $\bar{U} > (48)^{1/2}\mathcal{D}_{12}/R_o$. An experimental check is that calculated values of the physical $\mathcal{D}_{12}$ must be independent of $\bar{U}$.

In addition to the normal instrumentation errors for flow, temperature, <u>etc</u>. the gas-chromatography method has an appreciable error contribution from the measurement of the peak width at half height of the dispersion profile. Values of $w_{1/2}$ have been obtained with precisions of about 1%, but in terms of $\mathcal{D}_{12}$ this level of precision is degraded because of the quadratic relationship between $w_{1/2}$ and $\mathcal{D}_{12}$.

Possible uncertainties of gas chromatography are as follows:

(1) Entrance effects caused by the injection of a finite volume of sample into the carrier gas. It is in principle impossible to inject a delta function of sample into the carrier, though in practice the time of injection may be quite short and the sample volume small. Entrance effects can be accounted for by a short correction tube of precisely the same diameter as the regular long-tube.[35] The use of both a long-tube and a short-tube also corrects for effects of stagnant volumes associated with injection and detection devices, and connections along the tube. Instead of two columns, two detectors in one column may be used to eliminate entrance effects.[36,37] In other studies ingenious sample injection devices have been used. The sample volume should be less than about 1% of the tube volume.

(2) Nonsymmetrical dispersion characteristics caused by disturbances to the velocity profile. These disturbances may be due to variations in the carrier gas flow rate or to rough tube surfaces. Some tubes are made in coil form, and bending the tube tends to produce higher velocities on the inside radius

than the outside radius. The magnitude of this effect depends on the radius of curvature of the coil. At the detector the concentration curve may be skewed for values of $\mathcal{D}_{eff}/\bar{U}L \geq 0.01$.

(3) Small pressure drops caused by viscous flow in long tubes and by interferences to flow due to detector(s) immersed in the stream.

(4) End effects caused by the detection of the sample dispersion characteristics. Detector elements may disturb the concentration profile, may not measure point values of the concentration profile, but a finite amount of sample, and may not give a linear response to concentration.

The inaccuracies of determinations of $\mathcal{D}_{12}$ by gas chromatography have been reported to be about 1 to 2%. The reliability of these results is best estimated by comparisons with $\mathcal{D}_{12}$ by other methods. At a temperature of 300°K comparisons show deviations up to 4%, with an average deviation of about 2%. At temperatures up to 500°K the deviations are within 5%. Thus, results by gas chromatography are considered to have the same overall level of reliability as the point-source method, that is, uncertainties within 5%.

e. Evaporation Tube

In 1873 Stefan developed the evaporation-tube method, which is useful for determinations of $\mathcal{D}_{12}$ for vapor-gas mixtures. The method has been extensively used by other investigators, and until recently these studies have produced almost all the values of $\mathcal{D}_{12}$ for vapor-gas mixtures. The idea of the method is simple. The evaporation rate of a liquid which partially fills a tube is controlled by diffusion through the stagnant gas which fills the rest of the tube. The diffusion coefficient can be determined from observations of the (slow) loss of liquid from the tube at constant temperature and pressure.

In this method the liquid to be volatilized is placed at one end of a vertical tube, the other end of which is open. The tube is a cylinder of uniform cross section, and usually with the approximate dimensions of 5 to 10 mm in diameter and 10 to 20 cm in length. From the gas-liquid interface, vapor diffuses through the gas to the mouth of the tube. At the interface the mixture composition depends on the vapor pressure of the liquid. Across the tube outlet gas flows and carries the vapor away. The rate of liquid loss is observed over long periods of about half a day in order to determine values of $\mathcal{D}_{12}$.

The same procedure is applicable to the volatilization of a solid in place of a liquid in the evaporation tube.

The evaporation-tube method involves a simple experimental technique, but the studies are restricted to narrow intervals of temperature which are strongly dependent on the volatility of the substance to be tested. The evaporation-tube results for $\mathcal{D}_{12}$ are available for hundreds of different gas pairs. The studies are listed in chronological order in Table 7. Of these publications, which comprise more than 70 articles, about one-third have been published since 1960.

The simple theory for the evaporation-tube method involves the usual assumptions of constant pressure and temperature, constant $\mathcal{D}_{12}$, one-dimensional diffusion, axial symmetry, and the absence of convection effects. It also involves additional assumptions as follows:

(1) Quasi-steady-state conditions. This assumption means that the composition gradients between the liquid level and the tube outlet are constant. Since the rate of evaporation is slow the gas-vapor column in the tube changes little in height, and even though

there are steady losses of liquid due to evaporation the diffusion path can be approximated as constant. A constant liquid level could be maintained, for example, by adding liquid to the evaporation tube at a rate equal to losses due to evaporation. Under quasi-steady-state conditions the flux of vapor is constant.

(2) Gas insolubility. The gas does not dissolve into the liquid. It follows from these assumptions that the gas in the tube is stagnant (zero flux); that is, the net flux in the tube consists only of vapor. The fundamental diffusion equations, Eqs. (2.1-3) and (2.1-4), then become

$$J_1 = -n \mathcal{D}_{12}(\partial x_1/\partial z) + x_1(J_1+J_2) \quad , \qquad (3.2\text{-}19)$$

$$J_2 = 0 \quad , \qquad (3.2\text{-}20)$$

where subscript 1 denotes the vapor and subscript 2 the gas. The boundary conditions of the system are that the vapor concentrations are constant at the gas-liquid interface, $(x)_0$, and at the outlet of the tube $(x)_L$. In Eq. (3.2-19) the vapor velocity profile has been implicitly assumed to be flat, in accordance with the assumption of one-dimensional diffusion. The integration of Eq. (3.2-19) gives

$$J_1 = (n\mathcal{D}_{12}/L)\ln\left[\frac{1-(x)_0}{1-(x)_L}\right] \quad , \qquad (3.2\text{-}21)$$

where the axial distance is measured from the gas-liquid interface, $z = 0$, and at the tube outlet $z = L$. The experimental procedure does not require knowledge of the vapor composition as a function of distance, but only the net loss of vapor from the tube. Since the liquid level or the diffusion path length actually changes slowly, the flux of vapor can be related to this change by

$$(dL/dt) = J_1 M/N_0 \rho_{liq} \quad , \qquad (3.2\text{-}22)$$

where M is the molecular weight of the liquid, N_0 is Avogadro's number, and ρ_{liq} is the density of the liquid. In order to obtain a final expression useful for the calculation of $\mathcal{D}_{12}$, the following additional assumptions are made:

(1) The vapor concentration at the gas-liquid interface, $(x)_0$, corresponds to the equilibrium vapor pressure at the liquid surface temperature.

(2) The vapor concentration at the outlet of the tube, $(x)_L$, is zero. This means that the carrier gas (supplied free of vapor) removes all the vapor away from the outlet.

(3) The gases and vapors are ideal, so that compositions may be expressed in terms of partial pressures.

Under these assumptions, and when Eq. (3.2-21) is substituted in Eq. (3.2-22) and integrated, the expression for $\mathcal{D}_{12}$ is

$$\mathcal{D}_{12} = \frac{(L_2^2 - L_1^2)}{2(t_2 - t_1)} \left(\frac{RT}{p}\right) \left(\frac{\rho_{liq}}{M}\right) \ell n \left(\frac{p - p_s}{p}\right) \quad , \qquad (3.2\text{-}23)$$

where p is the total pressure, p_s is the vapor pressure, R is the ideal gas constant, and subscripts 1 and 2 on L and t denote the initial and final times for the observations of the evaporation losses during an interval of diffusion. In some studies the weight loss of liquid is measured instead of the change in height.

The quasi-steady-state condition will be approached within 1% for diffusion times greater than $L^2/2\mathcal{D}_{12}$; to satisfy this condition experiments are usually run for several hours.[39]

The calculated values of $\mathcal{D}_{12}$ should be corrected for end effects caused by surface tension at the gas-liquid interface and turbulence at the tube outlet. End effects are related to the accurate determinations of the length of the diffusion path. The principal factor is turbulence which arises from interference by the end of the tube to the carrier gas flow. To avoid the effects of turbulence the gas flow rate can be empirically adjusted - not too great to cause large eddy currents, and not too small to cause a non-zero vapor concentration at the tube outlet. The presence of eddy currents will effectively shorten the diffusion path length. An end correction can be made by a graphical procedure. The calculated values of $\mathcal{D}_{12}$ are plotted as a function of the reciprocal of the observed length of the diffusion path L; the corrected values of $\mathcal{D}_{12}$ are taken at the extrapolated point, $1/L = 0$.

In addition to the normal instrumentation errors, the possible uncertainties of the evaporation-tube method are as follows. The evaluation of the term $\ell n[(p-p_s)/p)]$ indicates that small changes in pressure and temperature will cause large uncertainties in $\mathcal{D}_{12}$.[38] Significant variations in barometric pressure and in system temperature may occur since evaporation-tube experiments usually run for many hours. For example, if the partial pressure p_s is 25 Torr then for variations of ± 10 Torr in total pressure the variation of $\ell n[(p-p_s)/p)]$ is 1.4% for total pressures at about 1 atmosphere. The variations of the liquid surface temperatures may be even more critical because of the sensitivity of the vapor pressure to small temperature changes. For precise results the variation in temperature of the liquid should be no greater than $\pm 0.1°K$.

Other possible uncertainties for the evaporation-tube method are as follows:

(1) Convection effects caused by the direction of vapor diffusion.[39] For example, water-air values of $\mathcal{D}_{12}$ may differ by about 2% depending on whether the water is placed in the bottom or at the top of the tube. Additional convection effects are dependent on the diameter of the evaporation-tube, and a possible error of 4% is indicated

if the diameter of the tube is large.

(2) Nonequilibrium conditions may exist because of excessive rates of evaporation and super-cooling at the surface of the liquid.

(3) The contamination of the liquid by trace amounts of impurities may significantly effect the evaporation rate. In a study of water-air, for example, any traces of oil in the air could accumulate on the surface of the water. Another contamination process is due to gases dissolved in the liquid. In one case this effect caused differences of about 5% in values of $\mathcal{D}_{12}$.[40] This shows that liquids should be degassed prior to their use.

(4) The equilibrium values used in the formulas for $\mathcal{D}_{12}$ may themselves have significant uncertainties, especially equilibrium values that have been found in handbooks where the reliability of the data has not been specified.

(5) Many vapors are non-ideal gases, and any deviations from ideal-gas behavior affect the accurate specification of the mixture composition.[41]

(6) The assumption of a flat velocity profile is not strictly correct, since a parabolic profile develops as the vapor moves away from the gas-liquid interface. However, the maximum effect is only 1.4% in deviations of the radial concentration from a uniform (flat) value.[42,43]

At best, the reliabilities of $\mathcal{D}_{12}$ by the evaporation-tube method are several percent. The following cases are illustrative. In thirteen independent studies for the system water-air, the standard deviation is 7.5% at 298°K, and at higher temperatures the scatter in the data is even greater.[44] In another review of $\mathcal{D}_{12}$ for water-air, four of twelve studies were dropped from the calculation of the average value because they seemed obviously in error.[39] There are only a few other gas pairs, H_2-H_2O and benzene-air, which have several independent measurements useful for the estimation of reliability. These results are also disappointing.

3.3 Minor Experimental Methods

In this section the six minor methods listed in Table 2 are briefly discussed. The minor methods could probably yield more reliable values of $\mathcal{D}_{12}$ by further developments of each. The studies using these methods are listed in Table 8, and this listing contains the reference information for this section unless special footnotes are given in the text.

a. Open Tube

If the top of a gas container is opened, the gas will diffuse into the surrounding atmosphere, and analysis of the composition of the remaining gas after a known time permits the determination of $\mathcal{D}_{12}$. In 1882 the first such measurements were independently made by von Obermayer and by Waitz. The theory of the experiment was described by Stefan in 1871.[45] Detailed analyses of the method and these early studies have been published.[46,47] The open-tube method has recently been revived and improved by Frost.

b. Back Diffusion

Harteck and Schmidt in 1933 performed the first low-temperature determinations of $\mathcal{D}_{12}$, down to 20°K, for a mixture of para-hydrogen in normal hydrogen. The method is an ingenious steady-state flow technique in which one component diffuses upstream against the second flowing component. The composition at one or more upstream points can be used to determine $\mathcal{D}_{12}$. Back diffusion can be used for diffusion measurements at extreme temperatures, low or high, as well as for "tagged" molecules. The description of the original method has been translated, in part, into English.[2]

c. Capillary Leak

The capillary-leak method is suitable for measurements of $\mathcal{D}_{12}$ over a large range of temperatures because it involves no moving parts. In 1942 this method was first used by Klibanova _et_ _al_. to determine $\mathcal{D}_{12}$ at high temperatures, up to 1533°K. In 1967 De Paz _et_ _al_. determined the self-diffusion coefficient of Ar at low temperatures, down to 78°K. Except for the results by De Paz _et_ _al_., the precision and reliability of $\mathcal{D}_{12}$ obtained by the capillary-leak method have been poor.

d. Unsteady Evaporation

An alternate evaporation-tube method was developed by Arnold in 1944. His purpose was to obtain a quantitative basis for calculations of unsteady-state vaporization of a liquid into a gas, a process of industrial importance. The equations obtained also furnished a basis for relatively quick determinations of $\mathcal{D}_{12}$ for vapor-gas mixtures. Measurements could be made in minutes, not in hours as required by the Stefan evaporation tube. The reliability of the unsteady-evaporation method is probably slightly better than for the evaporation tube, but more meaningful comparisons are not possible because of the meager data available. A somewhat similar technique was used earlier by Mackenzie and Melville with bromine vapor. Other unsteady-evaporation studies are also listed in Table 8.

e. Diffusion Bridge

This is a steady-state flow method in which two gas streams flow across opposite ends of a hollow capillary tube or opposite faces of a porous septum, and the emerging streams are analyzed. The flow rates are controlled, and adjusted to produce any desired pressure difference across the capillary. The ends of the capillary are generally maintained at equal total pressures, thus in the capillary, or septum, there is uniform pressure and no viscous flow. The advantage of the diffusion bridge is that no valves are required in the zone of the apparatus where diffusion occurs, so that the method is amenable to operation over wide temperature ranges.

The diffusion bridge has been used only once with a capillary, to obtain absolute values of $\mathcal{D}_{12}$ down to 1.74°K. This work was done by Bendt in 1958.

The diffusion bridge has been used frequently with a porous septum, to obtain relative values of $\mathcal{D}_{12}$ up to 882°K. These studies require the calibration of the porosity of the system by means of independently published values of $\mathcal{D}_{12}$.

f. Dissociated Gases

Direct measurements of the diffusion of highly reactive species such as free radicals and valence-unsaturated atoms are difficult, but are needed for basic understanding of many phenomena in chemical reactions and at high temperatures. There are a variety of techniques, both of the flow and non-flow types, that have been used to measure the diffusion of H, N, and O atoms in different gases. The results for $\mathcal{D}_{12}$ extend over a temperature range of 202° to 873°K. Dissociated gases were first studied in 1959 by Wise and by Krongelb and Strandberg. The technique by Morgan and Schiff minimizes uncertainties due to chemical reactions; the complete neglect of reactions in calculations of $\mathcal{D}_{12}$ introduces less than 1% error. The results for dissociated gases, as might be expected, are not very reproducible; the results are scattered by about 10% or more for many gas pairs.

In some cases there are indirect methods available for the determination of $\mathcal{D}_{12}$ for dissociated gases which probably give more reliable results than the present direct methods. For example, $\mathcal{D}_{12}$ for $H - H_2$ can be obtained from measurements of the mixture viscosity,[48] and $\mathcal{D}_{12}$ for $N - N_2$ and $O - O_2$ at $T > 1000°K$ can be obtained from molecular-beam scattering experiments and semi-empirical quantal calculations.[49]

3.4 Miscellaneous Experimental Methods

This section briefly describes several miscellaneous methods that have been used to determine values of $\mathcal{D}_{12}$. A list of these studies is given in Table 9, and this listing contains the reference information for this section unless special footnotes are given in the text. The listing is not comprehensive, as a complete enumeration of all miscellaneous methods used at some time or another would be both futile and boring. The miscellaneous methods listed have both general applicability and experimental ingenuity.

a. Droplet Evaporation

Observations of the rate of evaporation of a small sphere of volatile material may be utilized to determine vapor-gas $\mathcal{D}_{12}$. The applicable theory is similar to the evaporation tube. Droplet evaporation studies have been made for water, for heavy organic chemicals, and for iodine in air.

b. Dufour Effect

When different gases mix, a small temperature gradient is set up; this is called the Dufour effect or the diffusion thermoeffect. The asymptotic time decay of the temperature gradient can be used to determine $\mathcal{D}_{12}$, and results are available for about 10 gas pairs at 293°K. These results have varied agreements, within 10%, with $\mathcal{D}_{12}$ obtained by the major methods.

c. Thermal Separation Rate

The inverse of the Dufour effect is thermal diffusion, in which an imposed temperature gradient causes the components of a mixture to separate. The rate at which an initially uniform mixture separates under an imposed temperature gradient can be used to determine $\mathcal{D}_{12}$. The results, however, are not very reliable. The principal uncertainties arise from averaging the temperatures of the imposed temperature gradient and geometrical factors of the apparatus.

d. Kirkendall Effect

In solids, the net drift of inert markers placed near a diffusion interface is called the Kirkendall effect. A similar effect exists in gases, and the speed of the marker motion can be used to determine $\mathcal{D}_{12}$. The marker is located in a tube connected in parallel to a two-bulb apparatus. A value of $\mathcal{D}_{12}$ has been obtained for He-Ar at 303°K, which is in excellent agreement with directly determined $\mathcal{D}_{12}$.

Smoke particles suspended in a diffusing gas mixture can also be used as Kirkendall markers.[50,51]

The diffusion pressure-effect, discussed in Chapter 2, Section 2.1, part a, is closely related to the Kirkendall effect. It could therefore also be used to determine values of $\mathscr{D}_{12}$, but this has not yet been done.

e. Sound Absorption

The passage of a sound wave through a gas mixture produces a local partial separation of the components, caused mostly by pressure diffusion. The remixing by diffusion is out of phase with the sound wave, and the absorption of an ultrasonic wave in a gas mixture is stronger than in either pure component. The excess absorption depends on $\mathscr{D}_{12}$, which in principle can then be determined.[52] This method has been tested on the gas pair He-Ar up to temperatures of $\sim$5000°K. The agreement of these results with other available data is good at 300°K, but poor between 1255° and 4990°K. These measurements are difficult to perform and the results at high temperatures are scattered.

f. Cataphoresis

A dc-discharge in a gas mixture causes a partial separation of components. The phenomenon, which also occurs in solutions, is called cataphoresis. The separation disappears by diffusion after the discharge is stopped, and $\mathscr{D}_{12}$ can be calculated from the rate of disappearance of the separation. At present this method has been used only for the gas pairs Ne-Ar from 300° to 650°K; the results are in good agreement with other direct measurements.

g. Resonance Methods

The principle of all resonance methods is to "tag" some of the molecules in a gas, and then follow their dispersion due to diffusion. The tags used have been such things as the orientation of nuclear spin (nuclear magnetic resonance), the population of magnetic sublevels in the ground state (optical pumping), or a metastable excited electronic state (mercury band fluoresence). The names in parentheses indicate the groupings for the studies listed in Table 9. The nuclear magnetic resonance technique has been used to determine $\mathscr{D}_{12}$ at very low temperatures, down to 20°K for mixtures of ortho- and para-hydrogen, and down to 1.13°K for the self-diffusion coefficient of ^{3}He. The optical pumping technique has been used to determine $\mathscr{D}_{12}$ of alkali metal vapors (Na, Rb, and Cs) in various other gases.

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4. TREATMENT OF DATA

In this chapter are outlined the procedures used to evaluate the entire body of experimental data, and the derivation of semi-empirical approximations for the composition dependence and temperature dependence of $\mathcal{D}_{12}$.

The original data, published over the last one hundred years, were compiled by author and by gas pair. Bibliography I lists the references to experimental studies by author, and the gas pairs investigated and the experimental methods are noted. A cross-listing of $\mathcal{D}_{12}$ by gas pair is given in Table 16, in which the temperature range is also noted. In addition, there are bibliographies for short-range and long-range interaction data, and for measured mixture viscosities.

4.1 Reliability Estimates

The critical evaluation of the reliability of $\mathcal{D}_{12}$ from direct measurements included the following factors:

(1) experimental method,

(2) reproducibility of $\mathcal{D}_{12}$ by different experimental methods or laboratories,

(3) precision and number of measurements from a given laboratory,

(4) temperature dependence measured.

For indirect measurements, the reliability of $\mathcal{D}_{12}$ considered the reported accuracy of other transport property data of mixtures - viscosity, thermal conductivity, and thermal diffusion factor - and of molecular beam measurements. Whenever collision integral ratios were employed in intermediate calculations of $\mathcal{D}_{12}$, their reported accuracy was considered as well as the choice of the potential model.

For all measurements, the results of the more recent studies were not assumed to be necessarily more accurate than those from earlier studies. All the data for each gas pair were carefully inspected for discrepancies and systematic errors with the aid of large-scale graphs. From these it became apparent that the small composition dependence of $\mathcal{D}_{12}$ had to be taken into account, and that compact means of summarizing values of $\mathcal{D}_{12}$ over large temperature ranges were necessary.

The assignment of reliability estimates to experimental data always involves a large measure of subjective judgment.[1-3] Even after consideration of such things as reproducibility and internal consistency, external consistency for different types of apparatus and for different workers in different laboratories, and so on, the final decisions are nevertheless based heavily on the judgments of the evaluators. An attempt has been made to be conservative, in order that there shall be a high probability that the "true" value of a diffusion coefficient lies within the specified range of uncertainty. An attempt has also been made to be fair and not arbitrarily downgrade good measurements, but is is quite possible that a particular $\mathcal{D}_{12}$ may be more accurate than is implied by the specified uncertainty limits which are given in Chapter 5.

4.2 Correction for Composition Dependence

Even though the magnitude of the composition dependence of $\mathcal{D}_{12}$ is relatively small, from 0 to 5% for all gas pairs considered, the effect is sometimes greater than the uncertainty of experimental measurements. The formulas for the composition dependence according to the second approximation of the Chapman-Enskog theory (Section 2.4) are cumbersome to use, especially when thousands of data points must be considered. A simpler and more convenient formula of sufficient accuracy can be developed as follows.

The entire composition dependence of $\mathcal{D}_{12}$ is contained in the small term Δ_{12}, given in Section 2.4, which depends on both temperature and composition. The major complication of the Chapman-Enskog expression for Δ_{12} is its composition-dependent part (containing the P's and Q's). Previous work[4] indicates that the composition dependence can be adequately approximated by the formula,

$$\Delta_{12} \approx \zeta(6C_{12}^{*} - 5)^2 \frac{ax_1}{1+bx_1} \quad , \qquad (4.2\text{-}1)$$

where ζ is a numerical constant between 1 and 2, x_1 is the mole fraction of the heavy component, and a and b are

$$a = \frac{1}{10}(-S_2/Q_2)(P_1/S_1) \quad , \qquad (4.2\text{-}2)$$

$$b = (-S_2/Q_2)(Q_1/S_1)-1 \quad . \qquad (4.2\text{-}3)$$

The S_1 and S_2 occur in the expression for the thermal diffusion factor (see Section 2.7), which is related theoretically to Δ_{12}.[4] This formula is most accurate for $M_1 >> M_2$; the lower accuracy for $M_1 \approx M_2$ is not important, however, because Δ_{12} is negligible in such cases.

The expressions for a and b can be further simplified. It is obvious that

$$b + 1 = 10a(Q_1/P_1) \quad . \qquad (4.2\text{-}4)$$

Substitution for P_1 and Q_1 from Eqs. (2.4-1) and (2.4-3) yields

$$Q_1/P_1 = (\frac{5}{2} - \frac{6}{5}B_{12}^{*}) + 3(M_2/M_1)^2 + \frac{8}{5} A_{12}^{*} (M_2/M_1) \quad . (4.2\text{-}5)$$

An adequate approximation for the present purpose is to take $B_{12}^{*} = 5/4$ (Kihara approximation) and $A_{12}^{*} \approx 1.1$, which yields

$$b + 1 = 10a(1 + 1.8\ m + 3m^2) \quad , \qquad (4.2\text{-}6)$$

where

$$m = M_2/M_1 < 1 \quad . \qquad (4.2\text{-}7)$$

For small m, the major variation of a comes from the factor $(-S_2/Q_2)$ and can be represented by the simple expression

$$a \approx \frac{2^{1/2}}{8(1 + 1.8\ m)^2} \frac{\bar{\Omega}_{12}^{(1,1)}}{\bar{\Omega}_{22}^{(2,2)}} \quad . \qquad (4.2\text{-}8)$$

The factor $(1 + 1.8\ m)^2$ is an empirical representation of the various mass dependences, but the rest of the expression comes from theory. The collision integrals in a may be obtained either by calculation from a potential model or from experimental values of $\mathscr{D}_{12}$ and η_2, the viscosity of the light component, whereby Eq. (4.2-8) may be written as

$$a \approx \frac{3(1 + m)^{1/2}}{20(1 + 1.8\ m)^2} \frac{\eta_2 RT}{p\mathscr{D}_{12}M_2} \quad . \qquad (4.2\text{-}9)$$

The quantities a and b vary only weakly with temperature, and can usually be taken as constant.

The complete result for Δ_{12} is thus given by Eqs. (4.2-1), (4.2-6), (4.2-7), and (4.2-8) or (4.2-9). The value of C_{12}^* in Eq. (4.2-1) can be calculated from the Lennard-Jones (12-6) potential, and depends only on the temperature and the value of ε_{12}; the results are not too sensitive to the choice of the potential and the precise value of ε_{12} used. The value of ζ is unity according to the Chapman-Enskog second approximation; since this approximation seems to underestimate Δ_{12}, it is better to take ζ from experiment if accurate data are available. Values of ζ, a,b and ε_{12} are listed in Table 15 of Chapter 5.

Figure 3 shows a comparison of values of Δ_{12} calculated from the semi-empirical approximation with values calculated from the Chapman-Enskog expression. Two mass ratios are shown, which represent reasonable values for ordinary gas pairs like He-Ar and Ne-Ar; the potential parameters used in the calculations correspond to these two gas pairs. A high reduced temperature of $kT/\varepsilon_{12} = 10$ is used, for which Δ_{12} is large. The results for Δ_{12} are in agreement within the uncertainties of experimental measurements.

Values of the empirical constant ζ have been determined from measurements of the composition dependence of $\mathcal{D}_{12}$ for only fourteen systems (counting H_2 and D_2 as the same); namely, the ten noble-gas pairs,[5] H_2-N_2,[6] H_2-Ar and H_2-CO_2,[4] and He-N_2.[7] These are the only systems for which enough accurate data on composition dependence exist to justify assigning ζ a value other than 1.0. As can be seen from Table 15 of Chapter 5, the empirically determined values of ζ do lie between 1 and 2, as expected. An advantage of the semi-empirical formula is that improved experimental information on the composition dependence of $\mathcal{D}_{12}$ can be easily accommodated by adjustment of values of ζ.

Experimental $\mathcal{D}_{12}$ data were adjusted to refer to an equimolar composition according to the relation

$$\mathcal{D}_{12}(x_1 = 1/2) = \mathcal{D}_{12}(x_1) \left[\frac{1 + \Delta_{12}(x_1 = 1/2)}{1 + \Delta_{12}(x_1)}\right] , \quad (4.2\text{-}10)$$

where $\mathcal{D}_{12}(x_1)$ was the value measured at mole fraction x_1, and the Δ_{12} were calculated from Eq. (4.2-1) with the constants given in Table 15 of Chapter 5. An equimolar basis is a reasonable compromise between composition extremes, and required a minimum number of adjustments of the data.

4.3 Correlation for Temperature Dependence

The temperature dependence of $\mathcal{D}_{12}$ can be correlated by a semi-empirical equation which is applicable over a wide range of temperature. The theoretical background for the correlation, in terms of intermolecular forces, has been presented in Section 2.3; the equation itself is an empirical composite of terms corresponding to various types of contributions to the intermolecular forces. The equation correlates the temperature dependence of $\mathcal{D}_{12}$ within the experimental uncertainties of the experimental results with at most four adjustable parameters, and can be put into simplified form for data with low reliability or with limited temperature range.

The background is briefly as follows. Many previous correlations of the temperature dependence of $\mathcal{D}_{12}$ have been published.[8-34] These correlations have usually been restricted to fairly narrow temperature ranges between about 200° and 500°K, because of the lack of data at low and high temperatures. But recent results on $\mathcal{D}_{12}$ by direct measurements and by calculations from molecular-beam scattering experiments have significantly extended the temperature range, which now extends roughly from about 10,000° down to 80°K, or lower.

In Fig. 1 the insert shows the characteristic temperature dependence of $p\mathcal{D}_{12}$. Over a narrow range of temperatures a plot of $\ln(p\mathcal{D}_{12})$ versus $\ln T$ is essentially linear, as would be expected for an inverse-power potential, but over a large temperature range such a plot shows curvature. At low temperatures the curvature is caused by the increasing influence of the long-range attractive potential. At high temperatures the curvature is caused by the increasing "softness" of the repulsive potential at small separation distances, as would be expected for an exponential potential.

The foregoing features can be fitted by an equation of the form,

$$\ell n(p\mathscr{D}_{12}) = \ell n\ A + s\ \ell n\ T - \ell n[\ell n(\varphi_o/kT)]^2 - (S/T) - (S'/T^2) \quad , \qquad (4.3\text{-}1)$$

where A, s, φ_o, S, and S' are empirical constants, and k is the Boltzmann constant. The double logarithm term is taken from Eq. (2.3-4) and represents an exponential repulsion potential. The value of φ_o is taken from independent molecular-beam experiments,[35] and is not adjustable; however, its precise value is not critical for correlation purposes, because errors in φ_o are compensated for by values of s. The values of s are equal to or slightly greater than 3/2, as expected from theory. The terms containing S and S' are Sutherland-Reinganum terms, as in Eq. (2.3-11), and account for the attractive potential. For most gas pairs S' is not needed and can be taken as zero.

In many cases the values of $\mathscr{D}_{12}$ are not sufficiently precise to require the use of the double logarithm term in Eq. (4.3-1), and an adequate representation is given by

$$\ell n(p\mathscr{D}_{12}) = \ell n\ A + s\ \ell n\ T - (S/T) \quad . \qquad (4.3\text{-}2)$$

All the data could be correlated within the range of estimated experimental uncertainties by combinations of Eqs. (4.3-1) and (4.3-2).

An advantage of Eqs. (4.3-1) and (4.3-2) is that they are linear in all the adjustable constants (A, s, S, and S'), so that least-squares calculations are easy. A disadvantage is that the Sutherland-Reinganum terms for the effect of the long-range attraction do not permit the equations to be used at very low temperatures, where the London dispersion energy dominates. At present, no measurements of $\mathscr{D}_{12}$ seem to fall in this range, however. The equations are usable only for $kT/\varepsilon_{12} > 1$, and should never be extrapolated to low temperatures. At very low temperatures $\mathscr{D}_{12}$ has the asymptotic (classical) form,

$$p\mathscr{D}_{12} = AT^{11/6} \quad , \quad T \rightarrow 0 \quad , \tag{4.3-3}$$

where A is easily calculated if the London constant C is known.[36] The reliability of Eq. (4.3-3) has been discussed in Section 2.6, part a.

The values of the constants for Eqs. (4.3-1), (4.3-2), and (4.3-3) are given in Tables 12, 13, and 14, respectively, of Chapter 5. Details on their determination are presented in Chapter 5.

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5. RESULTS

In this chapter the recommended values of $\mathcal{D}_{12}$ for Standard Reference Data are presented. The reliability estimates of these $\mathcal{D}_{12}$ are given in Section 5.1. Then, in Section 5.2, the constants are listed for the correlations of $\mathcal{D}_{12}$ as a function of temperature and of composition. In Section 5.3 the deviations between data and the recommended values of $\mathcal{D}_{12}$ are illustrated by graphs. The detailed remarks on the critical evaluation of data for weighted least-squares calculations are given in Section 5.4.

This chapter summarizes the most reliable experimental results for binary gaseous diffusion coefficients. Gas pairs for which only limited or uncertain data exist are not included here, but these experimental measurements may be retrieved with the aid of Table 16.

5.1 Uncertainty Limits

The sources of reliable values of $\mathcal{D}_{12}$ are roughly as follows. For all gas pairs the most accurate results are at approximately 300°K, because of the existence of a large number of independent measurements by the most reliable experimental methods. Both closed-tube and two-bulb measurements are usually available at temperatures from 200° to 500°K, and several additional two-bulb measurements exist at lower temperatures. The temperature limits of $\mathcal{D}_{12}$ from direct experiment have been extended in both directions by the use of data on mixture viscosities, or, in a few instances, on thermal diffusion factors. These derived values of $\mathcal{D}_{12}$ have slightly less reliability than those near room temperature. The magnitude of the extended temperature range is variable, and depends on the particular gas pair. For several gas pairs data are available to about 1000°K, and in a few instances to higher temperatures, but less than 2000°K, from the point-source method. Values of $\mathcal{D}_{12}$ from 1000° to 10,000°K are derived mainly from molecular-beam measurements, which generally have the least reliability.

The gas pairs included as recommended Standard Reference Data systems can be grouped into three categories of reliability, as shown in Fig. 4. A gas pair in Group I, for instance, has uncertainty limits of $\pm 1\%$ in $\mathcal{D}_{12}$ at 300°K; the uncertainty increases to $\pm 5\%$ at 1000°K, and to $\pm 10\%$ at 10,000°K. The temperature dependences of the uncertainty limits are shown in Fig. 4, and the gas pairs assigned to each group are listed in Table 10. The borderline systems are assigned to the higher group, but are noted by a question mark. Table 10 lists the gas pairs in terms of one common member in a series of gas pairs; this leads to some duplication but is helpful for quick reference.

A miscellaneous group of gas pairs is also included because of possible special interest, and their uncertainty limits are listed in Table 11. The miscellaneous group contains mixtures with one component water, carbon dioxide, or dissociated gas (H, N, or O).

The gas pairs of Group I have the most reliable values of $\mathcal{D}_{12}$ for two principal reasons. First, results below 400°K are based on the very careful measurements of van Heijningen _et al_.. Second, at high temperatures, the values of $\mathcal{D}_{12}$ derived from molecular-beam scattering experiments for the noble gas pairs are more reliable than for diatomic or polyatomic gases, as discussed in Section 2.6, part b. In Group I the gas pairs Kr-Xe and H_2-N_2 are borderline systems, even though one is a noble gas pair and the other has more reliable data than any other except for He-Ar, because at temperatures above 1000°K they have uncertainty limits corresponding to Group II. This is due to the relatively large amount of scatter in the Kr-Xe results from molecular-beam measurements. For H_2-N_2 the potential was determined by use of the combination rules (Section 2.6, part c), not by direct molecular-beam measurements. Additional uncertainties arise for diatomic molecules which are not rigorously treated in the theoretical model.

The gas pairs of Group II have values of $\mathcal{D}_{12}$ which have been consistently verified by several independent studies and by different apparatus. There are, however, four borderline systems in Group II. Three of these, He-O_2, H_2-Ne, and N_2-Ar, do not have as many reliable measurements as the other gas pairs of Group II. The fourth gas pair, H_2-Kr, is a borderline system because the uncertainty limits are estimated to be ±4% at 90°K and ±3% at 500°K, which are limits slightly greater than the levels specified for Group II. Another gas pair, He-CO, is included in Group II because it has diffusion characteristics similar to He-N_2, or almost identical values of $\mathcal{D}_{12}$.

The gas pairs of Group III have a relatively small number of reliable measurements of $\mathcal{D}_{12}$ at about room temperature. At temperatures above 1000°K the values of $\mathcal{D}_{12}$ have relatively large uncertainties because large discrepancies exist in the molecular-beam measurements used to calculate values of $\mathcal{D}_{12}$, or because the beam measurements have been obtained from only one laboratory. Usually there are two laboratory sources for beam results.

The uncertainties in the miscellaneous systems are rather variable, as can be seen from Table 11. The uncertainties for H_2O-CO_2 look peculiar, but the higher accuracy at high temperatures is due to the existence of data by the point-source method.

5.2 Correlation Parameters

This section gives the correlation parameters for values of $\mathscr{D}_{12}$ as a function of temperature and of composition.

The diffusion coefficients were correlated as a function of temperature in accordance with the semi-empirical reference equations discussed in Section 4.3. The empirical constants for Eq. (4.3-1) are listed in Table 12, and for Eq. (4.3-2) in Table 13; there are seventy-four gas pairs in all. For Tables 12 and 13 the values of $\mathscr{D}_{12}$ were adjusted to refer to equimolar mixtures, with two exceptions. First, systems involving air refer to trace diffusion through a large excess of air (see Section 2.1, part b). Even when direct measurements were available for air, most of the constants were generated from the corresponding values of $\mathscr{D}_{12}$ for N_2 and O_2 according to Blanc's law. In this way more reliable data are used as the basis of the reference equations; the direct measurements were always compared to results by Blanc's law and found to be in agreement. Second, systems involving dissociated gases have data that are obtained from measurements of a trace atom diffusing through a mixture, or from calculations of $\mathscr{D}_{12}$ based on molecular-beam measurements. Since the uncertainties in both cases are greater than the composition dependence of $\mathscr{D}_{12}$, it was unnecessary to adjust these data to an equimolar composition.

In Tables 12 and 13 the gas pairs are ordered as follows: (1) mixtures of noble gases with noble gases arranged according to atomic weight of the lighter component, (2) mixtures of noble gases with other gases arranged according to the atomic weight of the noble gas, (3) other mixtures arranged according to the molecular weight of the lighter component, and (4) dissociated gases. Except for ^{3}He-^{4}He and H_2-D_2 isotopic mixtures are not included, since the self-diffusion coefficient is merely proportional to the viscosity.

The results given in Tables 12 and 13 cannot be extrapolated to low temperatures, for the form of Eqs. (4.3-1) and (4.3-2) is unsuitable when the long-range London dispersion energy dominates the interaction (see Section 2.6, part a). In such a case, values of $\mathscr{D}_{12}$ may be obtained from the classical asymptote, Eq. (4.3-3). The correlation constants for the classical asymptotes are given in Table 14; a total of twenty-four gas pairs are listed which have London dispersion constants available.

If estimates of $\mathscr{D}_{12}$ are required outside the temperature range of a reference equation, then care must be taken when extrapolations are made. At temperatures greater than 10,000°K, extrapolations are safer to make than at very low temperatures because of the form of the equations. However, at elevated temperatures an extrapolation will neglect the effects of inelastic collisions and internal excitation of molecules. When extrapolations have to be made at lower temperatures, both the reference equation and the low-temperature asymptote should be used to obtain two predictions of $\mathscr{D}_{12}$ at a given temperature. The larger value calculated is the better estimate of $\mathscr{D}_{12}$. This procedure neglects quantum effects, and unfortunately asymptotic constants are only available for about one-third of the gas pairs with recommended Standard Reference Data.

If values of $\mathscr{D}_{12}$ are required at pressures unequal to 1 atmosphere, then the reciprocal pressure relationship of $\mathscr{D}_{12}$ is used, as discussed in Section 2.2.

The values of $\mathcal{D}_{12}$ can be adjusted to a nonequimolar composition by the method developed in Section 4.2. The values of the constants of Eq. (4.2-1) are given in Table 15, using the same order of listing as described above for Tables 12 and 13. Included in Table 15 are a number of gas pairs in which D_2 replaces H_2. Omitted from this table are mixtures with dissociated gases and several systems for which the molecular weights of the gases are so close that the composition dependence is negligible. These systems are He-D_2, Ar-CO_2, N_2-CO, N_2-O_2, CO-O_2,CO-air, CO_2-N_2O, and CO_2-C_3H_8. Table 15 is convenient for making rapid estimates of the composition dependence of $\mathcal{D}_{12}$, or for correcting data to a specific composition, reliable to within the uncertainties of the experimental measurements.

5.3 Deviation Plots

The experimental diffusion coefficients are compared with correlated values of $\mathcal{D}_{12}$, and deviations are presented in a series of graphs, Figs. 5 to 81. Their sequence is in general accordance with the listing of gas pairs in Table 10. There are no deviation plots for the mixtures with dissociated gases and for several other gas pairs which have only meager data available. The deviation plots do not present all the data for a given gas pair; results obtained from miscellaneous experimental methods or published in graphical form have been omitted.

The deviation plots show general features of experimental values of $\mathcal{D}_{12}$ as follows. First, the overall consistency of the data is rather good, although some reported values of $\mathcal{D}_{12}$ show considerable scatter. Second, careful appraisal of the experimental data is necessary to obtain the most reliable estimate of $\mathcal{D}_{12}$. A random selection of a value of $\mathcal{D}_{12}$ from the literature could easily yield a result with an uncertainty of 5%, even though the original article would probably claim much less. Third, the results by the closed-tube and two-bulb methods are more consistent than others, and show no evidence of any systematic disagreement. This can be illustrated by the results for He-Ar and H_2-N_2 which are given in Figs. 6,7,18 and 19. Fourth, most values of $\mathcal{D}_{12}$ at temperatures above 1000°K are available only indirectly, that is from molecular-beam measurements. Fifth, in the approximate temperature range of 500° to 1000°K the point-source method has provided almost all the reliable data. Sixth, results from gas-chromatography measurements only supplement results by other methods for the gas pairs listed in Table 10; however, gas-chromatography measurements give the only reliable data for many other mixtures. Finally, it has obviously been difficult to make any diffusion coefficient measurement with an uncertainty less than 1%.

Detailed remarks on the deviation plots are as follows. A positive deviation means that an experimental value of $\mathcal{D}_{12}$ is greater than a value calculated from the reference equation. All values have been corrected to equimolar composition. Each deviation point has been plotted with a precision greater than 0.1% by means of a Calcomp plotter (model 563). When a number in parentheses is placed by a point, then this number specifies the magnitude of the deviation (which happens to be greater than the ordinate scale). The abscissa usually covers the temperature range from 63° to 10,000°K, but lower temperatures appear on a few graphs for ^{3}He-^{4}He and H_2-D_2. Along the abscissas, hash marks (≀≀) have been used to condense the temperature scale at elevated temperatures. Because of this break in the scale, points from similar sources are not connected by lines between 1000° and 10,000°K, as is done at lower temperatures.

For each gas pair the reference equation for $\mathcal{D}_{12}$ recommended as

Standard Reference Data is given with the deviation plot. These equations are exactly the same as in Tables 12 and 13. Occasionally, below an equation the parenthetical statement "(same as ...)" appears. This means that the diffusion characteristics of two gas pairs are so similar that one equation is suitable for the correlation of the data of both. Results by the closed-tube method are noted as "Loschmidt tube". The values of $\mathcal{D}_{12}$ calculated in this report from molecular-beam scattering experiments are referred to by one of the two laboratory sources, namely "Beam data of Amdur, et al." for data from the Massachusetts Institute of Technology, and "Beam data of Leonas, et al." for data from the Moscow State University. There are parenthetical notes in the legend, some of which indicate the following: (1) a prime author whose results were available only as reported by others; (2) two-bulb apparatus which has been used to produce both "relative" values of $\mathcal{D}_{12}$ and the usual "absolute" values (relative values of $\mathcal{D}_{12}$ are obtained by calibrating the apparatus against a mixture with known $\mathcal{D}_{12}$); (3) standard deviations that indicate significant internal scatter, as published; (4) the type of radioactive species used in some experimental determinations; (5) the basis of some values of $\mathcal{D}_{12}$, for instance, mixture viscosity.

5.4 Detailed Remarks

This section presents the detailed remarks on the critical evaluation and the correlations of $\mathcal{D}_{12}$. Reasons are given for the assignment of a gas pair into a particular category of reliability. Enough information is reported to allow the recovery and the verification of the reference equations; most of this information is presented in Tables 17 to 25. In these tables the sources of data noted by an asterisk are for values of $\mathcal{D}_{12}$ selected from large-scale graphs by eye. A selected value is a reasonable estimate in a small temperature region; that is, no published value of $\mathcal{D}_{12}$ was considered extraordinarily superior to other available measurements.

The general order of the detailed remarks is as follows. The remarks are divided into four sections corresponding to the four reliability groups of Table 10. The discussions contain the following information: (1) weights for values of $\mathcal{D}_{12}$ used in the least-squares calculations of the reference equations, (2) intermolecular potentials obtained from molecular-beam experiments which were used to calculate $\mathcal{D}_{12}$ at elevated temperatures, and (3) special comments.

For this chapter, most references are to be found in the Bibliography by author, and are not given at the end of the chapter.

a. Group I (Deviation Plots, Figs. 5 to 20)

Weights and Potentials. Similar weights were assigned to values of $\mathcal{D}_{12}$ to correlate the data of Group I. The accurate measurements (at equimolar composition) of van Heijningen *et al*. (1966,1968) were weighted unity; almost every other determination of $\mathcal{D}_{12}$ was disregarded at temperatures below 400°K for the ten noble gas pairs and H_2- N_2. At 1000° and 10,000°K selected values, which were based on molecular-beam measurements, were weighted 1/5 and 1/10, respectively; these values are listed in Table 17. These are logarithms of $\mathcal{D}_{12}$ which were read from the large-scale graphs and used directly in the calculations. At 1000°K, the selected diffusion coefficients were obtained by extrapolation of $\mathcal{D}_{12}$ calculated from beam results down to room temperature, and extrapolation of data between 295° and 400°K up to elevated temperatures. The selected values of $\mathcal{D}_{12}$ were taken to lie between these two extrapolations. At

10,000°K, the selected values approximate the mean of $\mathscr{D}_{12}$ based on the molecular-beam measurements of Amdur et al. and Leonas et al.

The above weighting policy had a few exceptions as follows. First, for He - Kr the weight of the datum at 295°K was increased from unity to two; otherwise the calculated deviations would have exceeded the uncertainty limits of Group I. An additional point at 77°K ($\mathscr{D}_{12}$ = 0.0607) was used in the least-squares calculations, and it was weighted 3/10. This value of $\mathscr{D}_{12}$ was obtained from calculations based on the temperature dependence of the thermal diffusion factor by Annis et al. (1968) and normalized to the 295°K datum by van Heijningen et al. (1968). Second, for H_2- N_2 an additional datum at 562°K ($\log_{10}T = 2.750$, $\log_{10}\mathscr{D}_{12} = 0.365$) was included with a weight of 1/3 in the least-squares calculations. This point was used in order to improve the interpolation between the highest temperature (295°K) result by van Heijningen et al. (1966) and the selected point at 1000°K.

At temperatures greater than about 1000°K, values of $\mathscr{D}_{12}$ were based on intermolecular potentials obtained from molecular-beam scattering experiments. Leonas et al. performed beam experiments for each of the ten noble gas pairs, and also determined potentials for H_2- H_2 and N_2- N_2 which lead to an H_2- N_2 potential by application of the combination rules given in Section 2.6, part c. Independent molecular-beam measurements have also been made by Amdur et al. for He - Ar, Ne - Ar, and the pairs He - He, Ne - Ne, Ar - Ar, Kr - Kr, and Xe - Xe. By application of the combination rules, this information also yielded potentials for all the noble gas pairs. Amdur et al. also measured potentials for He - H_2 and He - N_2, from which the H_2- N_2 potential was obtained. In Table 18 the potential functions are listed which were used to calculate the deviation points shown on Figs. 5 to 20.

Special Comments. The lower temperature limits for He - Ne and Ne - Ar might have been extended to temperatures beyond the results established by van Heijningen et al. For He - Ne a datum was available at 20.4°K, and for Ne - Ar a datum at 65°K; both values of $\mathcal{D}_{12}$ are based on mixture viscosity (Weissman and Mason, 1962 b).After consideration of the reliability of the viscosity data, of A^*_{12}, and of the need for a quantum correction, these values of $\mathcal{D}_{12}$ were not used to extend the lower temperature limits of the reference equations for these systems.

The systems Kr - Xe and H_2- N_2 are borderline because of the greater uncertainties in their molecular-beam potentials.

In two-bulb measurements for noble gas pairs there are apparent systematic errors in results by three independent investigators:

(1) van Heijningen et al. (1968),

(2) Malinauskas (1965, 1966, 1968),

(3) Srivastava (1959), Srivastava and Barua (1959), and Srivastava and Srivastava (1959).

The magnitudes of the errors are usually a few percent or less, and are discussed relative to the more accurate work by van Heijningen et al. The results by Malinauskas are slightly lower for the lighter gas pairs (He - Ne, He - Ar, He - Kr, He - Xe, and Ne - Ar) and higher for the heavier gas pairs (Ne - Kr, Ne - Xe, Ar - Kr, Ar - Xe, and Kr - Xe). A cause for this trend could not be found. The results by Srivastava are all below those by van Heijningen et al., except those for He - Xe, which are high. The lack of internal scatter in these measurements by Srivastava is presumably due to smoothing the data.

An interesting result for the gas pairs of Group I is that values of $\mathcal{D}_{12}$ by direct measurements are in reasonable agreement with $\mathcal{D}_{12}$

determined from other transport properties, as shown in Table 19. Here direct measurements by van Heijningen *et al*. (1966,1968) are compared with $\mathscr{D}_{12}$ calculated from mixture viscosity and thermal conductivity data, reported by Weissman and Mason (1962 b) and by Weissman (1965). For Table 19 the deviations were extracted from results given in the deviation plots for Group I. A relative index of reliability has also been computed, which is defined as the average absolute deviation of the results by van Heijningen *et al*. divided into the average absolute value of the other deviations, and it is given in the bottom row of Table 19. These results indicate that $\mathscr{D}_{12}$ can be well predicted from other transport property data at about room temperature. The diffusion coefficients calculated from the most accurate mixture viscosity data available (Kestin *et al*.) appear to be better than they should; that is, the viscosity-derived $\mathscr{D}_{12}$ are really less reliable than the direct measurements of $\mathscr{D}_{12}$ because the uncertainties in the A^*_{12} values are no less than 1%. The other mixture-viscosity sources yield $\mathscr{D}_{12}$ only as reliable as the Group III uncertainty limits. However, diffusion coefficients calculated from available mixture thermal conductivities fall outside the range of Group III, or the average deviation is greater than 3% at about 300°K. This occurs because thermal conductivity measurements have much larger uncertainties than viscosity data, and not from any inadequacies of the theoretical formula.

b. Group II (Deviation Plots, Figs. 21 to 46)

Weights and Potentials. The equimolar values of $\mathscr{D}_{12}$ and their weights used in the least-squares calculations are presented in Table 20. The potential functions obtained from molecular-beam measurements are summarized in Table 21. A few systems have direct molecular-beam measurements, but most gas pairs of Group II have potentials that were obtained by the combination rules. The calculated potential functions are listed on the left-hand side of Table 21, and the potentials from direct molecular-beam measurements are listed on the right-hand side. No potentials are given for He - CO_2, H_2 - air, H_2 - CO_2, and N_2 - CO_2 because when this work was done, there were no molecular-beam measurements available with air or CO_2.

The potentials by Amdur et al. for He - CO, He - 0_2, and H_2- CO were calculated by combination rules from measurements obtained in different apparatuses. The potential energy ranges for these measurements were not the same. Thus, the derived potentials are applicable over a smaller temperature range, and are also considered slightly less reliable than results obtained from a single apparatus.

The potential for ^{3}He - ^{4}He was taken the same as for ^{4}He - ^{4}He, and that for H_2- D_2 the same as for H_2- H_2; that is, potentials were assumed identical for isotopic pairs. This is only an approximation, but is sufficiently accurate for the present purposes.[1,2]

Special Comments. For the seventeen gas pairs of Group II special comments are as follows.

^{3}He - ^{4}He. This gas pair is exceptional because its assignment into Group II is based mainly on the reliability of values of $\mathcal{D}_{12}$ calculated from viscosity measurements. In some instances, viscosity data for ^{4}He - ^{4}He (Becker and Misenta, 1955; Coremans et al., 1958 a; Rietveld et al., 1959) were used along with the appropriate reduced-mass correction factor and quantum-corrected values of A^*_{12} and $\Omega^{(1,1)*}$. At high temperatures values of $\mathcal{D}_{12}$ were calculated from the ^{4}He viscosity data by Kalelkar and Kestin (1969). The direct measurements of $\mathcal{D}_{12}$ by Bendt (1958) are in good agreement with the low-temperature results calculated from viscosity. There are other direct measurements for ^3He - ^4He by DuBro (1969), which are not shown on the deviation plots because they were unavailable until recently. DuBro used a two-bulb method and covered the temperature range of 76.5° to 344°K. The average absolute deviation of his results from the reference equation is 2.6%.

In the temperature range of 14.4° to 90°K the form of Eq. (4.3-1) was not sufficiently flexible for curve-fitting purposes; that is, the data (obtained from mixture viscosities) were considered more accurate than the uncertainty specified for a Group II system in that temperature range. For temperatures between 14.4 and 90°K a simple power function was calculated by the method of least squares, in which 27 points were weighted equally. The result fits the data with an average absolute deviation of 1.1% and a standard deviation of 1.4%.

$\underline{He - N_2}$. This gas pair has many reliable measurements by different major experimental methods which allow it to be a Group II system, see Figs. 23 and 24.

$\underline{He - CO \text{ and } H_2 - CO}$. The diffusion coefficients of He - CO and H_2 - CO can be well approximated by those for He - N_2 and H_2- N_2 (Group I), respectively, because CO and N_2 are isosteric molecules and He - N_2 and H_2- N_2 have more reliable measurements than do He - CO and H_2 - CO. A comparison of the reference equations with the reliable measurements by Ivakin and Suetin (1964 a,b) for He - CO and H_2- CO shows deviations less than about 2%. The H_2 - CO system was not assigned to Group I, as is H_2- N_2, because of possible uncertainties due to the lack of measurements at low temperatures and asymmetry between the CO and N_2 molecules which may lead to small differences between the potentials.

$\underline{He - O_2}$. This gas pair is a borderline system of Group II. He - O_2 was assigned to Group II on the strength of the two-bulb measurements by

Paul and Srivastava (1961 a) and the consistency of its $\mathcal{D}_{12}$ with those of He - N_2 and He - air.

He - air and H_2 - air. These two gas pairs are grouped together because their reference equations have been calculated by application of Blanc's law, Eq. (2.1-7); that is, He - air was obtained from He - N_2 and He - O_2, and H_2 - air from H_2- N_2 and H_2- O_2. The reliable determinations of $\mathcal{D}_{12}$ have been by the closed-tube method; namely, for He - air the room temperature point by Fedorov et al. (1966) and for H_2 - air the point by Ivakin and Suetin (1964 b). Each of these direct measurements is in excellent agreement with the specified reference equations. However, since Blanc's law introduces a small uncertainty into the results, the H_2- air system was not assigned to Group I along with H_2- N_2, but to Group II.

Each lower temperature limit for the reference equations of He - air and H_2- air has been set by data of He - O_2 (244°K) and H_2- O_2 (252°K), respectively. But the lower temperature limit for He - N_2 is 77°K, and for H_2- N_2 it is 65°K. In order to estimate $\mathcal{D}_{12}$ for He - air at lower temperatures, if necessary, it is suggested that the ratio of $\mathcal{D}_{12}$ for He - O_2 to He - N_2 at room temperature be taken as a constant, independent of temperature, and a similar procedure used for H_2- air. By applying this ratio the correlation range can be extended for He - O_2 and H_2 - O_2, as well as He - air and H_2- air to the lower temperature limits for He - N_2 (77°K) and for H_2- N_2 (65°K).

He - CO_2. The few closed-tube measurements by Holsen and Strunk (1964) and by Ivakin and Suetin (1964 b), plus one two-bulb measurement by Annis et al. (1969), established He - CO_2 as a Group II gas pair.

H_2- He. This gas pair is in Group II primarily on the basis of reliable closed-tube measurements by Amdur and Malinauskas (1965), by

Rumpel (1955), and by Bunde (1955).

H_2- Ne. This gas pair is a borderline Group II system because only one set of direct measurements of $\mathcal{D}_{12}$ is available, obtained by the two-bulb method (Paul and Srivastava, 1961 c).

H_2- Ar. There are many independent determinations of $\mathcal{D}_{12}$ for H_2- Ar, but with a number of significant discrepancies, see Figs. 32 and 33. The most reliable results are probably by Westenberg and Frazier (1962). The usually reliable closed-tube measurements by Ivakin and Suetin (1964 b) seem to give too steep a temperature dependence for $\mathcal{D}_{12}$. The only direct molecular-beam measurement for H_2- Ar gives a potential which is suspected to be too great (Colgate *et al*., 1969). Thus the molecular-beam potential for H_2- Ar was obtained by application of the combination rules, see Table 21.

H_2- Kr. The gas pair H_2- Kr was placed in Group II on the basis of the measurements by Fedorov *et al*. (1966) and Annis *et al*. (1968). The temperature dependence of the thermal diffusion factor (Annis *et al*., 1968) was used to extend the correlation temperature range from room temperature down to 77°K. Near the temperature limits of the reference equation the data may not be as reliable as the Group II uncertainty limits, thus H_2- Kr is considered a borderline system.

H_2- D_2. Most of the $\mathcal{D}_{12}$ for H_2- D_2 have been calculated from viscosity measurements of the H_2 isotopes or mixtures thereof, see Figs. 36 and 37. At low temperatures the viscosity-derived $\mathcal{D}_{12}$ are considered more reliable than the direct measurements. None of the direct measurements of $\mathcal{D}_{12}$ were actually used for curve-fitting purposes. In the least-squares calculations the value of *s* of Eq. (4.3-1) turned out to be 1.4883 which was rounded to 1.500 to agree with the theoretical lower limit for the rigid-sphere model, and the remaining correlation con-

stants were determined on the basis that $s = 1.500$.

H_2- CO_2. The most reliable results for this gas pair are closed-tube measurements by Loschmidt (1870 b), Boyd *et al.* (1951), and Ivakin and Suetin (1964 b). The results by Vyshenskaya and Kosov (1959) extend to 1083°K, but these have not been used to establish the upper temperature limit of the reference equation because the data were not considered sufficiently reliable, see Fig. 41. The viscosity-derived $\mathcal{D}_{12}$ reported by Weissman (1964) were normalized to the datum by Boyd *et al.* (1951) because the points seemed systematically high by about 5%.

N_2- Ar. The N_2- Ar gas pair is in Group II on the basis of the measurements of Paul and Srivastava (1961 b) and one point-source measurement of Westenberg and Walker (1957); however, these results cover only a small temperature range, so that this is a borderline system.

N_2- CO. On the basis of two-bulb measurements by Winn (1950) and closed-tube measurements by Amdur and Shuler (1963), the gas pair N_2- CO was placed in Group II. Since N_2 and CO are isosteric molecules, the N_2- N_2, CO - CO, and N_2- CO data could all be used for $\mathcal{D}_{12}$. Determinations of $\mathcal{D}_{12}$ from molecular-beam measurements have been published by Belyaev and Leonas (1966) and Amdur and Mason (1958) for N_2- N_2, and are in excellent agreement with the present calculations, see Fig. 44. The viscosity-derived $\mathcal{D}_{12}$ reported by Weissman and Mason (1962 b) seemed systematically low, thus the published results were normalized to the datum at 319°K by Amdur and Shuler (1963).

N_2- CO_2. This gas pair is a Group II system on the basis of the measurements by Boyd *et al.* (1951), Walker and Westenberg (1958 a), Walker *et al.* (1960), and Pakurar and Ferron (1964,1966). The results of Vyshenskaya and Kosov (1959) drop well below the recommended $\mathcal{D}_{12}$ at high temperatures; a similar trend was evident for H_2- CO_2.

c. Group III (Deviation Plots, Figs. 47 to 75)

Weights and Potentials. The equimolar values of $\mathcal{D}_{12}$ and their weights used in the least-squares calculations are presented in Table 22. The potential functions obtained from molecular-beam measurements are summarized in Table 23. Since there were no molecular-beam measurements available for systems with air or CO_2, no potentials are listed for the systems air - (Ar, CH_4, CO, SF_6) and CO_2 - (Ar, CO, O_2, air, N_2O, SF_6). The potentials for most of the Group III gas pairs required the application of the combination rules. At present, Leonas et al. have not performed beam experiments with CH_4 and SF_6, but both of these gases have been used in experiments by Amdur et al. Thus there are no independent confirmations of the potentials of gas pairs containing CH_4 or SF_6. The directly measured potential for He - CH_4 was used to derive a potential for H_2- CH_4; but the CH_4- Ar potential was used for the heavier gas pairs with methane; CH_4 - (N_2,O_2,SF_6). If the He - CH_4 potential had been used for CH_4- N_2, etc., inconsistent results would have been obtained. Apparently the small helium atom "sees" some of the structure of the CH_4 molecule, and the He - CH_4 potential is not generally suitable for combination-rule calculations based on the assumption of spherically-symmetric potentials (Mason and Amdur, 1964). The combination-rule potentials by Amdur et al. for H_2- O_2, CH_4- O_2, CO - Kr, CO - O_2, and N_2- O_2 were obtained from two different apparatuses. Such results from "mixed" apparatus are reliable over a smaller temperature range than potentials obtained from the same equipment.

Special Comments. For the thirty-two gas pairs of Group III the special comments are as follows.

Ar - CH_4. The closed-tube measurements by Arnold and Toor (1967) were considered sufficiently reliable to establish Ar - CH_4 as a Group III system.

Ar - CO. The consistency of the closed-tube measurement by Ivakin and Suetin (1964 a) and $\mathscr{D}_{12}$ by molecular-beam measurements with the results for Ar - N_2 (Group II) were the bases for placing Ar - CO into Group III.

Ar - O_2. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$ available, obtained by the two-bulb method (Paul and Srivastava, 1961 a).

Ar - air. There are no direct measurements available, and the results were calculated by Blanc's law.

Ar - CO_2. The more reliable measurements for Ar - CO_2 are by Ivakin and Suetin (1964 b) and by Pakurar and Ferron (1964,1966). The results by Pakurar and Ferron appear to have an unusual amount of scatter, which is due to difficult point-source measurements at temperatures above 1000°K.

Ar - SF_6. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the closed-tube method (Ivakin and Suetin, 1964 b).

H_2- Xe. The two-bulb measurements by Paul and Srivastava (1961 c) and the $\mathscr{D}_{12}$ from mixture viscosity data calculated by Weissman and Mason (1962 b) are considered equally reliable.

H_2- CH_4. The closed-tube measurements by Boyd et al. (1951) and by Arnold and Toor (1967) agree within about 1%. These results are at room temperature; values of $\mathscr{D}_{12}$ at temperatures up to 523°K were obtained from mixture viscosity data by Weissman and Mason (1962 b).

H_2- O_2. The reference equation essentially splits the difference between the high-temperature results by Walker and Westenberg (1960) and by Weissman and Mason (1962 b). For this gas pair the usually reliable point-source results by Walker and Westenberg are considered possibly somewhat high. This conclusion is based on a comparison with the results of H_2- N_2 (Group I), which are expected to be similar. Spontaneous ignition occurred at about 920°K in the point-source measurements.

H_2- SF_6. The most reliable measurement for this gas pair is by Boyd et al. (1951); other reliable determinations by the closed-tube method are by Strehlow (1953) and by Ivakin and Suetin (1964 a,b).

CH_4- He. The reliable direct measurements of $\mathscr{D}_{12}$ are primarily from recent open-tube studies by Frost (1967) and by Rhodes and Amick (1967).

CH_4- N_2. The two-bulb measurements by Mueller and Cahill (1964) were considered sufficient to place this gas pair into Group III.

CH_4- O_2. The only direct measurements available are those of Walker and Westenberg (1960) by the point-source technique. Spontaneous ignition occurred at about 1020°K. The results are not inconsistent with those of the similar system CH_4- N_2.

CH_4- air. There are no direct measurements available, and the results were calculated by Blanc's law.

CH_4- SF_6. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the closed-tube method (Manner, 1967).

N_2- Ne. This gas pair has reliable values of $\mathscr{D}_{12}$ only from mixture viscosity measurements by DiPippo et al. (1967).

N_2- Kr. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the two-bulb method (Durbin and Kobayashi, 1962).

N_2- Xe. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the two-bulb method (Paul and Srivastava, 1961 b).

N_2- O_2. The most reliable results are the closed-tube measurements by Lonius (1909) and the values calculated from mixture viscosity by

Weissman and Mason (1962 b). The results reported by Giddings and Seager (1962) are omitted from the deviation plot, Fig. 63, because of difficulties with the mixture composition analysis.

$\underline{N_2 - SF_6}$. The most reliable direct measurements are by Ivakin and Suetin (1964 b), obtained by the closed-tube method.

CO - Kr. This gas pair has only one set of direct measurements of $\mathcal{D}_{12}$, obtained by the two-bulb method (Singh et al., 1967). Since CO and N_2 are isosteric molecules, the reference equation for N_2- Kr was used for CO - Kr, and the data agree, see Fig. 65.

$\underline{CO - O_2}$. Since CO and N_2 are isosteric molecules, the reference equation for $N_2 - O_2$ was used for $CO - O_2$. The most reliable results are considered to be from mixture viscosity (Weissman and Mason, 1962 b). However, this judgment implies that the usually more reliable measurements by Loschmidt (1870 b) and by Walker and Westenberg (1960) are somewhat high.

CO - air. There are no direct measurements available, and the results were calculated by Blanc's law.

$\underline{CO - CO_2}$. Since CO and N_2 are isosteric molecules, the reference equation for $N_2- CO_2$ (Group II) can be used for $CO - CO_2$; a slightly more precise correlation, however, is given for $CO - CO_2$ based only on its direct measurements. The direct measurements for $CO - CO_2$ are in the temperature range of 282° to 473°K. The use of the $N_2- CO_2$ reference equation will extend the higher temperature limit to 1800°K, which is a significant advantage.

$\underline{CO - SF_6}$. This gas pair has direct measurements of $\mathcal{D}_{12}$ which were obtained by the closed-tube method (Ivakin and Suetin, 1964 a,b).

$\underline{O_2 - CO_2}$. The reference equation is based primarily on results of point-source measurements (Walker and Westenberg, 1960); at room temperature the results by closed-tube studies give slightly higher values of $\mathcal{D}_{12}$ (Loschmidt, 1870 b; Wretschko, 1870).

O_2- SF_6. This gas pair has reliable closed-tube measurements by Ivakin *et al*. (1968), which, however, probably have somewhat too great a temperature dependence for $\mathcal{D}_{12}$ over 297 to 408°K.

CO_2- air. Even though there are many direct measurements available, the reference equation for CO_2- air was calculated from Blanc's law. Of the direct measurements, the most reliable are considered to be closed-tube measurements by Loschmidt (1870 a,b), by Coward and Georgeson (1937), and by Holsen and Strunk (1964). The open-tube measurements by Klibanova *et al*. (1942), which cover the temperature range of 290 to 1533°K, are not considered as reliable as results by Blanc's law with data from the point-source method measurements by Walker (1958) and by Pakurar and Ferron (1964, 1966) for N_2- CO_2, and by Walker and Westenberg (1960) for O_2- CO_2.

CO_2- N_2O. This gas pair has several closed-tube measurements which agree within about 2% at room temperature (Loschmidt, 1870 b; Boardman and Wild, 1937; Wall and Kidder, 1946; Amdur *et al*. , 1952); values of $\mathcal{D}_{12}$ from mixture viscosity were used to extend the temperature range to 550°K. The placement of CO_2- N_2O into Group III, and not Group II, was decided upon because of the limited temperature range of the direct measurements and the uncertainties in $\mathcal{D}_{12}$ obtained from mixture viscosity for cylindrical molecules.

CO_2- SF_6. This gas pair has reliable closed-tube measurements by Ivakin and Suetin (1964 b).

SF_6- He. This gas pair has a few direct measurements, of which those by Ivakin and Suetin (1964 b) and by Fedorov *et al*. (1966) are considered to be the most reliable.

<u>SF_6-air</u>. There are no direct measurements available, and the results were calculated from Blanc's law.

d. Miscellaneous (Figs. 76 to 81)

<u>Weights and Potentials</u>. The values of $\mathcal{D}_{12}$ and their weights used in the least-squares calculations are presented in Table 24. Except for mixtures with dissociated gases the values of $\mathcal{D}_{12}$ are at equimolar composition. The data for mixtures with dissociated gases were not corrected to equimolar composition because the experimental uncertainties are greater than the composition dependence of $\mathcal{D}_{12}$. The potential functions obtained from molecular-beam measurements are summarized in Table 25, which are available only for the mixtures with dissociated gases and not the other systems of the miscellaneous group. This information may be helpful for the prediction of $\mathcal{D}_{12}$ at high temperatures, but the potentials were not used to calculate points for deviation plots as was done for the other gas pairs in Groups I, II, and III. There are no deviation plots for mixtures with dissociated gases because of the large uncertainties in the data available.

<u>Special Comments</u>. For the fourteen gas pairs of the miscellaneous group the special comments are as follows.

<u>H_2O-N_2</u>. This gas pair has direct measurements obtained only by the evaporation-tube method. The most reliable results are considered to be by O'Connell <u>et</u> <u>al</u>. (1969), in which the H_2O diffused downwards through N_2. In the other studies the H_2O was located below the N_2, or the lighter component below the heavier, which would have possible adverse effects due to convection. Because values of $\mathcal{D}_{12}$ are available only from evaporation-tube studies, it may be noted that the temperature range is limited, 282° to 373°K.

$\underline{H_2O-O_2}$. The correlation consists of two power functions ($\mathscr{D}_{12} = AT^s$) each applicable over a specific temperature interval, and which pass through a common datum. The generation of this type of correlation is explained as follows. At high temperatures, 390° to 1070°K, the point-source measurements are considered to be reliable (Walker and Westenberg, 1960); however at low temperatures, 308° to 352°K, the H_2O-O_2 evaporation-tube studies are considered too uncertain. But, at low temperatures, data by O'Connell _et al_. (1969) for H_2O-N_2 were systematically increased by 1% to obtain values of $\mathscr{D}_{12}$ for H_2O-O_2. This slight adjustment was based on calculations for transport properties of polar-gas mixtures.[3] But all results for H_2O-O_2 could not be well correlated by a single equation, either in the form of a Sutherland equation, Eq. (4.3-2), or the more complex correlation function of Eq. (4.3-1). The simplest reliable curve-fit of the data is two power functions, one each for the low- and high-temperature ranges. The equation at high temperatures was obtained by least-squares calculations of the point-source measurements. This equation was forced to pass through a point at the intermediate temperature of 450°K, obtained from an extrapolation of the low-temperature equation.

$\underline{H_2O\text{-air}}$. The reference equation for this air-system is the only one in this report which was not determined by Blanc's law. The reason is that the available data for H_2O-N_2 and H_2O-O_2 have too much scatter, and a verification of a correlation by Blanc's law from direct measurements for H_2O-air would not be significant. The reference equation for H_2O-air is based on the synthesis of results by O'Connell _et al_. (1969) for H_2O-N_2 and by Walker and Westenberg (1960) for H_2O-O_2. Since air is approximately 80% N_2, the reference equation for H_2O-N_2 extrapolated to 450°K, was assumed directly applicable at low temperatures. For temperatures between 450° and 1070°K, the reference equation for H_2O-O_2 was systematically reduced 1%. The more reliable direct measurements are shown in Fig. 78. The large deviations are due to expected uncertainties in results obtained by evaporation-tube studies. There are many other experimental determinations for H_2O-air, which are listed in Table 16.

<u>H_2O-CO_2</u>. This gas pair has reliable direct measurements by Ferron (1967), obtained by the point-source method over the temperature range of 1058 to 1640°K, and evaporation-tube studies at about 310 to 350°K, obtained by Schwertz and Brow (1951) and by Crider (1956). In the least-squares calculations the value of <u>s</u> of Eq. (4.3-2) turned out to be 1.473 which was adjusted to 1.500 to agree with the theoretical lower limit for the rigid-sphere model.

<u>CO_2-Ne</u>. This gas pair has limited data, consisting of a two-bulb measurement at about room temperature plus information on the temperature dependence of the thermal diffusion factor which was used to calculate $\mathcal{D}_{12}$ between 195° to 625°K.

<u>$CO_2-C_3H_8$</u>. The closed-tube measurements by Wall and Kidder (1946) and the values of $\mathcal{D}_{12}$ from mixture viscosity by Weissman (1964) are considered sufficient to include this system.

Mixtures with dissociated gases were all correlated by power functions of the form $\mathcal{D}_{12} = AT^s$. The reference equations were calculated from two points, one at about room temperature, and the other at temperatures greater than 1000°K. Deviation plots are not given for mixtures with dissociated gases. The special comments emphasize the discrepancies among the various values of $\mathcal{D}_{12}$ obtained by different experiments.

<u>H-He</u>. This gas pair has only one direct measurement by Khouw <u>et</u> <u>al</u>. (1969), obtained at 275°K. The values of $\mathcal{D}_{12}$ obtained from molecular-beam measurements by Amdur and Mason (1956) and by Belyaev and Leonas

(1967 b,c) were essentially averaged; the difference in $\mathcal{D}_{12}$ between these results is approximately 25% at all temperatures.

H-Ar. The direct measurements by Wise (1959) and by Khouw et al. (1969), near room temperature, differ by about 10%. At elevated temperatures, values of $\mathcal{D}_{12}$ are available only from one potential, (Mason and Vanderslice, 1958), and these results when compared with the reference equation are high by about 25% at 2000°K and low by about 20% at 10,000°K.

$H-H_2$. For this gas pair the most reliable values of $\mathcal{D}_{12}$ are considered to be from mixture viscosity (Browning and Fox, 1964). The other determinations of $\mathcal{D}_{12}$ are considered to be less reliable (Wise, 1961; Weissman and Mason, 1962; Khouw et al. 1969; Sancier and Wise, 1969). The relative measurements by Wise (1961) are at temperatures from 293 to 719°K. At room temperature, the discrepancies are within ±10%. The recommended values of $\mathcal{D}_{12}$ above 1000°K are based on molecular-beam measurements. These results were derived from measurements by Amdur et al. and use of the combination rules, which were considered to be much more reliable than the direct beam measurement by Belyaev and Leonas (1967 b,c). The latter yield $\mathcal{D}_{12}$ which are too high in comparison with both the low-temperature data, and results of other beam measurements.

$N-N_2$, $O-N_2$, $O-O_2$. For these gas pairs, with similar diffusion characteristics, the results by Morgan and Schiff (1964) are considered to be the most reliable. For $O-O_2$, at about room temperature, the measurements by Krongelb and Strandberg (1959) and by Walker (1961) are within 10% (below) those by Morgan and Schiff; the measurements by Yolles and Wise (1969) and by Yolles et al. (1970) are about 20% below those of Morgan and Schiff. The results by Walker may be low due to the neglect of chemical reaction effects. At temperatures between 1000° and 10,000°K the differences between $\mathcal{D}_{12}$ from molecular-beam measurements for

$N-N_2$, $O-N_2$, $O-O_2$ (Belyaev and Leonas, 1966 c) were so small that these results were grouped together.

O-He, O-Ar. There are two direct measurements for each of these gas pairs. In comparison with the results by Morgan and Schiff (1964), which are considered the more reliable, the results by Yolles and Wise (1968) are low by about 35% for O-He, and high by about 30% for O-Ar. The values of $\mathcal{D}_{12}$ at elevated temperatures were based on a single laboratory source for the potentials (Leonas et al.).

REFERENCES

[1] Mason, E. A., I. Amdur, and I. Oppenheim, J. Chem. Phys. 43, 4458 (1965)

[2] Diller, D. E. and E. A. Mason, J. Chem. Phys. 44, 2604 (1966)

[3] Mason, E. A. and L. Monchick, J. Chem. Phys. 36, 2746 (1962)

Table 1. Quantum Effects on Diffusion Coefficients and on Collision Integral Ratio A_{12}^* in Terms of the deBoer Parameter, Λ^* , and the Reduced Temperature, $T^* \equiv kT/E$.[a]

	$[\mathscr{D}_{12}]_1$(QUANTAL)/$[\mathscr{D}_{12}]_1$(CLASSICAL)						A_{12}^*(QUANTAL)/A_{12}^*(CLASSICAL)				
T^* \ Λ^*	0	0.5	1.0	1.5	2.0	3.0	0.5	1.0	1.5	2.0	3.0
0.1	0	1.001	1.001	0.641	0.741	2.924	1.014	1.043	0.970	0.921	0.966
0.2	0	1.004	0.899	0.813	1.046	2.494	1.010	1.042	0.979	0.956	1.046
0.5	0	1.002	0.991	1.069	1.297	1.883	1.007	0.994	0.957	0.956	1.021
1.0	0	1.009	1.032	1.105	1.224	1.444	0.999	0.984	0.971	0.978	1.018
1.5	0	1.008	1.031	1.080	1.150	1.269	0.999	0.990	0.987	0.994	1.025
2.0	0	1.006	1.025	1.060	1.105	1.182	1.000	0.994	0.996	1.003	1.028
3.0	0	1.004	1.016	1.035	1.060	1.101	1.001	0.997	1.002	1.008	1.027

[a] Calculated from a Lennard-Jones (12-6) potential.[43]

Table 2. Classification of Experimental Methods

Name	Primary Investigator(s)	Reliability
	Major	
Closed Tube	Loschmidt (1870 a,b)	Good
Evaporation Tube	Stefan (1873)	Poor
Two-Bulb Apparatus	Ney and Armistead (1947)	Good
Point Source	Walker and Westenberg (1958)	Average
Gas Chromatography	Giddings and Seager (1960)[a]	Average
	Minor	
Open Tube	von Obermayer (1882); Waitz (1882)	Average
Back Diffusion	Harteck and Schmidt (1933)	Average
Capillary Leak	Klibanova _et_ _al_. (1942)	Poor
Unsteady Evaporation	Arnold (1944)	Fair
Diffusion Bridge	Bendt (1958)	Average
Dissociated Gases	Wise (1959); Krongelb and Strandberg (1959)	Poor
	Miscellaneous	
Droplet Evaporation	Langmuir (1918); Katan (1969)	?
Dufour Effect	Waldmann (1944)	?
Thermal Separation Rate	Nettley (1954)	?
Kirkendall Effect	McCarty and Mason (1960)	?
Sound Absorption	Holmes and Tempest (1960)	?
Cataphoresis	Hogervorst and Freudenthal (1967)	?
Resonance Methods	See text	?

Table 2. (Continued)

[a] In 1960 four independent gas chromatography studies were submitted for publication; for details see Section 3.2, part d.

Table 3. Determinations of $\mathcal{D}_{12}$ by the Closed-Tube Method.[a]

Author(s)	Date	Author(s)	Date
Loschmidt	1870 a,b	Strehlow	1953
Wretschko	1870	Bunde	1955
von Obermayer	1880,'82 b, '83,'87	Rumpel	1955
Rutherford and Brooks	1901	Amdur and Schatzki	1957,'58
Schmidt	1904	Suetin _et al_.	1960
Lonius	1909	Suetin and Ivakin	1961
Wintergerst	1930	Amdur and Shuler	1963
Harteck and Schmidt	1933	Holsen and Strunk	1964
Boardman and Wild	1937	Ivakin and Suetin	1964 a,b
Coward and Georgeson	1937	Suetin	1964
Hirst and Harrison	1939	Amdur and Beatty	1965
Braune and Zehle	1941	Amdur and Malinauskas	1965
Groth and Harteck	1941	Cordes and Kerl	1965
Heath _et al_.	1941	Ljunggren	1965
Groth and Sussner	1944	Reichenbacher _et al_.	1965
Wall and Kidder	1946	Fedorov _et al_.	1966
Hutchinson	1947	Kosov and Abdullina	1966
Boyd _et al_.	1951	Arnold and Toor	1967
Timmerhaus and Drickamer	1951	Manner	1967
Amdur _et al_.	1952	Ivakin _et al_.	1968

[a] Complete reference information is given in Bibliography I.

Table 4. Determinations of $\mathscr{D}_{12}$ by the Two-Bulb Method.[a]

Author(s)	Date	Author(s)	Date
Ney and Armistead	1947	Srivastava and Srivastava	1962
Winn and Ney	1947	Srivastava	1962
Winn	1948	Golubev and Bondarenko	1963
Hutchinson	1949	Schäfer and Reinhard	1963
Winn	1950	Srivastava and Srivastava	1963
Schäfer *et al*.	1951	Wendt *et al*.	1963
Visner	1951 a,b	Bondarenko and Golubev	1964
Winter	1951	Mason *et al*.	1964 a,b
DeLuca	1954	Miller and Carman	1964
Schäfer and Moesta	1954	Mueller and Cahill	1964
Andrew	1955	Watts	1964
Schäfer and Schuhmann	1957	Brown and Murphy	1965
Saxena and Mason	1959	Malinauskas	1965
Schäfer	1959	Mason *et al*.	1965
Srivastava and Srivastava	1959	Watts	1965
Srivastava	1959	Chakraborti and Gray	1966
Srivastava and Barua	1959	Kosov and Novosad	1966 a
Miller and Carman	1961	Malinauskas	1966
Paul and Srivastava	1961 a,b,c	Paul and Watson	1966
Weissman *et al*.	1961	Saran and Singh	1966
Zmbov and Knežević	1961	Srivastava and Saran	1966 a,b
Durbin and Kobayashi	1962	van Heijningen *et al*.	1966
Paul	1962	Vučić and Milojević	1966
Srivastava and Paul	1962	Singh *et al*.	1967

Table 4. (Continued)

Author(s)	Date
Annis *et al*.	1968
Malinauskas	1968
Mathur and Saxena	1968
Singh and Srivastava	1968
van Heijningen *et al*.	1968
Annis *et al*.	1969
DuBro	1969
Malinauskas and Silverman	1969
Vugts *et al*.	1969
Weissman	1969
Humphreys and Mason	1970

[a] Complete reference information is given in Bibliography I.

Table 5. Determinations of $\mathcal{D}_{12}$ by the Point-Source Method.[a]

Author(s)	Date
Westenberg and Walker[b]	1957
Walker	1958
Walker and Westenberg	1958 a,b,'59,'60
Walker *et al*.	1960
Westenberg and Frazier	1962
Ember *et al*.	1962,'64
Pakurar and Ferron	1964,'65,'66
Walker and Westenberg	1966
Ferron	1967
Walker and Westenberg	1968

[a] Complete reference information is given in Bibliography I.

[b] Preliminary note about the development of the point-source method.

Table 6. Determinations of $\mathcal{D}_{12}$ by the Gas-Chromatography Method.[a]

Author(s)	Date
Giddings and Seager	1960
Bohemen and Purnell	1961
Bournia *et al.*	1961
Fejes and Czárán	1961
Giddings and Seager	1962
Knox and McLaren	1963
Seager *et al.*	1963
Barr and Sawyer	1964
Knox and McLaren	1964
Evans and Kenney	1965
Fuller and Giddings	1965
Huber and van Vught	1965
Chang	1966
Arnikar, *et al.*[b]	1967 a,b
Fuller and Giddings	1967
Giddings	1967
Hargrove and Sawyer	1967
Giddings	1968
Zhukhovitskiĭ *et al.*	1968
Arnikar and Ghule	1969
Fuller *et al.*	1969
Wasik and McCulloh	1969

[a] Complete reference information is given in Bibliography I.

[b] Packed chromatography column.

Table 7. Determinations of $\mathcal{D}_{12}$ by the Evaporation-Tube Method[a]

Author(s)	Date	Author(s)	Date
Stefan	1873	Brookfield *et al*.	1947
Baumgartner	1877 a,b	Klotz and Miller	1947
Guglielmo	1881,'82	Goryunova and Kuvshinski	1948
Winkelmann	1884 a,b,c '85,'88, '89	Gush	1948
		McMurtie and Keyes	1948
Stefan	1889,'90	Hippenmeyer	1949
Gribojedoff	1893	Schwertz and Brow	1951
Houdaille	1896	Cvetanović and LeRoy	1952
Naccari	1909	Kimpton and Wall	1952
Mache	1910	Schlinger *et al*.	1952
Naccari	1910	Cummings and Ubbelohde	1953
Vaillant	1911	Rossié	1953
Pochettino	1914	Lee and Wilke	1954
LeBlanc and Wuppermann	1916	Bose and Chakraborty	1955
Mack	1925	Carmichael *et al*.	1955 a,b
Summerhays	1930	Cummins and Ubbelohde	1955
Trautz and Ludwig	1930	Cummings *et al*.	1955
Trautz and Ries	1931	Narsimhan	1955
Ackermann	1934	Raw	1955
Gilliland	1934	Crider	1956
Trautz and Müller	1935	Call	1957
Chambers and Sherwood	1937	Clarke and Ubbelohde	1957
Schirmer	1938	Richardson	1959

Table 7 (Continued)

Author(s)	Date
Altshuller and Cohen	1960
Hudson *et al.*	1960
Jorgeson and Watts	1961
Reamer and Sage	1963
Grieveson and Turkdogan	1964
Heinzelmann *et al.*	1965
Kohn and Romero	1965
Stevenson	1965
Brockett	1966
Mehta	1966
Ben-Aim *et al.*	1967
Byrne *et al.*	1967
Galloway and Sage	1967
Getzinger and Wilke	1967
Krol *et al.*	1967
Mikhailov and Kochegarova	1967
Nafikov and Usmanov	1967
Pryde and Pryde	1967
Yuan and Cheng	1967
Khomchenkov *et al.*	1968
Lugg	1968
Mrazek *et al.*	1968
O'Connell *et al.*	1968
Spencer *et al.*	1969

[a] Complete reference information is given in Bibliography I.

Table 8. Determinations of $\mathscr{D}_{12}$ by Minor Experimental Methods.[a]

Author(s)	Date
a. Open Tube	
von Obermayer	1882 a
Waitz	1882
von Obermayer	1883, '87
Toepler	1896
Foch	1913
Barus	1924 a,b
Currie	1960
Frost	1967
Kaufmann	1967
Rhodes and Amick	1967
Zhukhovitskiĭ *et al.*	1968
b. Back Diffusion	
Harteck and Schmidt	1933
Van der Held and Miesowicz	1937
Spier	1939, '40
c. Capillary Leak	
Klibanova *et al.*	1942
Kosov	1957
Vyshenskaya and Kosov	1959, '65
Kosov and Karpushin	1966
De Paz *et al.*	1967
d. Unsteady Evaporation	
Mullaly and Jacques	1924
Mackenzie and Melville	1932, '33
Arnold	1944
Fairbanks and Wilke	1950
Nelson	1956
de Nordwall and Flowers	1958
Nikolaev and Aleskovskiĭ	1964
Petit	1965
Grob and El-Wakil	1969
e. Diffusion Bridge	
Buckingham	1904
Wicke and Kallenbach	1941
Weisz	1957
Bendt[b]	1958
Scott and Cox	1960
Evans *et al.*	1961
Wicke and Hugo	1961
Scott and Dullien	1962
Evans *et al.*	1962, '63
Coates and Mian	1967
Henry *et al.*	1967

Table 8. (Continued)

Author(s)	Date
Mian	1967
Reist	1967
Zhalgasov and Kosov[b]	1968
Ellis and Holsen	1969
Hawtin *et al.*	1969
Schneider and Schäfer	1969
f. Dissociated Gases	
Krongelb and Strandberg	1959
Wise	1959, '61
Walker	1961
Young	1961
Morgan and Schiff	1964
Yolles and Wise	1968
Khouw *et al.*	1969
Sancier and Wise	1969
Yolles *et al.*	1970

[a] Complete reference information is given in Bibliography I.

[b] The investigations by Bendt involved an apparatus with a capillary, and by Zhalgasov and Kosov a bundle of capillaries; all other diffusion-bridge apparatuses used porous septums.

Table 9. Determinations of $\mathcal{D}_{12}$ by Miscellaneous Experimental Methods.[a]

Author(s)	Date
a. Droplet Evaporation	
Langmuir	1918
Topley and Whytlaw-Gray	1927
Houghton	1933
Bradley *et al*.	1946
Bradley and Shellard	1949
Birks and Bradley	1949
Bradley	1951
Bradley and Waghorn	1951
Katan[b]	1969
b. Dufour Effect	
Waldmann	1944,'47
Mason *et al*.	1967
c. Thermal Separation Rate	
Nettley	1954
van Itterbeek and Nihoul	1957
Lonsdale and Mason	1957
Saxena and Mason	1959
Weissman *et al*.	1961
Wendt *et al*.	1963
d. Kirkendall Effect	
McCarty and Mason	1960
Mason	1961
e. Sound Absorption	
Holmes and Tempest	1960
Carey *et al*.	1966
Carey *et al*.	1968
f. Cataphoresis	
Freudenthal	1966
Hogervorst and Freudenthal	1967
g. Resonance Methods	
(Nuclear Magnetic Resonance)	
Luszczynski *et al*.	1962
Lipsicas	1962
Hartland and Lipsicas	1963
Luszczynski *et al*.	1967
(Optical Pumping)[c]	
Franzen	1959
Bernheim	1962
McNeal	1962
Anderson and Ramsey	1963
Legowski	1964
Ramsey and Anderson	1964
Bernheim and Korte	1965

Table 9. (Continued)

Author(s)	Date
Gozzini *et al*.	1967
(Mercury Band Fluorescence)	
Coulliette	1928
Biondi	1953
McCoubrey	1954
McCoubrey and Matland	1954
Matland and McCoubrey	1955
McCoubrey and Matland	1956

[a] Complete reference information is given in Bibliography I.

[b] This is a clever combination of a uniform-pressure experiment with a porous membrane and the evaporation-tube method; the motion of an evaporating liquid bead in a tube is used to determine the diffusion coefficient.

[c] A recent review article has been published by Violino (1968).

Table 10. Grouping of Systems According to Uncertainty Limits of $\mathcal{D}_{12}$.

Group I	He-(Ne,Ar,Kr,Xe)
	Ne-(He,Ar,Kr,Xe)
	Ar-(He,Ne,Kr,Xe)
	Kr-(He,Ne,Ar,Xe?)
	H_2- N_2?
Group II	^{3}He - ^{4}He
	He-(H_2,N_2,CO,O_2?,air,CO_2)
	H_2-(He,Ne?,Ar,Kr?,D_2,CO,air,CO_2)
	N_2-(Ar?,CO,CO_2)
Group III	Ar-CH_4,CO,O_2,air,CO_2,SF_6)
	H_2-(Xe,CH_4,O_2,SF_6)
	CH_4-(He,Ar,H_2,N_2,O_2,air,SF_6)
	N_2-(Ne,Kr,Xe,CH_4,O_2,SF_6)
	CO-(Ar,Kr,O_2,air,CO_2,SF_6)
	O_2-(Ar,H_2,CH_4,N_2,CO,CO_2,SF_6)
	CO_2-(Ar,CO,O_2,air,N_2O,SF_6)
	SF_6-(He,Ar,H_2,CH_4,N_2,CO,O_2,air,CO_2)
Miscellaneous Systems	H_2O-(N_2,O_2,air,CO_2)
	CO_2-(Ne,H_2O,C_3H_8)
	H-(He,Ar,H_2)
	N-N_2
	O-(He,Ar,N_2,O_2)

Table 11. Uncertainty Limits for Systems of the Miscellaneous Group.

System	T, °K	Uncertainty, ±%
H_2O-N_2	282 to 373	4
H_2O-O_2	282 to 1070	7
H_2O-air	282 to 1070	5 to 10
H_2O-CO_2	296 to 1640	10 to 7
CO_2-Ne	195 to 625	3 to 5
$CO_2-C_3H_8$	298 to 550	3 to 5
$H-H_2$	∿300	5
	>1,000	30
$H-N_2,O-N_2,O-O_2$	∿300	10
	>1,000	25
H-He,H-Ar, O-He,O-Ar	∿300	15
	>1,000	30

Table 12. Correlation Parameters of Eq. (4.3-1) for $\mathcal{D}_{12}$.

System	$10^3 A$	s	$10^{-8}\frac{\varphi_0}{k}$	S	S'	T	Group
	$\frac{\text{atm-cm}^2}{\text{sec}(°\text{K})^s}$		°K	°K	$(°\text{K})^2$	°K	
$^3He-^4He$	32.4	1.501	0.0448	-0.9630	1.894	$1.74-10^4$	II
$^3He-^4He$	0.156	1.636	-	-	-	14.4 -90.0	-
He-Ne	25.41	1.509	0.212	1.87	-	$65-10^4$	I
He-Ar	15.21	1.552	0.410	1.71	-	$77-10^4$	I
He-Kr	10.61	1.609	1.42	-32.65	2036.	$77-10^4$	I
He-Xe	7.981	1.644	4.02	-68.87	5416.	$169-10^4$	I
$He-H_2$	27.0	1.510	0.0534	-	-	$90-10^4$	II
$He-N_2$	15.8	1.524	0.265	-	-	$77-10^4$	II
He-CO	15.8	1.524	0.265	-	-	$77-10^4$	II
Ne-Ar	8.779	1.546	1.94	1.82	1170.	$90-10^4$	I
Ne-Kr	8.520	1.555	6.73	20.4	-	$112-10^4$	I
Ne-Xe	6.747	1.584	19.0	10.1	-	$169-10^4$	I
Ar-Kr	5.346	1.556	13.0	47.3	-	$169-10^4$	I
Ar-Xe	5.000	1.563	36.8	59.9	-	$169-10^4$	I
$Ar-H_2$	23.5	1.519	0.488	39.8	-	$242-10^4$	II
Kr-Xe	2.933	1.608	128	52.7	-	$169-10^4$	I
$Kr-H_2$	18.2	1.564	1.69	26.4	-	$77-10^4$	II
H_2-D_2	24.7	1.500	0.0636	6.072	38.10	$14-10^4$	II
H_2-N_2	15.39	1.548	0.316	-2.80	1067.	$65-10^4$	I
H_2-CO	15.39	1.548	0.316	-2.80	1067.	$65-10^4$	II
N_2-CO	4.40	1.576	1.57	-36.2	3825.	$78-10^4$	II

Table 13. Correlation Parameters of Eq. (4.3-2) for $\mathcal{D}_{12}$.

System	$10^5 A$ $\frac{\text{atm-cm}^2}{\text{sec}(°\text{K})^s}$	s	S °K	T °K	Group
$He-CH_4$	3.13	1.750	-	$298-10^4$	III
$He-O_2$	4.37	1.710	-	$244-10^4$	II
He-air	3.78	1.729	-	$244-10^4$	II
$He-CO_2$	3.31	1.720	-	200-530	II
$He-SF_6$	3.87	1.627	-	$290-10^4$	III
$Ne-H_2$	5.95	1.731	-	$90-10^4$	II
$Ne-N_2$	1.59	1.743	-	$293-10^4$	III
$Ne-CO_2$	1.07	1.776	-	195-625	misc.
$Ar-CH_4$	0.784	1.785	-	$307-10^4$	III
$Ar-N_2$	0.904	1.752	-	$244-10^4$	II
Ar-CO	0.904	1.752	-	$244-10^4$	III
$Ar-O_2$	0.977	1.736	-	$243-10^4$	III
Ar-air	0.917	1.749	-	$244-10^4$	III
$Ar-CO_2$	1.74	1.646	89.1	276-1800	III
$Ar-SF_6$	1.48	1.596	145.4	$328-10^4$	III
$Kr-N_2$	0.653	1.766	-	$248-10^4$	III
Kr-CO	0.653	1.766	-	$248-10^4$	III
$Xe-H_2$	3.68	1.712	16.9	$242-10^4$	III
$Xe-N_2$	0.470	1.789	-	$242-10^4$	III
H_2-CH_4	3.13	1.765	-	$293-10^4$	III
H_2-O_2	4.17	1.732	-	$252-10^4$	III

Table 13. (Continued)

System	10^5A $\frac{\text{atm-cm}^2}{\text{sec}(°K)^s}$	s	S °K	T °K	Group
H_2-air	3.64	1.750	-	252-10^4	II
H_2-CO_2	3.14	1.750	11.7	200-550	II
H_2-SF_6	7.82	1.570	102.3	298-10^4	III
CH_4-N_2	1.00	1.750	-	298-10^4	III
CH_4-O_2	1.68	1.695	44.2	294-10^4	III
CH_4-air	1.03	1.747	-	298-10^4	III
CH_4-SF_6	1.10	1.657	69.2	298-10^4	III
N_2-O_2	1.13	1.724	-	285-10^4	III
N_2-H_2O	0.187	2.072	-	282-373	misc.
N_2-CO_2	3.15	1.570	113.6	288-1800	II
N_2-SF_6	1.66	1.590	119.4	328-10^4	III
CO-O_2	1.13	1.724	-	285-10^4	III
CO-air	1.12	1.730	-	285-10^4	III
CO-CO_2	0.577	1.803	-	282-473	III
CO-SF_6	1.76	1.584	139.4	297-10^4	III
O_2-H_2O	0.189	2.072	-	282-450	misc.
	2.78	1.632	-	450-1070	misc.
O_2-CO_2	1.56	1.661	61.3	287-1083	III
O_2-SF_6	2.65	1.522	129.0	297-10^4	III
air-H_2O	0.187	2.072	-	282-450	misc.
	2.75	1.632	-	450-1070	misc.
air-CO_2	2.70	1.590	102.1	280-1800	III
air-SF_6	1.83	1.576	121.1	328-10^4	III

Table 13. (Continued)

System	10^5A $\frac{\text{atm-cm}^2}{\text{sec}(°\text{K})^s}$	s	S °K	T °K	Group
H_2O-CO_2	9.24	1.500	307.9	296-1640	misc.
CO_2-N_2O	0.281	1.866	-	195-550	III
$CO_2-C_3H_8$	0.177	1.896	-	298-550	misc.
CO_2-SF_6	0.140	1.886	-	328-472	III
H-He	14.2	1.732	-	$275-10^4$	misc.
H-Ar	1.45	1.597	-	$275-10^4$	misc.
$H-H_2$	11.3	1.728	-	$190-10^4$	misc.
$N-N_2$	1.32	1.774	-	$280-10^4$	misc.
O-He	4.68	1.749	-	$280-10^4$	misc.
O-Ar	0.751	1.841	-	$280-10^4$	misc.
$O-N_2$	1.32	1.774	-	$280-10^4$	misc.
$O-O_2$	1.32	1.774	-	$280-10^4$	misc.

Table 14. Classical Low-Temperature Asymptotic Values of $\mathscr{D}_{12}$, Eq. (4.3-3).

System	$10^6 A$ $\frac{\text{atm-cm}^2}{\text{sec}(°\text{K})^{11/6}}$	C [a] $e^2 a_o^5$	Λ^* [b]	ε/k [b] °K
He-Ne	31.2	3.0	1.32	23.7
He-Ar	20.3	9.6	0.86	40.2
He-Kr	17.9	13	0.80	39.0
He-Xe	15.6	19	0.68	46.5
He-CH_4	19.0	14	0.89	37
He-N_2	20.4	10	0.96	31
Ne-Ar	8.26	20	0.35	61.7
Ne-Kr	6.79	27	0.28	69.8
Ne-Xe	5.84	38	0.26	69.1
Ne-H_2	30.1	8.2	1.35	34
Ne-N_2	8.69	21	0.37	57
Ar-Kr	3.51	91	0.14	145
Ar-Xe	2.93	130	0.11	178
Ar-H_2	19.5	28	0.87	64
Ar-CH_4	5.27	98	0.22	130
Ar-N_2	4.93	69	0.21	107
Kr-Xe	2.00	190	0.08	197
Kr-H_2	17.1	40	0.75	80
Kr-N_2	3.91	96	0.16	132

Table 14. (Continued)

System	$10^6 A$ $\frac{\text{atm-cm}^2}{\text{sec}(°\text{K})^{11/6}}$	C [a] $e^2 a_o^5$	Λ^* [b]	ε/k [b] °K
$Xe-H_2$	15.1	58	0.67	87
$Xe-N_2$	3.29	140	0.14	145
H_2-CH_4	17.5	43	0.82	68
H_2-N_2	19.3	30	0.87	62.9
CH_4-N_2	5.54	100	0.23	120

[a] Dalgarno (1967).

[b] Based on the 12-6 potential; parameters for noble gas pairs and for N_2-N_2 from van Heijningen et al. (1966,1968), and for other gas pairs from Hirschfelder et al. (1954).

Table 15. Correlation Parameters for the Composition Dependence of $\mathcal{D}_{12}$ According to Eq. (4.2-1).

System	ζ	ε/k [a] °K	a	b	System	ζ	ε/k [a] °K	a	b
^{3}He-^{4}He	1.0	10.2	0.031	0.26	Ar-Kr	1.4	145	0.051	0.30
He-Ne	1.64	23.7	0.098	0.45	Ar-Xe	1.8	178	0.086	0.57
He-Ar	1.67	40.2	0.18	1.17	Ar-H_2	1.73	64	0.17	0.85
He-Kr	1.65	39.0	0.23	1.56	Ar-D_2	1.73	64	0.14	0.74
He-Xe	1.78	46.5	0.29	2.08	Ar-CH_4	1.0	130	0.046	0.02
He-H_2	1.0	18.4	0.033	-0.11	Ar-N_2	1.0	107	0.029	0.10
He-CH_4	1.0	37	0.14	0.25	Ar-CO	1.0	117	0.029	0.10
He-N_2	1.80	31	0.17	1.22	Ar-O_2	1.0	118	0.026	0.15
He-CO	1.0	34	0.16	1.19	Ar-air	1.0	109	0.029	0.11
He-O_2	1.0	34	0.17	1.11	Ar-SF_6	1.0	179	0.12	1.07
He-air	1.0	31	0.17	1.19	Kr-Xe	1.8	197	0.039	0.33
He-CO_2	1.0	44	0.23	1.74	Kr-H_2	1.0	80	0.21	1.14
He-SF_6	1.0	51	0.39	3.09	Kr-D_2	1.0	80	0.19	1.07
Ne-Ar	1.2	61.7	0.059	0.57	Kr-N_2	1.0	132	0.066	0.28
Ne-Kr	1.01	69.8	0.12	0.87	Kr-CO	1.0	145	0.066	0.28
Ne-Xe	1.25	69.1	0.17	1.31	Xe-H_2	1.0	87	0.25	1.53
Ne-H_2	1.0	34	0.10	0.26	Xe-D_2	1.0	87	0.23	1.43
Ne-D_2	1.0	34	0.078	0.16	Xe-N_2	1.0	145	0.10	0.56
Ne-N_2	1.0	57	0.043	0.65	H_2-D_2	1.0	33	0.042	0.12
Ne-CO_2	1.0	82	0.081	0.98	H_2-CH_4	1.0	68	0.15	0.94

Table 15. (Continued)

System	ζ	ε/k [a] °K	a	b	System	ζ	ε/k [a] °K	a	b
H_2-N_2	1.00	62.9	0.17	0.89	N_2-H_2O	1.0	266	0.020	-0.32
H_2-CO	1.0	61	0.16	0.88	N_2-CO_2	1.0	132	0.041	0.38
H_2-O_2	1.0	61	0.16	0.81	N_2-SF_6	1.0	154	0.14	1.04
H_2-air	1.0	57	0.16	0.87	CO-CO_2	1.0	145	0.041	0.38
H_2-CO_2	1.84	80	0.21	1.33	CO-SF_6	1.0	169	0.14	1.06
H_2-SF_6	1.0	93	0.33	2.33	O_2-H_2O	1.0	296	0.033	-0.03
D_2-CH_4	1.0	68	0.11	0.81	O_2-CO_2	1.0	147	0.037	0.44
D_2-N_2	1.00	62.9	0.13	0.76	O_2-SF_6	1.0	171	0.14	1.14
D_2-CO	1.0	61	0.13	0.74	air-H_2O	1.0	274	0.020	-0.34
D_2-O_2	1.0	61	0.13	0.66	air-CO_2	1.0	136	0.040	0.39
D_2-air	1.0	57	0.13	0.74	air-SF_6	1.0	159	0.14	1.06
D_2-CO_2	1.84	80	0.18	1.20	H_2O-CO_2	1.0	384	0.060	0.34
D_2-SF_6	1.0	93	0.31	2.26	CO_2-SF_6	1.0	222	0.088	0.60
CH_4-N_2	1.0	120	0.035	0.05					
CH_4-O_2	1.0	124	0.038	0.00					
CH_4-air	1.0	120	0.035	0.05					
CH_4-SF_6	1.0	188	0.12	0.50					

[a] Based on the 12-6 potential; parameters for noble-gas pairs and for H_2-N_2 from van Heijningen et al.(1966,1968), and for others from Hirschfelder et al. (1954).

Table 16. Experimental Determinations of $\mathscr{D}_{12}$ According to Gas Pair, Temperature Noted.

The order of listing in Table 16 is as follows: (i) mixtures of noble gases with noble gases arranged according to the atomic weight of the lighter component, (ii) mixtures of noble gases with other gases arranged according to the atomic weight of the noble gas component, (iii) dissociated gases, and (iv) other mixtures arranged according to the molecular weight of the lighter component.

a. Noble Gases

System	Reference	T, °K
^3He - ^3He	Luszczynski *et al*. (1962)	1.7 to 4.2
	Luszczynski *et al*. (1967)	1.13 to 4.22
^3He - ^4He	Bendt (1958)	1.74 to 296
	Weissman and Mason (1962 b)	2.64 to 4.25
	DuBro (1969)	77 to 344
^4He - ^4He	Amdur and Mason (1958)	1000 to 15,000
He - Ne	Srivastava and Barua (1959)	273 to 318
	Holmes and Tempest (1960)	298
	Weissman and Mason (1962 b)	20 to 523
	Weissman (1965)	291 and 302
	DiPippo *et al*. (1967)	293 and 303

Table 16. (Continued)

System	Reference	T, °K
	Malinauskas (1968)	273 to 394
	van Heijningen *et al*. (1968)	65 to 295
	DuBro (1969)	77 to 364
	Malinauskas and Silverman (1969)	273 to 394
He - Ar	Schmidt (1904)	286 to 292
	Lonius (1909)	286 to 295
	Strehlow (1953)	288 to 418
	Schäfer and Moesta (1954)	200 to 400
	Amdur and Mason (1958)	1000 to 15,000
	Walker (1958)	298
	Saxena and Mason (1959)	251 to 418
	Srivastava (1959)	273 to 318
	Walker and Westenberg (1959)	298 to 1063
	Holmes and Tempest (1960)	298
	Evans *et al*. (1961)	293
	Mason (1961)	303
	Suetin and Ivakin (1961)	287

Table 16. (Continued)

System	Reference	T, °K
	Evans *et al*. (1962)	298 and 373
	Giddings and Seager (1962)	298
	Weissman and Mason (1962 b)	72 to 473
	Evans *et al*. (1963)	298
	Golubev and Bondarenko (1963)	298 and 363
	Seager *et al*. (1963)	298 to 498
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 465
	Suetin (1964)	287
	Ljunggren (1965)	293
	Malinauskas (1965)	273 to 394
	Weissman (1965)	291 to 311
	Carey *et al*. (1966)	300
	Fedorov *et al*. (1966)	291
	Kosov and Karpushin (1966)	169 to 296
	Kosov and Karpushin (1966 a)	293
	Malinauskas (1966)	273 to 394
	Mason and Smith (1966)	334

Table 16. (Continued)

System	Reference	T,°K
	Coates and Mian (1967)	298 to 522
	DiPippo *et al*. (1967)	293 and 303
	Mian (1967)	298 to 522
	Carey *et al*. (1968)	300, 1255 to 4990
	Mathur and Saxena (1968)	270 to 350
	van Heijningen *et al*. (1968)	90 to 400
	Annis *et al*. (1969)	295
	DuBro (1969)	305 and 335
	Hawtin *et al*. (1969)	293 to 873
	Kalelkar and Kestin (1969)	298 to 993
	Schneider and Schäfer (1969)	273 to 1300
	Wasik and McCulloh (1969)	77 to 357
He - Kr	Srivastava and Barua (1959)	273 to 318
	Holmes and Tempest (1960)	298
	Durbin and Kobayashi (1962)	308
	Srivastava and Paul (1962)	305
	Weissman and Mason (1962 b)	291
	Watts (1964)	303

Table 16. (Continued)

System	Reference	T, °K
	Weissman (1965)	291 and 302
	Fedorov *et al*. (1966)	290
	Kestin *et al*. (1966)	293 and 303
	Malinauskas (1966)	273 to 394
	Mason and Smith (1966)	318
	Annis *et al*. (1968)	77 to 760
	van Heijningen *et al*. (1968)	112 to 400
	Kalelkar and Kestin (1969)	298 to 993
	Wasik and McCulloh (1969)	298 to 366
He - Xe	Srivastava (1959)	273 to 318
	Weissman and Mason (1962 b)	291 to 550
	Malinauskas (1965)	273 to 394
	Watts (1965)	303
	Weissman (1965)	291 to 311
	van Heijningen *et al*. (1968)	169 to 400
He - Rn	Hirst and Harrison (1939)	283 - 286
Ne - Ne	Groth and Sussner (1944)	293
	Winn (1950)	78 to 353

Table 16. (Continued)

System	Reference	T, °K
	Amdur and Mason (1958)	1000 to 15,000
Ne - Ar	Schäfer and Schuhmann (1957)	90 to 473
	Srivastava and Srivastava (1959)	273 to 318
	Weissman and Mason (1962 b)	72 to 523
	Weissman (1965)	291 to 311
	Freudenthal (1966)	300 to 600
	DiPippo *et al*. (1967)	293 and 303
	Hogervorst and Freudenthal (1967)	300 to 650
	Malinauskas (1968)	273 to 394
	van Heijningen *et al*. (1968)	90 to 400
	Malinauskas and Silverman (1969)	273 to 394
Ne - Kr	Srivastava and Srivastava (1959)	273 to 318
	Paul (1962)	302
	Weissman and Mason (1962 b)	291
	Watts (1964)	303
	Weissman (1965)	291 to 311
	Malinauskas (1968)	273 to 394
	Mathur and Saxena (1968)	270 to 350

Table 16. (Continued)

System	Reference	T, °K
	van Heijningen *et al*. (1968)	112 to 400
	Malinauskas and Silverman (1969)	273 to 394
Ne - Xe	Srivastava and Barua (1959)	273 to 318
	Weissman and Mason (1962 b)	291
	Watts (1965)	303
	Weissman (1965)	291 and 302
	Malinauskas (1968)	273 to 394
	van Heijningen *et al*. (1968)	169 to 400
	Weissman (1968 b)	328 to 873
	DuBro (1969)	304
	Malinauskas and Silverman (1969)	273 to 394
	Taylor *et al*. (1969)	328 to 873
	Weissman (1969)	305 to 925
Ne - Rn	Hirst and Harrison (1939)	290 and 293
Ar - Ar	Hutchinson (1947)	295
	Hutchinson (1949)	90 to 327
	Winn (1950)	78 to 353
	Amdur and Schatzki (1957)	273

Table 16. (Continued)

System	Reference	T, °K
	Amdur and Mason (1958)	1000 to 15,000
	De Paz *et al*. (1967)	76 to 294
	Vugts *et al*. (1969)	235 to 418
Ar - Kr	Schäfer and Schuhmann (1957)	200 to 473
	Srivastava and Srivastava (1959)	273 to 318
	Durbin and Kobayashi (1962)	248 and 308
	Paul (1962)	302
	Weissman and Mason (1962 b)	291
	Watts (1964)	303
	Weissman (1965)	291 to 311
	Fedorov *et al*. (1966)	291
	Malinauskas (1966)	273 to 394
	Ivakin *et al*. (1968)	297 to 407
	van Heijningen *et al*. (1968)	169 to 400
	Humphreys and Mason (1970)	77 to 600
Ar - Xe	Amdur and Schatzki (1957)	195 to 378
	Amdur and Schatzki (1958)	330
	Srivastava (1959)	273 to 318

Table 16. (Continued)

System	Reference	T,°K
	Weissman and Mason (1962 b)	291
	Malinauskas (1965)	273 to 394
	Watts (1965)	303
	Weissman (1965)	291 to 311
	van Heijningen *et al*. (1968)	169 to 400
Ar - Rn	Hirst and Harrison (1939)	282 and 286
Kr - Kr	Groth and Harteck (1941)	294 and 296
	Schäfer and Schuhmann (1957)	199 to 474
	Amdur and Mason (1958)	1000 to 15,000
	Miller and Carman (1961)	293
	Durbin and Kobayashi (1962)	308
	Paul (1962)	302
	Srivastava and Paul (1962)	305
	Wendt *et al*. (1963)	232 to 470
	Miller and Carman (1964)	293
	Watts (1964, 1965)	303
	Kamnev and Leonas (1966)	2000 to 10,000
	Saran and Singh (1966)	303
	Annis *et al*. (1969)	295

Table 16. (Continued)

System	Reference	T,°K
	DuBro (1969)	305 and 367
Kr - Xe	Weissman and Mason (1962 b)	291
	Watts (1964, 1965)	303
	Weissman (1965)	291 and 302
	Malinauskas (1966)	273 to 394
	van Heijningen _et_ _al_. (1968)	169 to 400
Xe - Xe	Groth and Harteck (1941)	292
	Visner (1951 a,b)	300
	Amdur and Schatzki (1957)	195 to 378
	Amdur and Mason (1958)	1000 to 15,000
	Watts (1965)	303
	Kamnev and Leonas (1966)	2000 to 10,000

Table 16. (Continued)

b. Noble Gases and Another Component

System	Reference	T,°K
He - H_2	Bunde (1955)	298
	Rumpel (1955)	298 to 358
	van Itterbeek and Nihoul (1957)	52 to 153
	Suetin _et_ _al_. (1960)	273
	Suetin and Ivakin (1961)	292
	Giddings and Seager (1962)	298
	Suetin (1964)	294
	Weissman and Mason (1962 b)	90 to 523
	Amdur and Malinauskas (1965)	195 to 374
	Giddings (1968)	298
	Kestin and Yata (1968)	293 and 303
He - TH	Amdur and Malinauskas (1965)	195 to 374
He - D_2	Ivakin and Suetin (1964 a)	295
He - T_2	Amdur and Malinauskas (1965)	195 to 374
He - CH_4	Carswell and Stryland (1963)	298
	Fuller and Giddings (1965)	373
	Frost (1967)	303 to 764

Table 16. (Continued)

System	Reference	T, °K
	Rhodes and Amick (1967)	302 to 627
He - NH_3	Giddings and Seager (1962)	297
	Srivastava (1962)	274 to 333
	Ivakin and Suetin (1964 a)	297
He - H_2O	Schwertz and Brow (1951)	307 to 352
	Lee and Wilke (1954)	298
He - Na	von Hartel *et al.* (1932)	655
	Anderson and Ramsey (1963)	427
	Ramsey and Anderson (1964)	428
	Gozzini *et al.* (1967)	273, 443
	Violino (1968)	427 to 443
He - C_2H_2	Suetin and Ivakin (1961)	290
	Suetin (1964)	290
He - C_2H_4	Frost (1967)	303 to 765
He - N_2	Rumpel (1955)	298 to 358
	Westenberg and Walker (1957)	293
	Walker (1958)	297 to 1124
	Walker and Westenberg (1958 a,b)	298 to 1200

Table 16. (Continued)

System	Reference	T, °K
	Paul and Srivastava (1961 b)	243 to 333
	Suetin and Ivakin (1961)	289
	Giddings and Seager (1962)	296 to 304
	Seager *et al*. (1963)	298 to 498
	Ivakin and Suetin (1964 b)	289 to 470
	Suetin (1964)	289
	Chang (1966)	244 to 311
	Kestin *et al*. (1966)	293 and 303
	Walker and Westenberg (1966)	297 to 1124
	Coates and Mian (1967)	299 to 500
	Frost (1967)	303
	Henry *et al*. (1967)	299
	Mian (1967)	299 to 500
	Zhukhovitskiĭ *et al*. (1968)	293
	Ellis and Holsen (1969)	297 to 882
	Hawtin *et al*. (1969)	293 to 873
	Wasik and McCulloh (1969)	77 to 370
He - CO	Ivakin and Suetin (1964 a)	296
	Ivakin and Suetin (1964 b)	296 to 470

Table 16. (Continued)

System	Reference	T, °K
He - C_2H_6	Frost (1967)	303 to 751
	Kaufmann (1967)	373 to 523
He - O_2	Paul and Srivastava (1961 a)	244 to 334
	Suetin and Ivakin (1961)	287
	Giddings and Seager (1962)	297
	Seager *et al*. (1963)	298 to 498
	Suetin (1964)	287
	Kestin and Yata (1968)	293 and 303
	Wasik and McCulloh (1969)	298 to 365
He - air	Suetin *et al*. (1960)	273
	Suetin and Ivakin (1961)	287
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 469
	Suetin (1964)	287
	Fedorov *et al*. (1966)	292
	Evans *et al*. (1969)	295
He - CH_3OH	Seager *et al*. (1963)	423 to 523

Table 16. (Continued)

System	Reference	T, °K
He - K	Bernheim and Korte (1965)	358
	Khomchenkov *et al*. (1968)	723
	Violino (1968)	358
	Ivanovskiĭ *et al*. (1969)	570 to 930
He - C_3H_6	Weissman (1964)	293 to 523
	Frost (1967)	303 to 764
He - CO_2	Lonsdale and Mason (1957)	260 to 358
	Saxena and Mason (1959)	250 to 404
	McCarty and Mason (1960)	303
	Suetin *et al*. (1960)	273
	Walker *et al*. (1960)	299
	Suetin and Ivakin (1961)	287
	Giddings and Seager (1962)	300
	Seager *et al*. (1963)	298 to 498
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 465
	Suetin (1964)	287
	Kosov and Novosad (1966 a)	294

Table 16. (Continued)

System	Reference	T, °K
	DiPippo *et al*. (1967)	293 and 303
	Kosov and Bogatyrev (1968)	290 to 430
	Annis *et al*. (1969)	295
He - C_3H_8	Frost (1967)	303 to 767
	Kaufmann (1967)	373 to 503
	Rhodes and Amick (1967)	303
He - C_2H_5OH	Lee and Wilke (1954)	298
	Seager *et al*. (1963)	423 to 523
	Hargrove and Sawyer (1967)	298
He - difluoromethane	Fuller *et al*. (1969)	431
He - 1-butene	Frost (1967)	303 to 522
He - 2-butene	Weissman (1964)	293 to 523
He - C_4H_{10}	Frost (1967)	303 to 751
	Rhodes and Amick (1967)	303 to 477
	Hargrove and Sawyer (1967)	298 to 473
He - acetone	Hargrove and Sawyer (1967)	298 to 473
He - 1-propanol	Seager et al. (1963)	423 to 523
He - 2-propanol	Seager et al. (1963)	423 to 523

Table 16. (Continued)

System	Reference	T, °K
He - 1, 1-difluoroethane	Fuller et al. (1969)	430
He - n-pentane	Hargrove and Sawyer (1967)	298 to 473
He - ether	Hargrove and Sawyer (1967)	298 to 473
He - 1-butanol	Seager et al. (1963)	423 to 523
He - benzene	Lee and Wilke (1954)	298
	Seager et al. (1963)	423 to 523
	Hargrove and Sawyer (1967)	298 to 473
He - 1-chloropropane	Fuller et al. (1969)	428
He - dichloromethane	Fuller et al. (1969)	428
He - Rb	Bernheim (1962)	323
	Violino (1968)	323 and 340
He - 3-pentanone	Barr and Sawyer (1964)	300
He - n-hexane	Hargrove and Sawyer (1967)	298 to 473
	Fuller and Giddings (1965)	417
He - 1-pentanol	Seager et al. (1963)	423 to 523
He - 1-chlorobutane	Fuller et al. (1969)	429
He - 2-chlorobutane	Fuller et al. (1969)	429
He - fluorobenzene	Fuller et al. (1969)	430

Table 16. (Continued)

System	Reference	T, °K
He - 1, 2-dichloroethane	Fuller *et al*. (1969)	427
He - n-heptane	Clarke and Ubbelohde (1957)	303
He - 2, 4-dimethylpentane	Clarke and Ubbelohde (1957)	303
He - 1-hexanol	Seager *et al*. (1963)	423 to 523
He - 1-fluorohexane	Fuller *et al*. (1969)	432
He - 1-chloropentane	Fuller *et al*. (1969)	428
He - bromoethane	Fuller *et al*. (1969)	428
He - 4-fluorotoluene	Fuller *et al*. (1969)	432
He - chlorobenzene	Fuller *et al*. (1969)	431
He - n-octane	Clarke and Ubbelohde (1957)	303
He - 2, 2,4-trimethylpentane	Clarke and Ubbelohde (1957)	303
He - trichloromethane	Fuller *et al*. (1969)	429
He - 1-bromopropane	Fuller *et al*. (1969)	428
He - 2-bromopropane	Fuller *et al*. (1969)	428
He - nitrobenzene	Lee and Wilke (1954)	298
He - Cs	Legowski (1964)	299
	Khomchenkov *et al*. (1968)	723

Table 16. (Continued)

System	Reference	T, °K
	Violino (1968)	299
He - 1-bromobutane	Fuller *et* *al*. (1969)	427
He - 2-bromobutane	Fuller *et* *al*. (1969)	427
He - iodomethane	Fuller *et* *al*. (1969)	431
He - SF_6	Suetin and Ivakin (1961)	291
	Suetin (1964)	291
	Ivakin and Suetin (1964 b)	291 to 464
	Evans and Kenney (1965)	293
	Fedorov *et* *al*. (1966)	291
He - iodoethane	Fuller *et* *al*. (1969)	428
He - bromobenzene	Fuller *et* *al*. (1969)	427
He - 2-bromo-1-chloropropane	Fuller *et* *al*. (1969)	427
He - 1-bromohexane	Fuller *et* *al*. (1969)	428
He - 2-bromohexane	Fuller *et* *al*. (1969)	428
He - 3-bromohexane	Fuller *et* *al*. (1969)	429
He - 1-iodopropane	Fuller *et* *al*. (1969)	430
He - 2-iodopropane	Fuller et al. (1969)	430

Table 16. (Continued)

System	Reference	T,°K
He - dibromomethane	Fuller *et al*. (1969)	428
He - 1-iodobutane	Fuller *et al*. (1969)	428
He - 2-iodobutane	Fuller *et al*. (1969)	427
He - hexafluorobenzene	Fuller *et al*. (1969)	429
He - UF_6	Ljunggren (1965)	293
He - As_4	Krol *et al*. (1967)	733 to 913
Ne - H_2	Paul and Srivastava (1961)	242 to 341
	Weissman and Mason (1962 b)	90 to 523
Ne - D_2	Weissman and Mason (1962 b)	293
Ne - NH_3	Srivastava (1962)	274 to 333
Ne - Na	Anderson and Ramsey (1963)	425
	Violino (1968)	425
Ne - N_2	DiPippo *et al*. (1967)	293 and 303
Ne - CO_2	Weissman *et al*. (1961)	242 to 427
	Breetveld *et al*. (1966, 1967)	293 and 303
	Annis *et al*. (1969)	295
Ne - Rb	Franzen (1959)	320
	Violino (1968)	340

Table 16. (Continued)

System	Reference	T, °K
Ne - Cs	Legowski (1964)	299
	Violino (1968)	317
Ne - UF_6	Ljunggren (1965)	293
Ar - H_2	Waldmann (1944, 1947)	293
	Strehlow (1953)	288 to 418
	Paul and Srivastava (1961 c)	242 to 341
	Suetin and Ivakin (1961)	291
	Westenberg and Frazier (1962)	295 to 1069
	Weissman and Mason (1962 b)	293 to 523
	Golubev and Bondarenko (1963)	298 to 363
	Ivakin and Suetin (1964 b)	291 to 473
	Mason *et al*. (1964)	294
	Suetin (1964)	291
	Cordes and Kerl (1965)	296
	Evans and Kenney (1965)	293
	Arnold and Toor (1967)	307
	Mason *et al*. (1967)	296
	Annis *et al*. (1969)	295

Table 16. (Continued)

System	Reference	T, °K
Ar - D_2	Ivakin and Suetin (1964 a)	297
Ar - T_2	Mason *et al*. (1964 a)	295
	Annis *et al*. (1969)	295
Ar - CH_4	Carswell and Stryland (1963)	298
	Arnold and Toor (1967)	307
Ar - NH_3	Ivakin and Suetin (1964 a)	295
	Srivastava and Srivastava (1962)	255 to 333
	DiPippo *et al*. (1967)	293 and 303
Ar - H_2O	O'Connell *et al*. (1969)	282 to 353
Ar - Na	von Hartel et al. (1932)	654
	Violino (1968)	(?)
Ar - acetylene	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
	Ivakin *et al*. (1968)	298 to 407
Ar - ethylene	Weissman (1964)	298
Ar - N_2	Waldmann (1944, 1947)	293
	Schäfer and Moesta (1954)	233 to 363
	Westenberg and Walker (1957)	293

Table 16. (Continued)

System	Reference	T, °K
	Paul and Srivastava (1961 b)	244 to 335
	DiPippo *et al*. (1967)	293 and 303
Ar - CO	Ivakin and Suetin (1964 a)	296
Ar - O_2	Waldmann (1944, 1947)	293
	Paul and Srivastava (1961 a)	243 to 334
	Scott and Dullien (1962)	293
Ar - air	Evans *et al*. (1969)	295
Ar - K	Khomchenckov *et al*. (1968)	723
	Ivanovskiĭ *et al*. (1969)	630 to 950
Ar - CO_2	Waldmann (1944, 1947)	293
	Suetin and Ivakin (1961)	289
	Ivakin and Suetin (1964 b)	289 to 473
	Holsen and Strunk (1964)	276 and 317
	Suetin (1964)	289
	Pakurar and Ferron (1964)	295 and 1181 to 1676
	Kestin *et al*. (1966)	293 and 303
	Kosov and Novosad (1966 a)	294
	Pakurar and Ferron (1966)	1132 to 1798

Table 16. (Continued)

System	Reference	T, °K
	Ferron (1967)	1100 to 1800
	Gurvich and Matizen (1968)	308
Ar - Cr	Grieveson and Turkdogan (1964)	1600
Ar - Fe	Grieveson and Turkdogan (1964)	1600
Ar - acetone	Hargrove and Sawyer (1967)	298 to 473
Ar - n-butane	Hargrove and Sawyer (1967)	298 to 473
Ar - Ni	Grieveson and Turkdogan (1964)	1600
Ar - Co	Grieveson and Turkdogan (1964)	1600
Ar - nitromethane	Byrne et al. (1967)	303
Ar - SO_2	Schäfer (1959)	263
Ar - Zn	Nikolaev and Aleskovskiĭ (1964)	1100 to 2600
Ar - n-pentane	Hargrove and Sawyer (1967)	298 to 473
Ar - ether	Hargrove and Sawyer (1967)	298 to 473
Ar - benzene	Hargrove and Sawyer (1967)	298 to 473
Ar - HBr	Mian (1967)	328 to 523
Ar - cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
Ar - methylcyclopentane	Cummings and Ubbelohde (1953, 1955)	289
Ar - Rb	Franzen (1959)	320

Table 16. (Continued)

System	Reference	T,°K
	Violino (1968)	320 and 340
Ar - n-hexane	Cummings and Ubbelohde (1953, 1955)	289
	Hargrove and Sawyer (1967)	298 to 473
Ar - 2,3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	289
Ar - 3-pentanone	Barr and Sawyer (1964)	300
Ar - toluene	Fairbanks and Wilke (1950)	294
Ar - monofluorobenzene	Byrne *et al*. (1967)	303
Ar - 2,4-dimethylpentene	Clarke and Ubbelohde (1957)	303
Ar - n-heptane	Clarke and Ubbelohde (1957)	303
Ar - n-octane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
Ar - 2,2,4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
Ar - Cs	Legowski (1964)	299
	Khomchenkov *et al*. (1968)	723
	Violino (1968)	299 and 317
Ar - SF_6	Suetin and Ivakin (1961)	287
	Suetin (1964)	287

Table 16. (Continued)

System	Reference	T,°K
	Ivakin and Suetin (1964 b)	287 to 472
	Evans and Kenney (1965)	293
Ar - 1-bromo-3-methylbutane	Byrne *et al*. (1967)	303
Ar - Br_2	Mackenzie and Melville (1933)	289
Ar - Hg	Spencer *et al*. (1969)	459 to 607
Ar - UF_6	Ljunggren (1965)	293
Ar - As_4	Krol *et al*. (1967)	853 to 913
Kr - H_2	Miller and Carman (1961)	293
	Mason *et al*. (1964 b)	296
	Miller and Carman (1964)	293
	Fedorov *et al*. (1966)	291
	Annis *et al*. (1968)	77 to 760
	Annis *et al*. (1969)	295
Kr - D_2	Mason *et al*. (1964 b)	255 to 362
	Annis *et al*. (1969)	295
Kr - T_2	Mason *et al*. (1964 b)	252 to 346
	Annis *et al*. (1969)	295
Kr - NH_3	Srivastava and Srivastava (1962)	255 to 333

Table 16. (Continued)

System	Reference	T, °K
Kr - C_2H_4	Durbin and Kobayashi (1962)	298
Kr - N_2	Durbin and Kobayashi (1962)	248 and 308
Kr - CO	Singh _et_ _al_. (1967)	274 to 319
Kr - NO	Singh _et_ _al_. (1967)	274 to 318
Kr - O_2	Ivakin _et_ _al_. (1967)	298 to 408
Kr - air	Reist (1967)	273
Kr - CO_2	Durbin and Kobayashi (1962)	308
	Kestin and Yata (1968)	293 and 303
Kr - acetone	Srivastava and Saran (1966 a)	284 to 313
Kr - SO_2	Saran and Singh (1966)	303
	Srivastava and Saran (1966 b)	274 to 318
Kr - C_2H_5Cl	Singh and Srivastava (1968)	275 to 318
Kr - $(C_2H_5)_2O$	Srivastava and Saran (1966 b)	274 to 318
Kr - CH_2Cl_2	Singh and Srivastava (1968)	278 to 318
Kr - Rb	Franzen (1959)	320
	Violino (1968)	320
Kr - $CHCl_3$	Srivastava and Saran (1966 a)	284 to 313

Table 16. (Continued)

System	Reference	T, °K
Kr - Hg	Nakayama (1968)	301
Kr - UF_6	Ljunggren (1965)	293
Xe - H_2	Paul and Srivastava (1961 c)	242 to 341
	Weissman and Mason (1962 b)	293 to 550
	Miller and Carman (1964)	293
Xe - NH_3	Srivastava (1962)	274 to 331
Xe - N_2	Paul and Srivastava (1961 b)	242 to 334
Xe - O_2	Paul and Srivastava (1961 a)	242 to 334
Xe - Rb	Franzen (1959)	320
	Violino (1968)	320
Xe - Hg	Nakayama (1968)	301
Xe - UF_6	Ljunggren (1965)	293

The list of studies for Rn mixtures is not comprehensive, and references to other studies are given by Hirst and Harrison (1939) and by Raabe (1968).

System	Reference	T, °K
Rn - H_2	Hirst and Harrison (1939)	288
Rn - air	Rutherford and Brooks (1901)	room temperature
	Hirst and Harrison (1939)	288

Table 16. (Continued)

System	Reference	T, °K
	Korpusov et al. (1964)	(?)
	Vučić and Milojević (1966)	(?)
	Raabe (1968)	299

Table 16. (Continued)

c. Dissociated Gases

System	Reference	T, °K
H - He	Khouw *et al*. (1969)	275
H - Ar	Wise (1959)	293 (assumed)
	Khouw *et al*. (1969)	275
H - H_2	Wise (1959)	293 (assumed)
	Wise (1961)	293 to 719
	Weissman and Mason (1962 a)	200 to 1000
	Browning and Fox (1964)	190 to 373
	Khouw *et al*. (1969)	202 to 364
	Sancier and Wise (1969)	293 to 719
N - N_2	Young (1961)	298 (assumed)
	Morgan and Schiff (1964)	280
O - He	Morgan and Schiff (1964)	280
	Yolles and Wise (1968)	298
O - Ar	Morgan and Schiff (1964)	280
	Yolles and Wise (1968)	298
O - Kr	Yolles and Wise (1968)	298

Table 16. (Continued)

System	Reference	T, °K
O - N_2	Morgan and Schiff (1964)	280
O - O_2	Krongelb and Strandberg (1959)	300
	Walker (1961)	298
	Morgan and Schiff (1964)	280
	Yolles and Wise (1968)	298
	Yolles *et al*. (1970)	298 to 873

Table 16. (Continued)

d. Other Mixtures

System	Reference	T, °K
H_2 - H_2	Harteck and Schmidt (1933)	20 to 293
	Lipsicas (1962)	56 to 90
	Hartland and Lipsicas (1963)	20
	Amdur and Beatty (1965)	195 to 353
	Mason *et al.* (1965)	295
	Reichenbacher *et al.* (1965)	297
	Annis *et al.* (1969)	295
H_2 - HD	Weissman and Mason (1962 b)	72 to 293
H_2 - TH	Reichenbacher *et al.* (1965)	297
	Amdur and Beatty (1965)	195 and 273
H_2 - D_2	Heath *et al.* (1941)	288
	Waldmann (1944, 1947)	293
	Bendt (1958)	14 to 296
	McCarty and Mason (1960)	303
	Weissman and Mason (1962 b)	72 to 293
	Ivakin and Suetin (1964 a)	296

Table 16. (Continued)

System	Reference	T, °K
	Diller and Mason (1966)	14 to 293
H_2 - DT	Reichenbacher *et* *al*. (1965)	297
H_2 - T_2	Amdur and Beatty (1965)	195 to 353
	Mason *et* *al*. (1965)	295
	Reichenbacher *et* *al*. (1965)	297
H_2 - CH_4	von Obermayer *et* *al*. (1883)	273 to 289
	Boyd *et* *al*. (1951)	298
	Fejes and Czárán (1961)	298
	Weissman and Mason (1962 b)	293 to 523
	Arnold and Toor (1967)	307
	Mason *et* *al*. (1967)	296
H_2 - NH_3	Bunde (1955)	298 to 358
	Schäfer (1959)	240 to 403
	Scott and Cox (1960)	273 to 533
	Ivakin and Suetin (1964 a)	297
	Weissman (1964)	293 to 523
	Pal and Barua (1967)	306 to 479

Table 16. (Continued)

System	Reference	T, °K
H_2 - H_2O	Guglielmo (1882)	291
	Winkelmann (1884 a, b)	323 and 366
	Winkelmann (1889)	293 and 372
	Mache (1910)	300 to 366
	Trautz and Müller (1935)	293 to 372
	McMurtie and Keyes (1948)	303 to 333
	Hippenmeyer (1949)	283 to 368
	Schwertz and Brow (1951)	307 to 353
	Crider (1956)	307 and 329
	Nelson (1956)	298 to 328
H_2 - Na	von Hartel and Polanyi (1930)	633
	von Hartel *et al.* (1932)	655
	Ramsey and Anderson (1964)	473
	Violino (1968)	473
H_2 - C_2H_2	Weissman (1964)	293 to 373
H_2 - N_2	Lonius (1909)	285 - 287
	Boardman and Wild (1937)	288

Table 16. (Continued)

System	Reference	T, °K
	Waldmann (1944, 1947)	293
	Schäfer _et_ _al_. (1951)	193 to 336
	Nettley (1954)	288
	Schäfer and Moesta (1954)	200 to 400
	Bunde (1955)	298 to 358
	van Itterbeek and Nihoul (1957)	137 and 153
	Weisz (1957)	293 (?)
	Vyshenskaya and Kosov (1959)	293 to 1083
	Giddings and Seager (1960)	293
	Scott and Cox (1960)	294 to 573
	Bohemen and Purnell (1961)	324
	Fejes and Czárán (1961)	298
	Suetin and Ivakin (1961)	289
	Giddings and Seager (1962)	297
	Scott and Dullien (1962)	293
	Weissman and Mason (1962 b)	82 to 523
	Bondarenko and Golubev (1964)	273 to 473

Table 16. (Continued)

System	Reference	T, °K
	Ivakin and Suetin (1964 b)	289 to 471
	Suetin (1964)	289
	Cordes and Kerl (1965)	296
	Vyshenskaya and Kosov (1965)	293 to 1083
	van Heijningen *et al.* (1966)	65 to 295
	Pal and Barua (1967)	307 to 478
	Kestin and Yata (1968)	293 and 303
	Schneider and Schäfer (1969)	273 to 1300
H_2 - CO	Loschmidt (1870 b)	293
	von Obermayer (1883)	282 - 285
	Weissman and Mason (1962 b)	195 to 523
	Ivakin and Suetin (1964 a)	296
	Ivakin and Suetin (1964 b)	296 to 471
H_2 - C_2H_4	von Obermayer (1883)	287
	Weissman (1964)	195 to 523
H_2 - NO	Weissman (1964)	273
H_2 - C_2H_6	von Obermayer (1883)	288

Table 16. (Continued)

System	Reference	T, °K
	Boyd *et al.* (1951)	298
	Fejes and Czárán (1961)	298
	Weissman (1964)	293 to 523
H_2 - O_2	Loschmidt (1870 a)	252 to 286
	Loschmidt (1870 b)	252 to 289
	Wretschko (1870)	294 and 297
	von Obermayer (1880)	286 and 335
	von Obermayer (1883)	281 to 291
	Lonius (1909)	284 to 288
	van Itterbeek and Nihoul (1957)	142 and 153
	Walker and Westenberg (1960)	295 to 900
	Weissman and Mason (1962 b)	294 to 550
	Walker and Westenberg (1966)	295 to 901
	Zhalgasov and Kosov (1968)	103 to 298
H_2 - air	von Obermayer (1883)	281 to 284
	Barus (1924 b)	297
	Kosov (1957)	289

Table 16. (Continued)

System	Reference	T, °K
	Suetin *et al.* (1960)	273
	Currie (1960)	285 to 309
	Suetin and Ivakin (1961)	292
	Ivakin and Suetin (1964 a)	297
	Suetin (1964)	292
	Evans *et al.* (1969)	295
H_2 - CH_3OH	Winkelmann (1885)	299 and 323
H_2 - HCl	Weissman (1964)	293 to 523
H_2 - K	Ivanovskiĭ *et al.* (1969)	680 to 830
H_2 - C_3H_6	Weissman (1964)	293 to 523
H_2 - CO_2	Loschmidt (1870 a)	273 and 286
	Loschmidt (1870 b)	273 to 289
	Wretschko (1870)	297
	von Obermayer (1880)	285 and 335
	von Obermayer (1882 a)	284 to 293
	von Obermayer (1883)	280 to 294
	Schmidt (1904)	288

Table 16. (Continued)

System	Reference	T, °K
	Lonius (1909)	286 to 294
	Boardman and Wild (1937)	288
	Waldmann (1944, 1947)	293
	Boyd *et al*. (1951)	298
	Schäfer *et al*. (1951)	252 to 308
	Lonsdale and Mason (1957)	259 to 358
	Saxena and Mason (1959)	250 to 368
	Vyshenskaya and Kosov (1959)	293 to 1083
	McCarty and Mason (1960)	303
	Suetin *et al*. (1960)	273
	Miller and Carman (1961)	293
	Suetin and Ivakin (1961)	292
	Wicke and Hugo (1961)	295
	Giddings and Seager (1962)	300
	Bondarenko and Golubev (1964)	323 and 363
	Ivakin and Suetin (1964 b)	292 to 473
	Miller and Carman (1964)	293

Table 16. (Continued)

System	Reference	T, °K
	Suetin (1964)	292
	Weissman (1964)	298 to 550
	Vyshenskaya and Kosov (1965)	293 to 1083
	Mason _et_ _al_. (1967)	296
	Annis _et_ _al_. (1969)	295
	Schneider and Schäfer (1969)	273 to 990
H_2 - N_2O	von Obermayer (1883)	283
	Weissman (1964)	300 to 550
H_2 - C_3H_8	Fejes and Czárán (1961)	298
	Weissman (1964)	273 to 550
H_2 - formic acid	Winkelmann (1885)	339 and 358
H_2 - C_2H_5OH	Baumgartner (1877 a)	291
	Winkelmann (1884 a)	314 and 340
	Winkelmann (1885)	323 and 337
	Trautz and Müller (1935)	340
H_2 - 2-butene	Weissman (1964)	293 to 523
H_2 - acetone	Trautz and Müller (1935)	296

Table 16. (Continued)

System	Reference	T, °K
H_2 - n-butane	Strehlow (1953)	288 to 430
	Fejes and Czárán (1961)	298
H_2 - acetic acid	Winkelmann (1885)	339 to 372
H_2 - n-propyl alcohol	Winkelmann (1885)	340 and 357
H_2 - nitromethane	Byrne _et_ _al_. (1967)	303
H_2 - SO_2	Loschmidt (1870 b)	286
	Schäfer (1959)	263 to 473
	Weissman (1964)	290 to 472
H_2 - ethyl formate	Winkelmann (1884 c)	294 and 319
H_2 - methyl acetate	Winkelmann (1884 c)	294 and 319
H_2 - propionic acid	Winkelmann (1885)	366 and 372
H_2 - n-butyl alcohol	Winkelmann (1885)	372
H_2 - i-butyl alcohol	Winkelmann (1885)	340 and 357
H_2 - ethyl ether	Stefan (1873)	292
	Baumgartner (1877 a)	290
	Winkelmann (1884 a)	284 and 293
	Trautz and Müller (1935)	273 and 293

Table 16. (Continued)

System	Reference	T, °K
	Weissman (1964)	288 to 486
H_2 - CS_2	Baumgartner (1877 a)	290
	Baumgartner (1877 b)	268 to 311
H_2 - benzene	Trautz and Ludwig (1930)	296
	Trautz and Ries (1931)	296
	Trautz and Müller (1935)	296
	Hudson et al. (1960)	311
H_2 - pyridine	Hudson et al. (1960)	318
H_2 - 2:3-dimethylbuta-1:3-diene	Cummings et al. (1955)	288
H_2 - hexa-1:5-diene	Cummings et al. (1955)	288
H_2 - thiophen	Hudson et al. (1960)	302
H_2 - cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
	Hudson et al. (1960)	289
H_2 - 2:3-dimethylbut-2-ene	Cummings et al. (1955)	288
H_2 - methyl cyclopentane	Cummings and Ubbelohde (1953, 1955)	289
H_2 - piperidine	Hudson et al. (1960)	315
H_2 - Rb	McNeal (1962)	343

Table 16. (Continued)

System	Reference	T, °K
	Violino (1968)	343
H_2 - 2:3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	289
H_2 - n-hexane	Cummings and Ubbelohde (1953, 1955)	289
H_2 - n-butyric acid	Winkelmann (1885)	372
H_2 - i-butyric acid	Winkelmann (1885)	371
H_2 - ethyl acetate	Winkelmann (1884 c)	319
H_2 - methyl propionate	Winkelmann (1884 c)	319 and 340
H_2 - propyl formate	Winkelmann (1884 c)	319 and 340
H_2 - n-amyl alcohol	Winkelmann (1885)	372
H_2 - active amyl alcohol	Winkelmann (1885)	372
H_2 - tetrahydrothiophen	Hudson *et al.* (1960)	318
H_2 - toluene	Fairbanks and Wilke (1950)	301
H_2 - monofluorobenzene	Byrne *et al.* (1967)	303
H_2 - n-heptane	Cummings *et al.* (1955)	303
	Clarke and Ubbelohde (1957)	303
H_2 - 2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303

Table 16. (Continued)

System	Reference	T, °K
H_2 - triethylamine	Mehta (1966)	298
H_2 - ethyl propionate	Winkelmann (1884 c)	340 and 363
	Fairbanks and Wilke (1950)	301
H_2 - methyl butyrate	Winkelmann (1884 c)	340 and 365
H_2 - methyl i-butyrate	Winkelmann (1884 c)	323 and 340
H_2 - i-valeric acid	Winkelmann (1885)	372
H_2 - n-hexyl alcohol	Winkelmann (1885)	372
H_2 - n-octane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
H_2 - 2:2:4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
H_2 - i-butyl acetate	Winkelmann (1884 c)	340 and 371
H_2 - ethyl butyrate	Winkelmann (1884 c)	340 and 370
H_2 - ethyl i-butyrate	Winkelmann (1884 c)	340 and 369
H_2 - propyl propionate	Winkelmann (1884 c)	370
H_2 - $CHCl_3$	Baumgartner (1877 a)	291
H_2 - CF_2Cl_2	Miller and Carman (1961, 1964)	293

Table 16. (Continued)

System	Reference	T, °K
H_2 - n-nonane	Cummings *et al.* (1955)	340
H_2 - i-butyl propionate	Winkelmann (1884 c)	371
H_2 - propyl butyrate	Winkelmann (1884 c)	371
H_2 - propyl i-butyrate	Winkelmann (1884 c)	370
H_2 - ethyl valerate	Winkelmann (1884 c)	371
H_2 - 1-bromobutane	Byrne *et al.* (1967)	303
H_2 - 2:3:3-trimethylheptane	Cummings and Ubbelohde (1953, 1955)	364
H_2 - n-decane	Cummings and Ubbelohde (1953, 1955)	364
H_2 - amyl propionate	Winkelmann (1884 c)	371
H_2 - i-butyl butyrate	Winkelmann (1884 c)	371
H_2 - i-butyl i-butyrate	Winkelmann (1884 c)	371
H_2 - propyl valerate	Winkelmann (1884 c)	371
H_2 - SF_6	Boyd *et al.* (1951)	298
	Strehlow (1953)	286 to 418
	Suetin and Ivakin (1961)	290
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290

Table 16. (Continued)

System	Reference	T, °K
	Evans and Kenney (1965)	293
H_2 - 1-bromo-3-methylbutane	Byrne *et al*. (1967)	303
H_2 - CCl_4	Trautz and Ries (1931)	296
	Trautz and Müller (1935)	296
H_2 - amyl i-butyrate	Winkelmann (1884 c)	371
H_2 - i-butyl valerate	Winkelmann (1884 c)	371
H_2 - Br_2	Mackenzie and Melville (1932)	286
	Mackenzie and Melville (1933)	290
H_2 - 1-iodopropane	Byrne *et al*. (1967)	303
H_2 - n-dodecane	Cummings and Ubbelohde (1953, 1955)	400
H_2 - Hg	Gaede (1915)	room temperature
	Spier (1939)	314 to 325
H_2 - di-n-butyl phthalate	Birks and Bradley (1949)	293
H_2 - UF_6	Ljunggren (1965)	293

Table 16. (Continued)

System	Reference	T, °K
HD - D_2	Weissman and Mason (1962 b)	72 to 293
D_2 - TH	Reichenbacher _et_ _al_. (1965)	297
	Amdur and Beatty (1965)	195 and 273
D_2 - DT	Reichenbacher _et_ _al_. (1965)	297
D_2 - T_2	Amdur and Beatty (1965)	195 to 353
	Mason _et_ _al_. (1965)	295
	Reichenbacher _et_ _al_. (1965)	297
D_2 - NH_3	Ivakin and Suetin (1964 a)	297
D_2 - N_2	Ivakin and Suetin (1964 a)	297
D_2 - CO	Ivakin and Suetin (1964 a)	296
D_2 - air	Ivakin and Suetin (1964 a)	297
D_2 - CO_2	Saxena and Mason (1959)	250 to 372
	Ivakin and Suetin (1964 a)	296
	Annis _et_ _al_. (1969)	295
D_2 - n-heptane	Clarke and Ubbelohde (1957)	303
D_2 - 2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
D_2 - n-octane	Clarke and Ubbelohde (1957)	303

Table 16. (Continued)

System	Reference	T, °K
D_2 - 2:2:4-trimethylpentane	Clarke and Ubbelohde (1957)	303
D_2 - SF_6	Ivakin and Suetin (1964 a)	296
T_2 - N_2	Mason *et al.* (1964)	297
	Annis *et al.* (1969)	295
T_2 - CO_2	Mason *et al.* (1964)	298
	Annis *et al.* (1969)	295
CH_4 - CH_4	Winn and Ney (1947)	293
	Winn (1950)	90 to 353
	Ember *et al.* (1964)	297
	Mueller and Cahill (1964)	298 to 383
CH_4 - NH_3	Weissman (1964)	288
CH_4 - H_2O	Schwertz and Brow (1951)	308 to 352
	Kimpton and Wall (1952)	298 to 333
	O'Connell *et al.* (1969) Table I has a misprint, the highest temperature is not 323°K but 328°K.	283 to 328

Table 16. (Continued)

System	Reference	T, °K
CH_4 - C_2H_2	Weissman (1964)	313 to 373
CH_4 - H_2	Mueller and Cahill (1964)	298 to 383
CH_4 - CO	Mueller and Cahill (1964)	298 to 383
CH_4 - C_2H_6	Weissman (1964)	293 to 523
	Chang (1966)	255 to 311
CH_4 - O_2	Walker and Westenberg (1960, 1966, 1968)	294 to 985
CH_4 - air	Coward and Georgeson (1937)	289 to 295
CH_4 - CO_2	von Obermayer (1887)	294 - 295
	Fejes and Czárán (1961)	298
	Ember *et al.* (1964)	297
	Weissman (1964)	298
	Kestin and Yata (1968)	293 and 303
	Hawtin *et al.* (1969)	293 to 873
	Weissman (1969)	293 to 370
CH_4 - C_3H_8	Weissman (1964)	293 to 523
	Chang (1966)	255 to 311
CH_4 - CH_3Cl	Manner (1967)	298 to 478

Table 16. (Continued)

System	Reference	T, °K
CH_4 - n-C_4H_{10}	Chang (1966)	255 to 311
	Kestin and Yata (1968)	293 and 303
CH_4 - Rb	McNeal (1962)	333
	Violino (1968)	333
CH_4 - n-hexane	Carmichael *et al.* (1955 b)	294 to 377
	Kohn and Romero (1965)	298 to 333
CH_4 - 3-methylpentane	Kohn and Romero (1965)	298 to 333
CH_4 - CF_4	Mueller and Cahill (1964)	298 to 383
CH_4 - n-heptane	Carmichael *et al.* (1955 a)	311 to 377
	Reamer and Sage (1963)	311 to 377
CH_4 - SF_6	Manner (1967)	298 to 478
CH_4 - Br_2	Mackenzie and Melville (1933)	289
NH_3 - NH_3	Paul and Watson (1966)	233 to 353
NH_3 - N_2	Bunde (1955)	298 to 358
	Ivakin and Suetin (1964 a)	295
	Weissman (1964)	293 to 523
NH_3 - CO	Ivakin and Suetin (1964 a)	295

Table 16. (Continued)

System	Reference	T, °K
NH_3 - C_2H_4	Weissman (1964)	293 to 523
NH_3 - air	Toepler (1896)	292
	Wintergerst (1930)	287 to 298
	Andrew (1955)	293
	Ivakin and Suetin (1964 a)	295
NH_3 - CH_3NH_2	Burch and Raw (1967)	273 to 673
NH_3 - O_2	Weissman (1964)	293 to 473
NH_3 - ethyl ether	Srivastava and Srivastava (1963)	288 to 338
	Pal and Bhattacharyya (1969)	299 to 373
NH_3 - SF_6	Ivakin and Suetin (1964 a)	297

Table 16. (Continued)

System	Reference	T, °K
$H_2O - H_2O$	Ferron (1967)	950 to 1400
$H_2O - N_2$	Hippenmeyer (1949)	273 to 368
	Schwertz and Brow (1951)	307 to 352
	Bose and Chakraborty (1955-56)	332 and 336
	Crider (1956)	329 and 349
	Nelson (1956)	298 to 328
	O'Connell *et al*. (1969)	282 to 373
$H_2O - C_2H_4$	Schwertz and Brow (1951)	308 to 353
	Kimpton and Wall (1952)	298
$H_2O - C_2H_6$	Kimpton and Wall (1952)	298
	Bose and Chakraborty (1955-56)	332 and 336
$H_2O - O_2$	Schwertz and Brow (1951)	308 to 352
	Walker and Westenberg (1960,1966)	390 to 1070
H_2O - air	Stefan (1871)	room temperature
	Guglielmo (1881,1882)	280 to 296
	Winkelmann (1884 a,b)	323 and 366
	Winkelmann (1888)	290 to 294
	Winkelmann (1889)	290 and 372
	Houdaille (1896)	273

Table 16. (Continued)

System	Reference	T.°K
	Brown and Escombe	286 to 290
	Mache (1910)	301 to 366
	Le Blanc and Wupperman (1916)	315
	Summerhays (1930)	289-290
	Houghton (1933)	293
	Ackermann (1934)	356 to 366
	Gilliland (1934)	299 to 332
	Trautz and Müller (1935)	281 to 373
	Schirmer (1938)	273 to 370
	Klibanova *et al*. (1942)	373 to 1493
	Brookfield *et al*. (1947)	298 to 318
	Kimpton and Wall (1952)	298
	Rossié (1953)	356 to 575
	Lee and Wilke (1954)	298
	Bose and Chakraborty (1955-56)	327 and 336
	Narsimhan (1955-56)	303
	Nelson (1956)	298
	Richardson (1959)	319

Table 16. (Continued)

System	Reference	T, °K
	Petit (1965)	293 to 301
H_2O - CH_3OH	Weissman (1968 a)	373
H_2O - H_2O_2	Weissman (1968 a)	443 to 513
H_2O - CO_2	Guglielmo (1882)	291
	Winkelmann (1884 a,b)	323 and 366
	Winkelmann (1889)	294-298 and 373
	Trautz and Müller (1935)	294 to 372
	Schwertz and Brow (1951)	307 to 352
	Rossie (1953)	433
	Crider (1956)	329 and 349
	Ember *et al*. (1964)	1000 to 1400
	Ferron (1967)	1000 to 1700
H_2O - C_3H_8	Kimpton and Wall (1952)	298
H_2O - C_2H_5OH	Weissman (1968 a)	373
H_2O - SO_2	Kimpton and Wall (1952)	298
H_2O - ethyl ether	Winkelmann (1884 a)	284 and 293
H_2O - CCl_2F_2	Lee and Wilke (1954)	298
D_2O - air	Kimpton and Wall (1952)	298 to 333

Table 16. (Continued)

System	Reference	T,°K
Na - N_2	von Hartel and Polyani (1930)	633
	von Hartel *et al*. (1932)	655
	Van der Held and Miesowicz (1937)	288
	Cvetanović and Le Roy (1952)	527
	Ramsey and Anderson (1964)	453
	Violino (1968)	453
Na - C_5H_{12}	von Hartel *et al*. (1932)	655
C_2H_2 - C_2H_2	Mueller and Cahill (1964)	298 to 383
C_2H_2 - C_2H_4	Mueller and Cahill (1964)	298 to 383
C_2H_2 - C_2H_6	Mueller and Cahill (1964)	298 to 383
C_2H_2 - O_2	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
C_2H_2 - air	Kosov (1957)	289
	Suetin and Ivakin (1961)	288
	Suetin (1964)	288
C_2H_2 - C_3H_6	Weissman (1964)	313 to 373
HCN - air	Klotz and Miller (1947)	273

Table 16. (Continued)

System	Reference	T, °K
N_2 - N_2	Winn (1948)	293
	Winn (1950)	78 to 353
	Winter (1951)	273 and 318
	DeLuca (1954)	273
N_2 - CO	Amdur and Mason (1958)	1000 to 15,000
	Belyaev and Leonas (1966)	2000 to 15,000
	Boardman and Wild (1937)	288-291
	Wicke and Hugo (1961)	295
	Weissman and Mason (1962 b)	300 to 550
	Amdur and Shuler (1963)	195 to 373
N_2 - C_2H_4	Ivakin and Suetin (1964 a)	296
	Boyd *et al.* (1951)	298
	Knox and McLaren (1963,1964)	291
	Mueller and Cahill (1964)	298 and 373
	Weissman (1964)	300 to 550
	Evans and Kenney (1965)	287 and 291
	Fuller and Giddings (1967)	303
	Fuller *et al.* (1969)	303
N_2 - NO	Weissman (1964)	293 and 373
N_2 - C_2H_6	Boyd *et al.* (1951)	298

Table 16. (Continued)

System	Reference	T, °K
	Fejes and Czárán (1961)	298
N_2 - O_2	von Obermayer (1880)	286-287 and 335
	von Obermayer (1882 b)	286-289
	Lonius (1909)	285-286
	Parker and Hottel (1936)	1157
	Waldmann (1944, 1947)	293
	Bohemen and Purnell (1961)	324
	Giddings and Seager (1962)	298
	Weissman and Mason (1962 b)	300 to 550
	Belyaev and Leonas (1966)	2000 to 15,000
	Arnikar *et al*. (1967 a,b)	298
N_2 - CH_3OH	Arnikar and Ghule (1969)	355
N_2 - HCl	Mian (1967)	324 to 523
N_2 - K	Ivanovskiĭ *et al*. (1969)	630 to 920
N_2 - CO_2	Parker and Hottel (1936)	1157
	Boardman and Wild (1937)	288-290
	Wicke and Kallenbach (1941)	273
	Waldmann (1944, 1947)	293

Table 16. (Continued)

System	Reference	T,°K
	Boyd *et al*. (1951)	298
	Schäfer et al. (1951)	252 to 308
	Westenberg and Walker (1957)	293
	Walker (1958)	296 to 1114
	Walker and Westenberg (1958 a)	298 to 1150
	Walker and Westenberg (1958 b)	300 to 1150
	Vyshenskaya and Kosov (1959)	293 to 1083
	Walker *et al*. (1960)	299
	Bohemen and Purnell (1961)	324
	Suetin and Ivakin (1961)	290
	Wicke and Hugo (1961)	295
	Giddings and Seager (1962)	293 to 299
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290
	Pakurar and Ferron (1964)	295, 1156 to 1653
	Weissman (1964)	293
	Vyshenskaya and Kosov (1965)	293 to 1083
	Kestin *et al*. (1966)	293 and 304

Table 16. (Continued)

System	Reference	T, °K
	Pakurar and Ferron (1966)	1081 to 1810
	Walker and Westenberg (1966)	296 to 1114
	Coates and Mian (1967)	301 to 525
	Ferron (1967)	1100 to 1800
	Mian (1967)	301 to 525
	Ellis and Holsen (1969)	298 to 880
	Schneider and Schäfer (1969)	273 to 1300
N_2 - C_3H_8	Fejes and Czárán (1961)	298
N_2 - C_2H_5OH	Bose and Chakraborty (1955-56)	327 and 331
	Arnikar *et al*. (1967 b)	353
	Arnikar and Ghule (1969)	355
N_2 - acetone	Mehta (1966)	298
	Arnikar *et al*. (1967 b)	353
	Arnikar and Ghule (1969)	353
N_2 - n-butane	Boyd *et al*. (1951)	298
	Fejes and Czárán (1961)	298
	Fuller and Giddings (1967)	302
	Hargrove and Sawyer (1967)	298

Table 16. (Continued)

System	Reference	T, °K
	Manner (1967)	298
	Fuller *et al*. (1969)	302
N_2 - i-butane	Boyd *et al*. (1951)	298
N_2 - i-propyl alcohol	Arnikar and Ghule (1969)	358
N_2 - nitromethane	Byrne *et al*. (1967)	303
N_2 - SO_2	Schäfer (1959)	263
N_2 - n-pentane	Arnikar and Ghule (1969)	353
N_2 - n-butylamine	Mehta (1966)	298
N_2 - methyl acetate	Arnikar *et al*. (1967 b)	353
	Arnikar and Ghule (1969)	358
N_2 - benzene	Bose and Chakraborty (1955-56)	326 and 332
	Hudson *et al*. (1960)	311
	Arnikar and Ghule (1969)	353
N_2 - pyridine	Hudson *et al*. (1960)	318
N_2 - HBr	Mian (1967)	336 to 525
N_2 - 2:3-dimethylbuta-1:3-diene	Cummings *et al*. (1955)	288
N_2 - thiophen	Hudson *et al*. (1960)	302
N_2 - cyclohexane	Cummings and Ubbelohde (1953,1955)	289

Table 16. (Continued)

System	Reference	T, °K
	Hudson *et al*. (1960)	289
N_2 - 2:3-dimethyl but-2-ene	Cummings *et al*. (1955)	288
N_2 - methyl cyclopentane	Cummings and Ubbelohde (1953,1955)	286
N_2 - piperidine	Hudson *et al*. (1960)	315
N_2 - Rb	McNeal (1962)	328
	Violino (1968)	328
N_2 - 3-pentanone	Barr and Sawyer (1964)	300
N_2 - 2:3-dimethylbutane	Cummings and Ubbelohde (1953,1955)	289
N_2 - n-hexane	Cummings and Ubbelohde (1953,1955)	289
	Huber and van Vught (1965)	353
	Arnikar *et al*. (1967 b)	353
N_2 - hexa-1:5-diene	Cummings *et al*. (1955)	288
N_2 - ethyl acetate	Arnikar and Ghule (1969)	355
N_2 - tetrahydrothiophen	Hudson *et al*. (1960)	319
N_2 - N_2O_4	Chambers and Sherwood (1937)	273 and 283
N_2 - monofluorobenzene	Byrne *et al*. (1967)	303
N_2 - n-heptane	Cummings *et al*. (1955)	303
	Clarke and Ubbelohde (1957)	303

Table 16. (Continued)

System	Reference	T,°K
N_2 - 2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
N_2 - triethylamine	Mehta (1966)	298
N_2 - Cd	Spier (1940)	290-293
N_2 - n-octane	Cummings and Ubbelohde (1953,1955)	303
	Clarke and Ubbelohde (1957)	303
N_2 - 2:2:4-trimethylpentane	Cummings and Ubbelohde (1953,1955)	303
	Clarke and Ubbelohde (1957)	303
N_2 - n-nonane	Cummings *et al*. (1955)	340
N_2 - Cs	Violino (1968)	317
N_2 - 1-bromobutane	Byrne *et al*. (1967)	303
N_2 - 2:3:3-trimethylheptane	Cummings and Ubbelohde (1953,1955)	364
N_2 - n-decane	Cummings and Ubbelohde (1953,1955)	364
N_2 - SF_6	Suetin and Ivakin (1961)	290
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290
	Evans and Kenney (1965)	293
N - 1-bromo-3-methylbutane	Byrne *et al*. (1967)	303
N_2 - CCl_4	Bose and Chakraborty (1955-56)	323 and 330

Table 16. (Continued)

System	Reference	T,°K
N_2 - Br_2	Mackenzie and Melville (1932)	286
N_2 - 1-iodopropane	Byrne et al. (1967)	303
N_2 - n-dodecane	Cummings and Ubbelohde (1953,1955)	400
N_2 - Hg	Mullaly and Jacques (1924)	293
	Trautz and Müller (1935)	293
	Spier (1940)	292-298
	Nakayama (1968)	301
N_2 - I_2	Mullaly and Jacques (1924)	293
	Trautz and Müller (1935)	293
	Vyshenskaya and Kosov (1965)	452 to 873
N_2 - UF_6	Ljunggren (1965)	293
CO - CO	Amdur and Shuler (1963)	195 to 373
CO - C_2H_4	von Obermayer (1883)	290-296
	Weissman (1964)	300 to 550
CO - O_2	Loschmidt (1870 b)	294
	von Obermayer (1883)	287 and 335
	Walker and Westenberg (1960)	295 to 800
	Weissman and Mason (1962 b)	300 to 500

Table 16. (Continued)

System	Reference	T,°K
	Walker and Westenberg (1966)	295 to 796
$CO - CO_2$	Loschmidt (1870 b)	282 and 293
	von Obermayer (1887)	292
	Ivakin and Suetin (1964 a)	296
	Ivakin and Suetin (1964 b)	296 to 473
$CO - SF_6$	Ivakin and Suetin (1964 a)	297
	Ivakin and Suetin (1964 b)	297 to 473
$C_2H_4 - C_2H_4$	Mueller and Cahill (1964)	298 to 383
$C_2H_4 - C_2H_6$	Mueller and Cahill (1964)	298 to 383
$C_2H_4 - O_2$	Weissman (1964)	293 to 373
$C_2H_4 - CO_2$	von Obermayer (1887)	295
$C_2H_4 - Rb$	McNeal (1962)	333
	Violino (1968)	333
$NO - N_2O$	Weissman (1964)	550 to 700
$C_2H_6 - C_2H_6$	Mueller and Cahill (1964)	298 to 383
$C_2H_6 - C_3H_8$	Weissman (1964)	293 to 523
$C_2H_6 - Rb$	McNeal (1962)	333
	Violino (1968)	333

Table 16. (Continued)

System	Reference	T,°K
C_2H_6 - n-hexane	Carmichael et al. (1955 b)	294 to 377

Table 16. (Continued)

System	Reference	T, °K
$O_2 - O_2$	Winn (1950)	78 to 353
	Winter (1951)	273 and 318
	Belyaev and Leonas (1966)	2000 to 15,000
$O_2 - CO_2$	Loschmidt (1870 b)	287
	Wretschko (1870)	297
	von Obermayer (1882 a)	284-293
	Waldmann (1944,1947)	293
	Walker and Westenberg (1960)	297 to 1080
	Suetin and Ivakin (1961)	288
	Suetin (1964)	288
	Walker and Westenberg (1966)	296 to 1083
$O_2 - C_2H_5OH$	Bose and Chakraborty (1955-56)	327 and 331
O_2 - benzene	Trautz and Ludwig (1930)	296
	Trautz and Müller (1935)	296
	Bose and Chakraborty (1955-56)	326-332
	Hudson *et al.* (1960)	311
O_2 - pyridine	Hudson *et al.* (1960)	318
O_2 - thiophen	Hudson *et al.* (1960)	302

Table 16. (Continued)

System	Reference	T,°K
O_2 - cyclohexane	Cummings and Ubbelohde (1953,1955)	289
	Hudson *et al.* (1960)	289
O_2 - methyl cyclopentane	Cummings and Ubbelohde (1953,1955)	287
O_2 - piperidine	Hudson *et al.* (1960)	315
O_2 - 2:3-dimethylbutane	Cummings and Ubbelohde (1953,1955)	288
O_2 - n-hexane	Cummings and Ubbelohde (1953,1955)	289
O_2 - tetrahydrothiophen	Hudson *et al.* (1960)	319
O_2 - n-octane	Cummings and Ubbelohde (1953,1955)	303
O_2 - 2:2:4-trimethylpentane	Cummings and Ubbelohde (1953,1955)	303
O_2 - SF_6	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
	Ivakin *et al.* (1968)	297 to 408
O_2 - CCl_4	Trautz and Müller (1935)	296
	Bose and Chakraborty (1955-56)	323 and 330
O_2 - Br_2	Mackenzie and Melville (1932)	286
O_2 - UF_6	Ljunggren (1965)	293

Table 16. (Continued)

System	Reference	T, °K
air - O_2	von Obermayer (1882 b)	290 - 294
	von Obermayer (1887)	287 - 288
air - CH_3OH	Winkelmann (1885)	299 and 323
	Vaillant (1911)	283
	Altshuller and Cohen (1960)	298
	Stevenson (1965)	298
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Mrazek <u>et</u> <u>al</u>. (1968)	328
	Katan (1969)	295
air - H_2O_2	McMurtie and Keyes (1948)	333
air - CO_2	Loschmidt (1870 a)	252 to 299
	Loschmidt (1870 b)	252 to 291
	von Obermayer (1880)	218 - 285 and 335
	von Obermayer (1882 a)	284 - 292
	von Obermayer (1882 b)	283 to 298
	Waitz (1882 a, b)	290 - 292

Table 16. (Continued)

System	Reference	T, °K
	von Obermayer (1883)	280 - 283 and 335
	von Obermayer (1887)	281 - 298
	Toepler (1896)	291 -292
	Brown and Escombe (1900)	280 - 288
	Buckingham (1904)	300
	Foch (1913)	room temperature
	Coward and Georgeson (1937)	273
	Klibanova *et al.* (1942)	290 to 1533
	Andrew (1955)	291 to 293
	Kosov (1957)	291
	Suetin and Ivakin (1961)	289
	Holsen and Strunk (1964)	276 and 317
	Suetin (1964)	289
air - formic acid	Winkelmann (1885)	339 and 358
	Lugg (1968)	298
air - C_2H_5OH	Baumgartner (1877 a)	290
	Winkelmann (1884 a)	314 and 340

Table 16. (Continued)

System	Reference	T, °K
	Winkelmann (1885)	323 and 337
	Vaillant (1911)	283 to 291
	Le Blanc and Wuppermann (1916)	315 and 340
	Trautz and Müller (1935)	340
	Lee and Wilke (1954)	298
	Bose and Chakraborty (1955-56)	327 and 331
	Narsimhan (1955-56)	303
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Katan (1969)	295
air - acrylonitrile	Lugg (1968)	298
air - acetone	Goryunova and Kuvshinskiĭ (1948)	273
	Gush (1948)	323
	Richardson (1959)	293 to 328
	Stevenson (1965)	298
	Pryde and Pryde (1967)	295
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - allyl alcohol	Lugg (1968)	298
air - acetic acid	Winkelmann (1885)	339 to 372
	Pochettino (1914)	336 to 372
	Lugg (1968)	298
air - methyl formate	Griboiedov (1893)	289 - 295
	Pochettino (1914)	284 and 293
	Lugg (1968)	298
air - n-propyl alcohol	Winkelmann (1885)	340 and 357
	Vaillant (1911)	287
	Pochettino (1914)	288 to 355
	Lugg (1968)	298
air - i-propyl alcohol	Pochettino (1914)	288 and 333
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - ethylene diamine	Lugg (1968)	298
air - CNCl	Klotz and Miller (1947)	273
air - ethylene glycol	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - SO_2	Andrew (1955)	293
air - 2-methyl-1,3-butadiene	Altshuller and Cohen (1960)	288
air - Cl_2	Andrew (1955)	293
	Kosov (1957)	289
air - methylethylketone	Lugg (1968)	298
air - pentane	Lugg (1968)	298
air - dimethylformamide	Lugg (1968)	298
air - n-butylamine	Pochettino (1914)	334 - 335
	Lugg (1968)	298
air - i-butylamine	Pochettino (1914)	292 and 335
	Lugg (1968)	298
air - diethylamine	Pochettino (1914)	283 and 324
	Lugg (1968)	298
air - methyl acetate	Winkelmann (1884 c)	294 and 319
	Griboiedov (1893)	300 to 317
	Pochettino (1914)	283 to 324
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - ethyl formate	Winkelmann (1884 c)	294 and 319
	Pochettino (1914)	284 to 323
	Lugg (1968)	298
air - propionic acid	Winkelmann (1885)	366 and 372
	Pochettino (1914)	324 to 372
	Lugg (1968)	298
air - n-butyl alcohol	Winkelmann (1885)	372
	Pochettino (1914)	334 and 373
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - i-butyl alcohol	Winkelmann (1885)	340 and 357
	Pochettino (1914)	333 and 372
	Lugg (1968)	298
air - sec-butyl alcohol	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - tert-butyl alcohol	Pochettino (1914)	294 and 340
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - ethyl ether	Stefan (1873)	292
	Baumgartner (1877 a)	289
	Stefan (1889, 1890)	292
	Griboiedov (1893)	289 - 292
	Winkelmann (1884 a)	284 and 293
	Naccari (1909, 1910)	285 to 299
	Pochettino (1914)	283 to 303
	Trautz and Müller (1935)	288 - 293
	Pryde and Pryde (1967)	295
	Lugg (1968)	298
air - CS_2	Stefan (1873)	289
	Baumgartner (1877 a)	291
	Baumgartner (1877 b)	269 to 315
	Stefan (1889, 1890)	289
	Arnold (1944)	303
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - ethylene glycol-monomethyl ether	Lugg (1968)	298
air - propylene glycol	Lugg (1968)	298
air - allyl chloride	Lugg (1968)	298
air - benzene	Griboiedov (1893)	315 to 338
	Le Blanc and Wuppermann (1916)	315 and 340
	Lee and Wilke (1954)	298
	Bose and Chakraborty (1955-56)	321 to 332
	Narsimhan (1955-56)	303
	Altshuller and Cohen (1960)	300 to 334
	Jorgensen and Watts (1961)	308
	Heinzelmann *et al.* (1965)	308
	Stevenson (1965)	298
	Ben Aim *et al.* (1967)	298
	Getzinger and Wilke (1967)	308
	Nafikov and Usmanov (1967)	293 to 333
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297

Table 16. (Continued)

System	Reference	T, °K
	Katan (1969)	295
air - ethylene chlorohydrin	Lugg (1968)	298
air - cyclohexane	Goryunova and Kuvshinskiĭ (1948)	318
air - hexene	Altshuller and Cohen (1960)	293 and 303
air - dichloromethane	Lugg (1968)	298
air - methylpropylketone	Lugg (1968)	298
air - n-hexane	Schlinger _et_ _al_. (1952-53)	294 to 328
	Altshuller and Cohen (1960)	298 to 323
	Galloway and Sage (1967)	311
	Nafikov and Usmanov (1967)	293 to 333
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air - n-butyric acid	Winkelmann (1885)	372
	Pochettino (1914)	348 and 373
	Lugg (1968)	298
air - i-butyric acid	Winkelmann (1885)	371
	Pochettino (1914)	351 and 373

Table 16. (Continued)

System	Reference	T, °K
	Lugg (1968)	298
air - ethyl acetate	Winkelmann (1884 c)	319
	Pochettino (1914)	283 to 343
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
	Katan (1969)	295
air - methyl propionate	Winkelmann (1884 c)	319 and 340
	Griboiedov (1893)	316 and 332
	Pochettino (1914)	288 to 343
	Lugg (1968)	298
air - propyl formate	Winkelmann (1884 c)	319 and 340
	Pochettino (1914)	293 and 353
	Lugg (1968)	298
air - p-dioxane	Lugg (1968)	298
air - n-amyl alcohol	Winkelmann (1885)	372
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - active amyl alcohol	Winkelmann (1885)	372
air - sec-amyl alcohol	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - ethylene glycol-monoethyl ether	Lugg (1968)	298
air - toluene	Mack (1925)	298
	Gilliland (1934)	299 to 332
	Fairbanks and Wilke (1950)	301
	Narsimhan (1955-56)	303
	Altshuller and Cohen (1960)	298 and 318
	Stevenson (1965)	298
	Yuan and Cheng (1967)	310 to 343
	Lugg (1968)	298
air - n-butyl chloride	Grob and El-Wakil (1969)	297
air - aniline	Mack (1925)	298
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - furfural	Brookfield _et_ _al_. (1947)	298 to 323

Table 16. (Continued)

System	Reference	T, °K
air - fluorobenzene	Grob and El-Wakil (1969)	297
air - mesityl oxide	Lugg (1968)	298
air - $COCl_2$	Klotz and Miller (1947)	273
air - 1,1-dichloroethane	Lugg (1968)	298
air - 1,2-dichloroethane	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air - n-heptane	Schlinger et al. (1952-53)	294 to 361
	Altshuller and Cohen (1960)	290 and 338
	Stevenson (1965)	298
	Galloway and Sage (1967)	339 - 350
	Nafikov and Usmanov (1967)	293 to 353
	Grob and El-Wakil (1969)	297
air - triethylamine	Lugg (1968)	298
air - i-butyl formate	Pochettino (1914)	298 to 358
	Lugg (1968)	298
air - ethyl propionate	Winkelmann (1884 c)	340 and 363
	Pochettino (1914)	283 to 366

Table 16. (Continued)

System	Reference	T, °K
	Fairbanks and Wilke (1950)	301
	Lugg (1968)	298
air - methyl n-butyrate	Winkelmann (1884 c)	340 and 365
	Pochettino (1914)	295 to 372
	Lugg (1968)	298
air - methyl i-butyrate	Winkelmann (1884 c)	323 and 340
	Pochettino (1914)	285 and 353
	Lugg (1968)	298
air - n-propyl acetate	Le Blanc and Wuppermann (1916)	315 and 340
	Pochettino (1914)	283 to 372
	Lugg (1968)	298
air - i-propyl acetate	Lugg (1968)	298
air - valeric acid	Pochettino (1914)	355 and 373
air - i-valeric acid	Winkelmann (1885)	372
	Pochettino (1914)	344 and 373
	Lugg (1968)	298
air - 2-ethyl-1-butanol	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - n-hexyl alcohol	Winkelmann (1885)	372
	Lugg (1968)	298
air - i-propylether	Lugg (1968)	298
air - methyl-2-pentanol	Lugg (1968)	298
air - benzonitrile	Lugg (1968)	298
air - phenylethylene	Lugg (1968)	298
air - diethylene glycol	Lugg (1968)	298
air - ethyl benzene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air - m-xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air - o-xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air - p-xylene	Pochettino (1914)	294 to 373
	Lugg (1968)	298
air - benzyl alcohol	Lugg (1968)	298
air - ethyl bromide	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
	Grob and El-Wakil (1969)	297
air - propylene dichloride	Lugg (1968)	298
air - 1-octene	Altshuller and Cohen (1960)	313 and 370
air - chlorobenzene	Le Blanc and Wuppermann (1916)	315 and 340
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air - ethyl cyanoacetate	Lugg (1968)	298
air - n-octane	Mack (1925)	298
	Galloway and Sage (1967)	364
	Nafikov and Usmanov (1967)	293 to 353
	Lugg (1968)	298
air - n-amyl formate	Pochettino (1914)	310 to 373
	Lugg (1968)	298
air - i-amyl formate	Pochettino (1914)	322 and 373
	Lugg (1968)	298
air - n-butyl acetate	Pochettino (1914)	325 and 373
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - i-butyl acetate	Winkelmann (1884 c)	340 and 371
	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air - n-caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air - i-caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air - diacetone alcohol	Lugg (1968)	298
air - ethyl n-butyrate	Winkelmann (1884 c)	340 and 370
	Pochettino (1914)	315 and 373
	Lugg (1968)	298
air - ethyl i-butyrate	Winkelmann (1884 c)	340 and 369
	Pochettino (1914)	332 and 373
	Lugg (1968)	298
air - methyl valerate	Pochettino (1914)	319 to 373
	Lugg (1968)	298
air - propyl propionate	Winkelmann (1884 c)	370

Table 16. (Continued)

System	Reference	T, °K
	Pochettino (1914)	326 to 373
air - n-heptyl alcohol	Lugg (1968)	298
air - $CHCl_3$	Baumgartner (1877 a)	292
	Goryunova and Kuvshinskiĭ (1948)	273
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Mrazek _et_ _al_. (1968)	323
air - mesitylene	Pochettino (1914)	334 and 373
	Lugg (1968)	298
air - n-propyl benzene	Pochettino (1914)	325 to 372
	Lugg (1968)	298
air - i-propyl benzene	Pochettino (1914)	333 to 372
	Lugg (1968)	298
air - pseudo-cumene	Lugg (1968)	298
air - benzoic acid	Yuan and Cheng (1967)	413 to 433
air - n-propyl bromide	Pochettino (1914)	294 and 336
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T,°K
air - i-propyl bromide	Pochettino (1914)	292 and 325
	Lugg (1968)	298
air - nitrobenzene	Lee and Wilke (1954)	298
	Lugg (1968)	298
air - benzyl chloride	Pochettino (1914)	357 and 372
	Lugg (1968)	298
air - o-chlorotoluene	Pochettino (1914)	338 to 371
	Lugg (1968)	298
air - m-chlorotoluene	Pochettino (1914)	338 and 371
	Lugg (1968)	298
air - p-chlorotoluene	Pochettino (1914)	333 to 373
	Lugg (1968)	298
air - n-C_9H_{20}	Nafikov and Usmanov (1967)	293 to 353
air - napthalene	Mack (1925)	298
air - bromochloromethane	Lugg (1968)	298
air - n-amyl acetate	Lugg (1968)	298
air - n-butyl propionate	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T,°K
air - i-butyl propionate	Winkelmann (1884 c)	371
	Pochettino (1914)	329 to 373
	Lugg (1968)	298
air - ethyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	324 to 372
	Lugg (1968)	298
air - methyl-n-caproate	Lugg (1968)	298
air - n-propyl n-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air - n-propyl i-butyrate	Winkelmann (1884 c)	370
	Pochettino (1914)	339 to 373
	Lugg (1968)	298
air - i-propyl i-butyrate	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air - octyl alcohol	Lugg (1968)	298
air - butylether	Lugg (1968)	298

Table 10. (Continued)

System	Reference	T, °K
air - trichloro-ethylene	Lugg (1968)	298
air - 1,1,1-trichlorethane	Lugg (1968)	298
air - 1,1,2-trichlorethane	Lugg (1968)	298
air - diethylene glycol-monoethyl ether	Lugg (1968)	298
air - p-cymene	Lugg (1968)	298
air - i-phorone	Lugg (1968)	298
air - toluene-2,4-diisocyanate	Lugg (1968)	298
air - n-$C_{10}H_{22}$	Altshuller and Cohen (1960)	313 to 422
	Nafikov and Usmanov (1967)	313 to 353
air - dichloroethylether	Lugg (1968)	298
air - amyl propionate	Winkelmann (1884 c)	371
	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air - i-butyl-n-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	348 to 373
	Lugg (1968)	298
air - i-butyl-i-butyrate	Winkelmann (1884 c)	371

Table 16. (Continued)

System	Reference	T, °K
	Pochettino (1914)	348 and 373
	Lugg (1968)	298
air - propyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	343 to 373
	Lugg (1968)	298
air - SF_6	Suetin and Ivakin (1961)	290
	Suetin (1964)	290
air - ethylene glycol monoethylether acetate	Lugg (1968)	298
air - p-tert-butyltoluene	Lugg (1968)	298
air - triethylene glycol	Lugg (1968)	298
air - benzyl acetate	Lugg (1968)	298
air - methyl salicylate	Brookfield _et_ _al_. (1947)	298 and 323
air - CCl_4	Bose and Chakraborty (1955-56)	323 and 330
	Narsimhan (1955-56)	303
	Richardson (1959)	315 to 335
	Getzinger and Wilke (1967)	308
	Pryde and Pryde (1967)	295

Table 16. (Continued)

System	Reference	T, °K
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air - diphenyl	Mack (1925)	298
	Gilliland (1934)	491
air - n-$C_{11}H_{24}$	Nafikov and Usmanov (1967)	333 and 353
air - ethyl iodide	Grob and El-Wakil (1969)	297
air - amyl n-butyrate	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air - amyl i-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	357 and 373
	Lugg (1968)	298
air - i-butyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	353 and 373
	Lugg (1968)	298
air - Br_2	Andrew (1955)	293
	Brockett (1966)	301
	Lugg (1968)	298

Table 16. (Continued)

System	Reference	T, °K
air - safrole	Pochettino (1914)	350 and 373
air - i-safrole	Pochettino (1914)	336 and 373
air - eugenol	Pochettino (1914)	359 and 372
air - i-eugenol	Pochettino (1914)	358 and 372
air - chlorpicrin	Lugg (1968)	298
air - CCl_3NO_2	Klotz and Miller (1947)	298
air - tetrachloroethylene	Lugg (1968)	298
air - 1,1,2,2-tetrachlorethane	Lugg (1968)	298
air - n-propyl iodide	Pochettino (1914)	304 to 373
	Lugg (1968)	298
air - i-propyl iodide	Pochettino (1914)	324 to 352
	Lugg (1968)	298
air - n-$C_{12}H_{26}$	Nafikov and Usmanov (1967)	333 and 353
air - anthracene	Mack (1925)	372
air - triethyl phosphate	Lugg (1968)	298
air - benzidine	Mack (1925)	372
air - ethylene dibromide	Call (1957)	273 to 293

Table 16. (Continued)

System	Reference	T, °K
	Lugg (1968)	298
air - Hg	Gaede (1915)	room temperature, 413 and 473
	Gilliland (1934)	614
	Trautz and Müller (1935)	413 and 473
	Mikhailov and Kochegarova (1967)	not available
	Lugg (1968)	298
air - pentachloroethane	Lugg (1968)	298
air - diethyl phthalate	Lugg (1968)	298
air - n-$C_{16}H_{34}$	Bradley and Shellard (1949)	288 to 308
air - 1,2-dibromo-3-chloropropane	Lugg (1968)	298
air - n-$C_{17}H_{36}$	Bradley and Shellard (1949)	288 to 313
air - bromoform	Lugg (1968)	298
air - I_2	Langmuir (1918)	293
	Mack (1925)	298
	Topley and Whytlaw-Gray (1927)	287 to 303
	Trautz and Müller (1935)	287 to 303
	de Nordwall and Flowers (1958)	298

Table 16. (Continued)

System	Reference	T, °K
air - n-$C_{18}H_{38}$	Bradley and Shellard (1949)	288 to 313
air - S_8	Bradley (1951)	303
air - tributyl phosphate	Lugg (1968)	298
air - di-n-butyl phthalate	Bradley *et al.* (1946)	293
	Birks and Bradley (1949)	288 to 313
	Lugg (1968)	298
air - tetraethyl pyrophosphate	Lugg (1968)	298
air - bis-2-ethylhexyl phosphate	Lugg (1968)	298
air - $(C_7H_{15})_3CH$	Bradley and Waghorn (1951)	298 to 308
air - diisooctyl phthalate	Lugg (1968)	298
air - butyl stearate	Bradley *et al.* (1946)	293
air - tri-orthocresol phosphate	Lugg (1968)	298
air - $(C_{10}H_{21})_3CH$	Bradley and Waghorn (1951)	298 to 308
air - n-$C_{16}F_{34}$	Bradley and Waghorn (1951)	288 to 303

Table 16. (Continued)

System	Reference	T, ^{o}K
CH_3OH - CO_2	Winkelmann (1885)	299 and 323
CH_3OH - 1-butanol	Weissman (1968 a)	423
H_2S - ethyl ether	Pal and Bhattacharyya (1969)	298 to 373
HCl - DCl	Braune and Zehle (1941)	293-296
HCl - CO_2	Weissman (1964)	291
HCl - Br_2	Mackenzie and Melville (1933)	288
C_3H_6 - C_3H_8	Weissmann (1964)	313 to 373

Table 16. (Continued)

System	Reference	T, ^{o}K
CO_2 - CO_2	Winn (1950)	195 to 353
	Timmerhaus and Drickamer (1951)	296-297
	Winter (1951)	273 and 318
	Amdur *et al*. (1952)	195 to 363
	Miller and Carman (1961)	293
	Ember *et al*. (1962)	297, 1180 to 1680
	Schäfer and Reinhard (1963)	233 to 513
	Wendt *et al*. (1963)	248 to 362
	Ember *et al*. (1964)	297
	Miller and Carman (1964)	293
	Pakurar and Ferron (1964)	295, 1250 to 1650
	Pakurar and Ferron (1965)	1103 to 1944
	Ferron (1967)	300 to 1900
	Annis *et al*. (1969)	295
CO_2 - N_2O	Loschmidt (1870 b)	288 and 293
	von Obermayer (1880)	283-285 and 335
	von Obermayer (1882 b)	283-287

Table 16. (Continued)

System	Reference	T, ^{o}K
	Boardman and Wild (1937)	286-287
	Wall and Kidder (1946)	298
	Amdur et al. (1952)	195 to 363
	Weissman (1964)	300 to 550
	Kosov and Abdullina (1966)	298
CO_2 - ethylene oxide	Wall and Kidder (1946)	298
CO_2 - C_3H_8	Weissman (1964)	300 to 550
	Wall and Kidder (1946)	298
CO_2 - formic acid	Winkelmann (1885)	339 and 358
CO_2 - C_2H_5OH	Winkelmann (1884 a)	314 and 340
	Winkelmann (1885)	323 and 337
	Trautz and Müller (1935)	315 and 340
CO_2 - acetic acid	Winkelmann (1885)	339 to 372
CO_2 - n-propyl alcohol	Winkelmann (1885)	340 and 357
CO_2 - SO_2	Schäfer (1959)	263 to 473
	Weissman (1964)	289

Table 16. (Continued)

System	Reference	T, ^{o}K
CO_2 - ethyl formate	Winkelmann (1884 c)	294 and 319
CO_2 - methyl acetate	Winkelmann (1884 c)	294 and 319
CO_2 - propionic acid	Winkelmann (1885)	366 and 372
CO_2 - n-butyl alcohol	Winkelmann (1885)	372
CO_2 - i-butyl alcohol	Winkelmann (1885)	340 and 357
CO_2 - ethyl ether	Baumgartner (1877 a)	291
	Winkelmann (1884 a)	283 and 293
	Trautz and Müller (1935)	290-293
CO_2 - CS_2	Baumgartner (1877 a)	290
	Baumgartner (1877 b)	267 to 313
CO_2 - n-butyric acid	Winkelmann (1885)	372
CO_2 - i-butyric acid	Winkelmann (1885)	371
CO_2 - ethyl acetate	Winkelmann (1884 c)	319
CO_2 - methyl propionate	Winkelmann (1884 c)	319 and 340
CO_2 - propyl formate	Winkelmann (1884 c)	319 and 340
CO_2 - n-amyl alcohol	Winkelmann (1885)	372
CO_2 - active amyl alcohol	Winkelmann (1885)	372

Table 16. (Continued)

System	Reference	T, °K
CO_2 - ethyl propionate	Winkelmann (1884 c)	340 and 363
CO_2 - methyl butyrate	Winkelmann (1884 c)	340 and 365
CO_2 - methyl i-butyrate	Winkelmann (1884 c)	323 and 340
CO_2 - i-valeric acid	Winkelmann (1885)	372
CO_2 - n-hexyl alcohol	Winkelmann (1885)	372
CO_2 - i-butyl acetate	Winkelmann (1884 c)	340 and 371
CO_2 - ethyl butyrate	Winkelmann (1884 c)	340 and 370
CO_2 - ethyl i-butyrate	Winkelmann (1884 c)	340 and 369
CO_2 - ethyl valerate	Winkelmann (1884 c)	371
CO_2 - propyl propionate	Winkelmann (1884 c)	370
CO_2 - $CHCl_3$	Baumgartner (1877 a)	291
CO_2 - i-butyl propionate	Winkelmann (1884 c)	371
CO_2 - propyl butyrate	Winkelmann (1884 c)	371
CO_2 - propyl-i-butyrate	Winkelmann (1884 c)	370
CO_2 - amyl propionate	Winkelmann (1884 c)	371
CO_2 - i-butyl butyrate	Winkelmann (1884 c)	371
CO_2 - i-butyl-i-butyrate	Winkelmann (1884 c)	371

Table 16. (Continued)

System	Reference	T, ^{o}K
CO_2 - propyl valerate	Winkelmann (1884 c)	371
CO_2 - SF_6	Suetin and Ivakin (1961)	291
	Suetin (1964)	291
	Ivakin and Suetin (1964 b)	291 to 472
CO_2 - amyl i-butyrate	Winkelmann (1884 c)	371
CO_2 - i-butyl valerate	Winkelmann (1884 c)	371
CO_2 - Br_2	Mackenzie and Melville (1932)	288
	Mackenzie and Melville (1933)	290
CO_2 - I_2	Vyshenskaya and Kosov (1965)	452 to 1275

Table 16. (Continued)

System	Reference	T, ^{o}K
N_2O - ethylene oxide	Wall and Kidder (1946)	298
N_2O - C_3H_8	Weissman (1964)	300 to 550
	Wall and Kidder (1946)	298
N_2O - Br_2	Mackenzie and Melville (1933)	290
C_3H_8 - n-hexane	Carmichael et al. (1955 b)	294 to 377
NO_2 - N_2O_4	Weissmann (1968 a)	303 to 343
C_2H_5OH - C_4H_9OH	Weissman (1968 a)	423
C_2H_5OH - CCl_2F_2	Lee and Wilke (1954)	298
$(CH_3)_2O$ - CH_3Cl	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
$(CH_3)_2O$ - SO_2	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH_3Cl - SO_2	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH_3Cl - C_2H_5Cl	Manner (1967)	298 to 419
1,3-butadiene - 1-butyne	Bournia et al. (1961)	300
C_3H_7OH - C_4H_9OH	Weissman (1968 a)	423

Table 16. (Continued)

System	Reference	T, ^oK
BF_3 - BF_3	Zmbov and Knežević (1961)	298 and 316
BF_3 - CCl_4	Raw (1955)	303
$(C_2H_5)_2O$ - $CHCl_3$	Weissman (1964)	293
C_6H_6 - CCl_4	Weissman (1964)	293
C_6H_6 - CCl_2F_2	Lee and Wilke (1954)	298
HBr - DBr	Braune and Zehle (1941)	294 - 296
cyclohexane - Rb	McNeal (1962)	323
	Violino (1968)	323
CH_2Cl_2 - CCl_4	Weissman (1964)	293 to 413
CF_4 - SF_6	Raw and Tang (1963)	303 to 342
i-octane - $C_6H_5NO_2$	Huber and van Vught (1965)	298
triethylamine - CCl_2F_2	Mehta (1966)	298
C_8H_{18} - C_7F_{16}	Weissman (1964)	303 and 323
BCl_3 - CCl_4	Raw (1955)	303
$CHCl_3$ - CCl_4	Weissman (1964)	293
Cl_2CF_2 - Cl_2CF_2	Miller and Carman (1961)	293
CCl_2F_2 - di-n-butyl phthalate	Birks and Bradley (1949)	293 and 303

Table 16. (Continued)

System	Reference	T, ^{o}K
Hg - Hg	Coulliette (1928)	338 to 376
	Biondi (1953)	350
	McCoubrey (1954)	473
	McCoubrey and Matland (1954)	473?
	Matland and McCoubrey (1955)	380 to 580
	McCoubrey and Matland (1956)	473
Hg - I_2	Mullaly and Jacques (1924)	292
$GeBr_2$ - $GeBr_4$	Jona (1965)	684
GeI_2 - GeI_4	Jona (1965)	684
UF_6 - UF_6	Ney and Armistead (1947)	297-301
	Brown and Murphy (1965)	273 to 344

Table 17. Selected High-Temperature Points for Curve-Fitting, Group I

System	$\log_{10}[\mathcal{D}_{12}(x = 1/2)]$	
	1000°K	10,000°K
He-Ne	0.930	2.680
He-Ar	0.785	2.555
He-Kr	0.700	2.510
He-Xe	0.630	2.435
Ne-Ar	0.410	2.150
Ne-Kr	0.320	2.065
Ne-Xe	0.250	2.000
Ar-Kr	0.070	1.810
Ar-Xe	0.010	1.730
Kr-Xe	-0.150	1.600
H_2-N_2	0.790	2.575

Table 18. Molecular-Beam Potentials, $\varphi(r) = Kr^s$, for Group I. [a]

	Amdur *et al.* [b]			Leonas *et al.* [c]		
System	K eV(Å)^s	s	Range Å	K eV(Å)^s	s	Range Å
He-Ne	38.3	7.97	1.52-1.86	10.3	5.61	1.3 -1.65
He-Ar	62.1	7.25	1.64-2.27	22.6	5.15	1.63-2.06
He-Ke	27.4	5.68	1.85-2.37	45.3	5.52	1.67-2.04
He-Xe	182	6.96	2.14-2.60	35.2	5.2	1.73-2.2
Ne-Ar	630	9.18	1.91-2.44	99.5	6.56	1.93-2.49
Ne-Kr	223	7.71	2.09-2.64	437	7.65	2.15-2.52
Ne-Xe	1480	8.98	2.39-2.87	210	6.76	2.0 -2.56
Ar-Kr	367	6.88	2.30-2.92	855	6.92	2.4 -3.1
Ar-Xe	2450	8.15	2.60-3.15	292	5.9	2.48-3.27
Kr-Xe	1060	6.70	2.72-3.37	875	7.1	2.44-3.0
H_2-N_2	191.0	7.19	1.96-2.46	88.1	6.63_5	1.84-2.50

[a] Complete reference information is given in Bibliography II.

[b] All results except He-Ar and Ne-Ar are calculated by combination rules.

[c] Only the potential for H_2-N_2 is calculated by combination rules.

Table 19. Values of $\mathcal{D}_{12}$ by Direct Measurement Compared with Those from Mixture Viscosity, and from Thermal Conductivity.

	Direct Measurement	Mixture Viscosity				Thermal Conductivity	
	van Heijningen *et al.*	Kestin	Trautz *et al.*	Thornton	van Itterbeek *et al.*	von Ubisch	Thornton
	(295°K)	(293°K)	(293°K)	(291°K)	(∿292°K)	(302°K)	(291°K)
System	Deviation from Reference Equations, Percent						
He-Ne	-0.8	-0.1_5	+2.3	+3.5	+1.4	+2.6	+18.8
He-Ar	-0.0_5	+0.1	+1.2	+1.0	+5.6	+4.9	+3.3
He-Kr	+1.0	+0.6	-	+3.2	-	+4.3	+9.1
He-Xe	+0.6	-	+0.9	+0.5	-	+1.3	+4.5
Ne-Ar	+0.1	-0.4	-0.1	-4.4	+1.8	+4.0	+6.7
Ne-Kr	-0.1	-	-	+1.5	-	+3.4	+0.45
Ne-Xe	+0.8	-	-	+0.1	-	+4.5	+2.1
Ar-Kr	+0.2	-	-	-0.3	-	+4.9	-4.7
Ar-Xe	+0.1	-	-	-1.1	-	+1.9	-2.8
Kr-Xe	$+0.0_5$	-	-	+0.2	-	-1.8	-0.2
H_2-N_2	+0.7	+0.5	-1.0	-	-0.8	-	-
|avg. dev.|	0.41	0.35	1.1	1.6	2.4	3.4	5.3 (3.8) [a]
index of reliability	1	1	3	4	6	8	13 (9) [a]

[a] Disregards large deviation (18.8%) of He-Ne.

Table 20. Diffusion Coefficients and Weights for Curve-Fitting, Group II.

System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note	System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note
$^3He-^4He$	2.64	-3.1325	1/4	a	$^3He-^4He$	473.15	0.5977	1	c
	4.15	-2.8125	1/4	a		573.15	0.7405	1	c
	1.74	-3.4789	1/4	b		673.15	0.8594	1	c
	2.00	-3.3665	1/4	b		773.15	0.9614	1	c
	2.31	-3.2396	1/4	b		873.15	1.0527	1	c
	2.66	-3.1355	1/4	b		1010.15	1.1550	1	c
	3.08	-3.0306	1/4	b		1121.15	1.2307	1	c
	3.96	-2.8386	1/4	b		2039	1.7127	1/4	d
	14.4	-1.9066	1	b		7746	2.7774	1/4	d
	19.6	-1.7012	1	b		3377	2.1038	1/4	e
	64.8	-0.8327	1	b		10,000	2.9983	1/4	e
	76.1	-0.7282	1	b		2444	1.8639	1/4	f
	192	-0.0742	1	b		10,000	2.9908	1/4	f
	296	0.2253	1	b		10,000	2.9895	1/4	g
	298.15	0.2550	1	c	$He-N_2$	77.2	-1.1331	2	h
	373.15	0.4214	1	c		251	-0.265	1	*

Table 20. (Continued)

System	T,°K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Weight	Note	System	T,°K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Weight	Note
$He-N_2$	317	-0.100	1	*	$He-CO_2$	200	-0.5229	1	k
	399	0.070	1	*		298.4	-0.2240	1	ℓ
	501	0.241	1	*	H_2-He	90.1	-0.7012	1	a
	631	0.405	1	*		194.7	-0.1264	1	m
	794	0.575	1	*		251.2	0.063	1	*
	1000	0.745	1	*		317	0.232	1	*
	3170	1.640	2/5	*		399	0.398	1	*
	10,000	2.530	1/5	*		501	0.567	1	*
He-CO	-	-	-	i		1000	1.080	1/3	*
$He-O_2$	317	-0.085	1	*		3170	1.970	1/3	*
	10,000	2.480	1	*		10,000	2.900	1/6	*
He-air	282	-0.1818	1	j	H_2-Ne	90.1	-0.8416	1	a
	355	-0.0119	1	j		9505	2.6599	1	n
	447	0.1584	1	j	H_2-Ar	251.2	-0.220	1	*
	1000	0.7582	1	j		317	-0.045	1	*
	10,000	2.4969	1	j		399	0.140	1	*

Table 20. (Continued)

System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note	System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note
H_2-Ar	501	0.320	1	*	H_2-D_2	17.04	-2.1945	1	q
	631	0.495	1	*		18.70	-2.1051	1	q
	794	0.670	1	*		20.32	-2.0329	1	q
	1000	0.845	1	*		90.0	-0.7721	1	q
	3170	1.710	1/3	*		26.09	-1.8097	1	r
	10,000	2.590	1/6	*		32.57	-1.6091	1	r
H_2-Kr	77.0	-1.270	1/5	*,ℓ		41.35	-1.4117	1	r
	100	-1.040	1/5	*,ℓ		48.06	-1.2832	1	r
	178	-0.563	1/5	*,ℓ		60.30	-1.1002	1	r
	290.7	-0.1688	1	o		70.32	-0.9851	1	r
	296.0	-0.1564	1	p		200.0	-0.1925	1	s
	562	0.334	1/5	*,ℓ		250.0	-0.0292	1	s
	3160	1.632	1/5	*		293.0	0.0864	1	s
	10,000	2.557	1/5	*		400	0.3181	1	s
H_2-D_2	14.12	-2.3675	1	q		500	0.4757	1	s
	15.47	-2.2832	1	q		763	0.7882	1	s

Table 20. (Continued)

System	T, °K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note
H_2-D_2	986	0.9741	1	s
	3313	1.9047	1	t
	5000	2.2305	1	t
	10,000	2.7796	1	t
H_2-CO	-	-	-	u
H_2-air	282	-0.1487	1	v
	355	0.0253	1	v
	447	0.1987	1	v
	1000	0.8048	1	v
	10,000	2.5635	1	v
H_2-CO_2	200.0	-0.5017	1	w
	298.15	-0.1898	1	x
	473.0	0.1673	1	y
N_2-Ar	316	-0.664	1	*
N_2-Ar	3160	1.088	1	*
N_2-CO	77.65	-1.7747	1/5	z
	194	-0.980	1/2	*
	251	-0.790	1	*
	316	-0.619	1	*
	398	-0.450	1	*
	562	-0.195	1/2	*
	1000	0.226	1/5	*
	10,000	1.979	1/10	*
N_2-CO_2	298.15	-0.7825	1	x
	447	-0.450	1/2	*
	708	-0.095	1/2	*
	1000	0.1553	1/2	aa
	1800	0.5832	1/2	aa

Table 20. (Continued)

[*] Selected value, see explanation in first part of Section 5.4.

[a] Weissman and Mason (1962 b).

[b] Bendt (1958)

[c] Calculated from viscosity data by Kalelkar and Kestin (1969).

[d] Calculated from molecular-beam potential by Amdur and Harkness (1954).

[e] Calculated from molecular-beam potential by Amdur *et al*. (1961 a).

[f] Calculated from molecular-beam potential by Belyaev and Leonas (1967 b).

[g] Calculated from molecular-beam potential by Kamnev and Leonas (1965 a).

[h] Wasik and McCulloh (1969).

[i] Reference equation of $He-N_2$ is suitable because of isoteric molecules.

[j] Calculated from reference equations for $He-N_2$ and $He-O_2$ according to Blanc's law, Eq. (2.1-7).

[k] Calculated from temperature dependence of thermal diffusion factor (Saxena and Mason, 1959) according to the iterative method by Annis *et al*. (1968), and results are normalized to measurement of $\mathcal{D}_{12}$ by Annis *et al*. (1969).

[ℓ] Annis *et al*. (1969).

[m] Amdur and Malinauskas (1965).

[n] Calculated from molecular-beam potential by Amdur *et al*., see Table 21.

Table 20. (Continued)

[o] Fedorov *et al.* (1966).

[p] Annis *et al.* (1968).

[q] Calculated from HD viscosity data by Becker and Misenta (1955).

[r] Calculated from HD viscosity data by Coremans *et al.* (1958 b).

[s] Calculated from H_2 viscosity data as summarized by Mason and Rice (1954).

[t] Calculated from molecular-beam potential by Amdur *et al.*, see Table 21.

[u] Reference equation of H_2-N_2 is suitable because of isoteric molecules.

[v] Calculated from reference equations for H_2-N_2 and H_2-O_2 according to Blanc's law, Eq. (2.1-7).

[w] Calculated from temperature dependence of thermal diffusion factor (Saxena and Mason, 1959) according to the iterative method by Annis *et al.* (1968), and results are normalized to measurement of $\mathscr{D}_{12}$ by Boyd *et al.* (1951).

[x] Boyd *et al.* (1951).

[y] Ivakin and Suetin (1964 b).

[z] Winn (1950).

[aa] Pakurar and Ferron (1966); Ferron (1967).

Table 21. Molecular-Beam Potentials, $\varphi(r) = Kr^s$, for Group II. [a,b]

System	Potential			Source			Reference
	K, eV(Å)^s	s	range, Å	system	K, eV(Å)^s	s	
$^3He-^4He$	4.71	5.94	1.27-1.59	$^4He-^4He$	4.71	5.94	Amdur and Harkness (1954)
	4.33	5.86	1.10-1.53	$^4He-^4He$	4.33	5.86	Belyaev and Leonas (1967 b)
He-N_2	74.3	7.06	1.79-2.29	direct measurement			Amdur *et al*. (1957)
	48.8	6.63	1.72-2.29	He-He	4.33	5.86	Belyaev and Leonas (1967 b)
				N_2-N_2	550	7.4	Belyaev and Leonas (1966 a)
He-CO	40.3	5.91	1.55-2.26	CO-Ar	551	6.99	Jordan *et al*. (1970)
				He-Ar	62.1	7.25	Amdur *et al*. (1954)
				Ar-Ar	849	8.33	Amdur and Mason (1954)
	92.24	7.045	1.705-2.225	He-He	4.33	5.86	Belyaev and Leonas (1967 b)
				CO-CO	1965	8.23	Belyaev *et al*. (1967)
He-O_2	99.5	7.26	1.47-2.08	He-Ar	62.1	7.25	Amdur *et al*. (1954)
				Ar-O_2	1360	8.34	Jordan *et al*. (1970)
				Ar-Ar	849	8.33	Amdur and Mason (1954)
	32.24	6.08	1.72-2.34	He-He	4.33	5.86	Belyaev and Leonas (1967 b)
				O_2-O_2	240	6.3	Belyaev and Leonas (1967 a)

Table 21. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s	s	
H_2-He	12.11	6.07	1.44-1.76	direct measurement			Amdur and Smith (1968)
	5.0	3.8	1.15-1.89	direct measurement			Belyaev and Leonas (1967 b)
H_2-Ne	98.55	8.095	1.685-2.03	He-He	4.71	5.94	Amdur and Harkness (1954)
				Ne-Ne	312	9.99	Amdur and Mason (1955 a)
				He-H_2	12.11	6.07	Amdur and Smith (1968)
	21	4.70	1.45-2.215	He-H_2	5	3.8	Belyaev and Leonas (1967 b)
				Ne-Ne	78	7.65	Kamnev and Leonas (1965 a)
				He-He	4.33	5.86	Belyaev and Leonas (1967 b)
H_2-Ar	160	7.38	1.81-2.44	He-H_2	12.11	6.07	Amdur and Smith (1968)
				He-Ar	62.1	7.25	Amdur _et al_. (1954)
				He-He	4.71	5.94	Amdur and Harkness (1954)
	49.1	5.96_5	$1.80-2.54_5$	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b)
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a)
H_2-Kr	70.37	5.81	$2.01_5-2.53_5$	He-H_2	12.11	6.07	Amdur and Smith (1968)
				Kr-Kr	159	5.42	Amdur and Mason (1955 b)
				He-He	4.71	5.94	Amdur and Harkness (1954)

Table 21. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s	s	
H_2-Kr	89.33	4.72	1.80-2.66	He-H_2	5	3.8	Belyaev and Leonas (1967 b)
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a)
				He-He	4.33	5.86	Belyaev and Leonas (1967 b)
H_2-D_2	31.55	6.19	1.62-1.96	He-H_2	12.11	6.07	Amdur and Smith (1968)
				He-D_2	12.27	6.06	Amdur and Smith (1968)
				He-He	4.71	5.94	Amdur and Harkness (1954)
	14.1	5.87	1.34-1.95	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b)
H_2-CO	107.4	5.81	1.89-2.17	Ar-CO	551	6.99	Jordan *et al*. (1970)
				He-H_2	12.11	6.07	Amdur and Smith (1968)
				He-Ar	62.1	7.25	Amdur *et al*. (1954)
	166.4	7.05	1.82_5-2.43_5	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b)
				CO-CO	1965	8.23	Belyaev *et al*. (1967)
N_2-Ar	755	7.78	2.28-2.83	direct measurement			Amdur *et al*. (1957)
	1050	8.16	2.12-2.67	direct measurement			Belyaev and Leonas (1967 a)
N_2-CO	2038	8.70	2.22-2.77	direct measurement			Belyaev *et al*. (1967)
	596	7.27	2.43-3.07	N_2-N_2	596	7.27	Amdur *et al*. (1957)

Table 21. (Continued)

[a] Potentials were not determined for air-(He,H_2) and CO_2-(He,H_2,N_2) because molecular-beam measurements were unavailable.

[b] Complete reference information is given in Bibliography II.

Table 22. Diffusion Coefficients and Weights for Curve-Fitting, Group III.[a]

System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
Ar-CH_4	307.15	-0.6655	b
	8660	1.9232	c
Ar-CO	-	-	d
Ar-O_2	316	-0.670	*
	3160	1.066	*
Ar-air	282	-0.7520	e
	355	-0.5784	e
	447	-0.4029	e
	1000	0.2095	e
	10,000	1.9576	e
Ar-CO_2	276.2	-0.8775	f
	317.2	-0.7820	f
	328.0	-0.7328	g
	348.0	-0.6819	g
	373.0	-0.6289	g
	410.0	-0.5528	g
Ar-CO_2	455.0	-0.4737	g
	473.0	-0.4401	g
	1100	0.2122	h
	1800	0.5752	h
Ar-SF_6	328.0	-1.0000	g
	348.0	-0.9508	g
	373.0	-0.8962	g
	410.0	-0.8210	g
	447.0	-0.7375	g
	472.0	-0.6981	g
	1000	-0.0985	i
	4640	1.0086	i
H_2-Xe	242.2	-0.3872	j
	274.2	-0.2941	j
	303.9	-0.2132	j
	341.2	-0.1244	j

Table 22. (Continued)

System	T, °K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note	System	T, °K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
H_2-Xe	293.2	-0.2277	k	H_2-SF_6	344.4	-0.2441	g
	400.0	0.0065	k		376.0	-0.1791	g
	500.0	0.1847	k		401.0	-0.1249	g
	550.0	0.2529	k		429.0	-0.0680	g
	2320	1.2989	i		473.0	0.0128	g
	8290	2.2856	i		1320	0.7185	i
H_2-CH_4	316	-0.092	*		7460	1.9782	i
	10,000	2.556	*	CH_4-He	316	-0.130	*
H_2-O_2	316	-0.050	*		3160	1.620	*
	3160	1.682	*	CH_4-N_2	316	-0.625	*
H_2-SF_6	298.15	-0.3788	ℓ		10,000	2.000	*
	286.2	-0.4023	m	CH_4-O_2	293.6_5	-0.6676	n
	306.9	-0.3391	m		395	-0.4168	n
	370.8	-0.1891	m		402	-0.3936	n
	418.0	-0.0768	m		408	-0.3990	n
	313.0	-0.3006	g		517	-0.2122	n

Table 22. (Continued)

System	T,°K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
CH_4-O_2	521	-0.2048	n
	534	-0.1993	n
	668	-0.0101	n
	669	-0.0031	n
	707	0.0378	n
	708	0.0418	n
	768	0.0831	n
	771	0.0973	n
	840	0.1523	n
	842	0.1467	n
	845	0.1399	n
	3550	1.250	*
	10,000	2.000	*
CH_4-air	282	-0.7077	e
	355	-0.5317	e
	447	-0.3551	e
CH_4-air	1000	0.2577	e
	10,000	2.0009	e
CH_4-SF_6	297.58	-0.9566	o
	357.92	-0.8119	o
	418.19	-0.6872	o
	477.87	-0.5784	o
	2045	0.5159	i
	5000	1.0212	i
N_2-Ne	293.15	-0.4989	p
	6090	1.7973	i
N_2-Kr	316	-0.770	*
	5620	1.438	*
N_2-Xe	316	-0.855	*
	7940	1.650	*
N_2-O_2	316	-0.638	*
	3160	1.086	*

Table 22. (Continued)

System	T, °K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note	System	T, °K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
N_2-SF_6	328.0	-0.9393	g	CO-CO_2	293.3_5	-0.7854	q
	348.0	-0.8861	g		293.1_5	-0.8027	q
	373.0	-0.8327	g		315.4	-0.7328	g
	410.0	-0.7471	g		348.0	-0.6536	g
	455.0	-0.6615	g		373.0	-0.5969	g
	473.0	-0.6364	g		410.0	-0.5302	g
	1000	-0.0640	i		455.0	-0.4449	g
	3960	0.9294	i		473.0	-0.4191	g
CO-Kr	-	-	d	CO-SF_6	296.8	-1.0521	g
CO-O_2	-	-	d		315.4	-0.9788	g
CO-air	282	-0.7077	e		348.0	-0.9031	g
	355	-0.5376	e		373.4	-0.8416	g
	447	-0.3686	e		410.0	-0.7595	g
	1000	0.2279	e		455.0	-0.6737	g
	10,000	1.9727	e		473.0	-0.6536	g
CO-CO_2	281.6_5	-0.8187	q		1000	-0.0640	i

Table 22. (Continued)

System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note	System	T,°K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
$CO\text{-}SF_6$	3960	0.9294	i	$O_2\text{-}CO_2$	864	0.0302	n
$O_2\text{-}CO_2$	286.9_5	-0.8069	q		867	0.0546	n
	287.1_5	-0.8097	q		874	0.0492	n
	287.1_5	-0.8041	q		1080	0.2084	n
	296.5_5	-0.7932	r		1081	0.2098	n
	287.8	-0.8125	s		1083	0.2049	n
	296	-0.8069	n	$O_2\text{-}SF_6$	297	-1.0044	t
	409	-0.5287	n		317	-0.9626	t
	419	-0.5229	n		340	-0.8894	t
	430	-0.4935	n		379	-0.7959	t
	596	-0.2457	n		408	-0.7144	t
	612	-0.2111	n		2930	0.658	i
	635	-0.1858	n		6310	1.212	i
	649	-0.1838	n	$CO_2\text{-air}$	282	-0.8300	e
	768	-0.0434	n		355	-0.6387	e
	770	-0.0297	n		501	-0.3636	e

Table 22. (Continued)

System	T, °K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note	System	T, °K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
CO_2-air	708	-0.0992	e	CO_2-N_2O	500.0	-0.5143	x
	1000	0.1575	e		550.0	-0.4401	x
	1590	0.4921	e	CO_2-SF_6	328.0	-1.1113	g
CO_2-N_2O	287.9_5	-0.9586	q		348.0	-1.0595	g
	287.9_5	-0.9582	q		373.0	-1.0088	g
	293.1_5	-0.9520	q		410.0	-0.9281	g
	288.1_5	-0.9706	u		447.0	-0.8446	g
	298.1_5	-0.9318	v		472.0	-0.8210	g
	194.8	-1.2790	w	SF_6-He	316	-0.345	*
	273.2	-0.9974	w		10,000	2.095	*
	312.8	-0.9052	w	SF_6-air	300	-1.0097	e
	312.8	-0.8887	w		500	-0.5901	e
	362.6	-0.7657	w		700	-0.3298	e
	300.0	-0.9318	x		1000	-0.0630	e
	400.0	-0.6946	x		10,000	1.5599	e

Table 22. (Continued)

[*] Selected value, see explanation in first part of Section 5.4.

[a] All listed values of $\mathcal{D}_{12}$ are weighted one; except in H_2-SF_6 for which the datum by Boyd *et al.* (1951) at 298.15°K is weighted ten.

[b] Arnold and Toor (1967).

[c] Mason and Amdur (1964).

[d] Reference equation of N_2-(gas) is suitable because of isoteric molecules.

[e] Calculated from reference equations for N_2-(gas) and O_2-(gas) according to Blanc's law, Eq. (2.1-7).

[f] Holsen and Strunk (1964).

[g] Ivakin and Suetin (1964 b).

[h] Pakurar and Ferron (1966) and Ferron (1967).

[i] Calculated from molecular-beam potential by Amdur *et al.*, see Table 23.

[j] Paul and Srivastava (1961 c).

[k] Weissman and Mason (1962 b).

[ℓ] Boyd *et al.* (1951); weight of datum ten for least-squares calculations.

[m] Strehlow (1953).

[n] Walker and Westenberg (1960, 1966).

[o] Manner (1967).

Table 22. (Continued)

[p] DiPippo *et al*. (1967).

[q] Loschmidt (1870 b).

[r] Wretschko (1870).

[s] Suetin and Ivakin (1961).

[t] Ivakin *et al*. (1968).

[u] Boardman and Wild (1937).

[v] Wall and Kidder (1946).

[w] Amdur *et al*. (1952).

[x] Weissman (1964).

Table 23. Molecular-Beam Potentials, $\varphi(r) = Kr^s$, for Group III.^{a,b}

System	Potential			Source			Reference
	$K, eV(Å)^s$	s	range, Å	system	$K, eV(Å)^s$	s	
Ar-CH_4	936	7.85	2.31-2.66	direct measurement			Mason and Amdur (1964)
Ar-CO	551	6.99	2.09-2.68	direct measurement			Jordan *et al*. (1970)
	580	7.14_5	2.28_5-3.03	CO-CO	1965	8.23	Belyaev *et al*. (1967)
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a)
Ar-O_2	1360	8.34	2.01-2.50	direct measurement			Jordan *et al*. (1970)
	5000	9.9	2.15-2.63	direct measurement			Belyaev and Leonas (1967 a)
Ar-SF_6	24.5×10^5	12.8	3.24-4.04	He-SF_6	1.86×10^5	11.48	Amdur (1967)
				He-Ar	62.1	7.25	Amdur *et al*. (1954)
				He-He	4.71	5.94	Amdur and Harkness (1954)
H_2-Xe	468.5	7.08_5	2.31-2.76_5	He-H_2	12.11	6.07	Amdur and Smith (1968)
				Xe-Xe	7.05×10^3	7.97	Amdur and Mason (1956 a)
				He-He	4.71	5.94	Amdur and Harkness (1954)
	51.7	4.04_5	1.84-2.67	He-H_2	5	3.8	Belyaev and Leonas (1967 b)
				Xe-Xe	463	6.35	Kamnev and Leonas (1966 a)
				He-He	4.33	5.86	Belyaev and Leonas (1967 b)
H_2-CH_4	1548	9.56	2.09-2.54	He-CH_4	602	9.43	Amdur *et al*. (1961 b)

Table 23. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s		
				$He-H_2$	12.11	6.07	Amdur and Smith (1968)
				He-He	4.71	5.94	Amdur and Harkness (1954)
H_2-O_2	265	7.16	1.81-1.99	$Ar-O_2$	1360	8.34	Jordan <u>et al</u>. (1970)
				$He-H_2$	12.11	6.07	Amdur and Smith (1968)
				He-Ar	62.1	7.25	Amdur <u>et al</u>. (1954)
	58.2	6.1	1.84-2.55	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b)
				O_2-O_2	240	6.3	Belyaev and Leonas (1967 a)
H_2-SF_6	$4.78x10^5$	11.61	3.04-3.53	$He-SF_6$	$1.86x10^5$	11.48	Amdur (1967)
				$He-H_2$	12.11	6.07	Amdur and Smith (1968)
				He-He	4.71	5.94	Amdur and Harkness (1954)
CH_4-He	602	9.43	1.92-2.37	direct measurement			Amdur <u>et al</u>. (1961 b)
CH_4-N_2	832	7.30	2.41-2.80	$Ar-N_2$	755	7.78	Amdur <u>et al</u>. (1957)
				$Ar-CH_4$	936	7.85	Mason and Amdur (1964)
				Ar-Ar	849	8.33	Amdur and Mason (1954)
CH_4-O_2	1500	7.86	2.14-2.47	$Ar-CH_4$	936	7.85	Mason and Amdur (1964)
				$Ar-O_2$	1360	8.34	Jordan <u>et al</u>. (1970)

Table 23. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s	s	
				Ar-Ar	849	8.33	Amdur and Mason (1954)
CH_4-SF_6	28.0x10^5	12.08	3.43-3.75	He-SF_6	1.86x10^5	11.48	Amdur (1967)
				Ar-CH_4	936	7.85	Mason and Amdur (1964)
				He-Ar	62.1	7.21	Amdur *et al.* (1954)
Ne-N_2	605	9.08_5	2.03_5-2.56	He-N_2	74.3	7.06	Amdur *et al.* (1957)
				He-He	4.71	5.94	Amdur and Harkness (1954)
				Ne-Ne	312	9.99	Amdur and Mason (1955 a)
	207	7.52_5	2.02-2.61_5	Ne-Ne	78	7.65	Kamnev and Leonas (1965 a)
				N_2-N_2	550	7.4	Belyaev and Leonas (1967 a)
N_2-Kr	432	6.80	2.36_5- 3.06_5	He-N_2	74.3	7.06	Amdur *et al.* (1957)
				Kr-Kr	159	5.42	Amdur and Mason (1955 b)
				He-He	4.71	5.94	Amdur and Harkness (1954)
	872	7.55	2.37-3.03_5	N_2-N_2	550	7.4	Belyaev and Leonas (1967 a)
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a)
N_2-Xe	2874	8.07_5	2.66-3.29_5	He-N_2	74.3	7.06	Amdur *et al.* (1957)
				He-He	4.71	5.94	Amdur and Harkness (1954)

Table 23. (Continued)

System	Potential			Source			Reference
	K, eV($\text{\AA}$)^s	s	range, $\text{\AA}$	system	K, eV($\text{\AA}$)^s	s	
				Xe-Xe	7050	7.97	Amdur and Mason (1956 a)
	505	6.87_5	2.41-3.07	N_2-N_2	550	7.4	Belyaev and Leonas (1967 a)
				Xe-Xe	463	6.35	Kamnev and Leonas (1966 a)
N_2-O_2	1630	8.15	2.16-2.52	Ar-O_2	1360	8.34	Jordan *et al*. (1970)
				He-N_2	74.3	7.06	Amdur *et al*. (1957)
				He-Ar	62.1	7.25	Amdur *et al*. (1954)
	330	6.8	2.34-3.05	direct measurement			Belyaev and Leonas (1967 a)
N_2-SF_6	29.3×10^5	12.60	3.39-4.06	He-SF_6	1.86×10^5	11.48	Amdur (1967)
				He-N_2	74.3	7.06	Amdur *et al*. (1957)
				He-He	4.71	5.94	Amdur and Harkness (1954)
CO-Kr	238	5.53_5	2.21-2.90_5	Ar-CO	551	6.99	Jordan *et al*. (1970)
				Ar-Ar	849	8.33	Amdur and Mason (1954)
				Kr-Kr	159	5.42	Amdur and Mason (1955 b)
	1648	7.96_5	2.35_5-2.97	CO-CO	1965	8.23	Belyaev *et al*. (1967)
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966)
CO-O_2	883	7.00	1.92-2.49	CO-Ar	551	6.99	Jordan *et al*. (1970)

Table 23. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s	s	
				O_2-Ar	1360	8.34	Jordan *et al*. (1970)
				Ar-Ar	849	8.33	Amdur and Mason (1954)
	687	7.26_5	2.32_5-3.03_5	CO-CO	1965	8.23	Belyaev *et al*. (1967)
				O_2-O_2	240	6.3	Belyaev and Leonas (1967 a)
CO-SF_6	16.5×10^5	11.22	3.32-3.77	He-SF_6	1.86×10^5	11.48	Amdur (1967)
				Ar-CO	551	6.99	Jordan *et al*. (1970)
				He-Ar	62.1	7.25	Amdur *et al*. (1954)
O_2-SF_6	40.7×10^5	12.53	3.24-3.59	He-SF_6	1.86×10^5	11.48	Amdur (1967)
				Ar-O_2	1360	8.30	Jordan *et al*.(1970)
				He-Ar	62.1	7.25	Amdur et al. (1954)
He-SF_6	1.86×10^5	11.48	2.87-3.36	direct measurement			Amdur (1967)

[a] Potentials were not determined for air-(Ar,CH_4,CO,SF_6) and CO_2-(Ar,CO,O_2,air,N_2O,SF_6) because molecular-beam measurements were unavailable.

[b] Complete reference information is given in Bibliography II.

Table 24. Diffusion Coefficients and Weights for Curve-Fitting, Miscellaneous Group.[a]

System	T,°K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note	System	T,°K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
H_2O-N_2	281.9	-0.6554	b	H_2O-O_2	1069	0.3879	d
	298.2	-0.5965	b		1069	0.3939	d
	327.5	-0.5158	b		1070	0.3928	d
	327.5	-0.5200	b	H_2O-air	-	-	e
	327.4	-0.5131	b	H_2O-CO_2	307.4_5	-0.6947	f
	353.2	-0.4436	b		328.5_5	-0.6757	f
	373.4	-0.4029	b		352.3_5	-0.6108	f
H_2O-O_2	450.0	-0.2261	c		328.6_5	-0.7033	g
	511	-0.1367	d		349.1_5	-0.5901	g
	529	-0.1146	d		1000	0.3617	h
	698	+0.0792	d		1100	0.4099	h
	715	0.1007	d		1200	0.4639	h
	722	0.1096	d		1300	0.5198	h
	908	0.2683	d		1400	0.5775	h
	917	0.2721	d		1500	0.6325	h
	921	0.2813	d		1600	0.6884	h

Table 24. (Continued)

System	T, °K	$\log_{10}[\ {}_{12}(x=1/2)]$	Note	System	T, °K	$\log_{10}[\ {}_{12}(x=1/2)]$	Note
H_2O-CO_2	1700	0.7482	h	H-Ar	282	0.075	*
CO_2-Ne	175	-0.9914	i		4620	2.015	*
	625	-0.0097	i	H-H_2	274	0.2667	m
CO_2-C_3H_8	298.1$_5$	-1.0665	j		10,000	2.966	*
	298.1$_5$	-1.0620	j	N-N_2	280	-0.5376	n
	300.0	-1.0492	k	O-N_2	2850	1.250	*,n
	400.0	-0.8097	k	O-O_2			
	500.0	-0.6364	k	O-He	316	0.043	*
	550.0	-0.5575	k		10,000	2.6665	o
H-He	275	0.3766	ℓ	O-Ar	316	-0.522	*
	4620	2.500	*		3760	1.457	*

Table 24. (Continued)

[*] Selected value, see explanation in first part of Section 5.4.

[a] All listed values of $\mathcal{D}_{12}$ are weighted one; except in H_2O-CO_2 for which the data between 1000° and 1700°K inclusive, are weighted two. For these systems, in almost all instances, the composition dependence of $\mathcal{D}_{12}$ is insignificant; maximum correction is 0.45% for H_2O-O_2 datum at 1070°K.

[b] O'Connell *et al.* (1969).

[c] Common point for low- and high-temperature correlations, see text (Section 5.4, part d).

[d] Walker and Westenberg (1960, 1966).

[e] See Section 5.4, part d.

[f] Schwertz and Brow (1951).

[g] Crider (1956).

[h] Ferron (1967); weight of datum doubled for least-squares calculations.

[i] Calculated from data on temperature dependence of thermal diffusion factor (Weissman *et al.*, 1961) and method of Annis *et al.* (1968). The reference equation lower temperature limit is reported at 195°K which is in agreement with the correct temperature limit of the thermal diffusion factor data; the equation was verified to be correct, even though a value of T of 175°K was used in the curve-fitting calculations.

[j] Wall and Kidder (1946).

Table 24. (Continued)

[k] Weissman (1964).

[ℓ] Morgan and Schiff (1964).

[m] Browning and Fox (1964).

[n] Morgan and Schiff (1946), average of results for $N\text{-}N_2$, $O\text{-}N_2$, and $O\text{-}O_2$.

[o] Calculated from potential of Leonas *et al.*, see Table 25.

Table 25. Molecular-Beam Potentials, $\varphi(r) = Kr^s$, for Miscellaneous Group.[a,b]

System	Potential			Source			
	$K, eV(\text{Å})^s$	s	range, Å	system	$K, eV(\text{Å})^s$	s	Reference
H-He	2.34	3.29	1.16-1.71	direct measurement			Amdur and Mason (1956 b)
	1.2	2.7	0.79-1.35	direct measurement			Belyaev and Leonas (1967 b)
H-Ar	31.6	4.49	2.1-3.0	as reported			Mason and Vanderslice (1958)
	6.26	1.99	1.32-1.88	He-H	1.2	2.7	Belyaev and Leonas (1967 b)
				He-Ar	22.6	5.15	Kamnev and Leonas (1965 a)
				He-He	4.33	5.86	Belyaev and Leonas (1967 b)
$H-H_2$	6.02	3.42	1.33-1.88	He-H	2.34	3.29	Amdur and Mason (1956 b)
				$He-H_2$	12.11	6.07	Amdur and Smith (1968)
				He-He	4.71	5.94	Amdur and Harkness (1954)
	0.91	4.15	1.00-1.24	direct measurement			Belyaev and Leonas (1967 b)
$N-N_2$	76.6	6.31	1.76-2.54	direct measurement			Belyaev and Leonas (1966 b)
O-He	38.0	7.99	1.20-1.60	Ar-O	239	8.09	Belyaev *et al*. (1967)
				He-He	4.33	5.86	Belyaev and Leonas (1967 b)
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a)
O-Ar	239	8.09	1.78-2.40	direct measurement			Belyaev *et al*. (1967)
$O-N_2$	22.5	5.0	2.00-2.48	direct measurement			Belyaev and Leonas (1966 b)

Table 25. (Continued)

System	Potential			Source			Reference
	K,eV(Å)^s	s	range, Å	system	K,eV(Å)^s	s	
$O-O_2$	13.25	4.4	2.05-2.46	direct measurement			Belyaev and Leonas (1966 b)

[a] Potentials were not determined for $H_2O-(N_2,O_2,air,CO_2)$ and $CO_2-(N_2O,C_3H_8)$ because molecular-beam measurements were unavailable.

[b] Complete reference information is given in Bibliography II.

Fig. 1. Qualitative Temperature Dependence of Diffusion Coefficients.

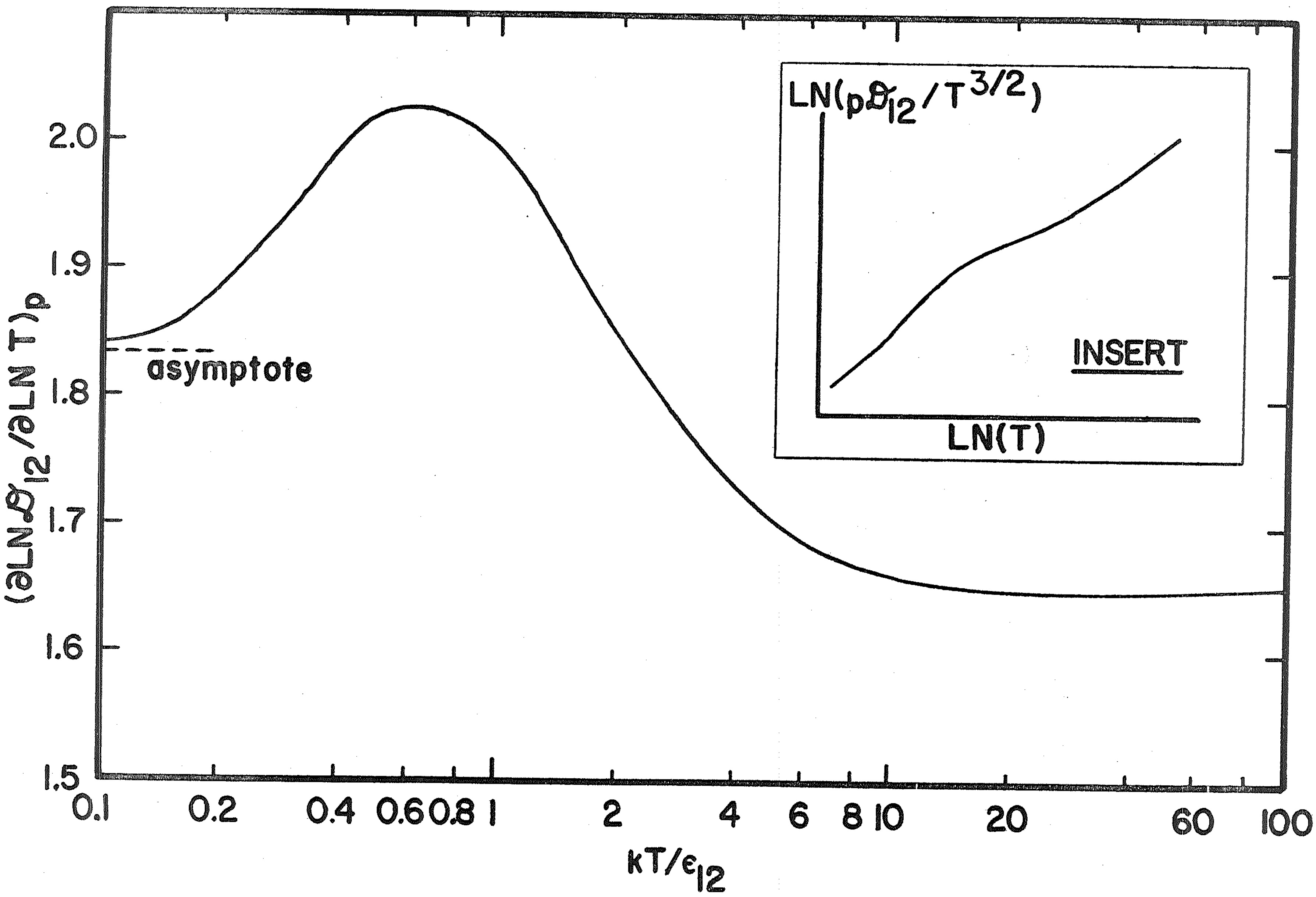

2.0
1.9
1.8
1.7
1.6
1.5
$(\partial LN\mathcal{D}_{12}/\partial LN\ T)_p$
asymptote
0.1
0.2
0.4
0.6
0.8
1
2
4
6
8
10
20
60
100
kT/ϵ_{12}
$LN(p\mathcal{D}_{12}/T^{3/2})$
LN(T)
INSERT

Fig. 2. Principal Experimental Methods
for Diffusion Coefficients.

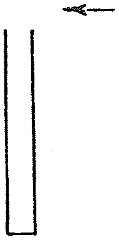
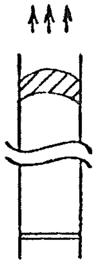
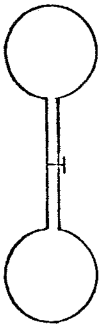
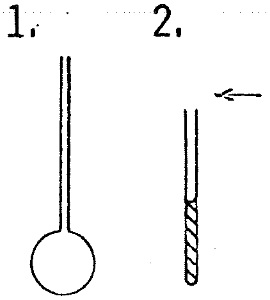
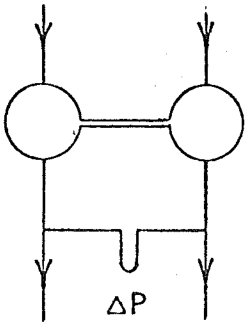
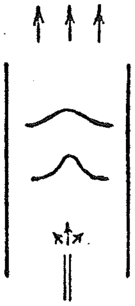
TYPE TIME BEHAVIOR	TWO-SIDED	ONE-SIDED	FLOW
UNSTEADY	 CLOSED TUBE	 OPEN TUBE	 PULSE SOURCE (GAS CHROMATOGRAPHY)
QUASI-STEADY	 TWO-BULB	 1. CAPILLARY LEAK 2. EVAPORATION TUBE	
STEADY	 DIFFUSION BRIDGE		 POINT SOURCE

Fig. 3. Composition Dependence of Δ_{12} at Two Mass Ratios (m). Eq. (2.2-10) is the exact formula of the Chapman-Enskog theory; Eq. (4.2-1) is the semi-empirical expression of this report.

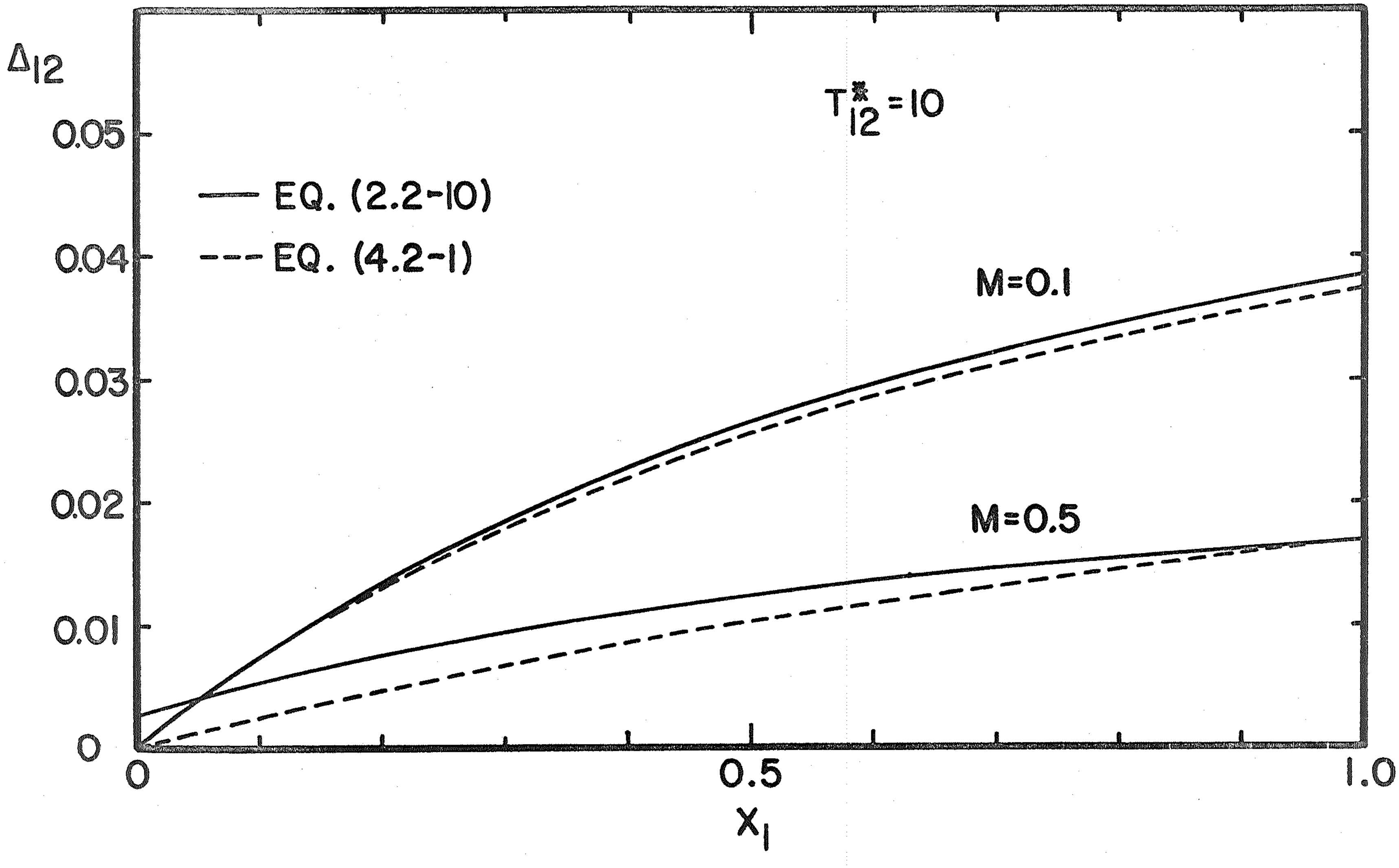
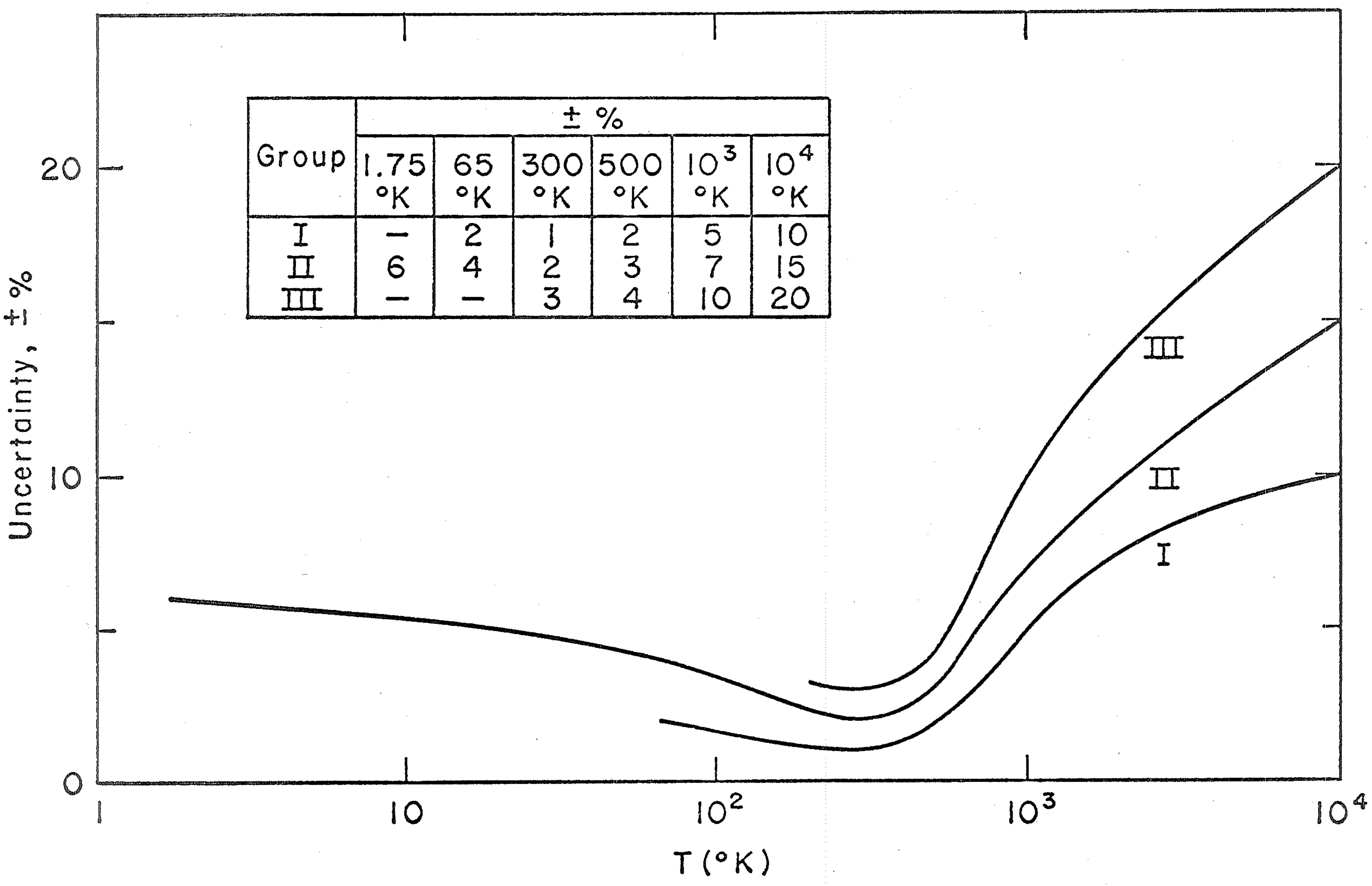

Δ_{12}
0.05
0.04
0.03
0.02
0.01
0
$T^*_{12}=10$
EQ. (2.2-10)
EQ. (4.2-1)
M=0.1
M=0.5
0
0.5
1.0
X_1

Fig. 4. Estimated Uncertainty Limits of $\mathscr{D}_{12}$ as a Function of Temperature.



Group	± %					
	1.75 °K	65 °K	300 °K	500 °K	10^3 °K	10^4 °K
I	—	2	1	2	5	10
II	6	4	2	3	7	15
III	—	—	3	4	10	20

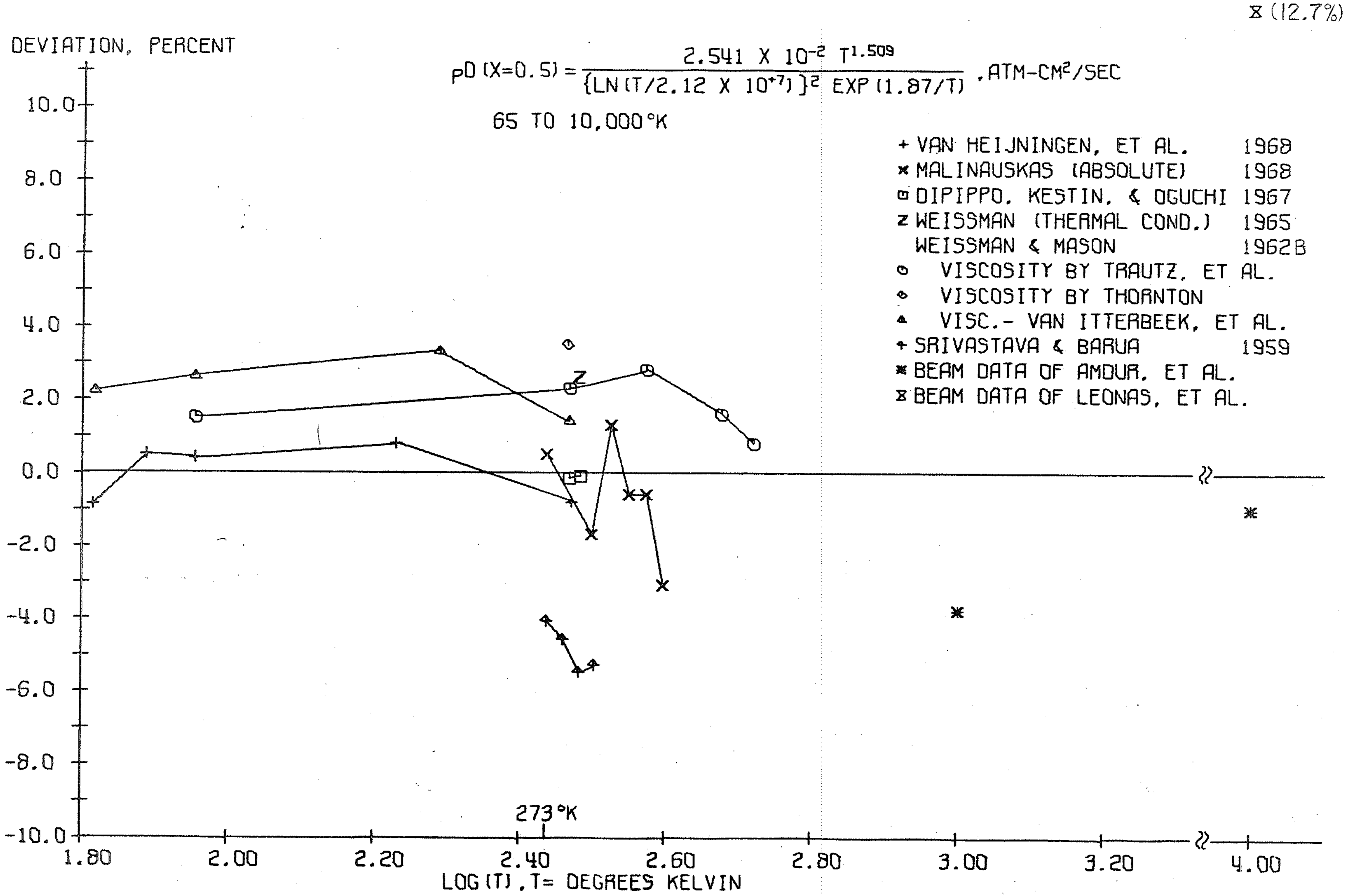


FIG. 5. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - NEON

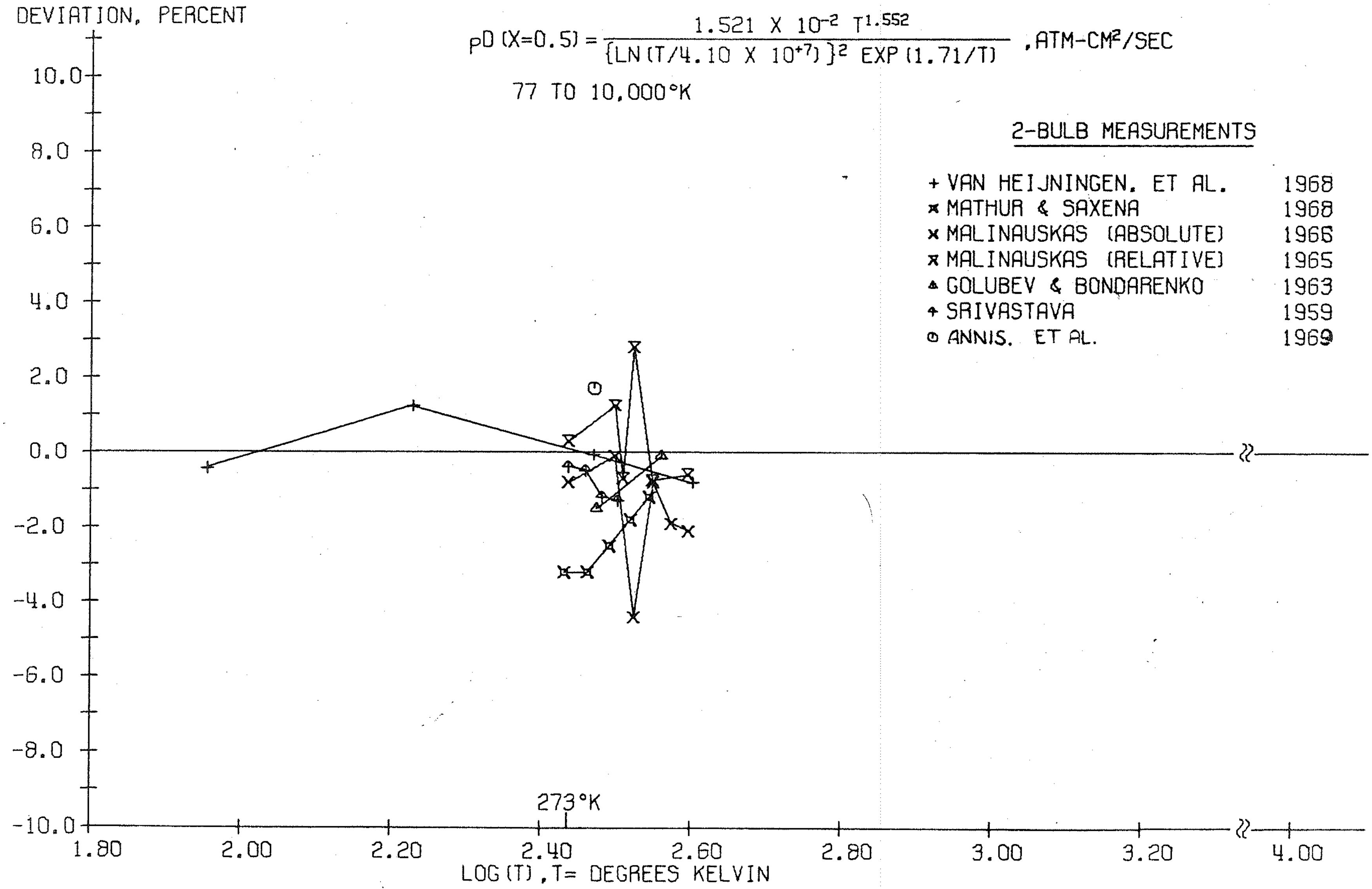


FIG. 6. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - ARGON

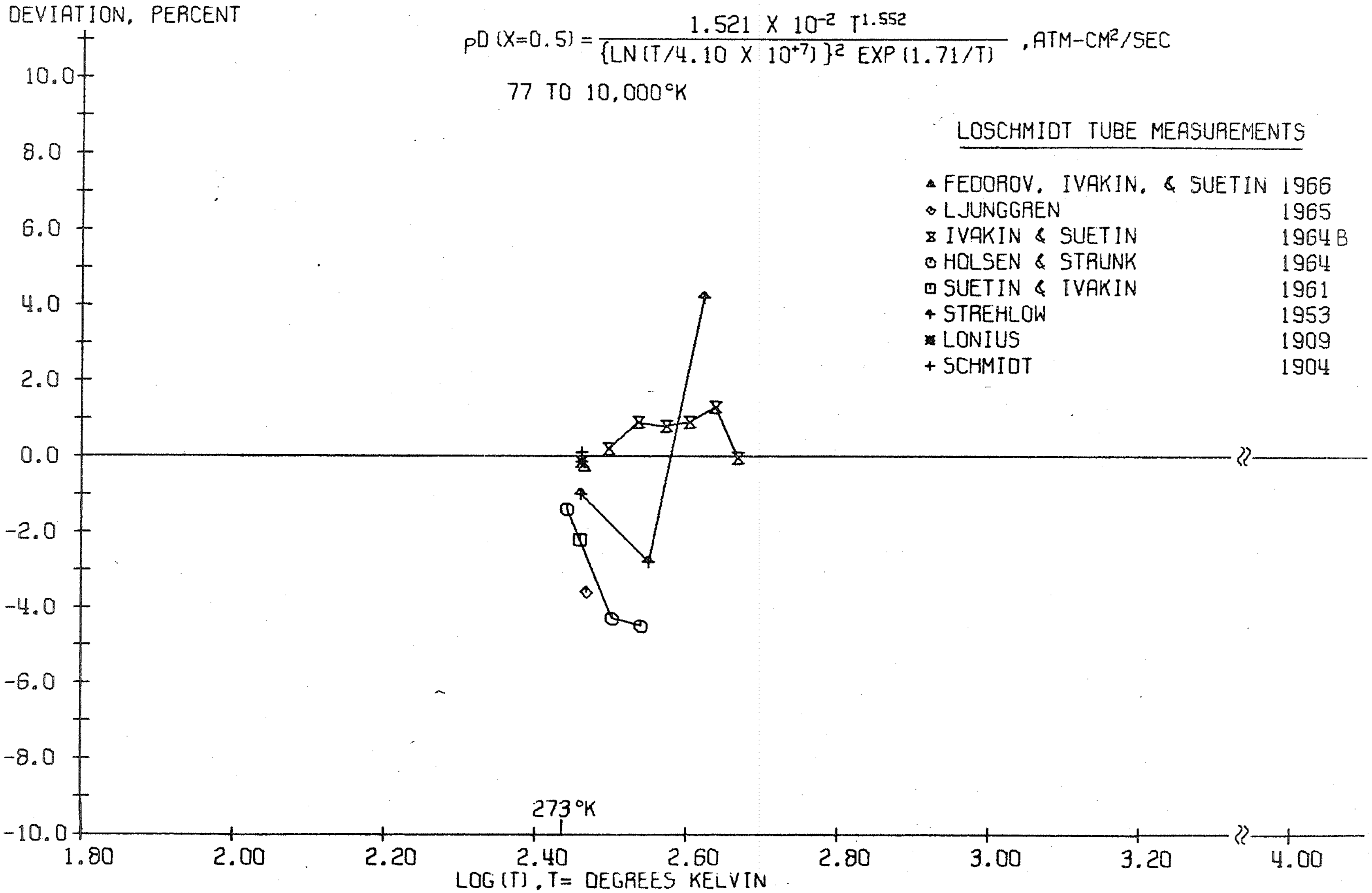


FIG. 7. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - ARGON

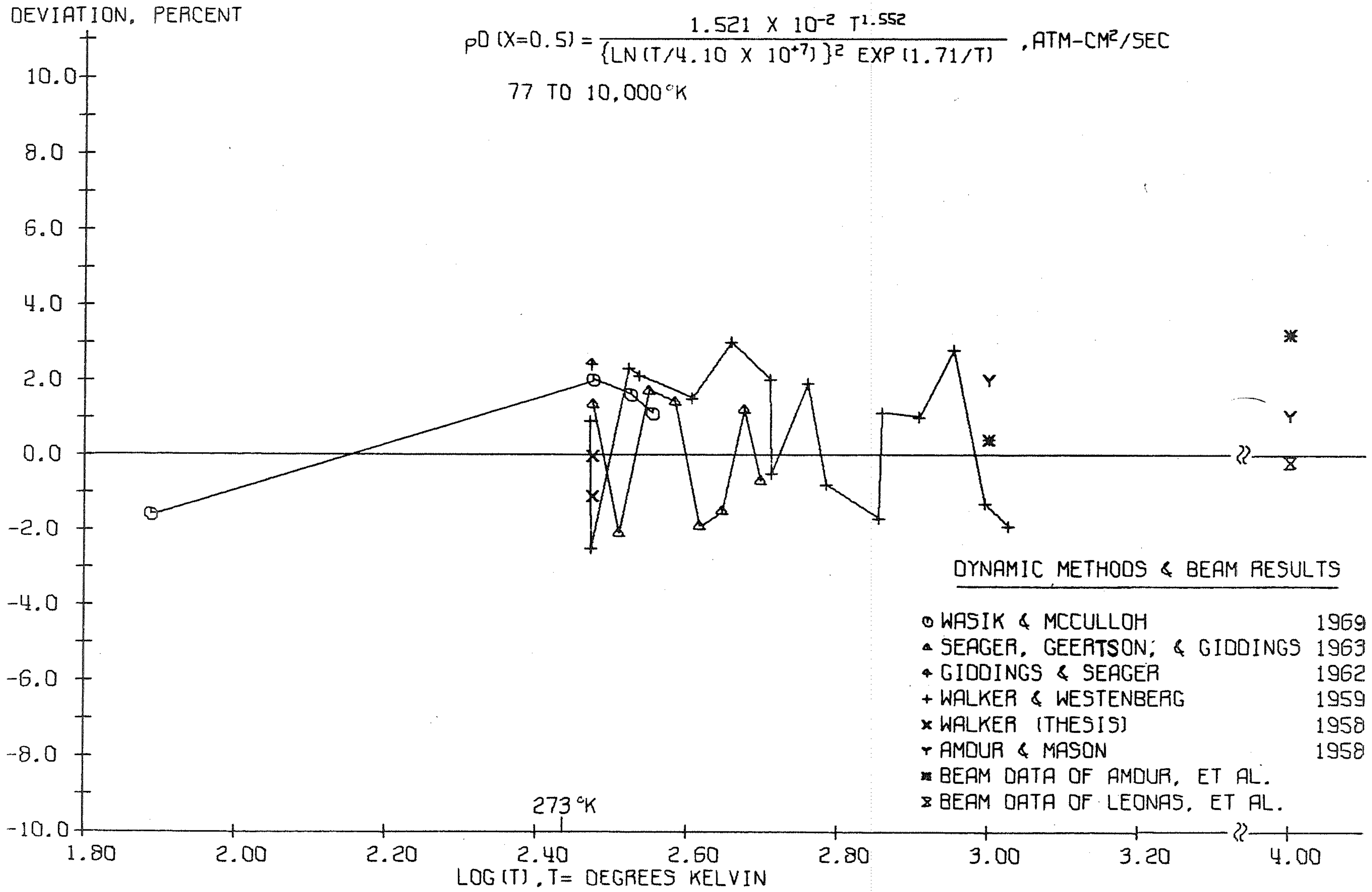


FIG. 8. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - ARGON

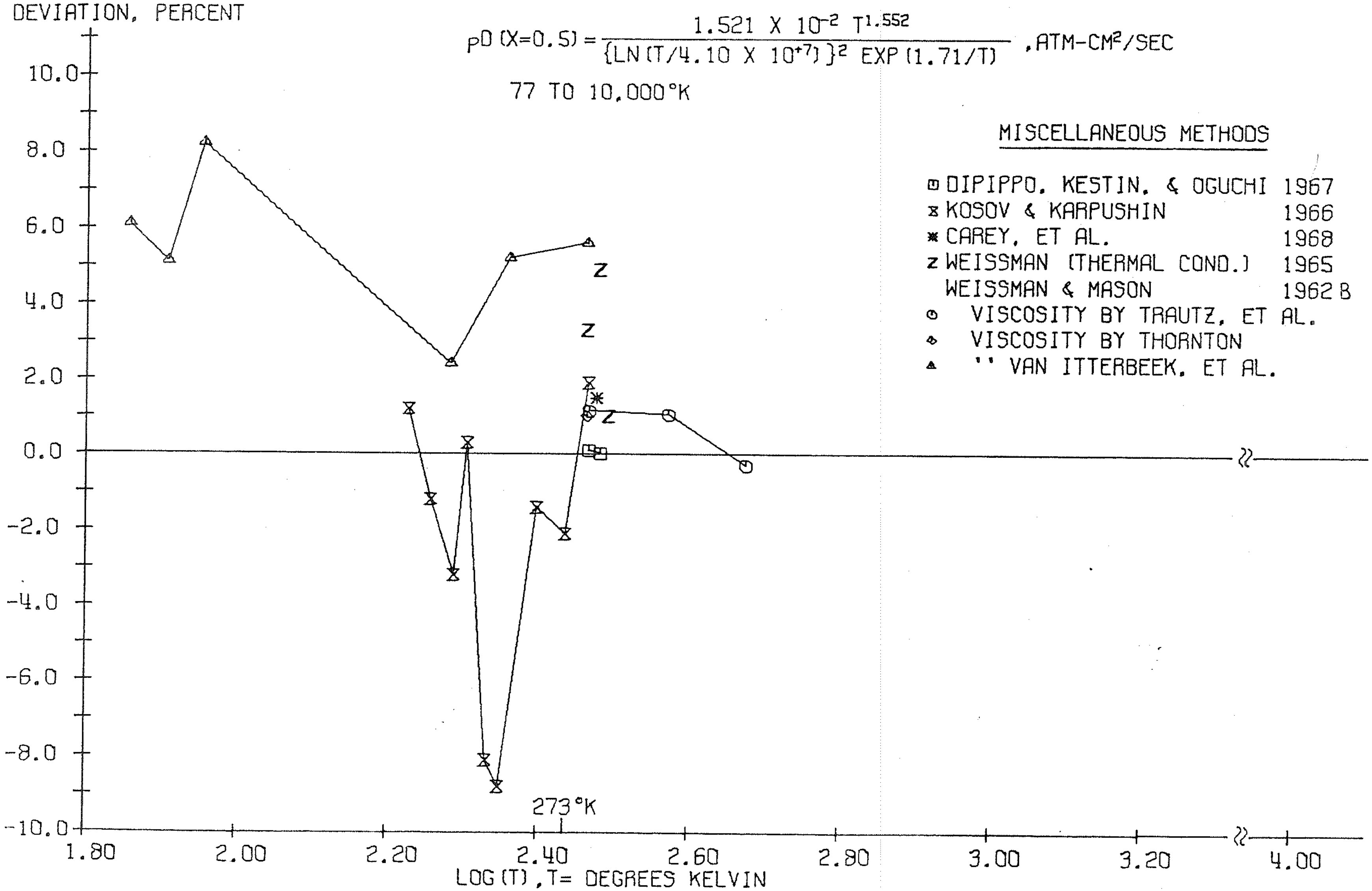


FIG. 9. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - ARGON

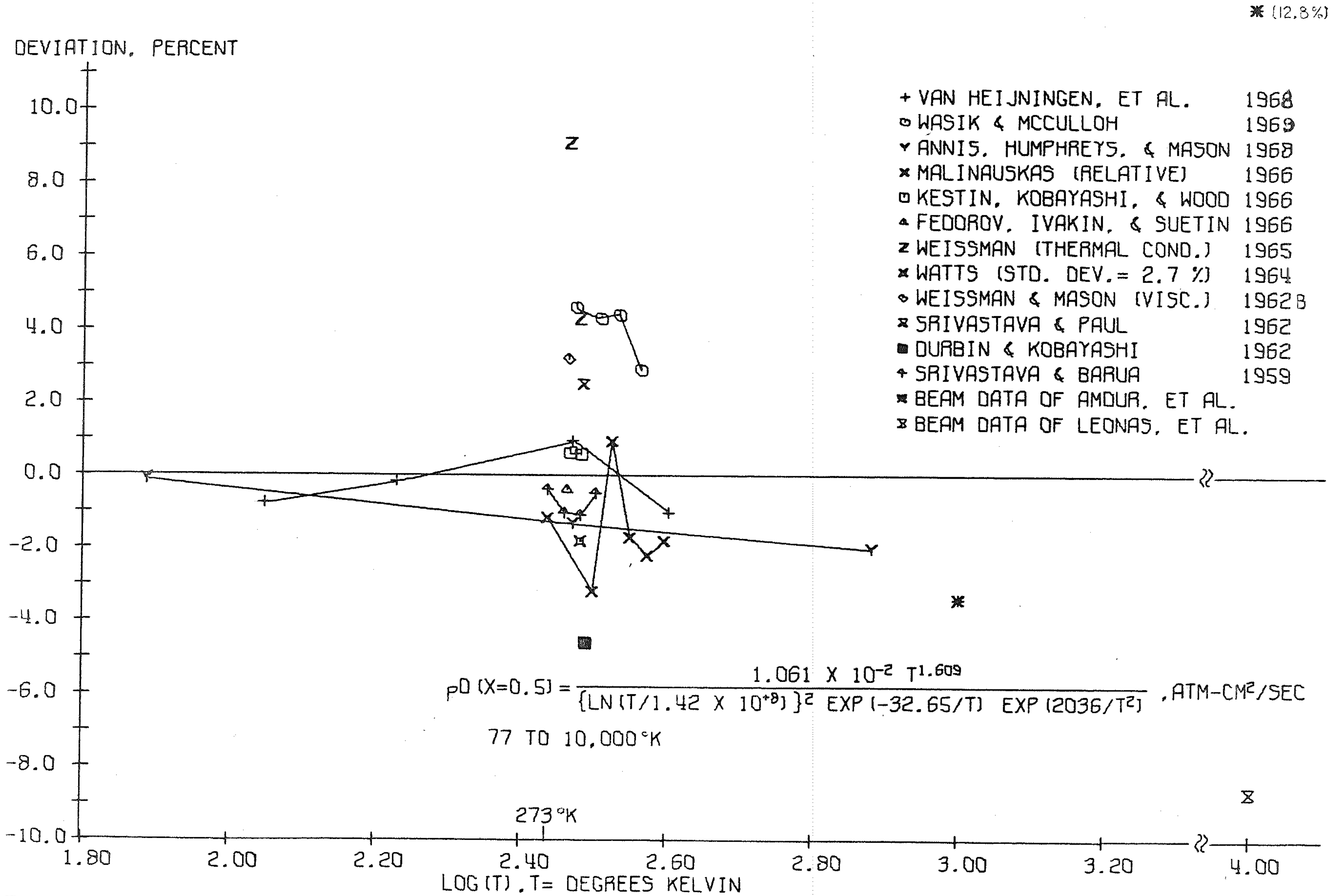


FIG. 10. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - KRYPTON

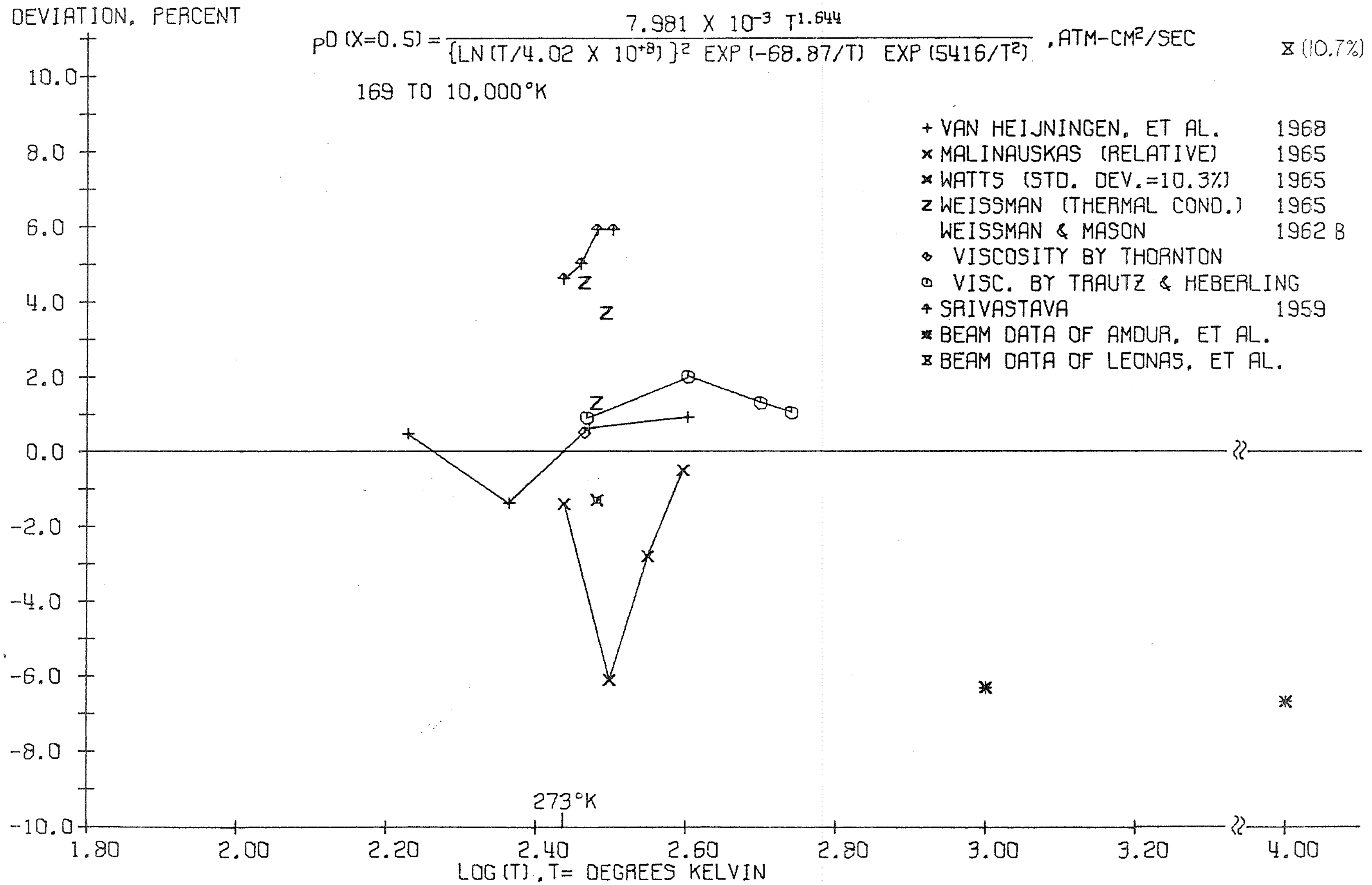


FIG. 11. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - XENON

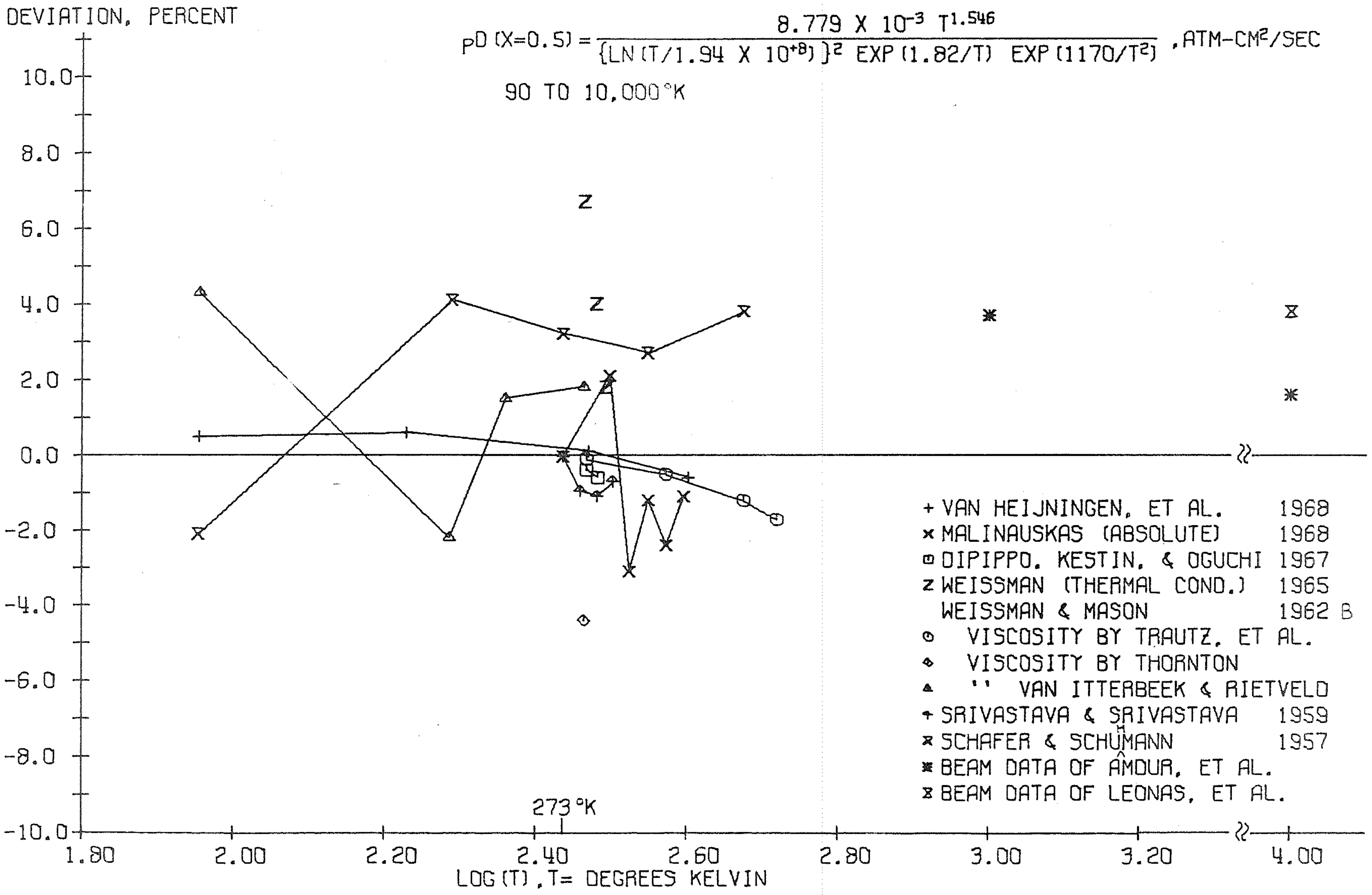


FIG. 12. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NEON – ARGON

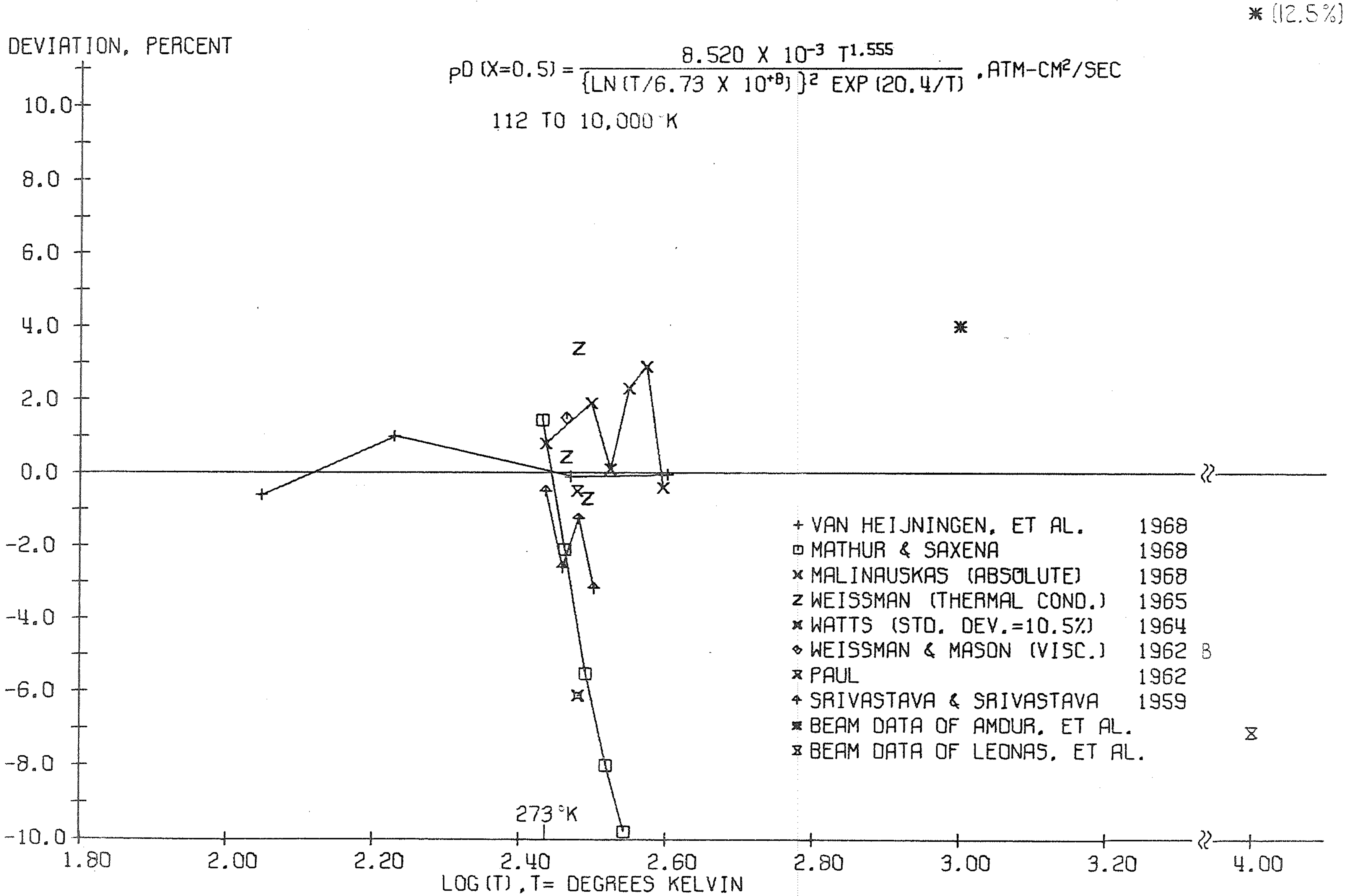


FIG. 13. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NEON - KRYPTON

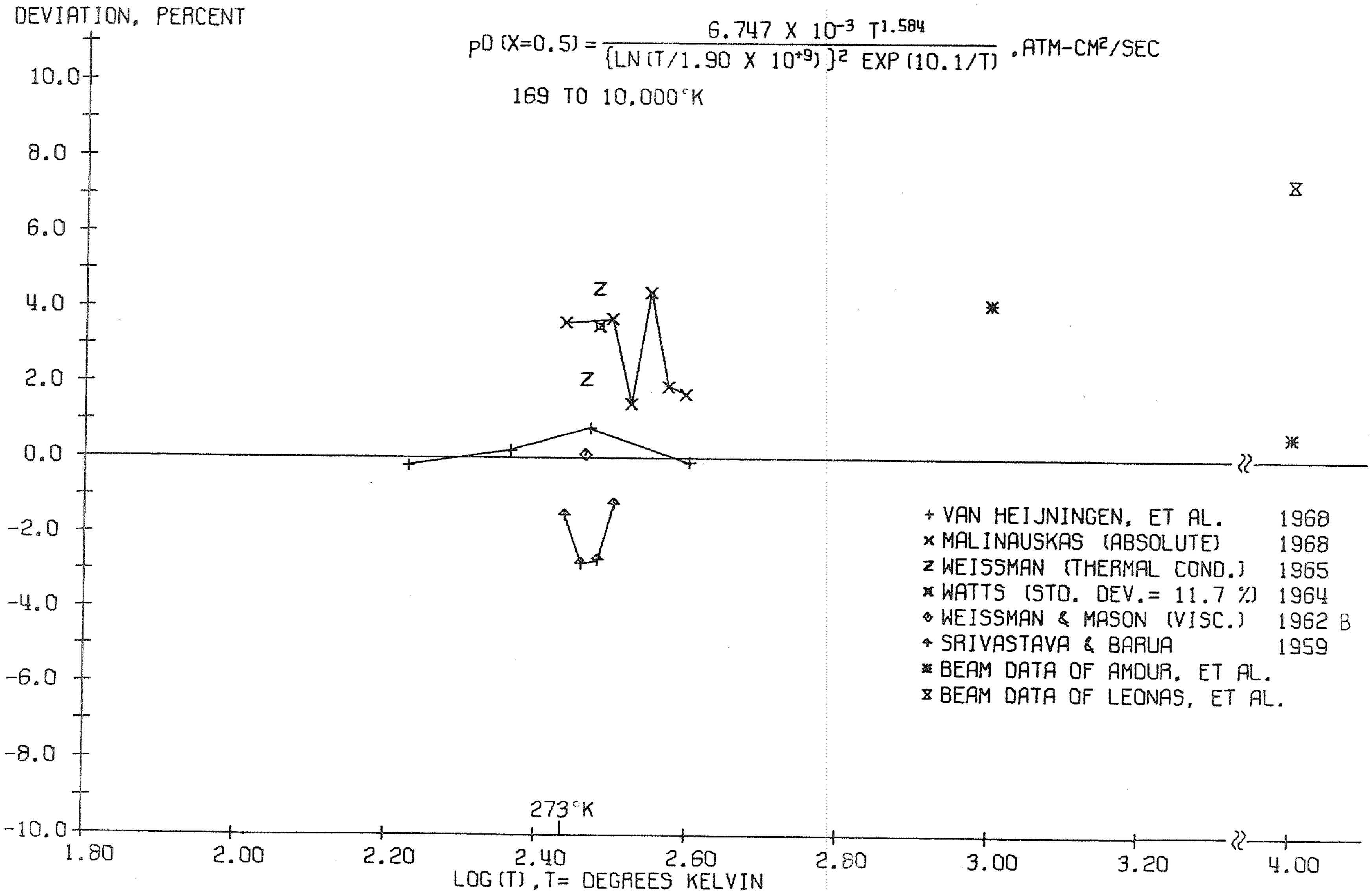


FIG. 14. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NEON - XENON

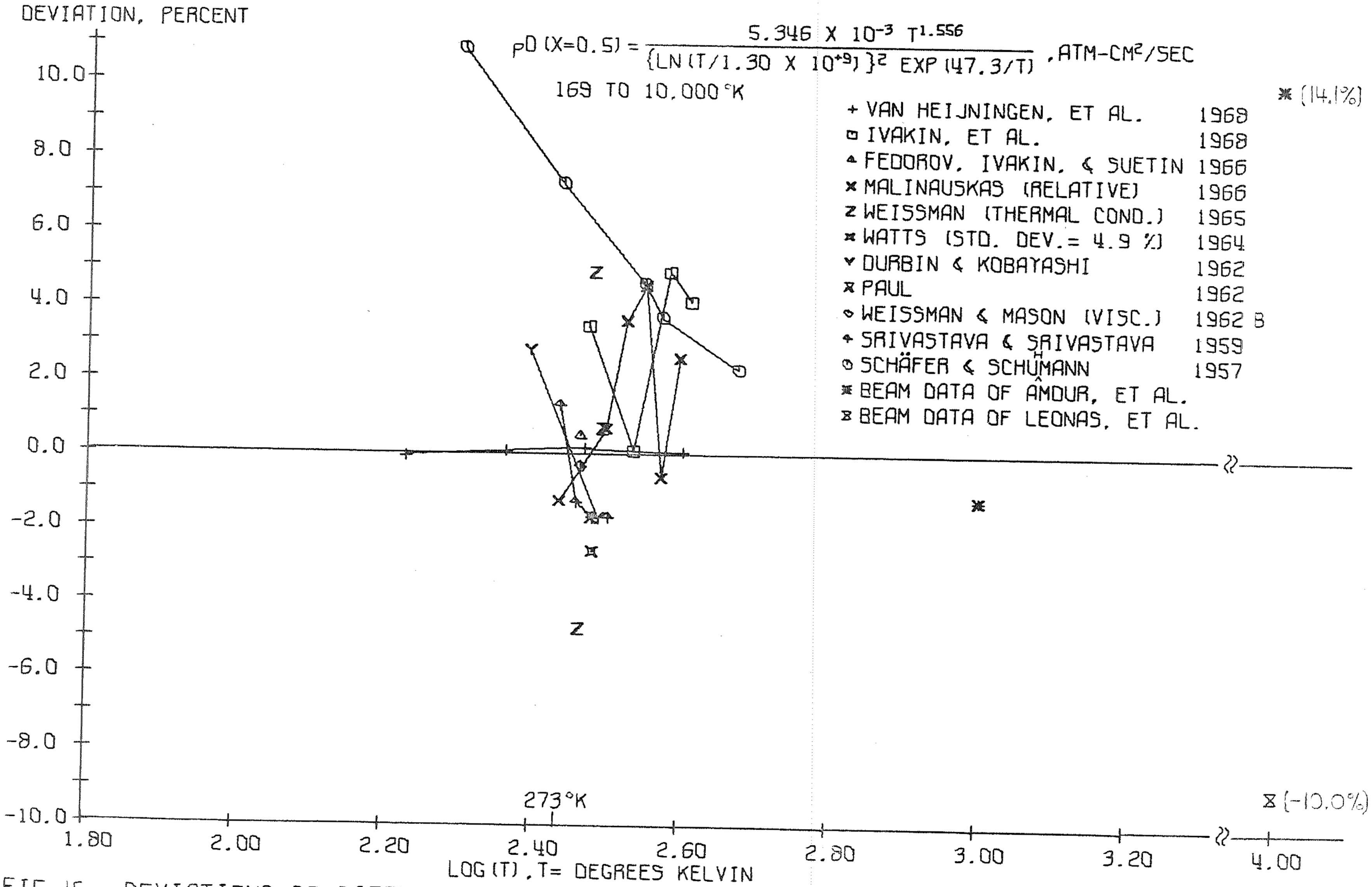


FIG. 15. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, ARGON - KRYPTON

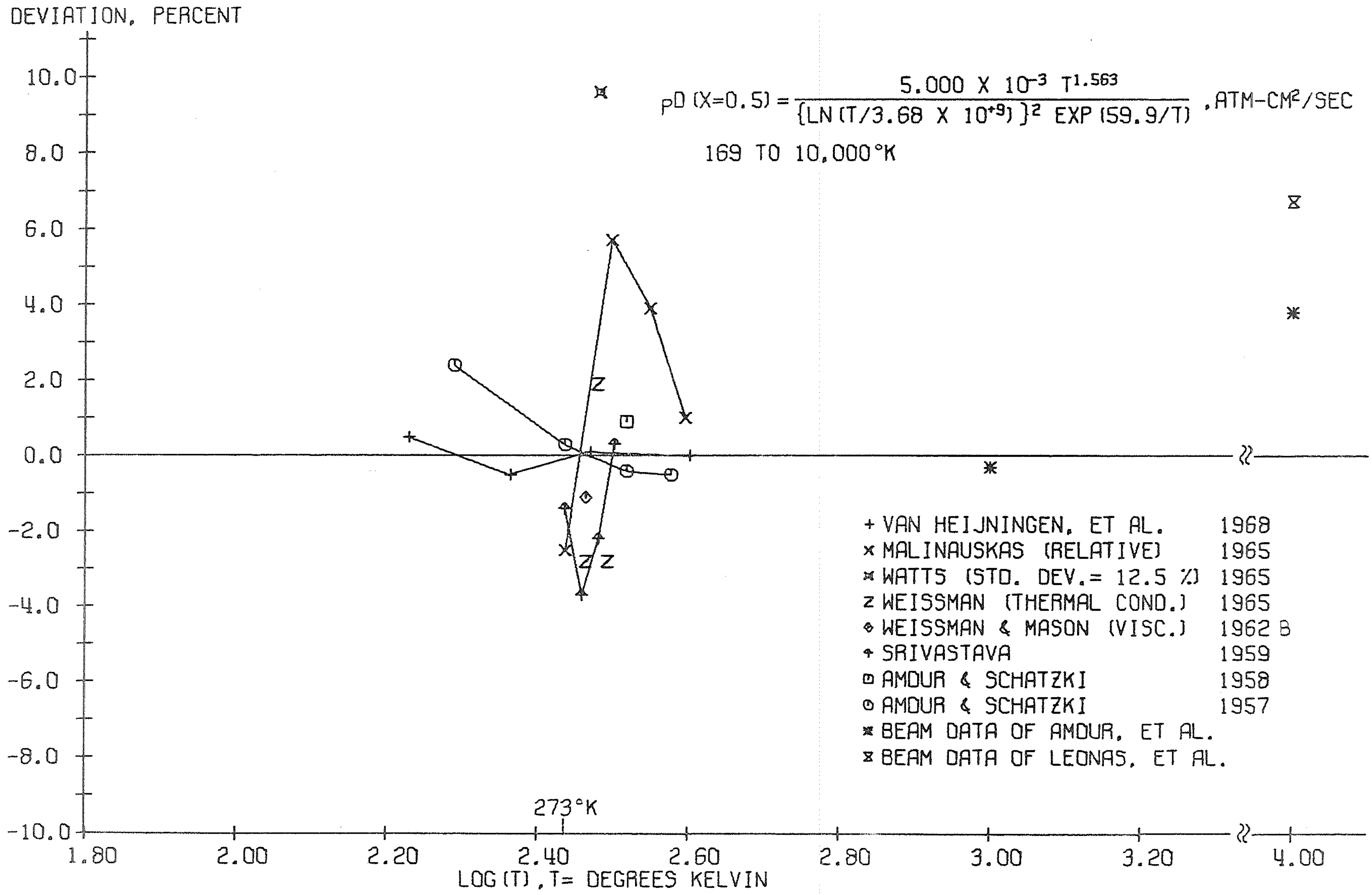


FIG. 16. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, ARGON - XENON

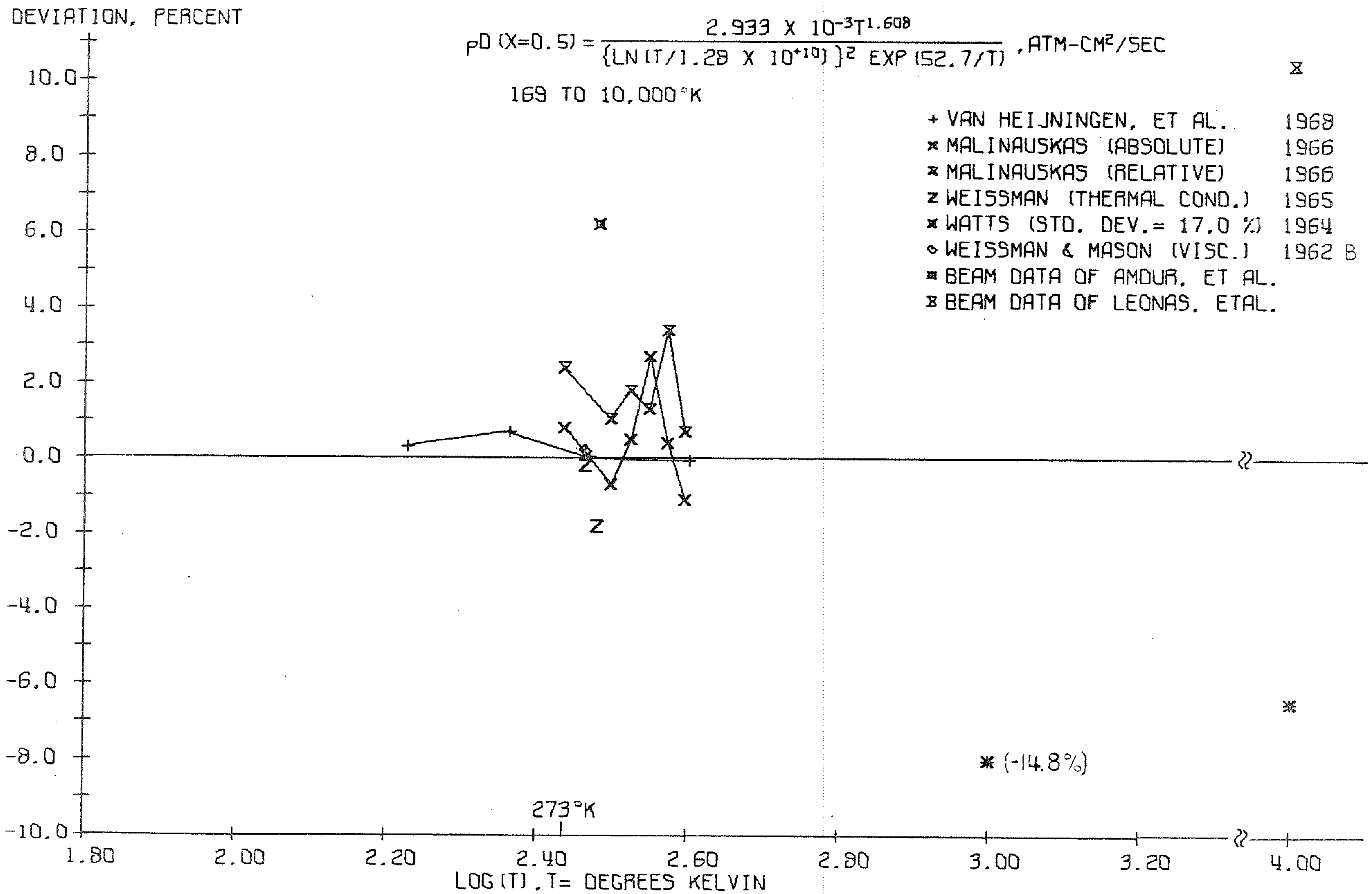


FIG. 17. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, KRYPTON - XENON

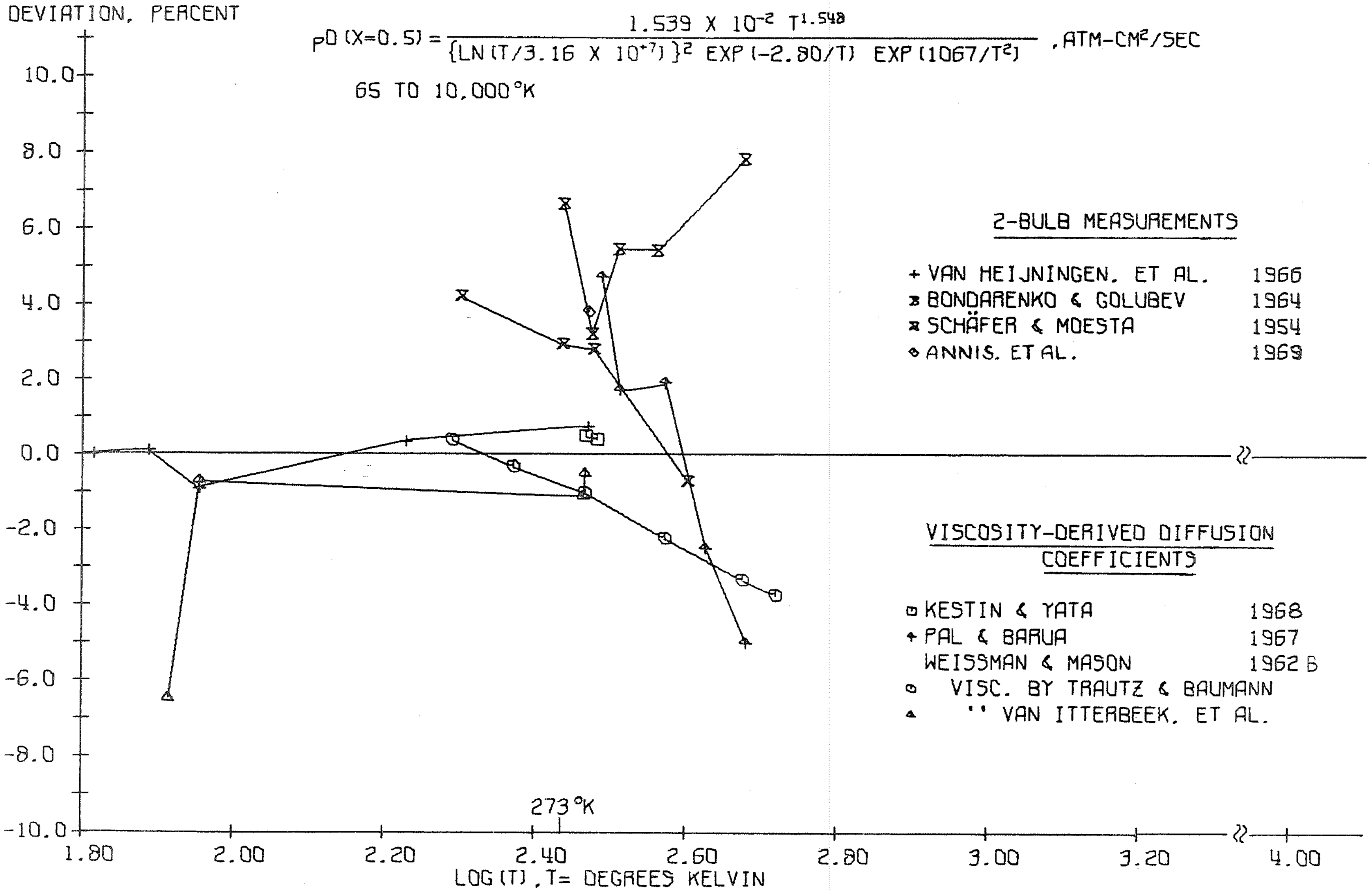


FIG. 18. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - NITROGEN

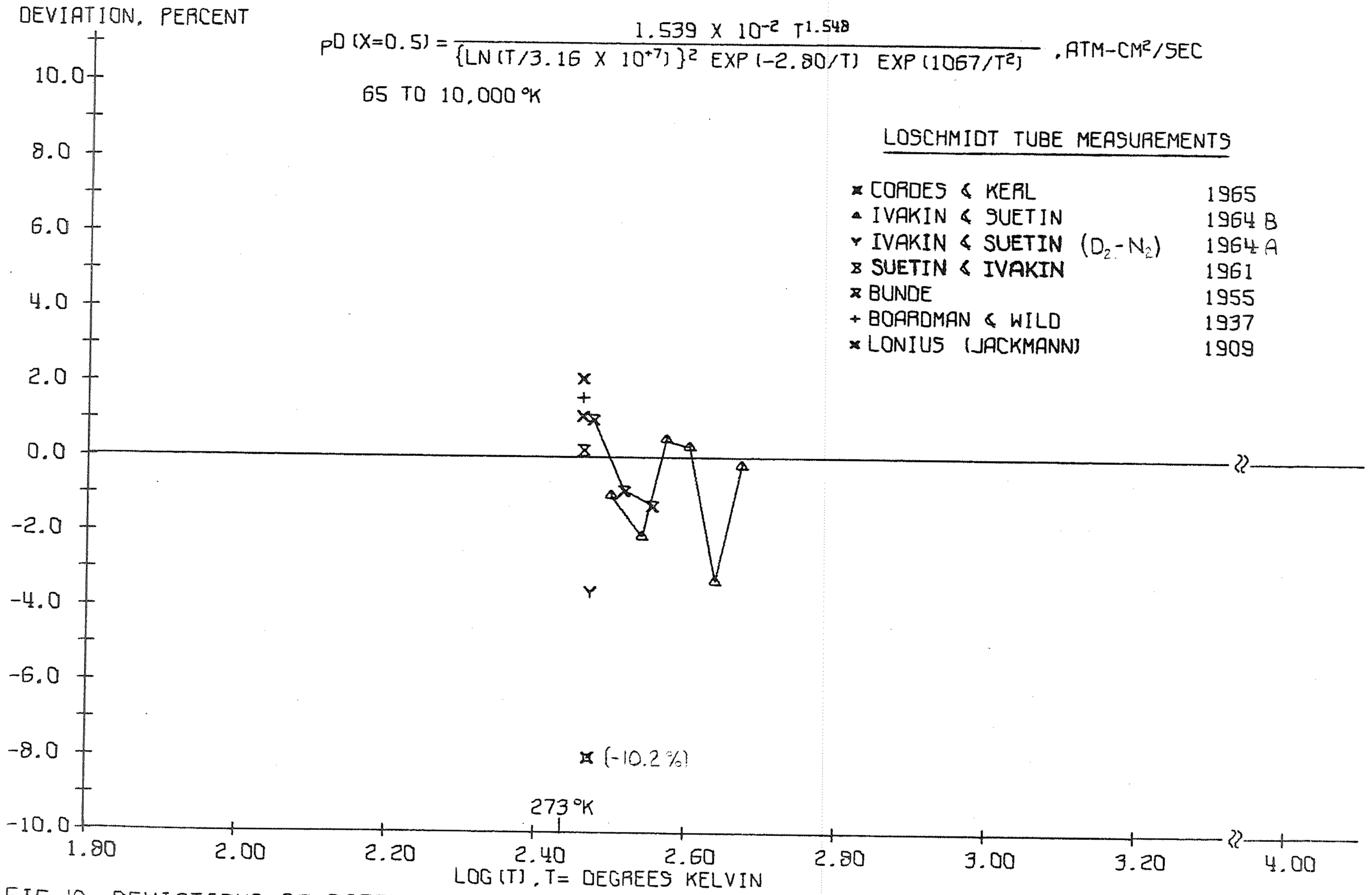


FIG. 19. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - NITROGEN

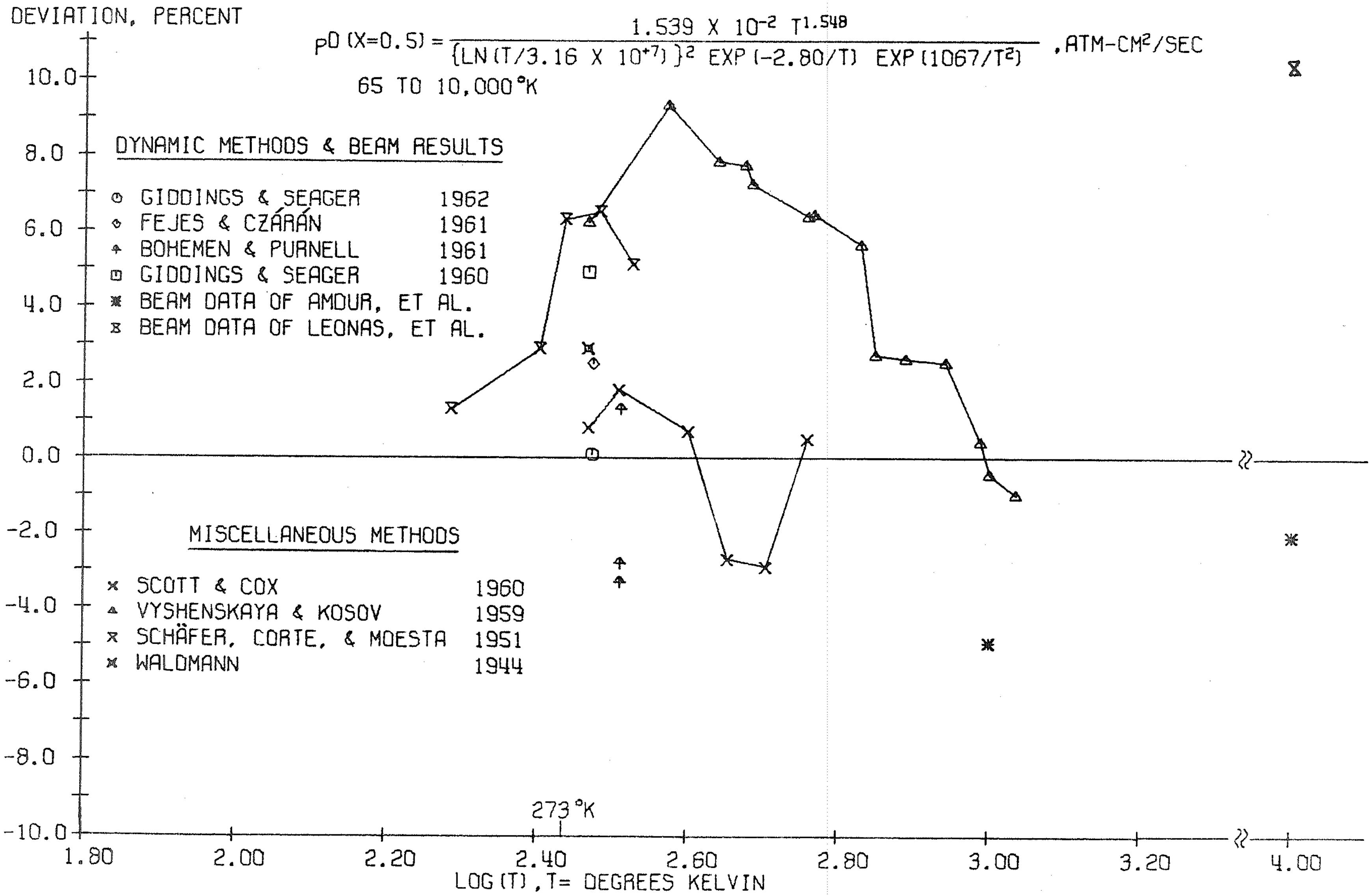


FIG. 20. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - NITROGEN

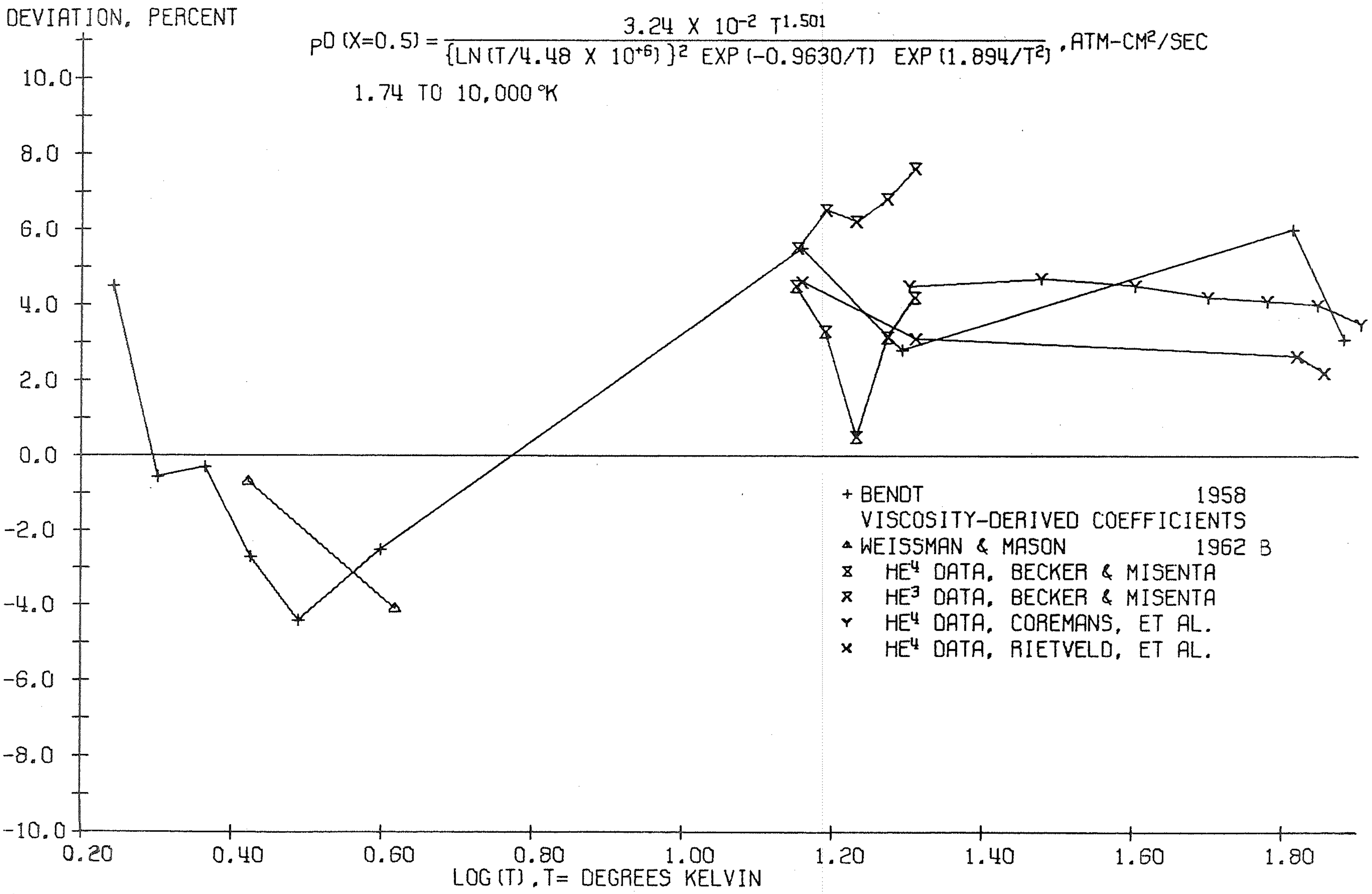


FIG. 21. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM-3 - HELIUM-4

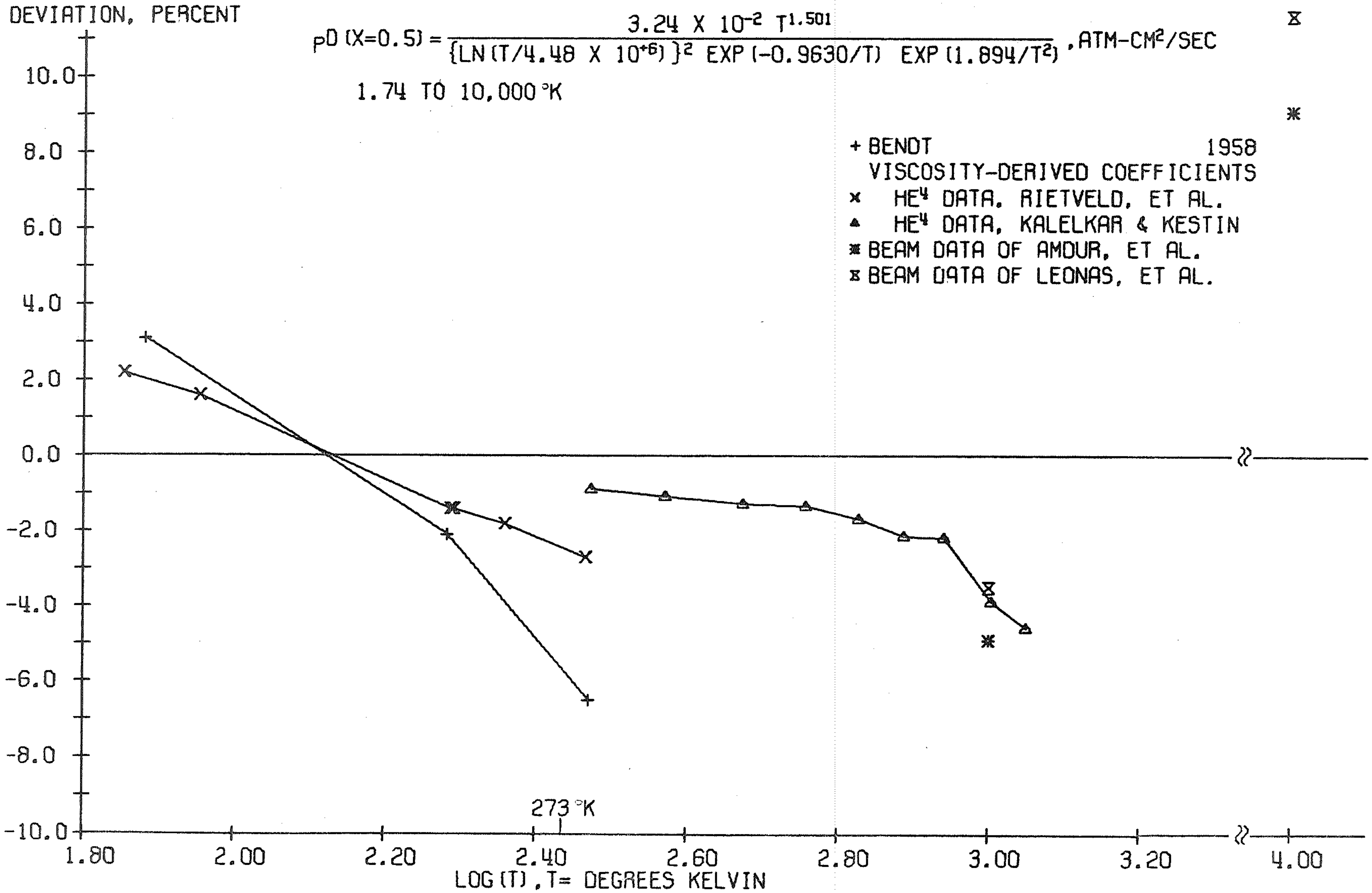


FIG. 22. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM-3 - HELIUM-4

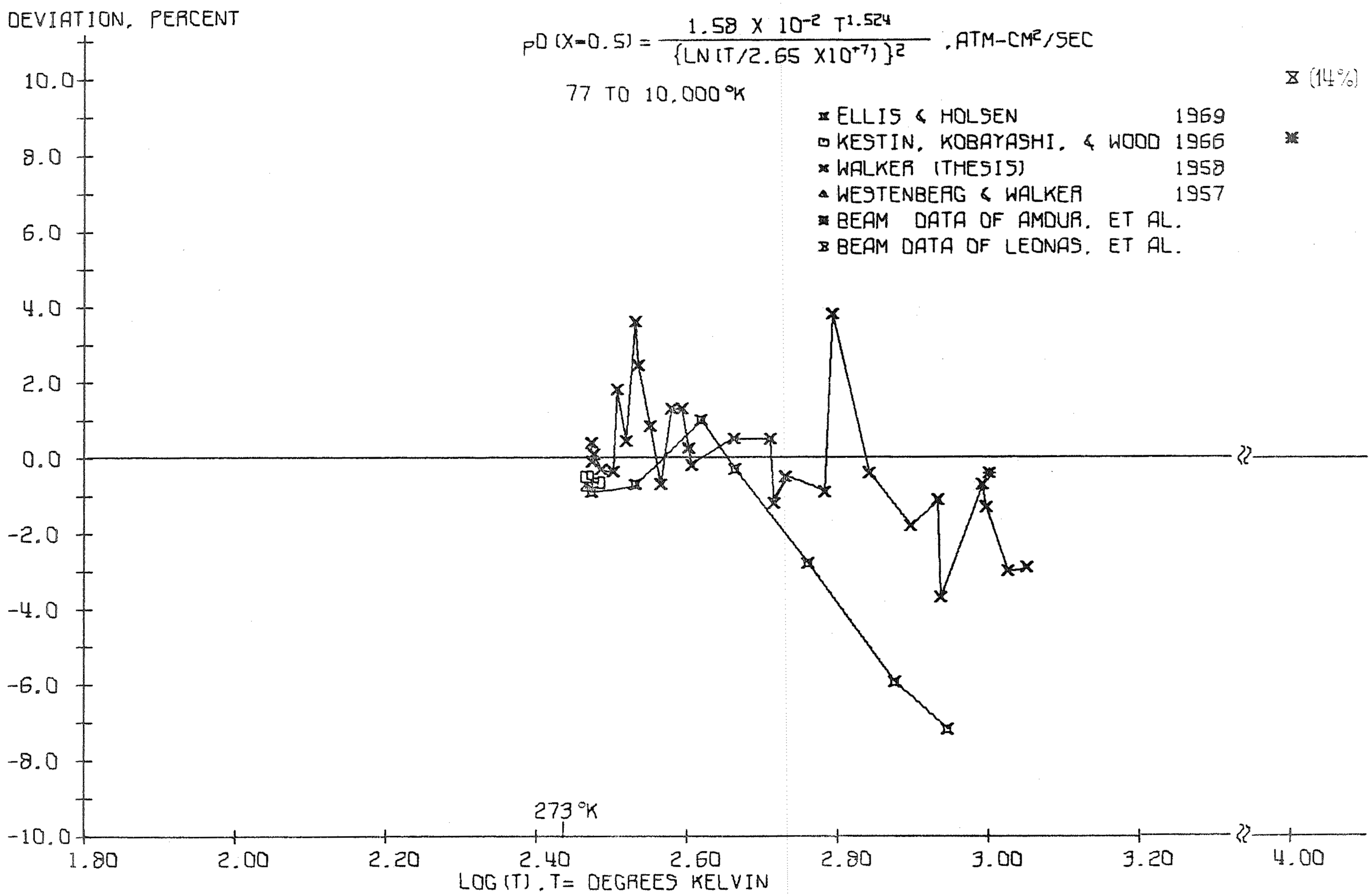


FIG. 23. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - NITROGEN

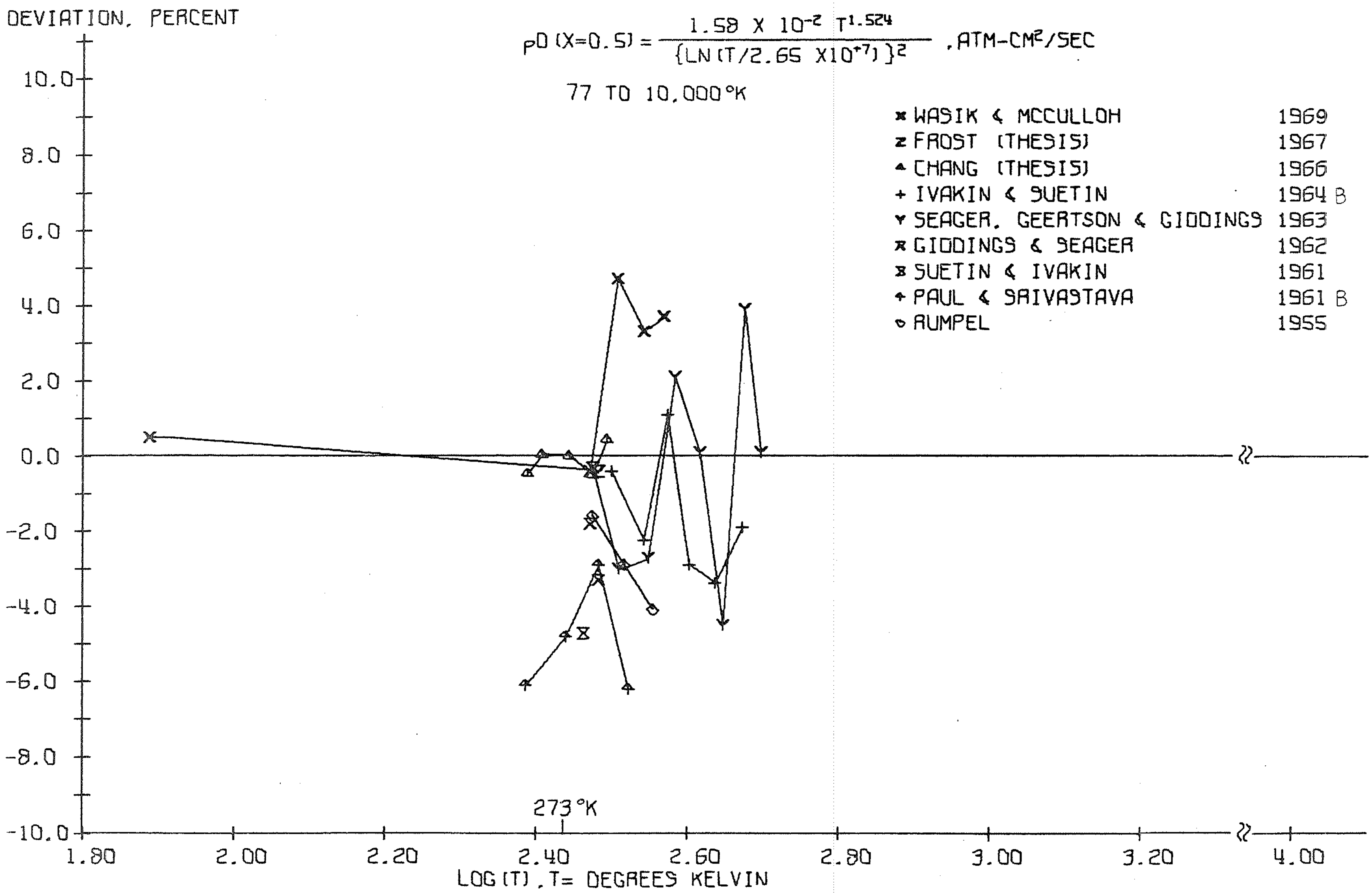


FIG. 24. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - NITROGEN

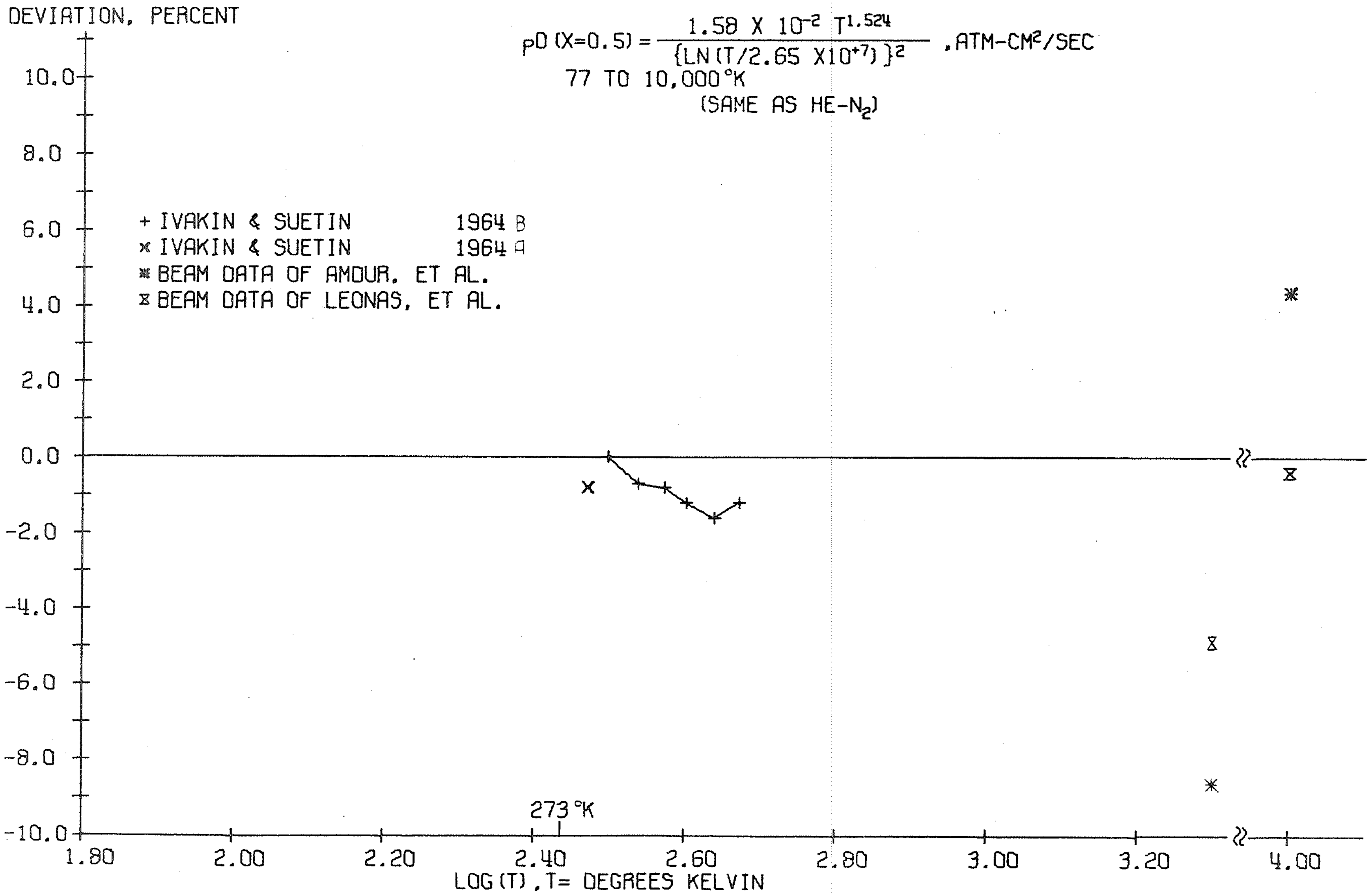


FIG. 25. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - CARBON MONOXIDE

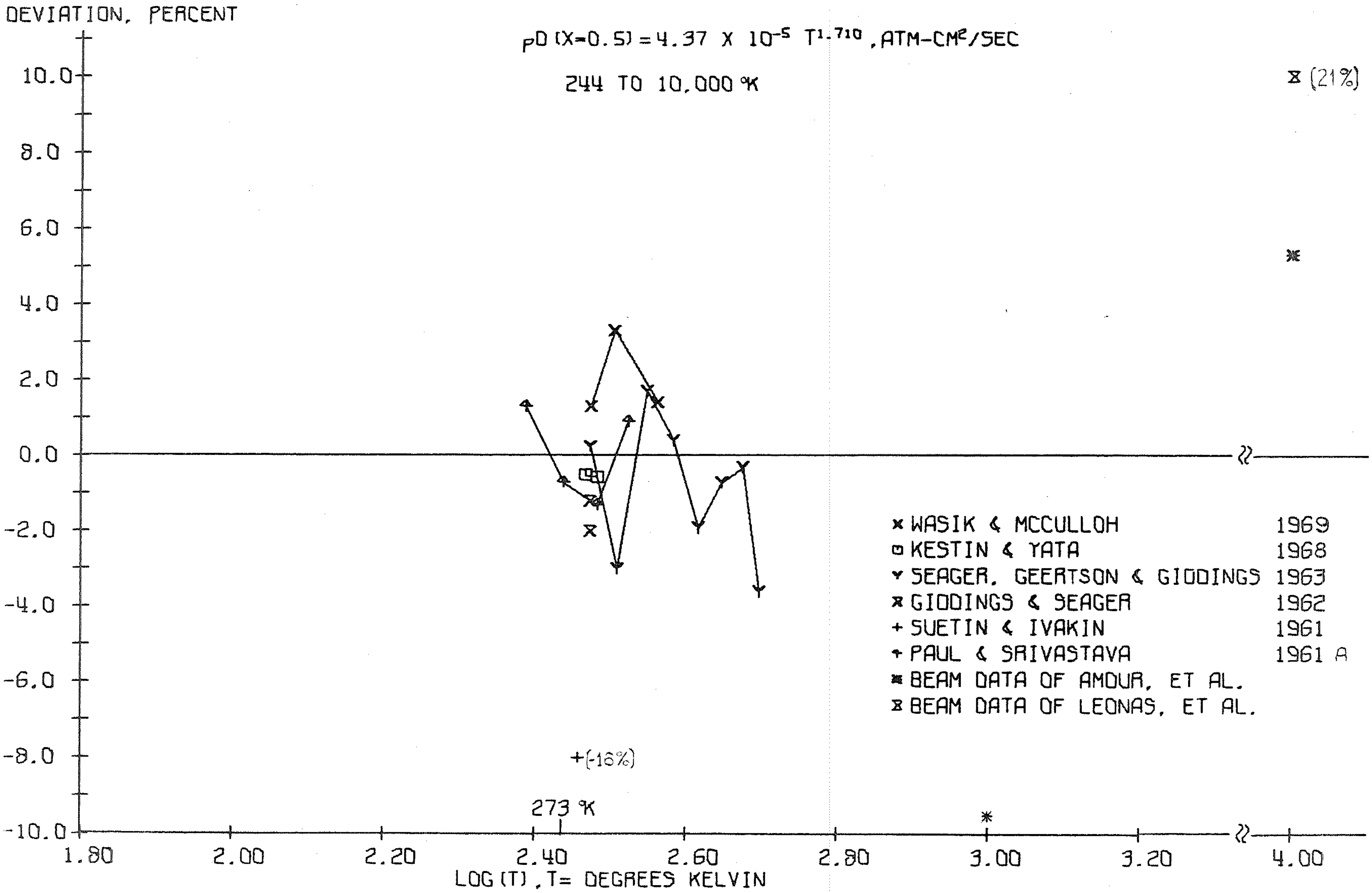


FIG. 26. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - OXYGEN

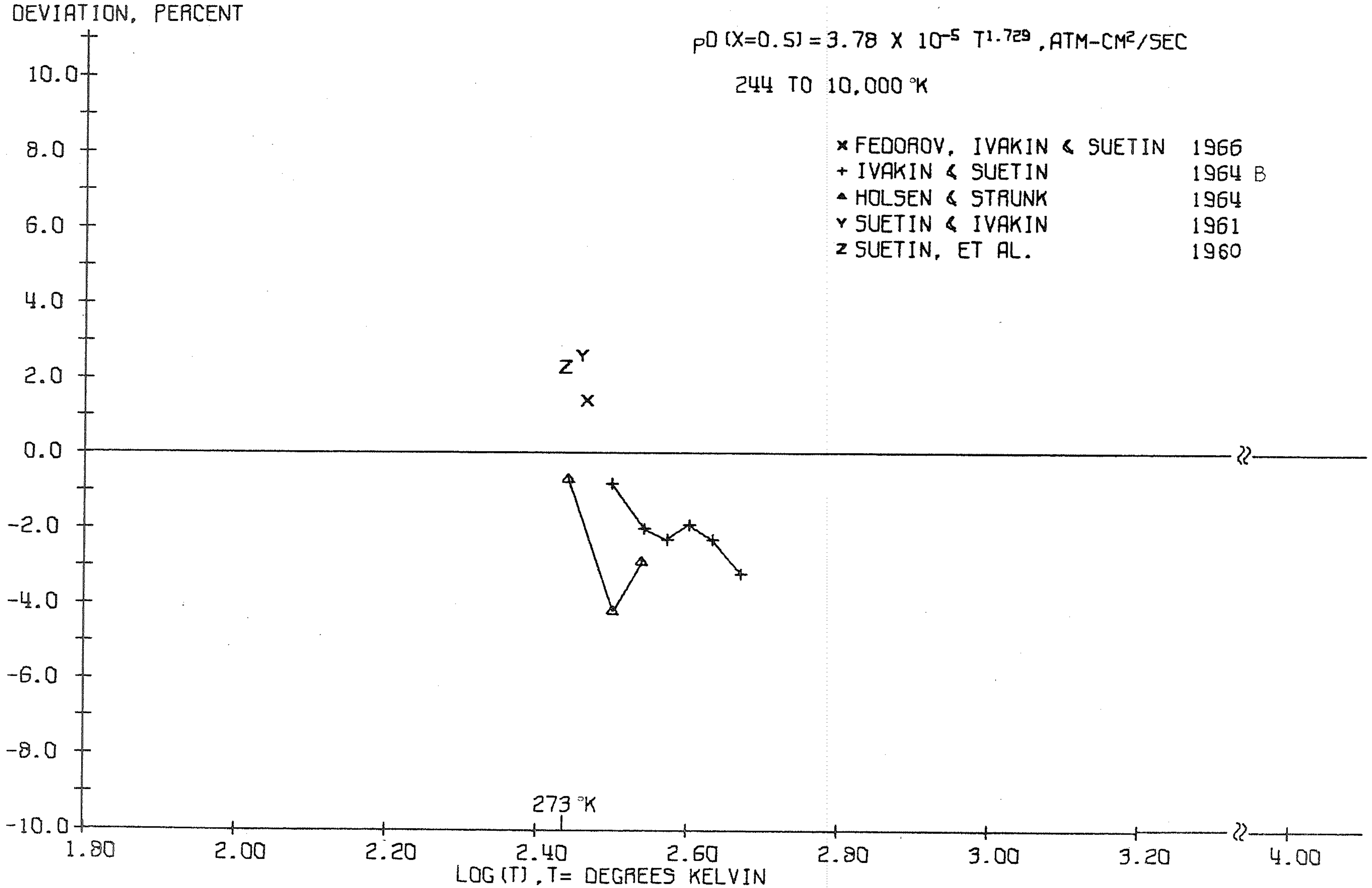


FIG. 27. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - AIR

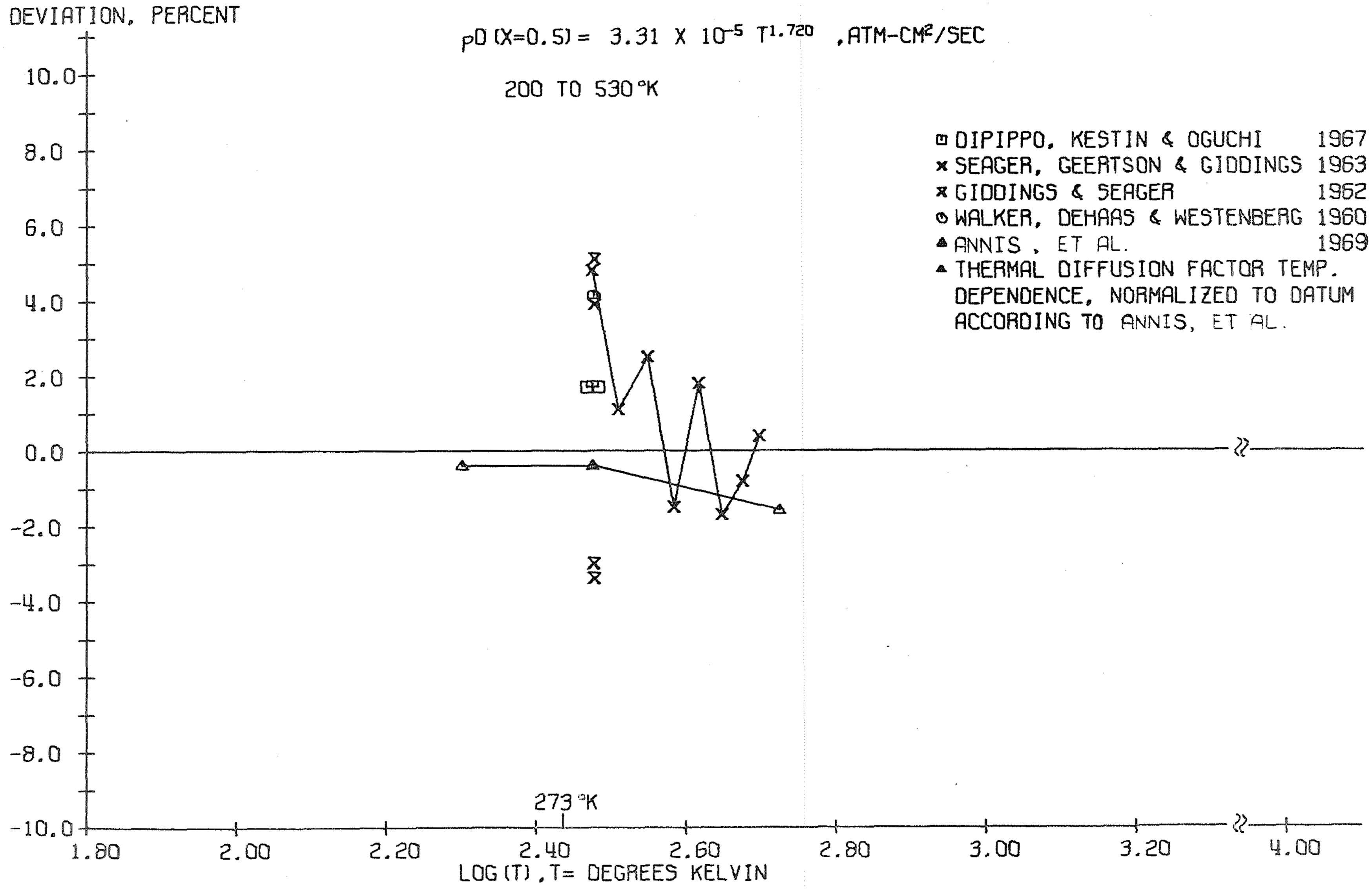


FIG. 28. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM – CARBON DIOXIDE

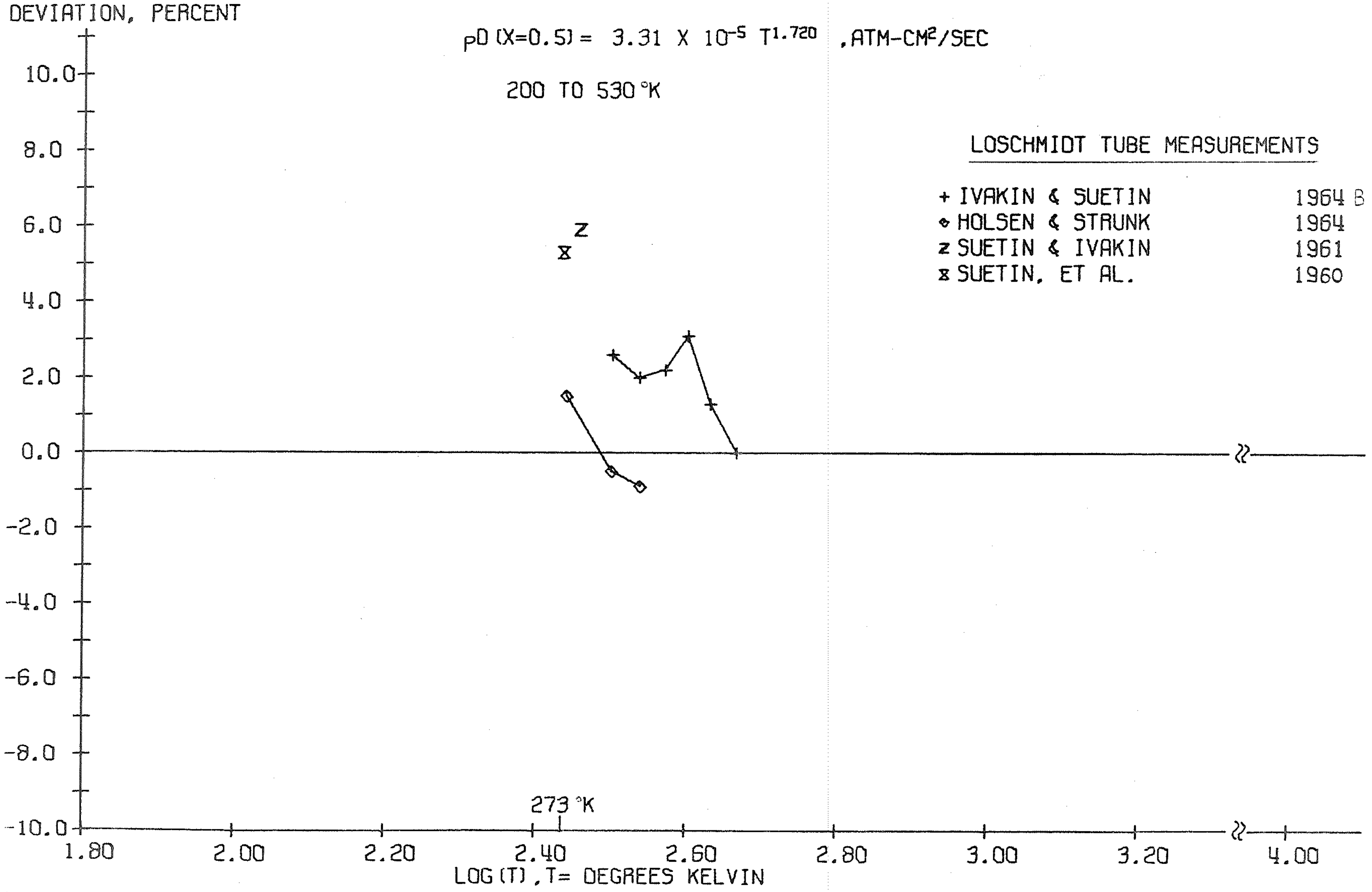


FIG. 29. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - CARBON DIOXIDE

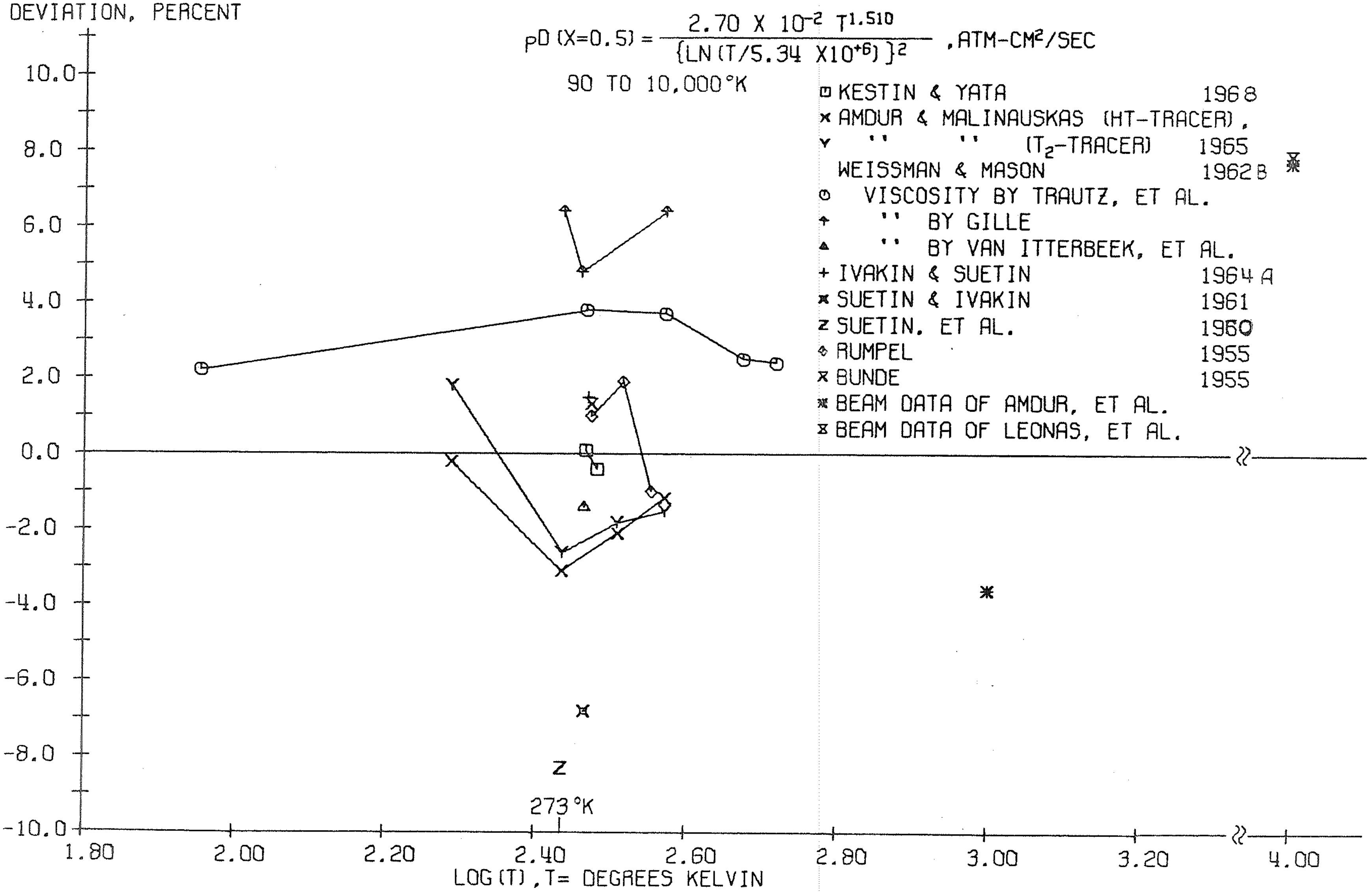


FIG. 30. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - HELIUM

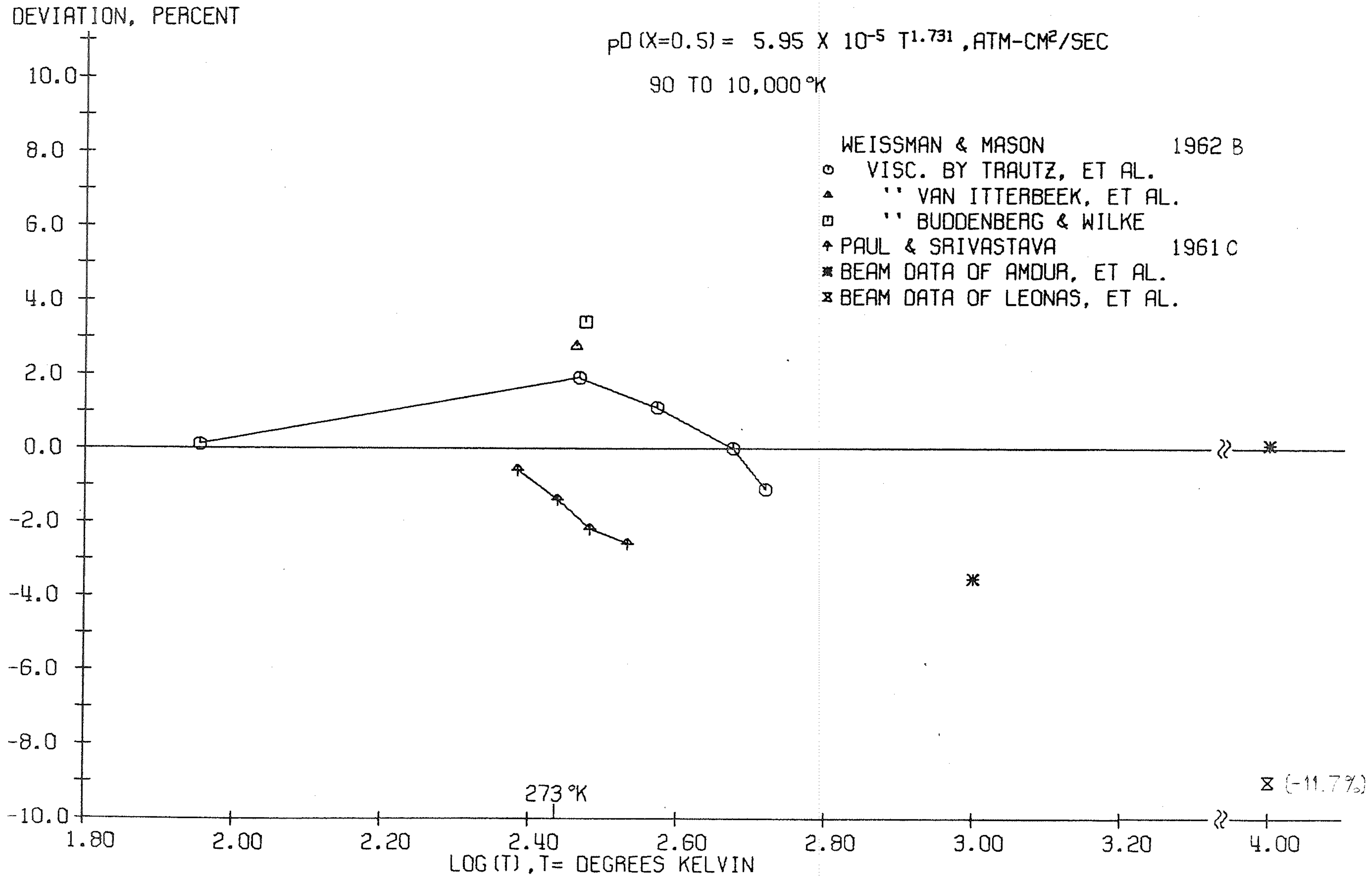


FIG. 31. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - NEON

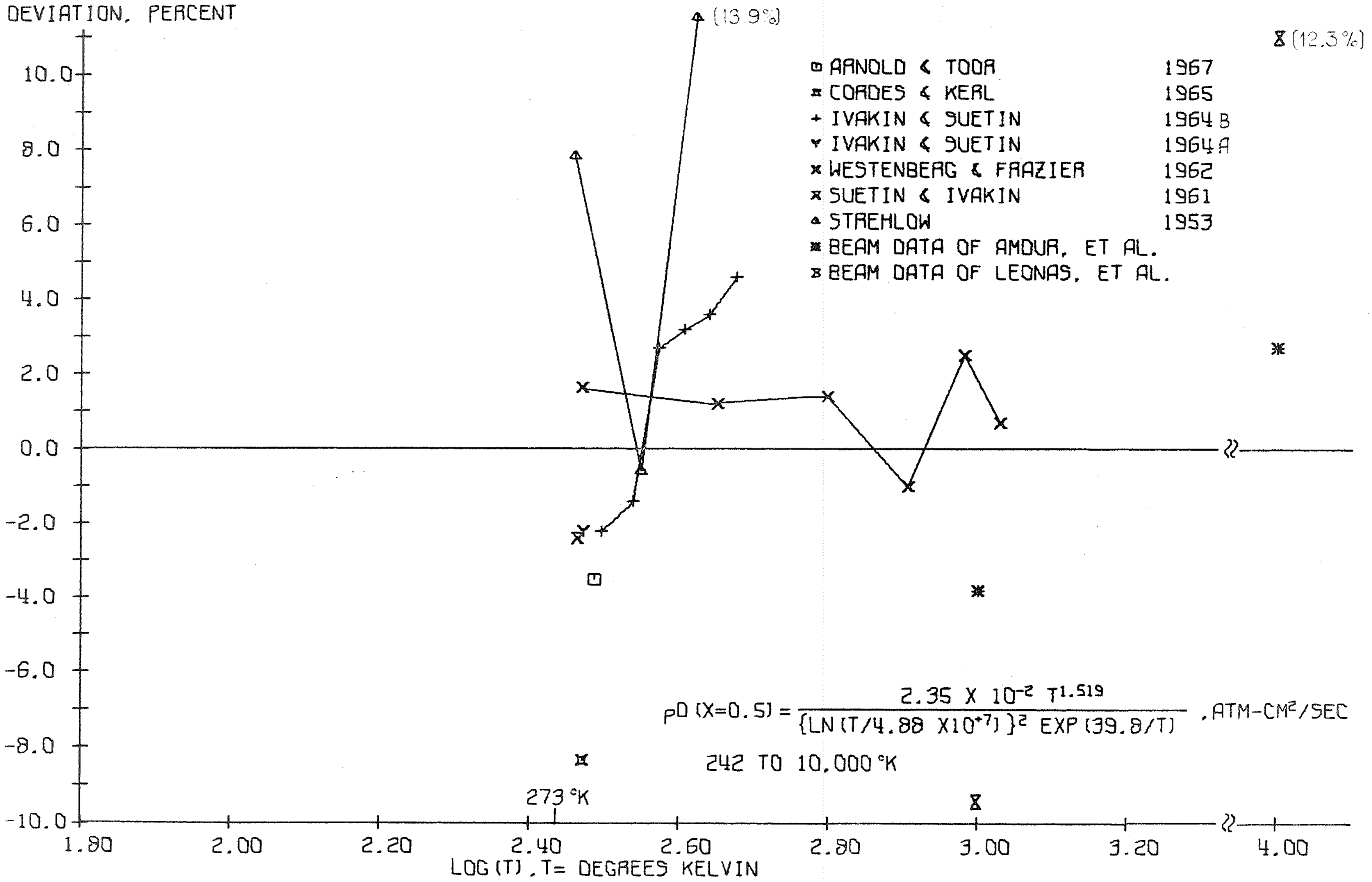


FIG. 32. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - ARGON

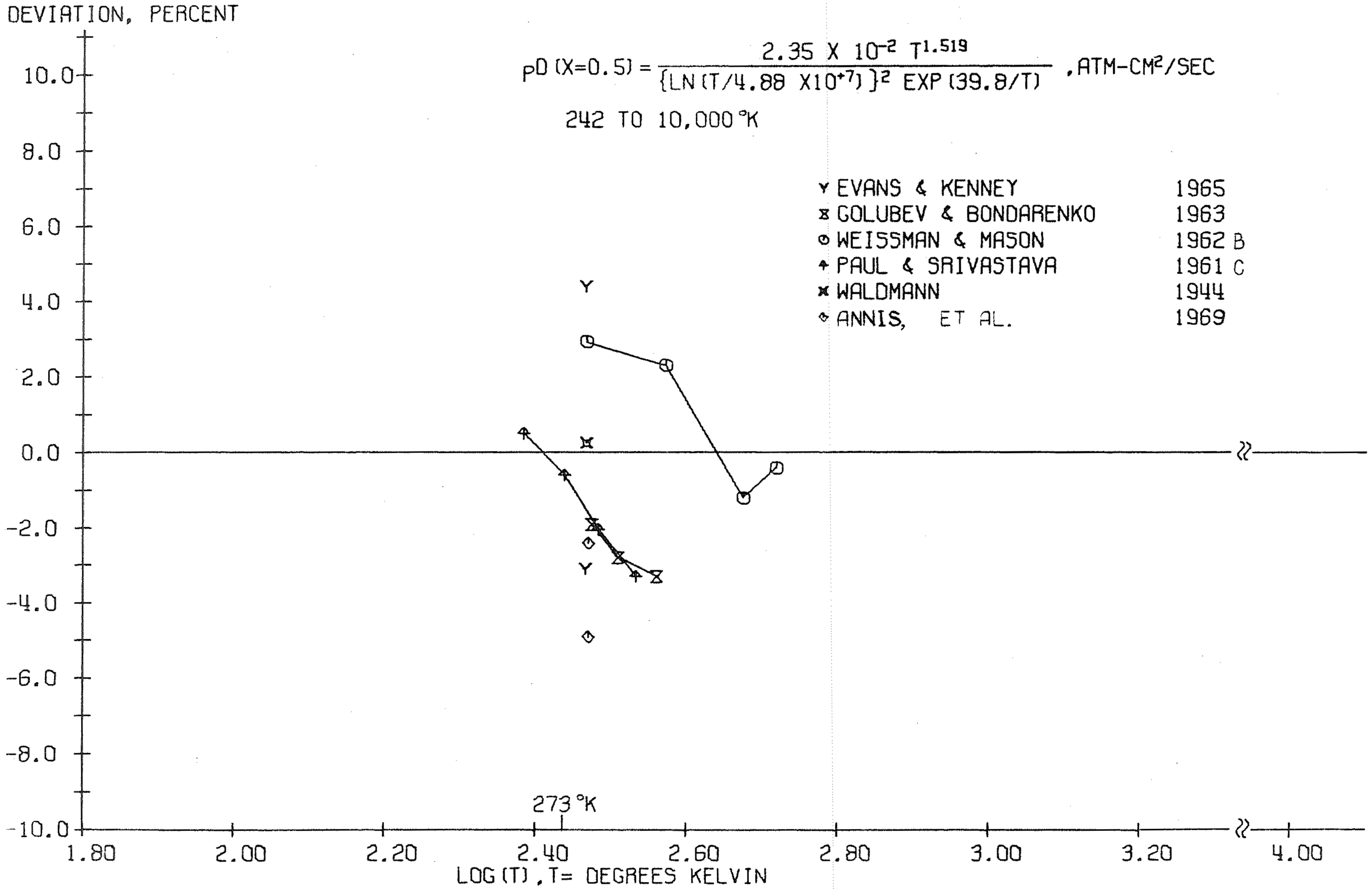


FIG. 33. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - ARGON

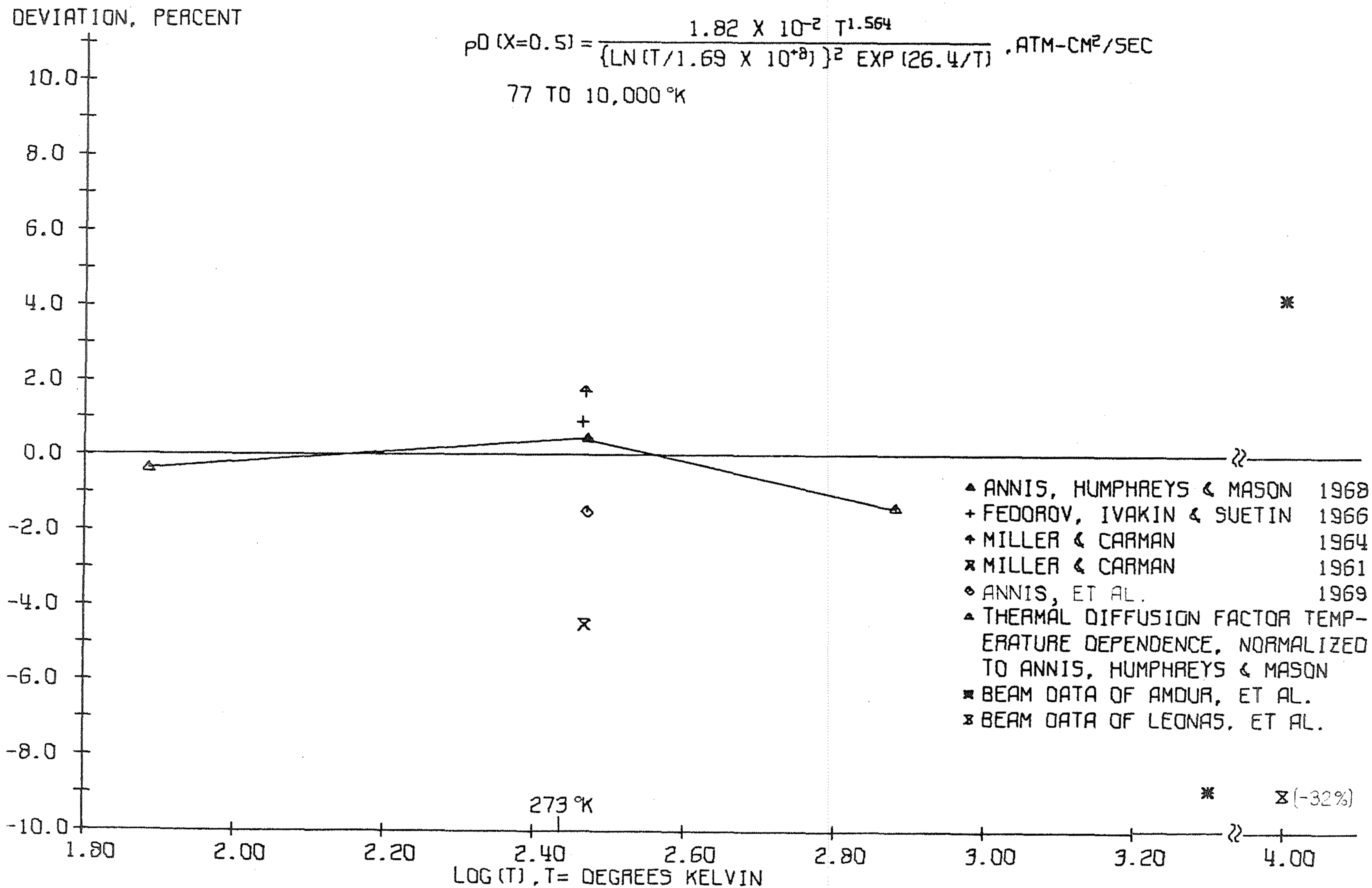


FIG. 34. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. HYDROGEN - KRYPTON

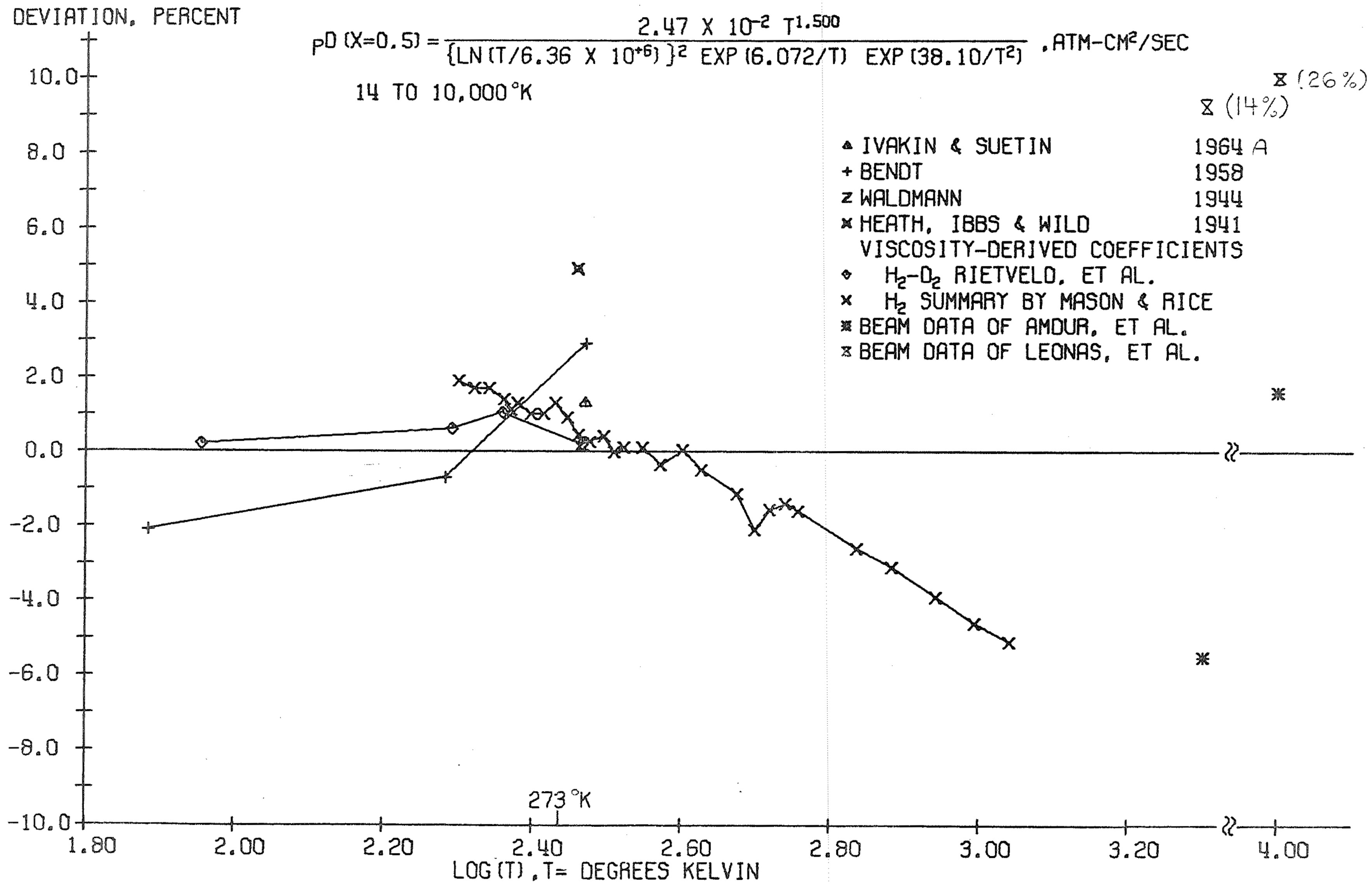


FIG. 35. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - DEUTERIUM

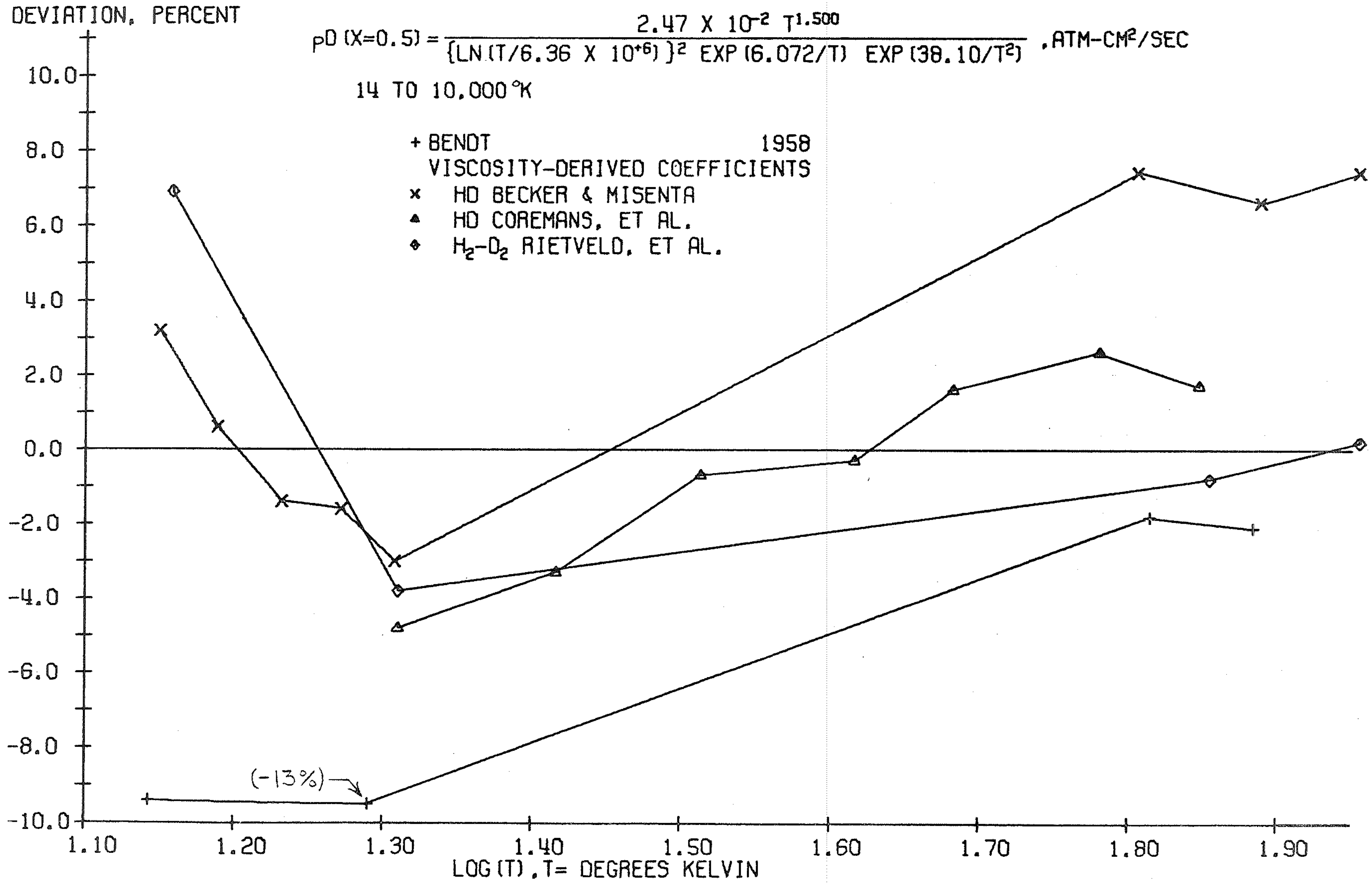


FIG.36. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - DEUTERIUM

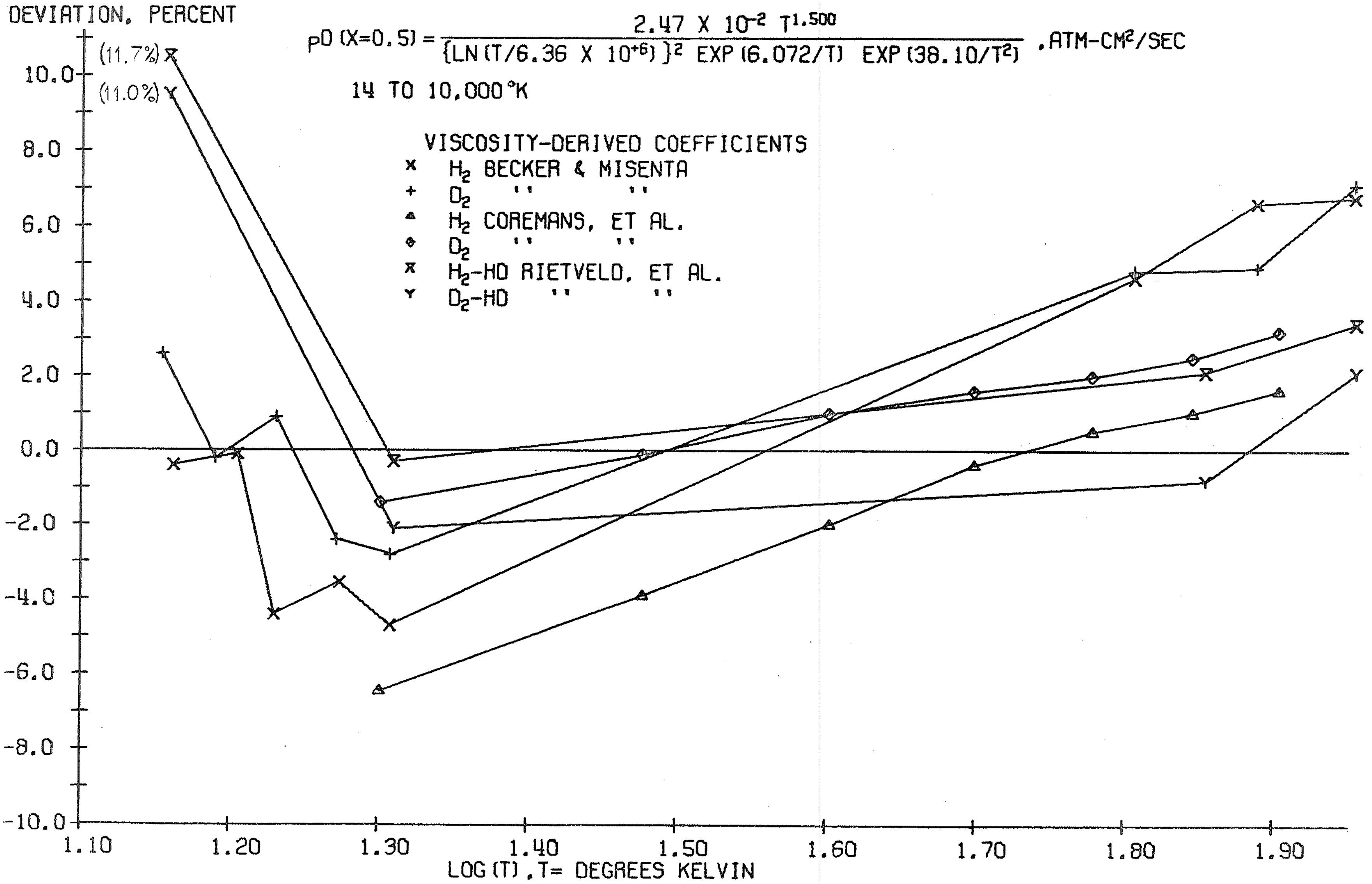


FIG. 37. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - DEUTERIUM

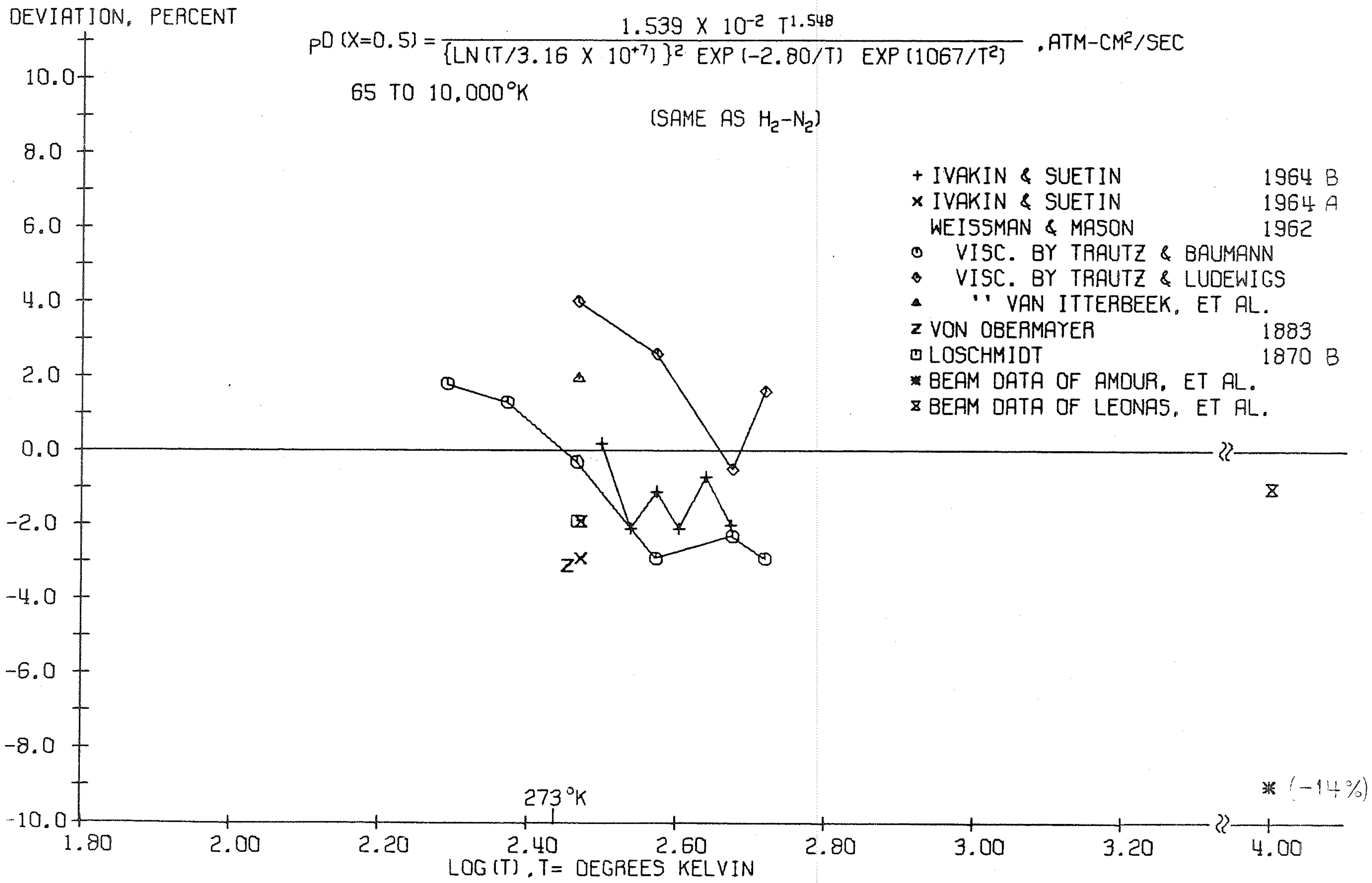


FIG.38. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - CARBON MONOXIDE

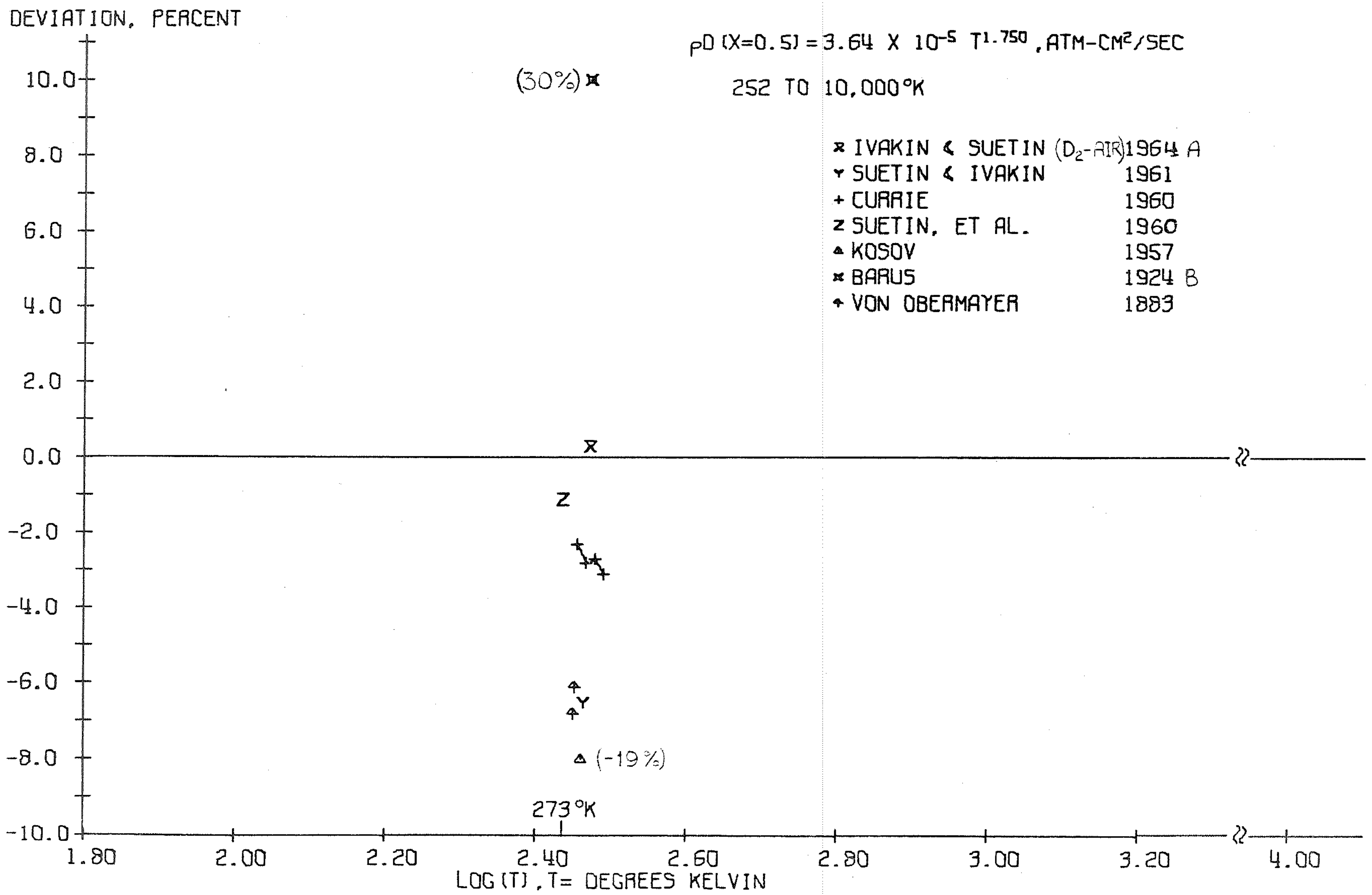


FIG. 39. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. HYDROGEN - AIR

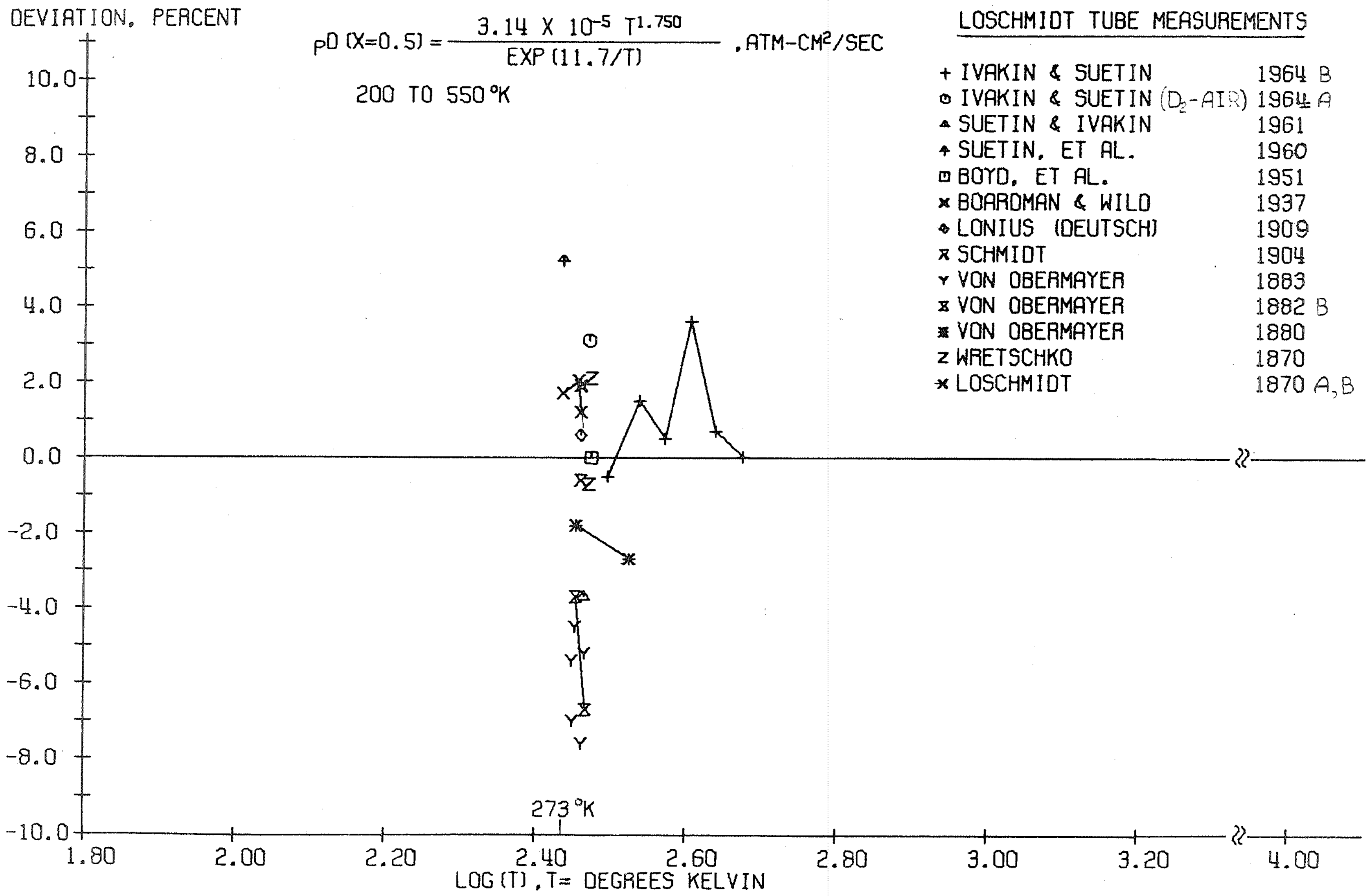


FIG. 40. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - CARBON DIOXIDE

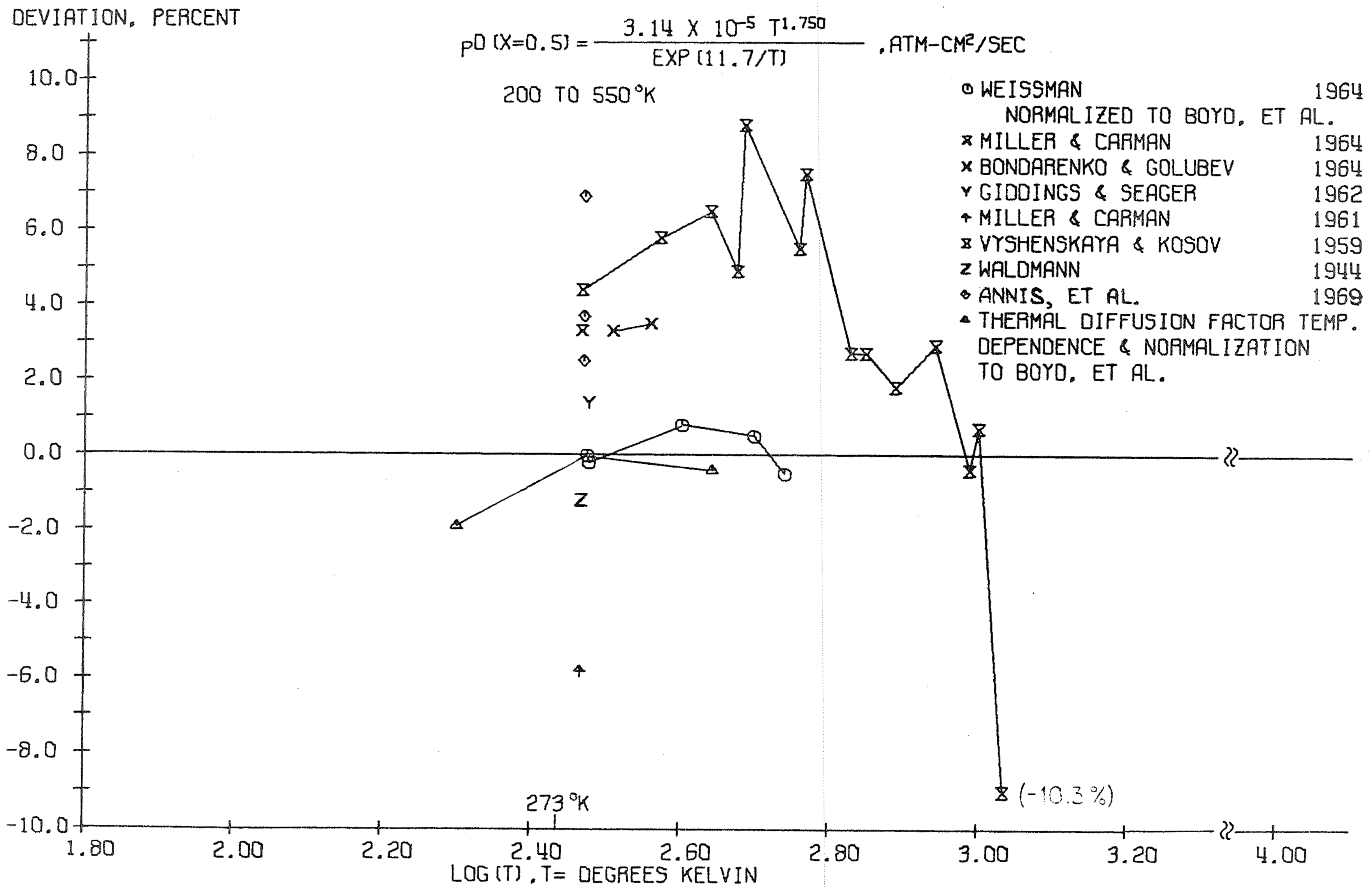


FIG. 41. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - CARBON DIOXIDE

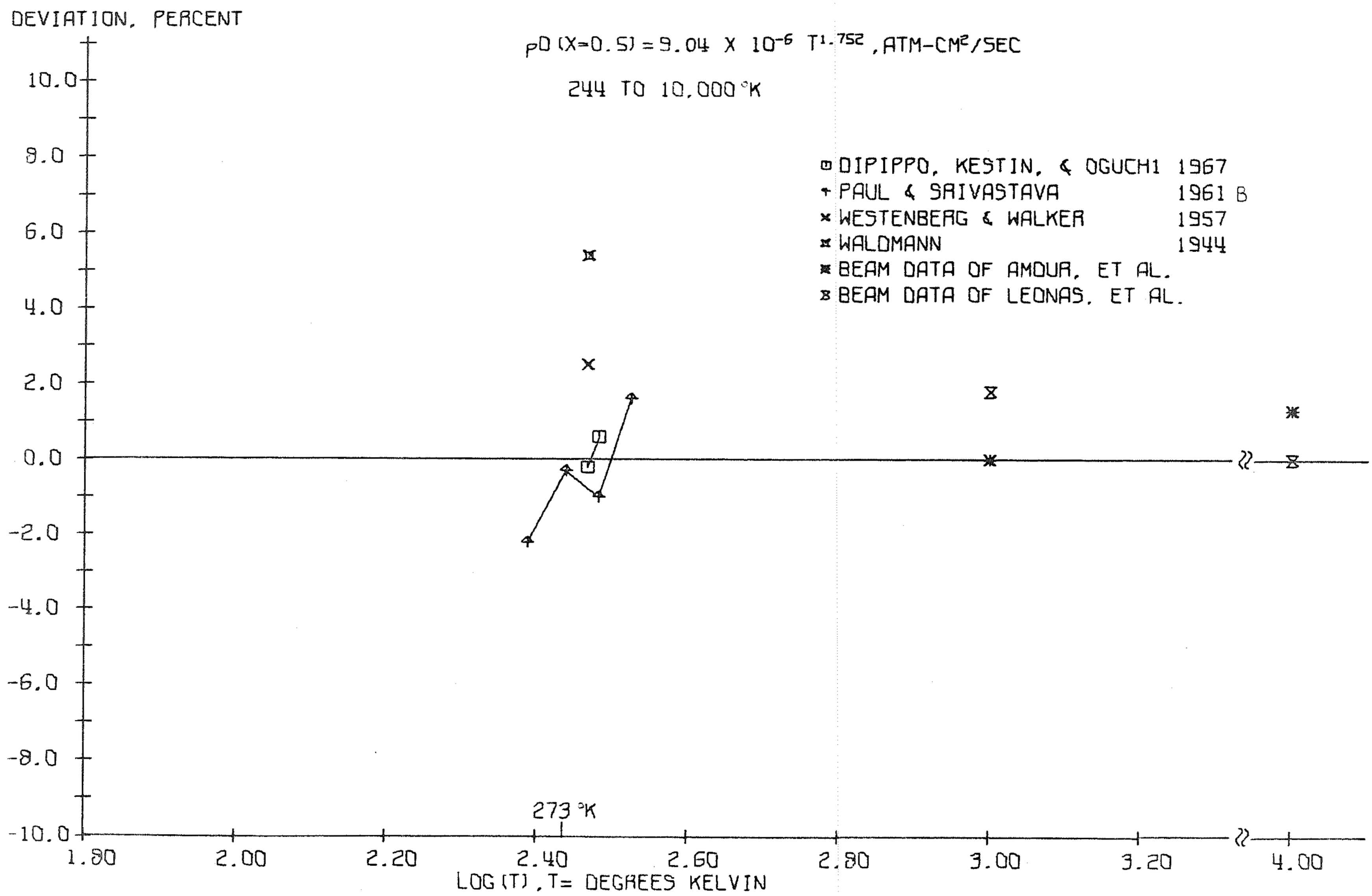


FIG. 42. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - ARGON

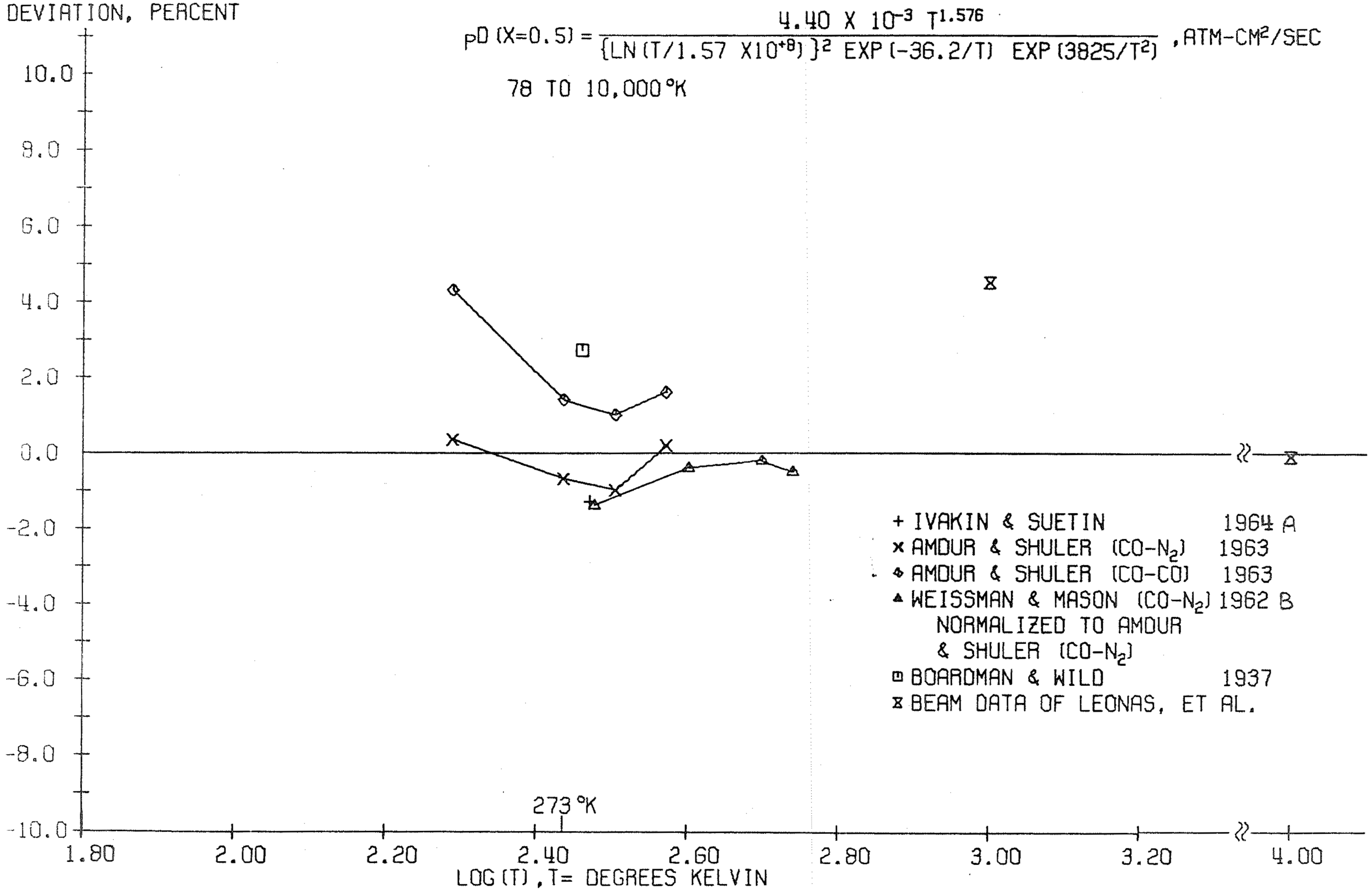


FIG. 43. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - NITROGEN

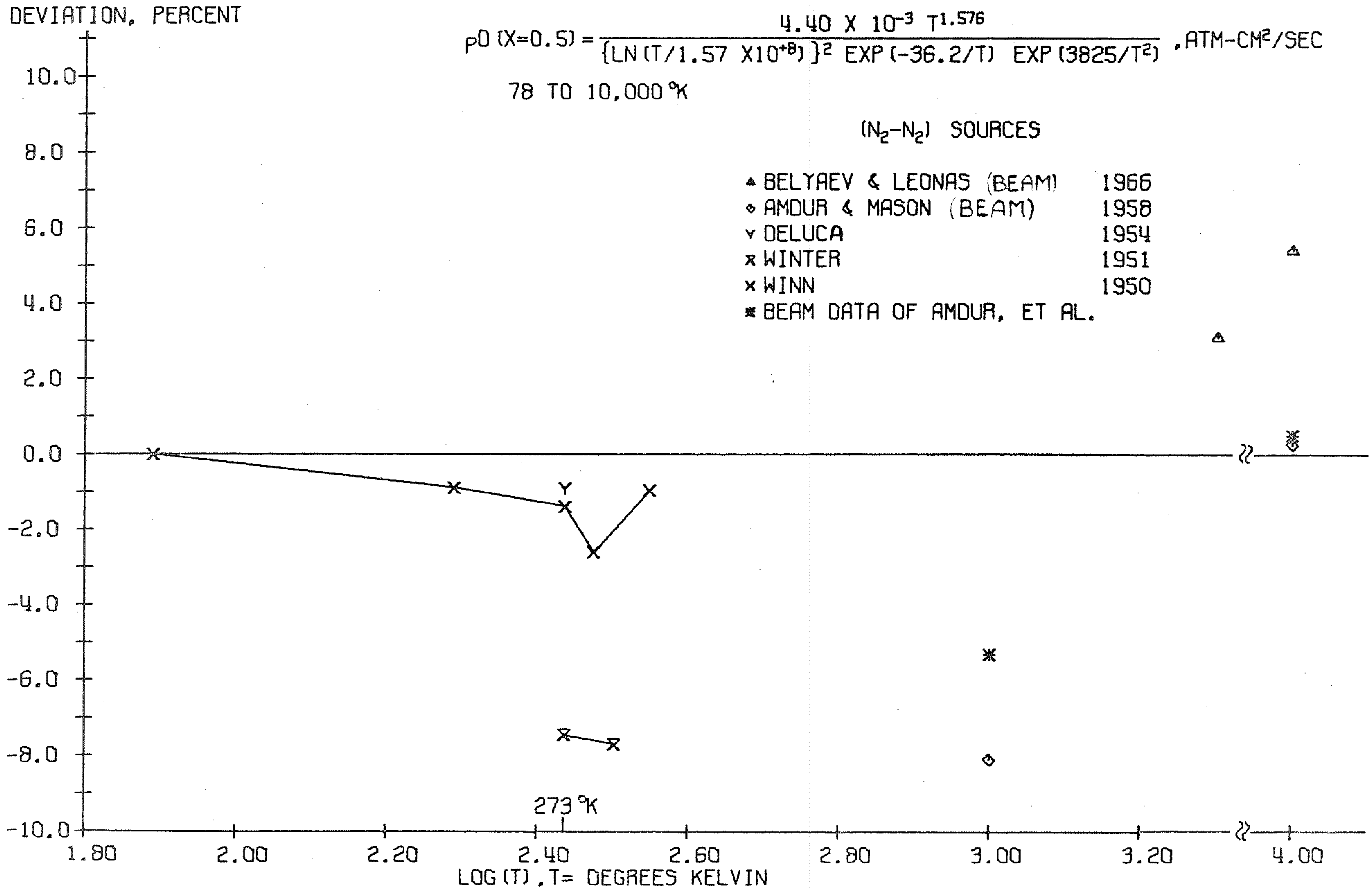


FIG. 44. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - NITROGEN

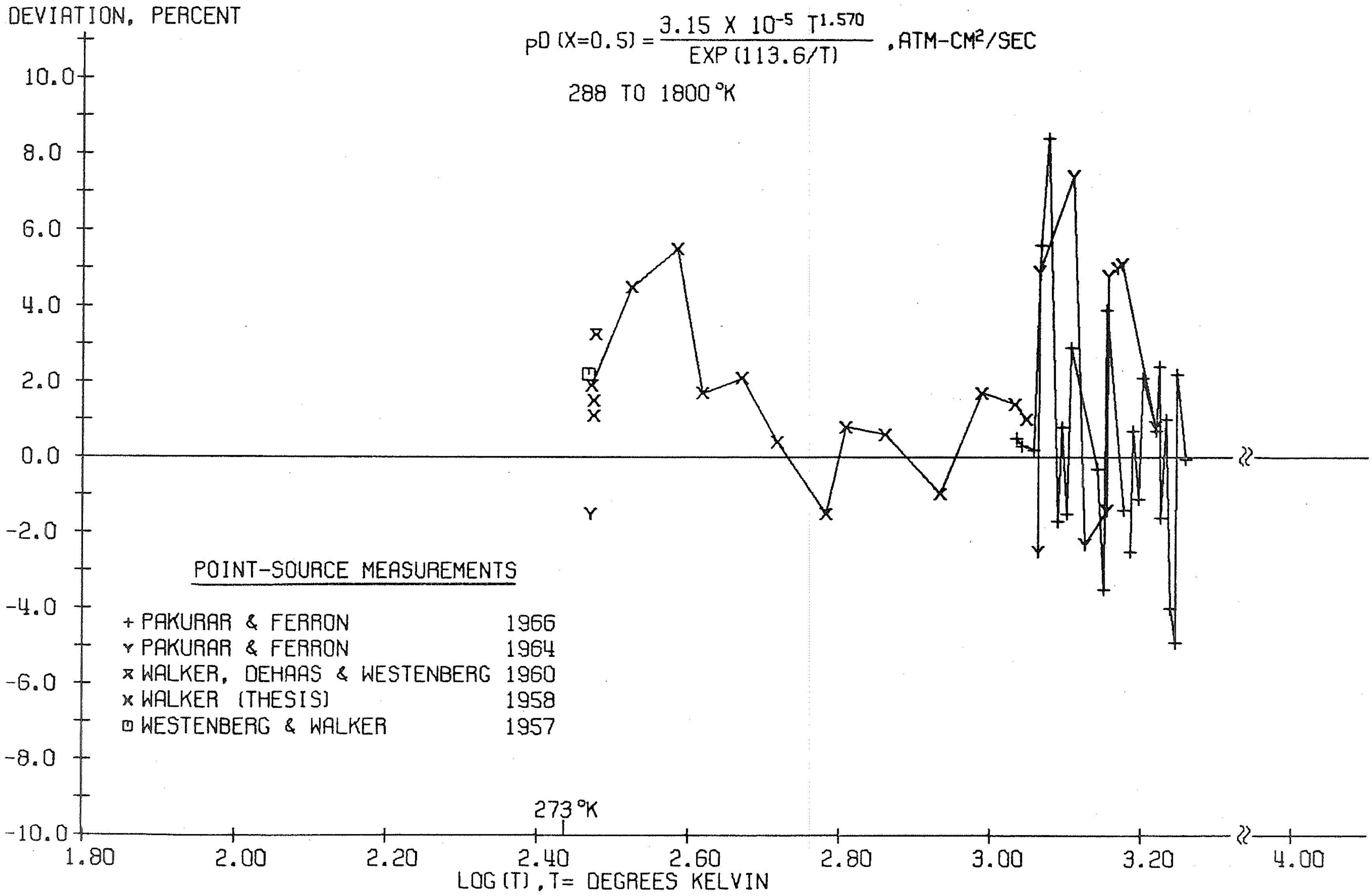


FIG. 45. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - CARBON DIOXIDE

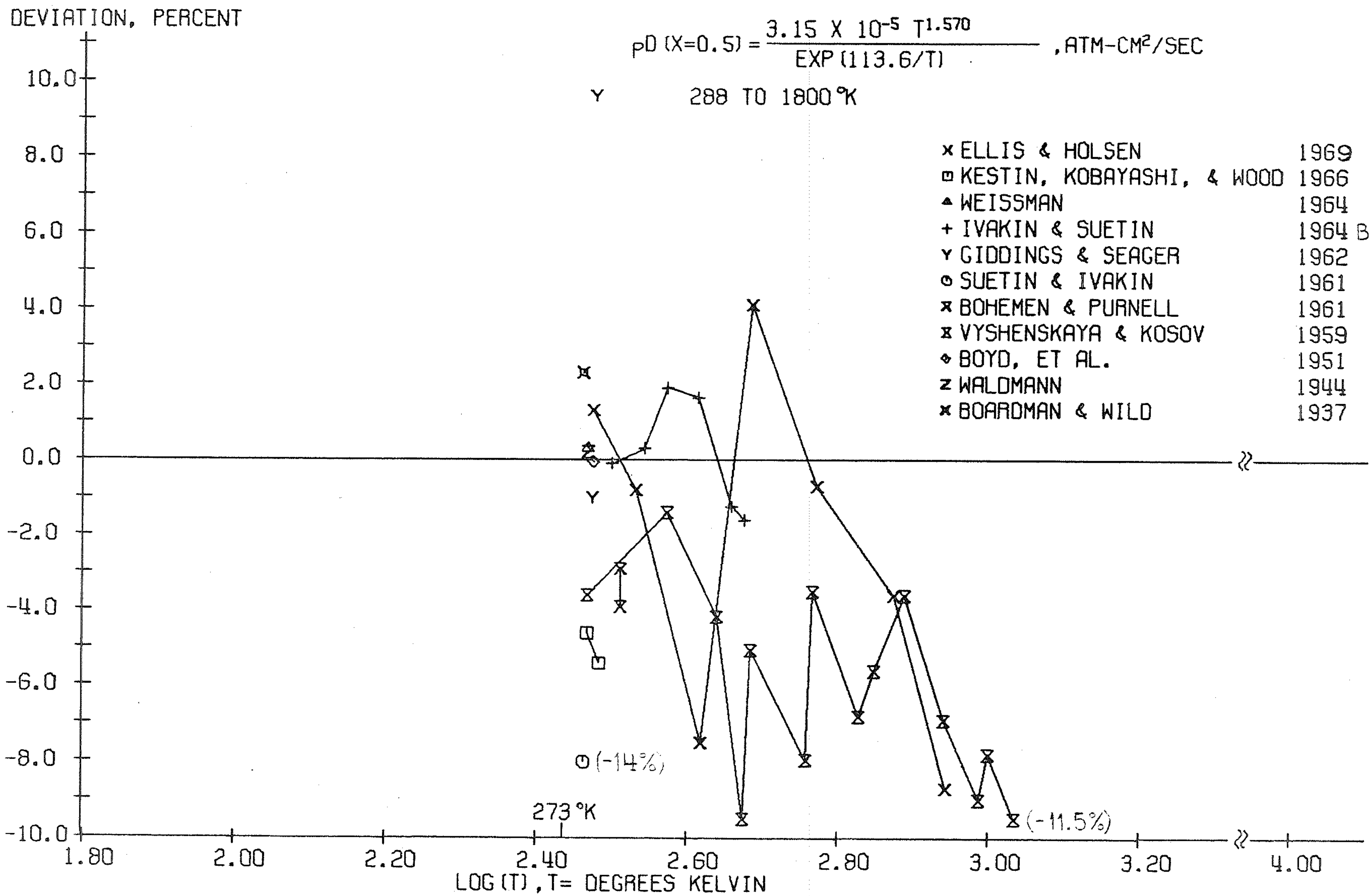


FIG. 46. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - CARBON DIOXIDE

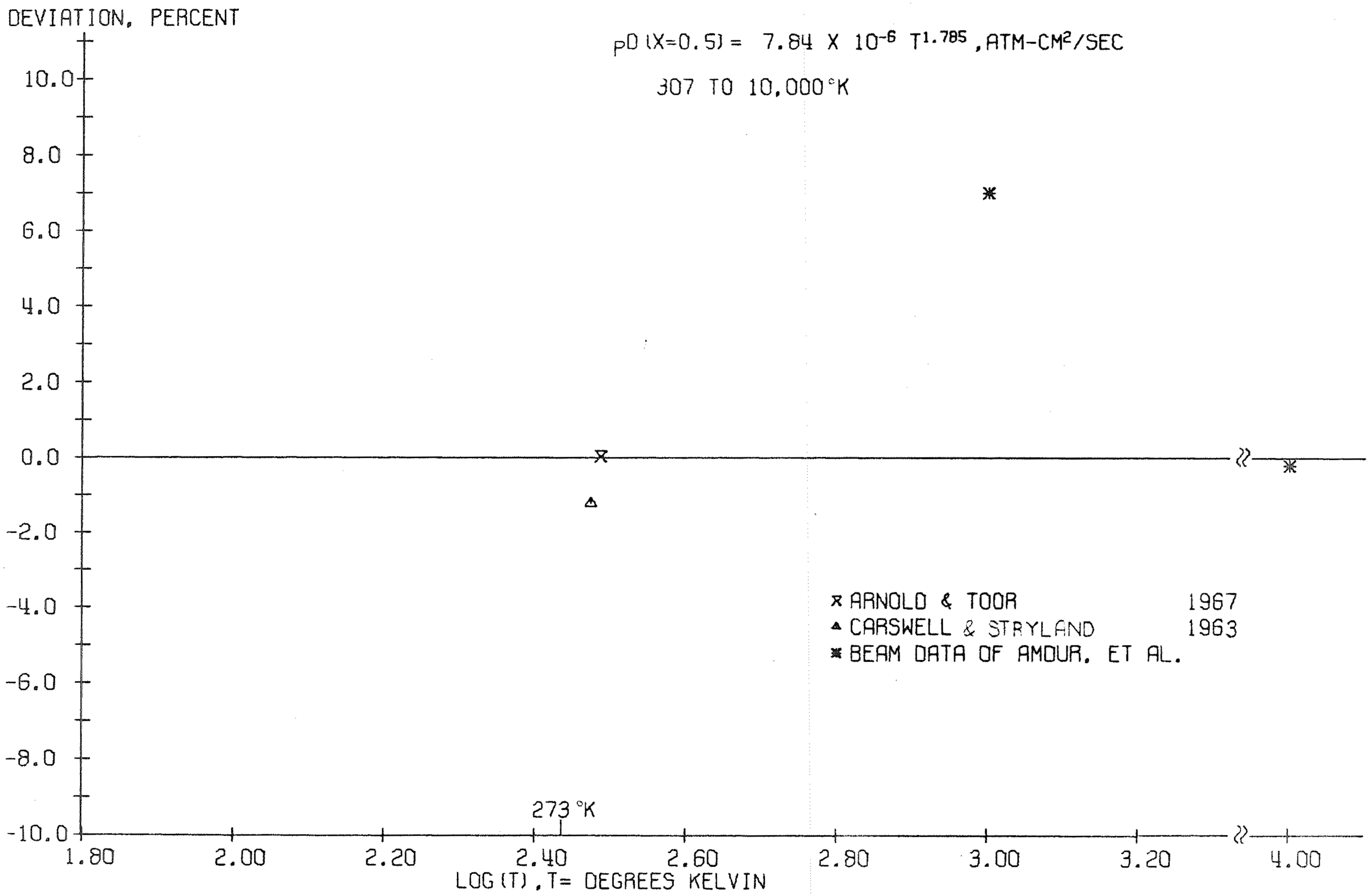


FIG. 47. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, METHANE - ARGON

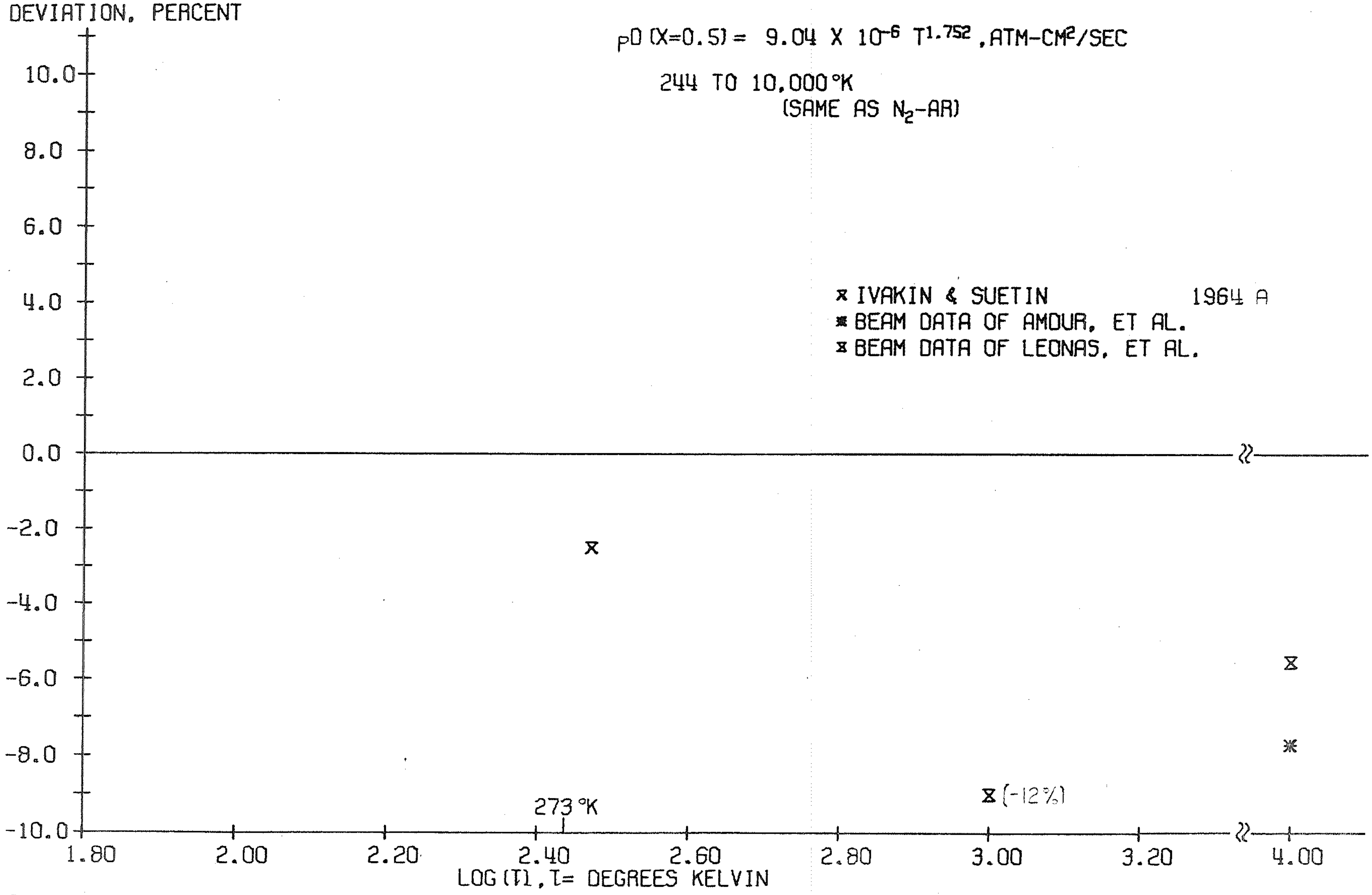


FIG. 48. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - ARGON

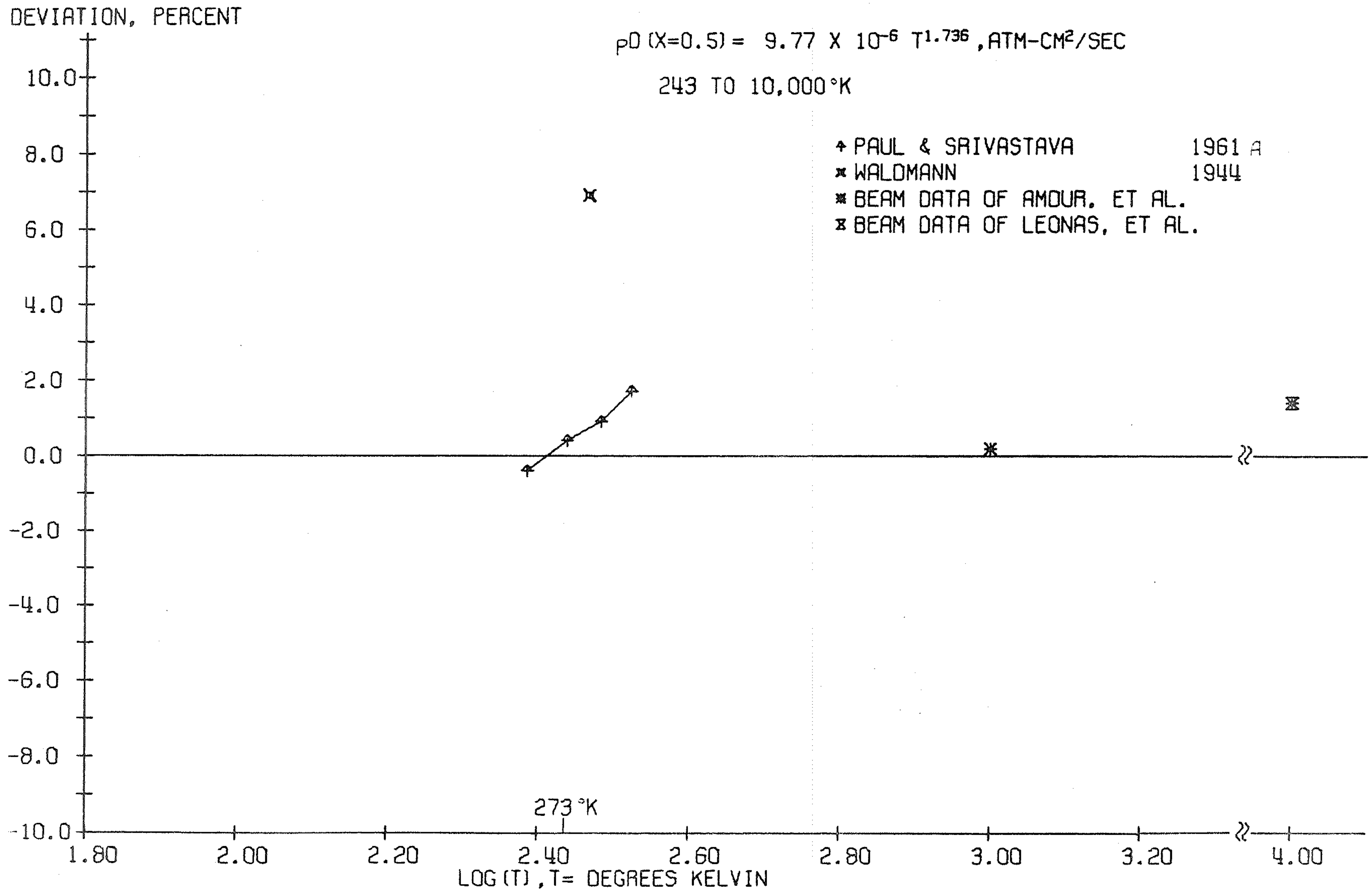


FIG. 49. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, OXYGEN - ARGON

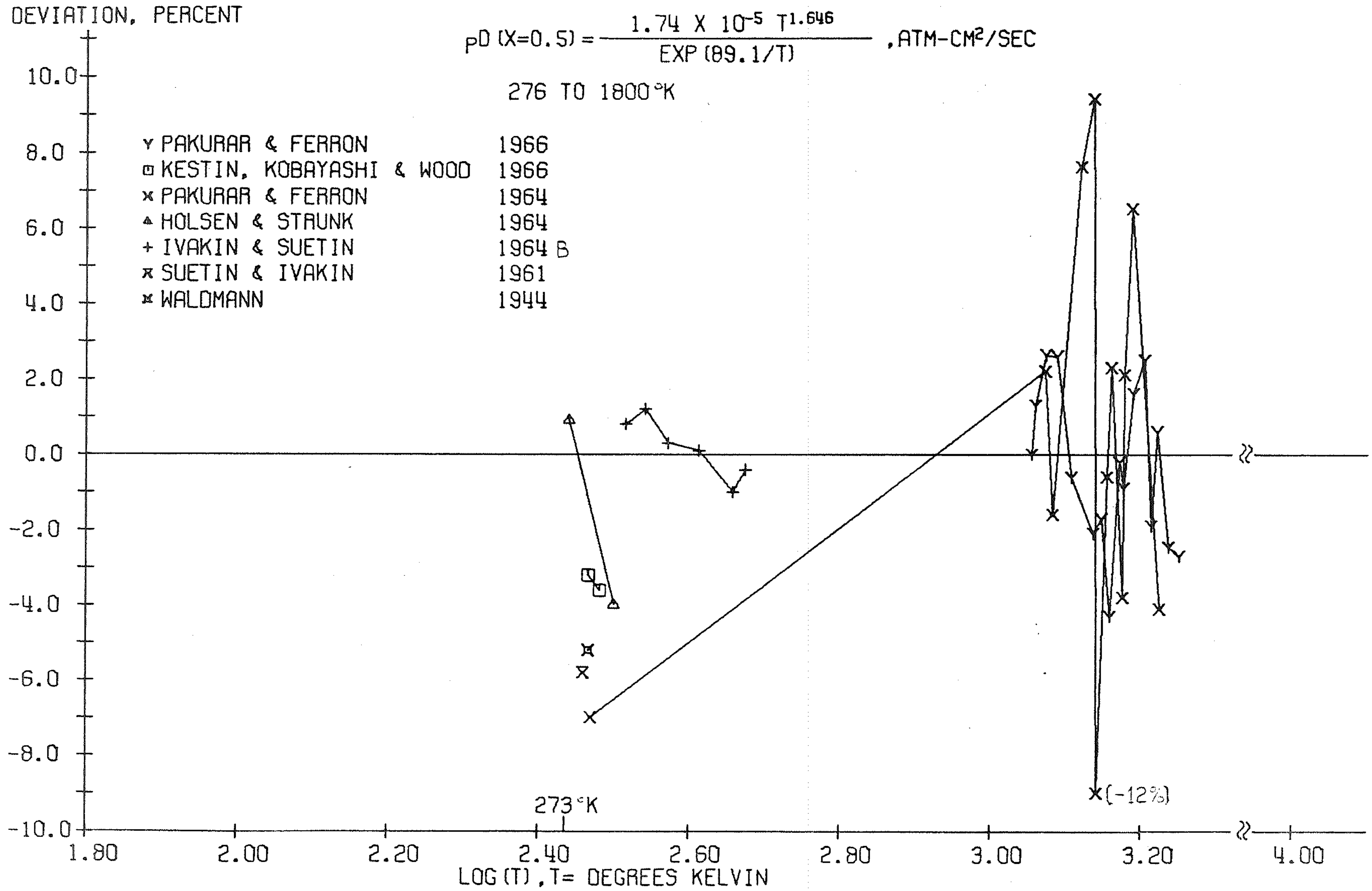


FIG. 50. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, ARGON - CARBON DIOXIDE

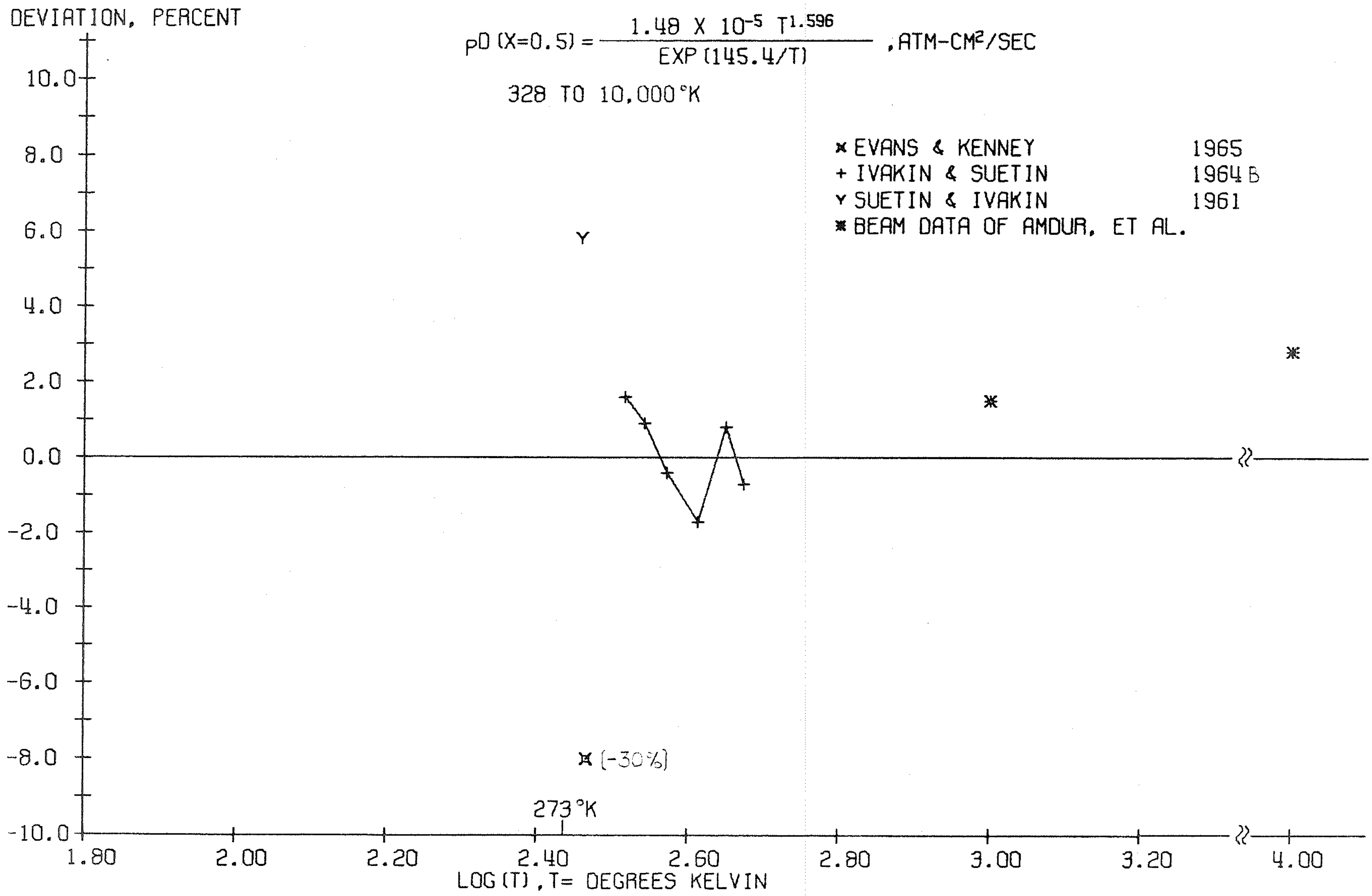


FIG. 51. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, ARGON - SF_6

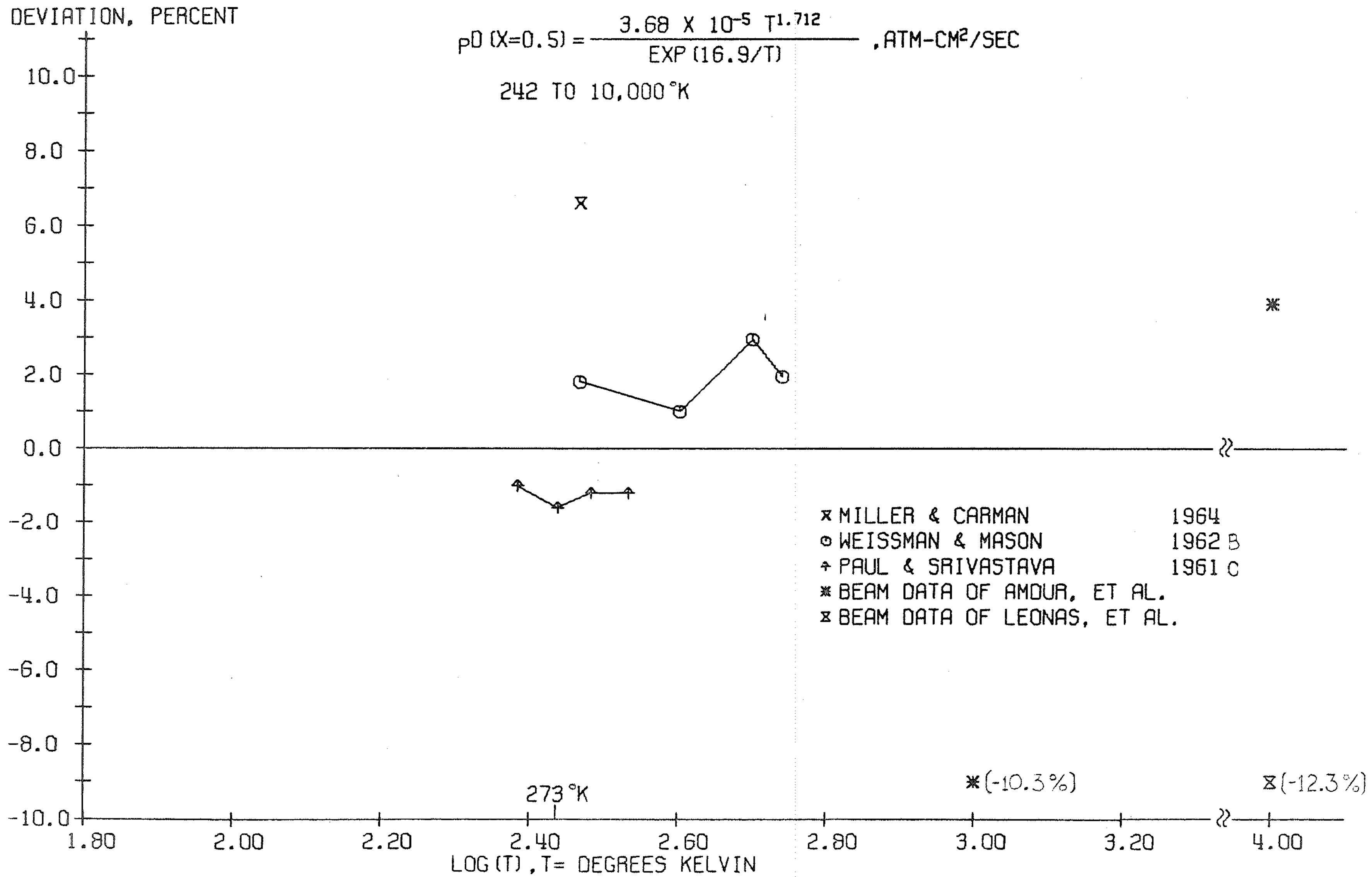


FIG. 52. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - XENON

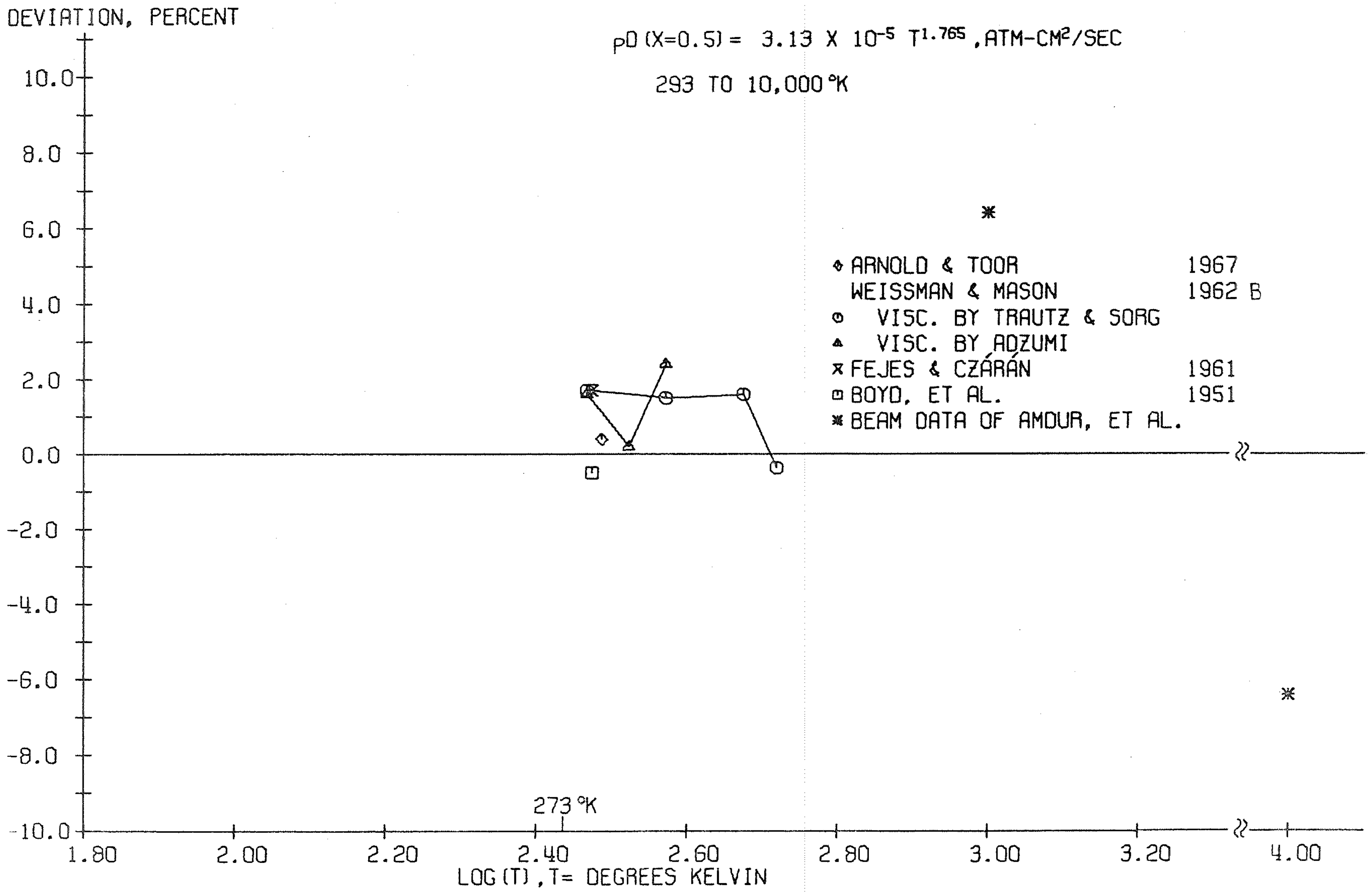


FIG. 53. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - METHANE

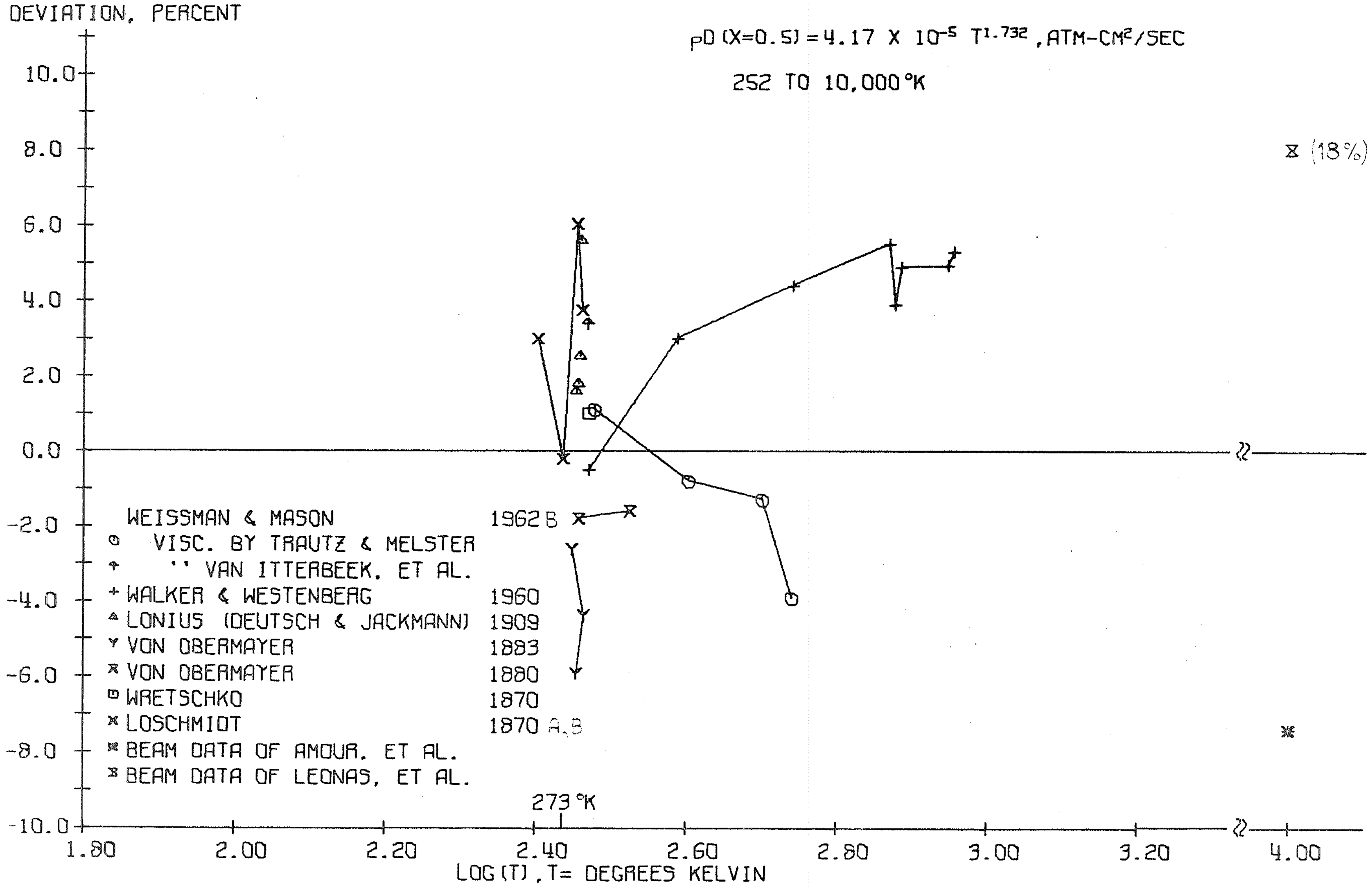


FIG. 54. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN - OXYGEN

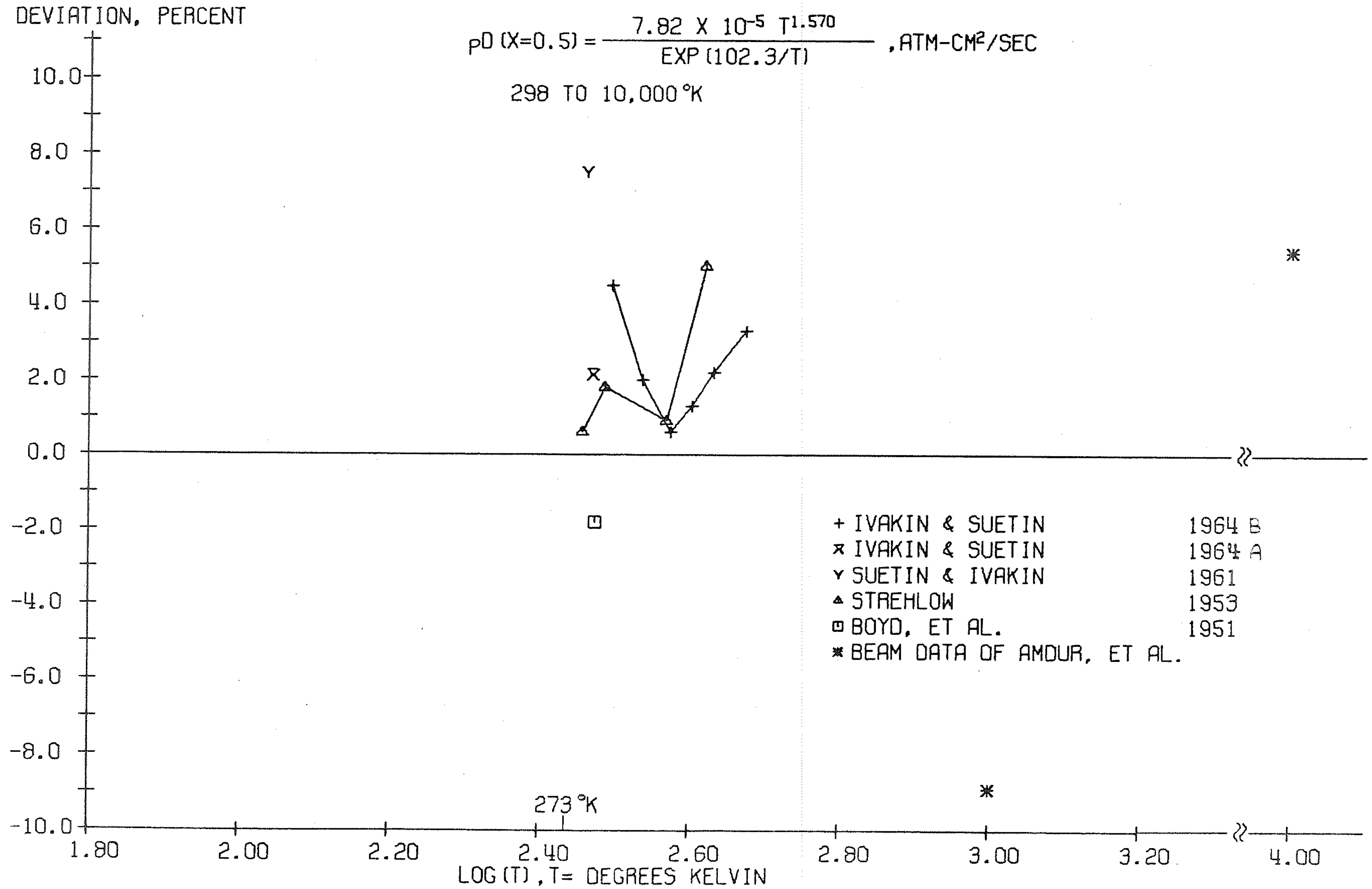


FIG. 55. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HYDROGEN – SF_6

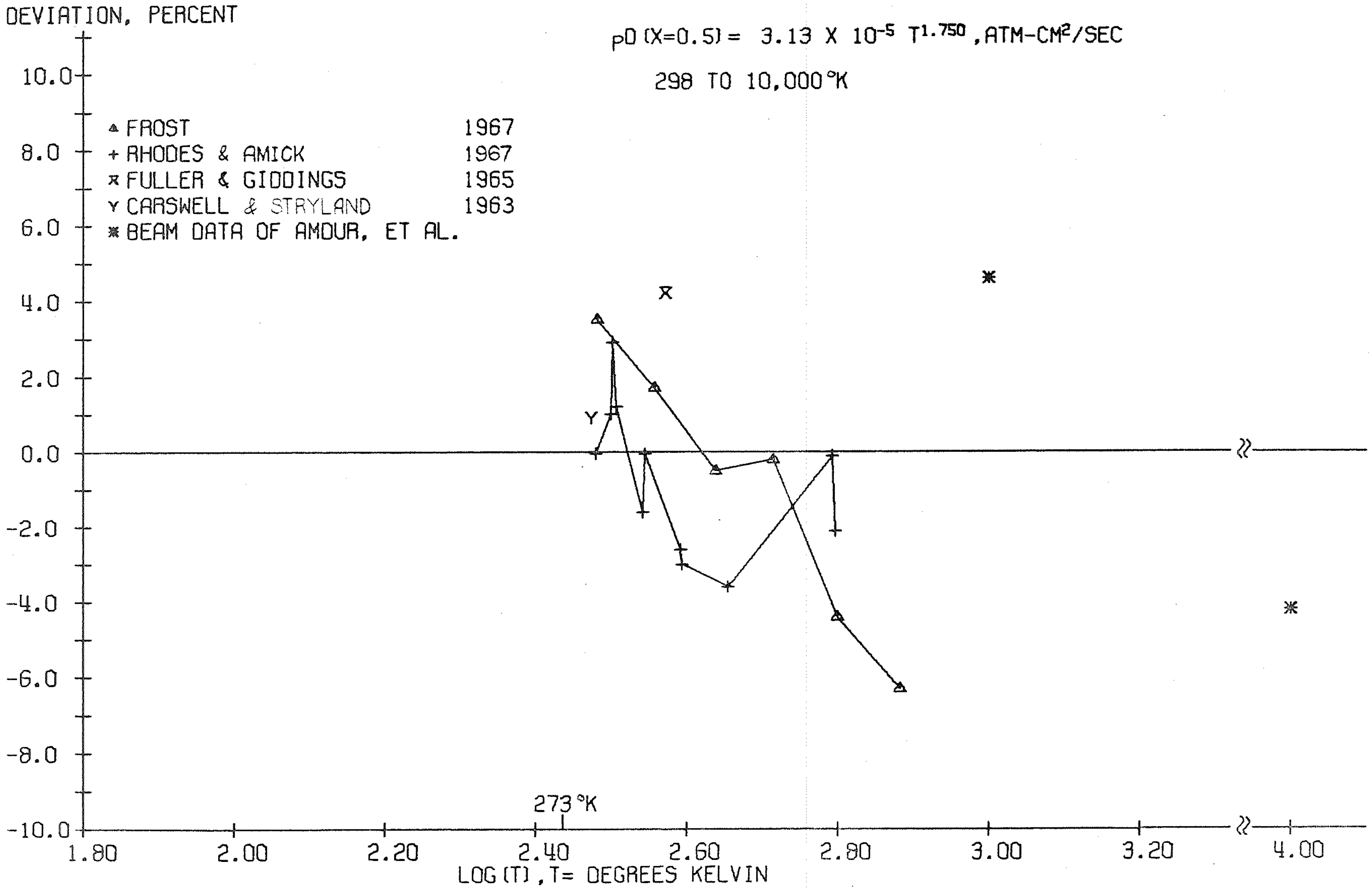


FIG. 56. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - METHANE

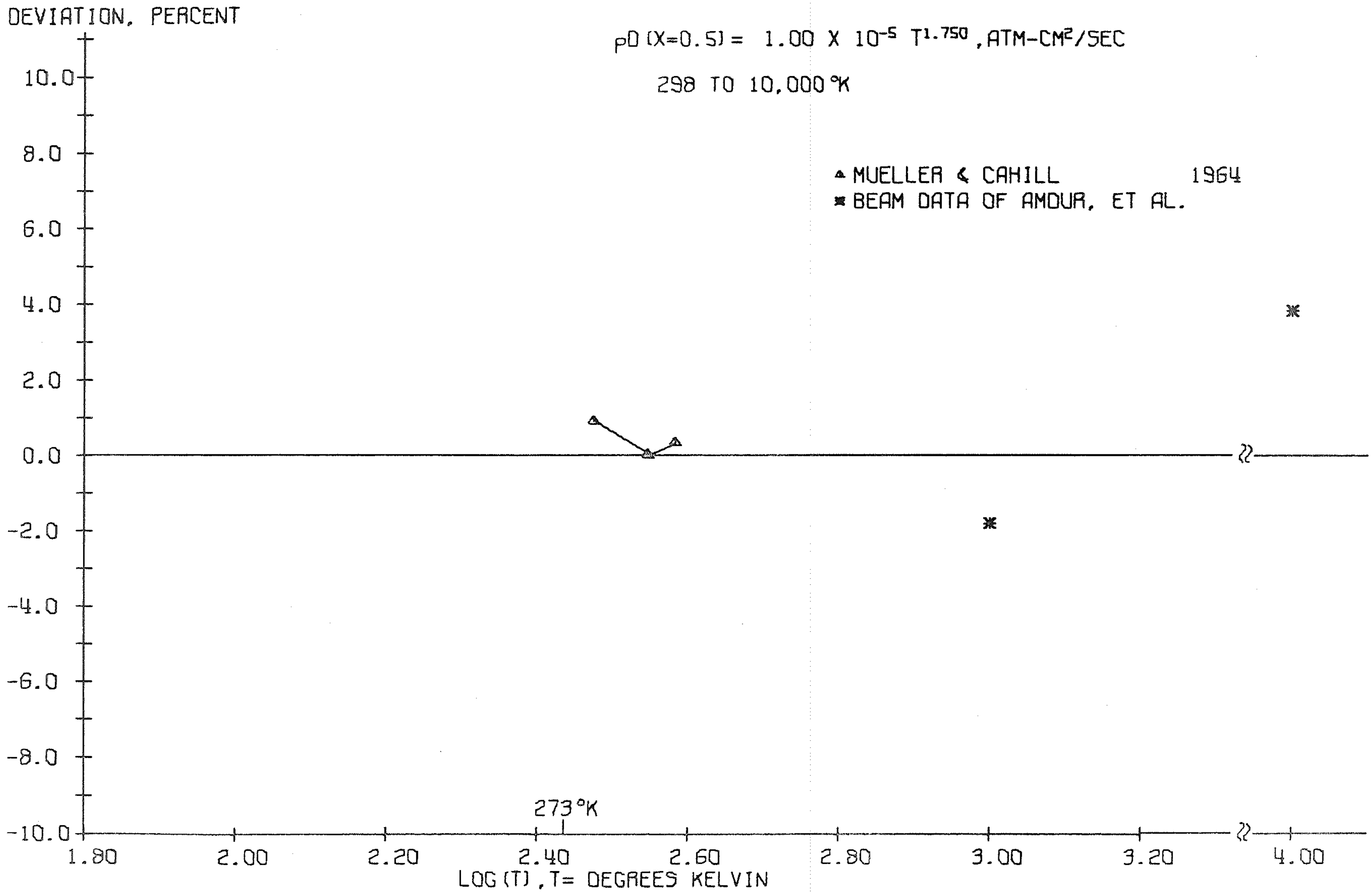


FIG. 57. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, METHANE - NITROGEN

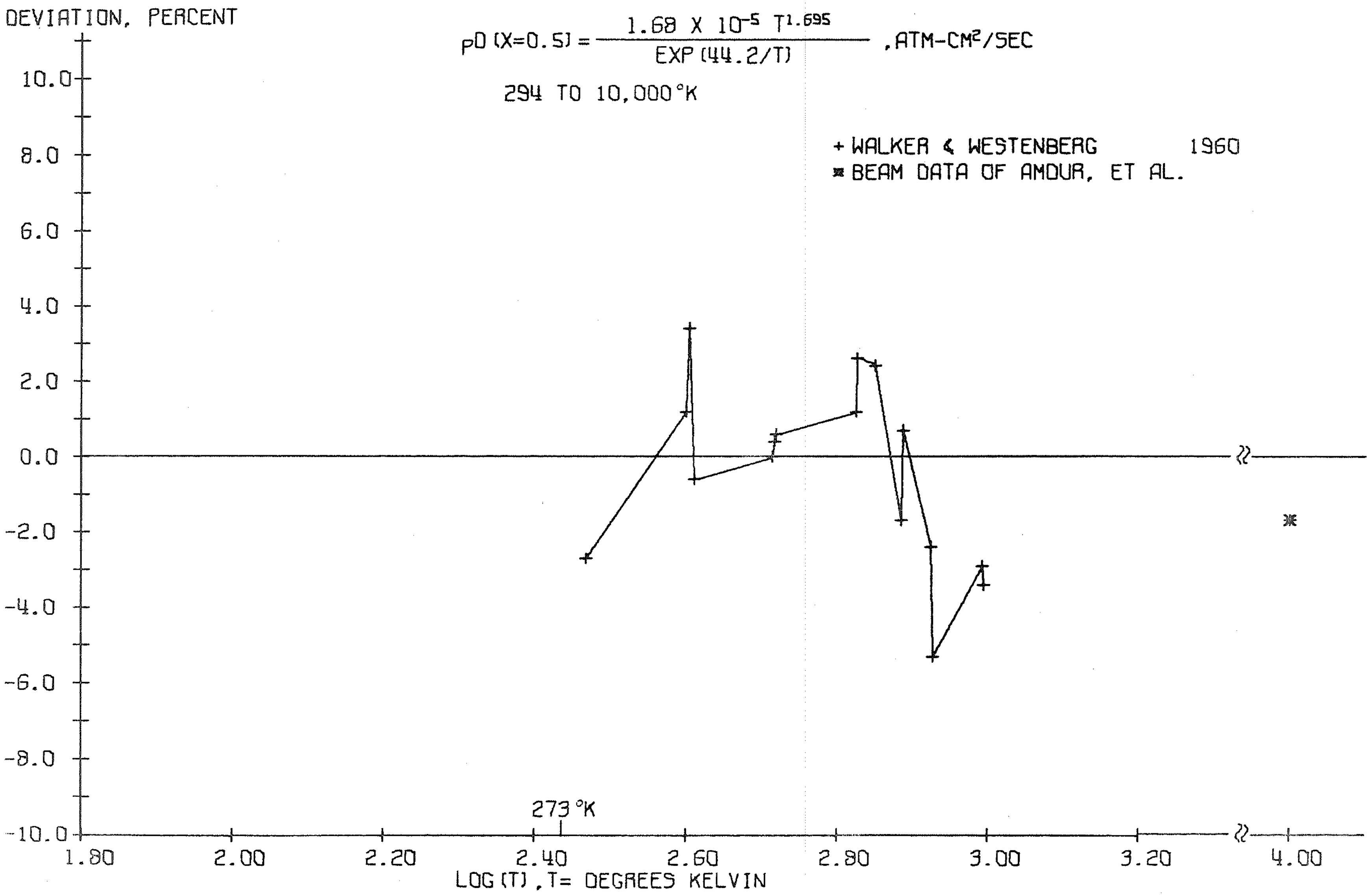


FIG. 58. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, METHANE - OXYGEN

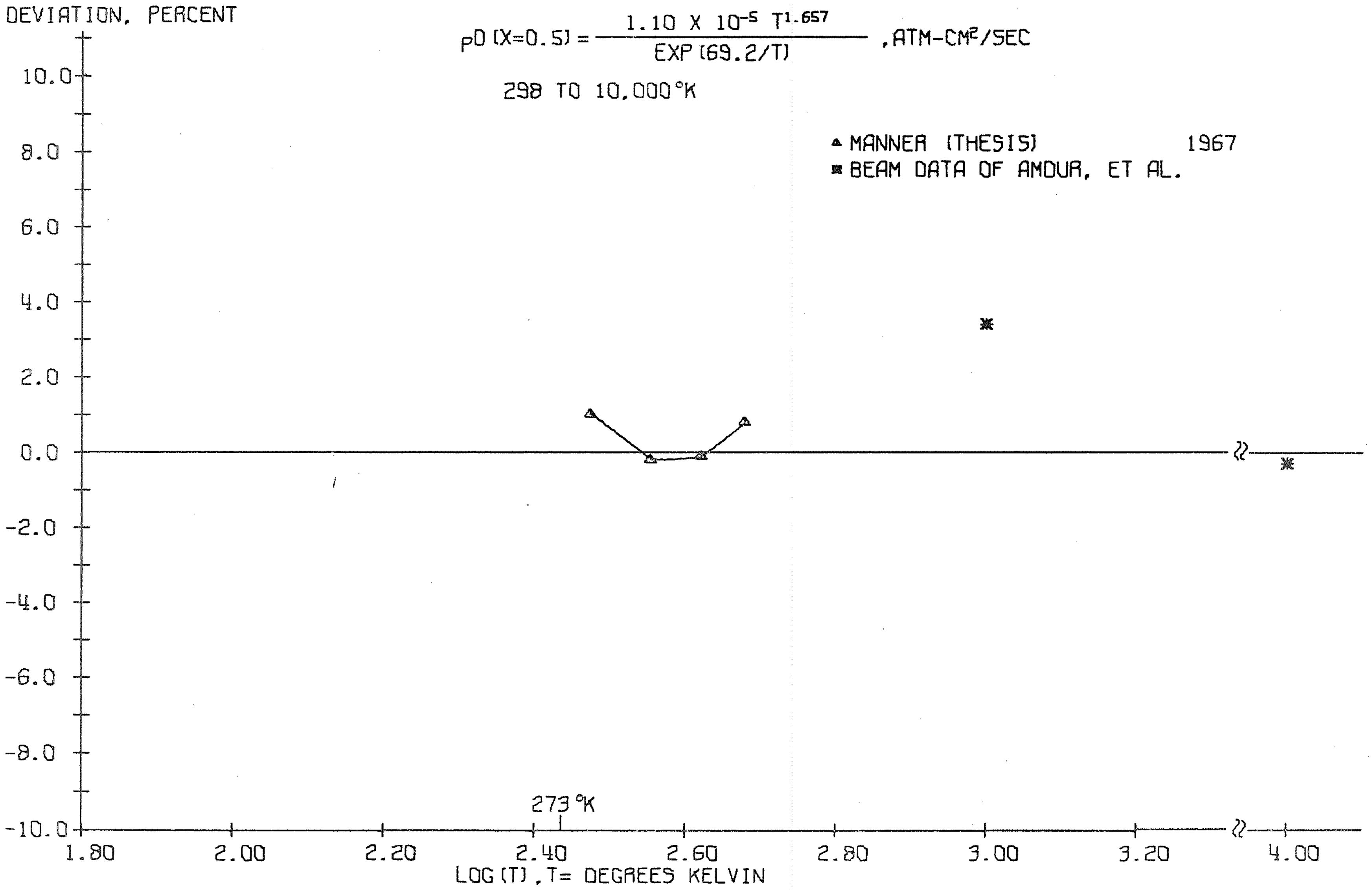


FIG. 59. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. METHANE - SF_6

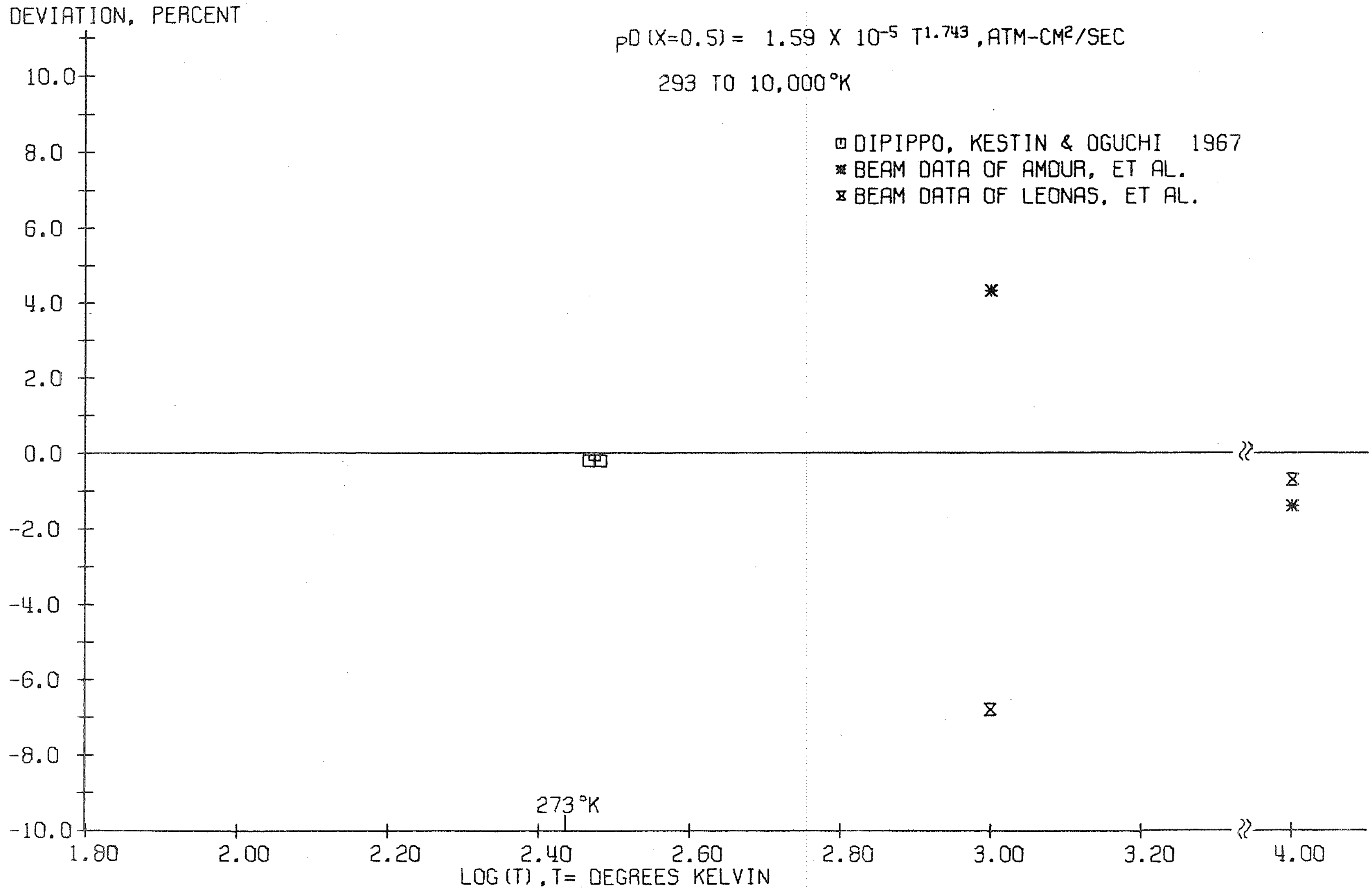


FIG. 60. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NEON - NITROGEN

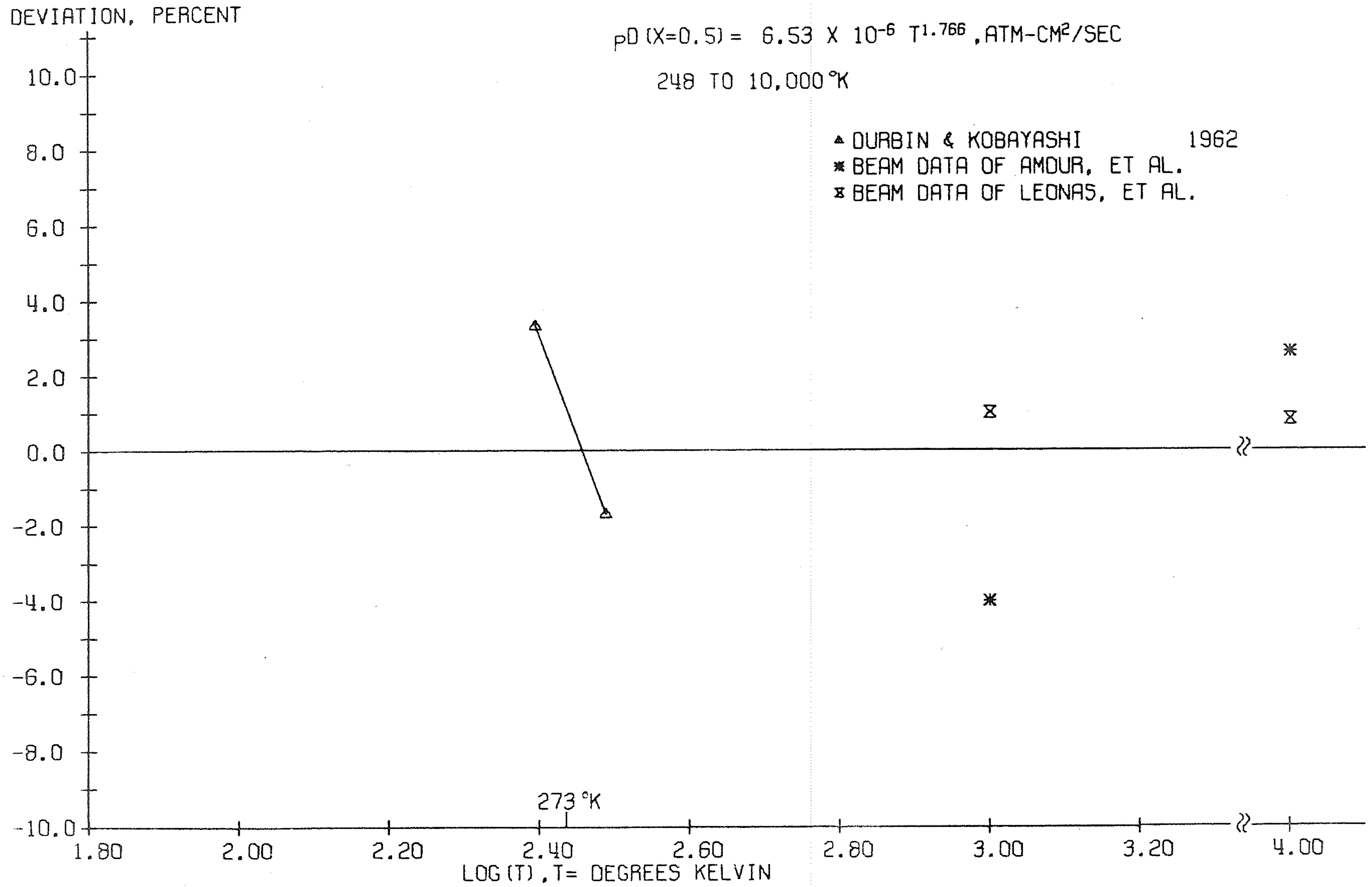


FIG. 61. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - KRYPTON

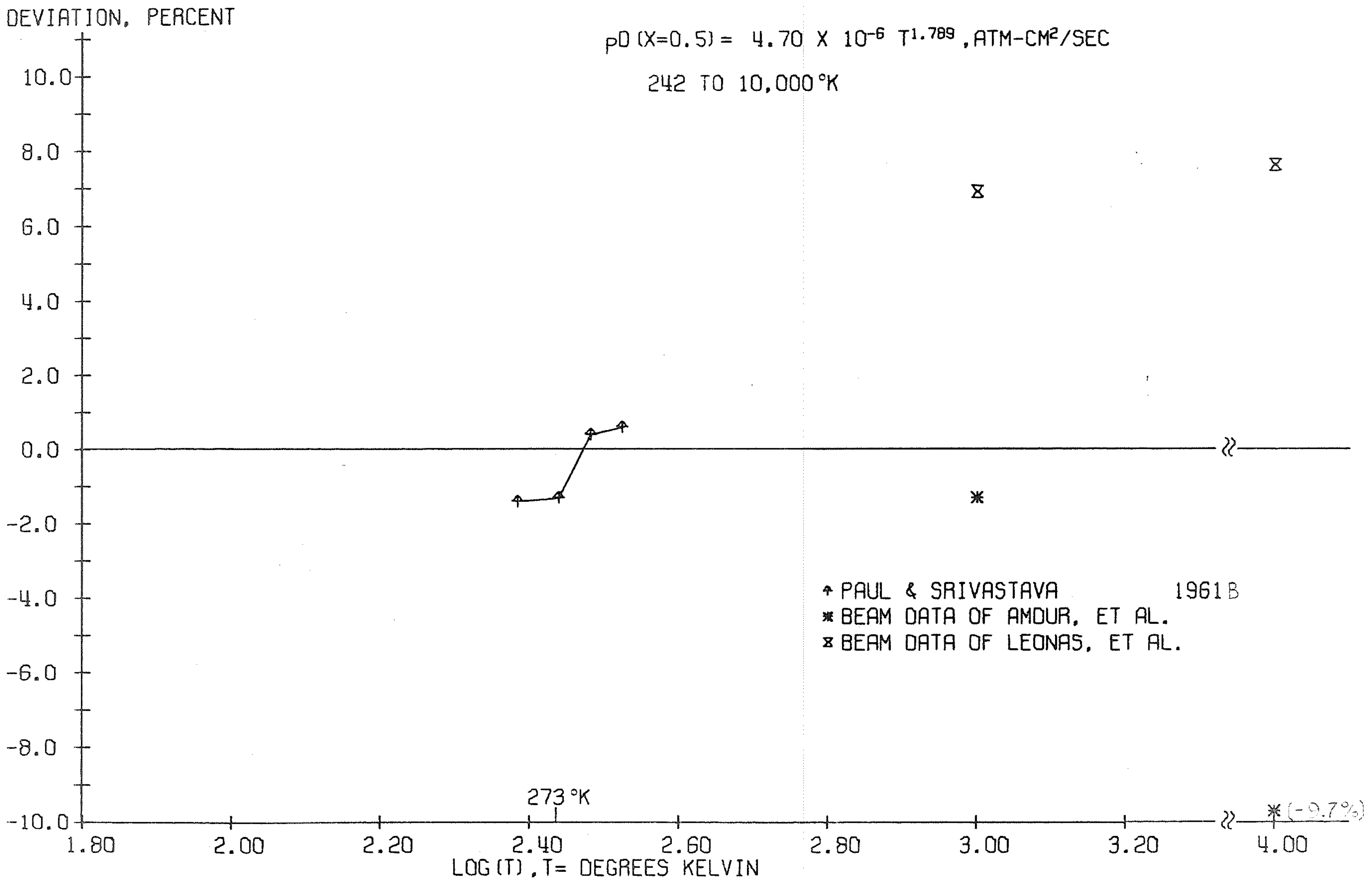


FIG. 62. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - XENON

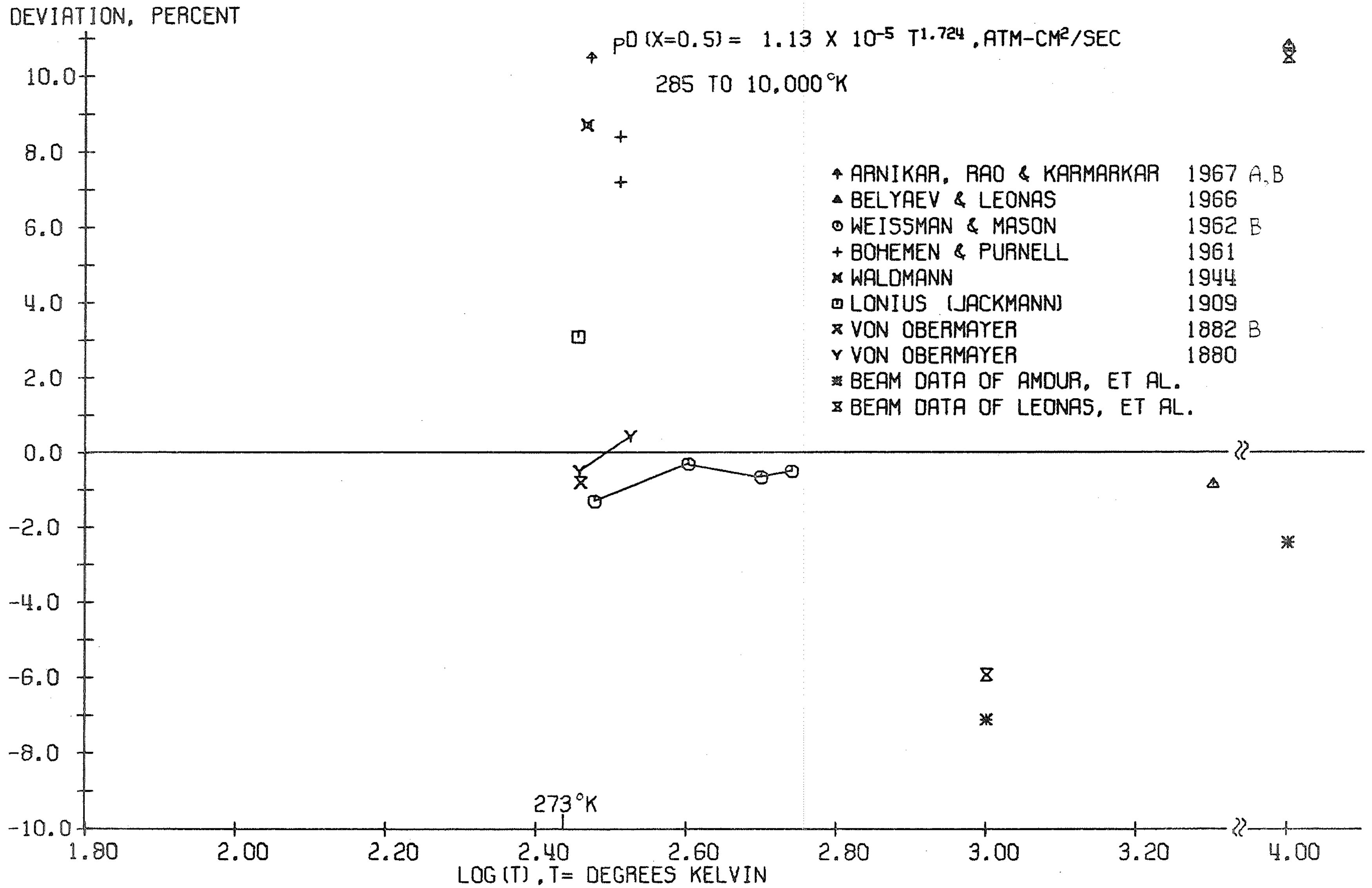


FIG. 63. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - OXYGEN

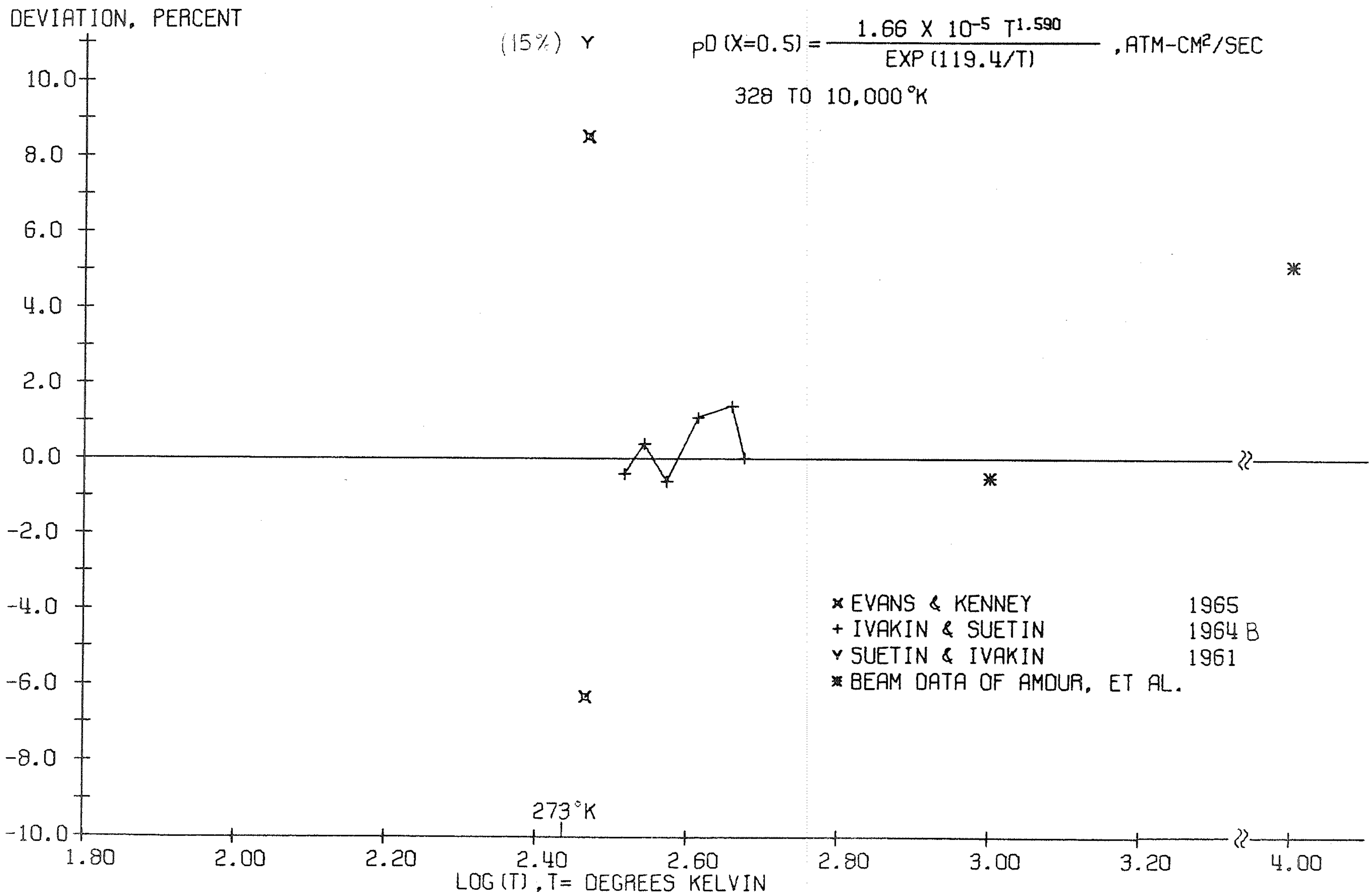


FIG. 64. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NITROGEN - SF_6

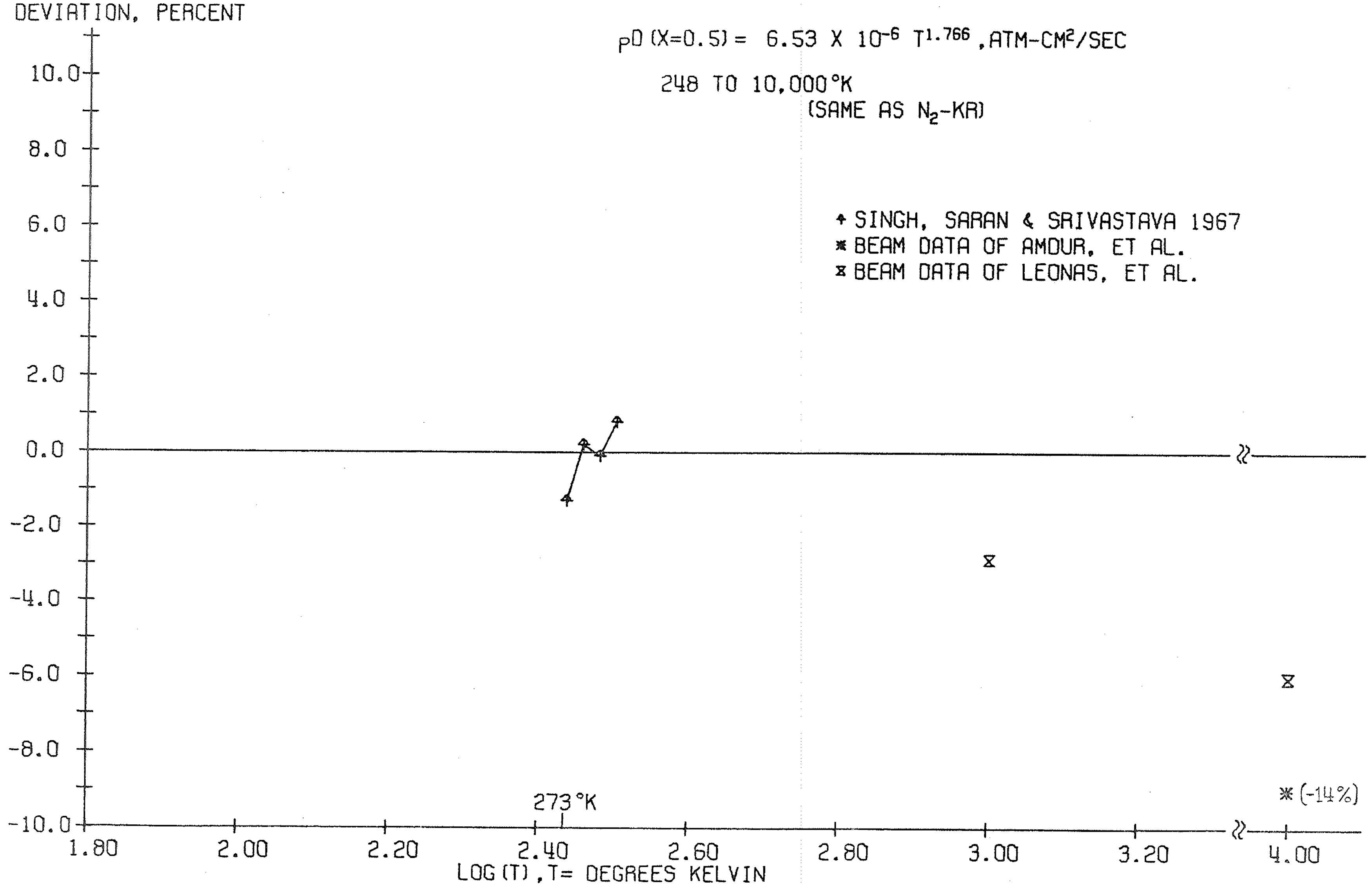


FIG. 65. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - KRYPTON

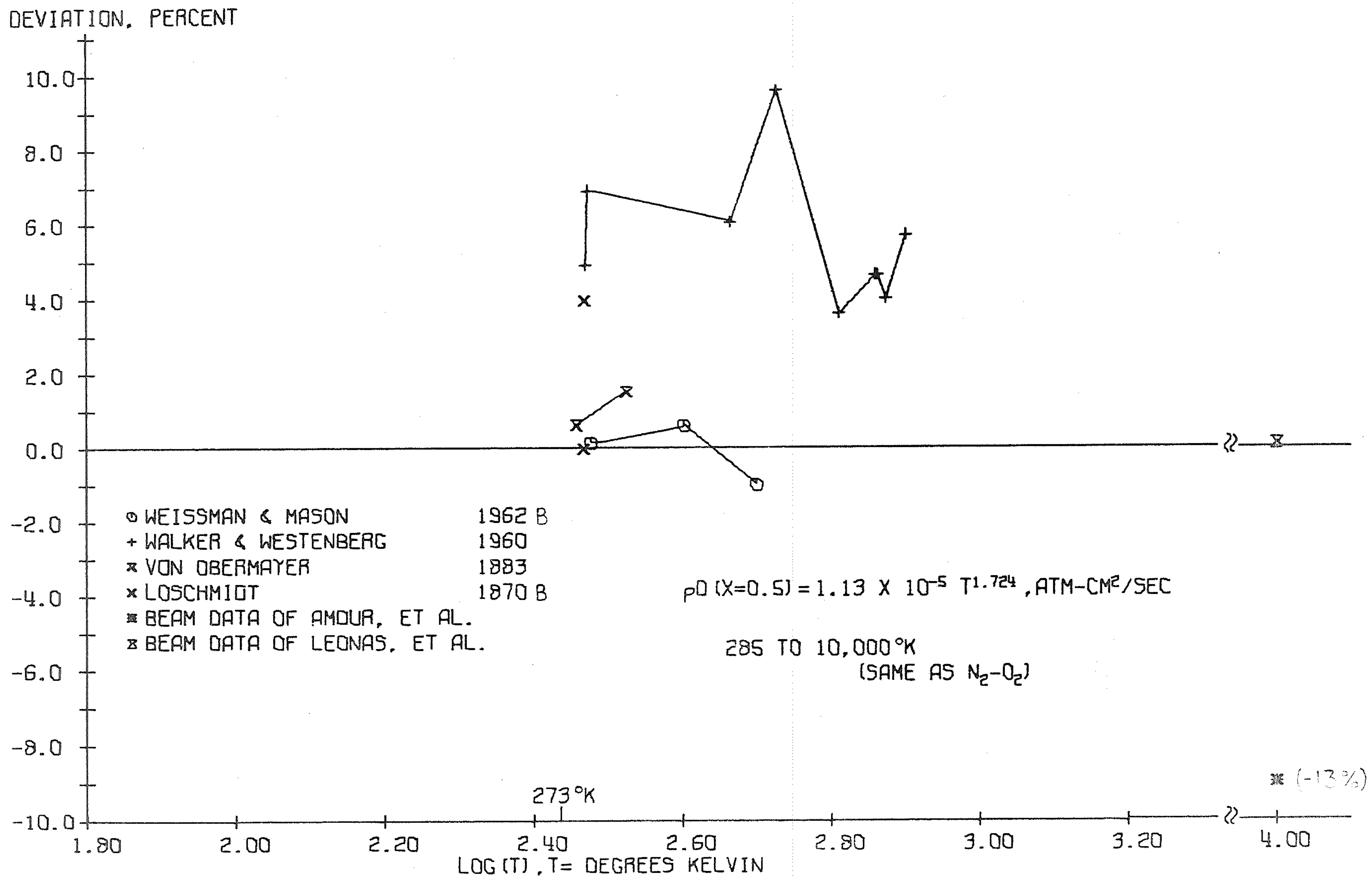


FIG. 66. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CO - OXYGEN

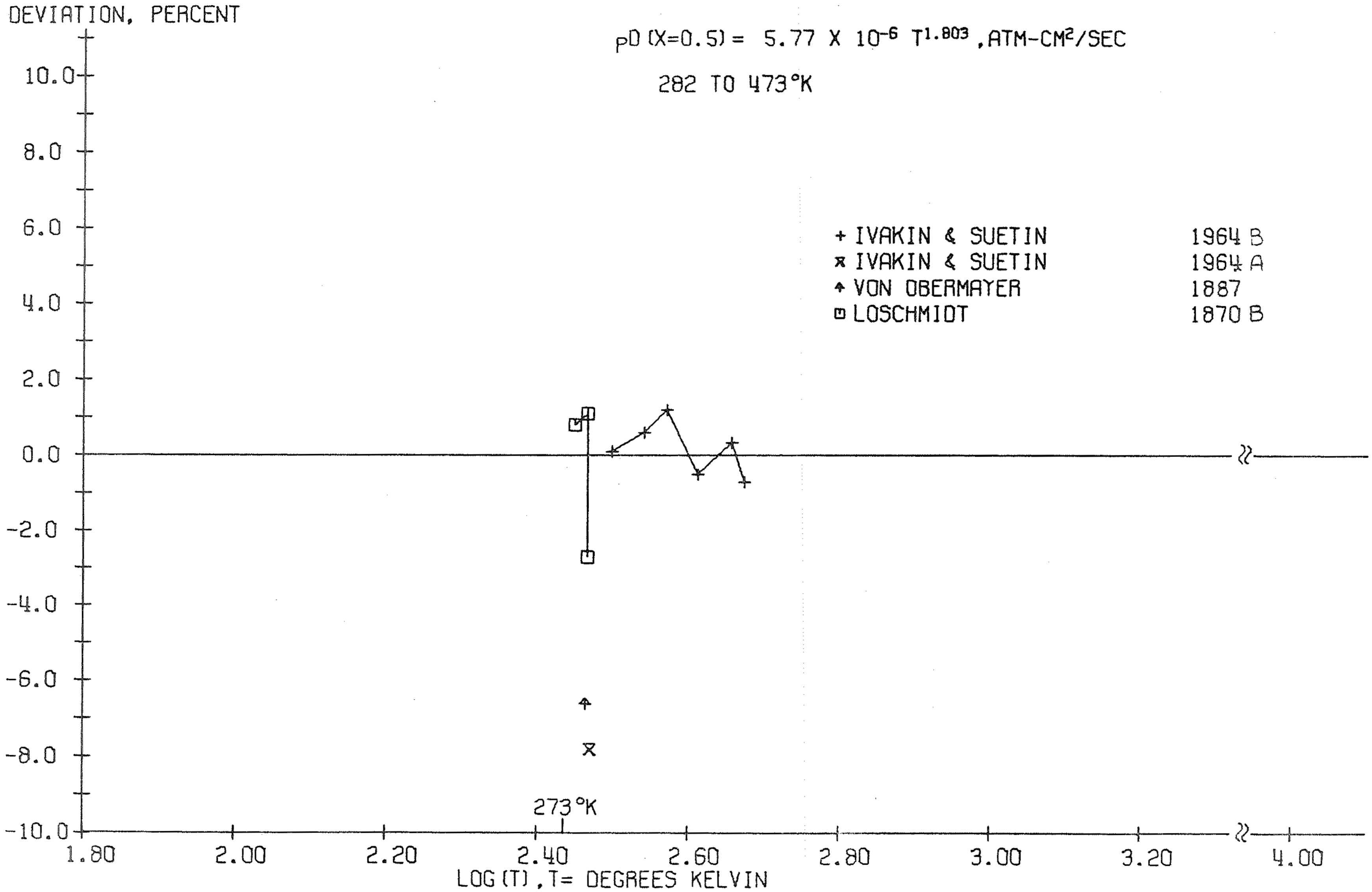


FIG.67. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - CO_2

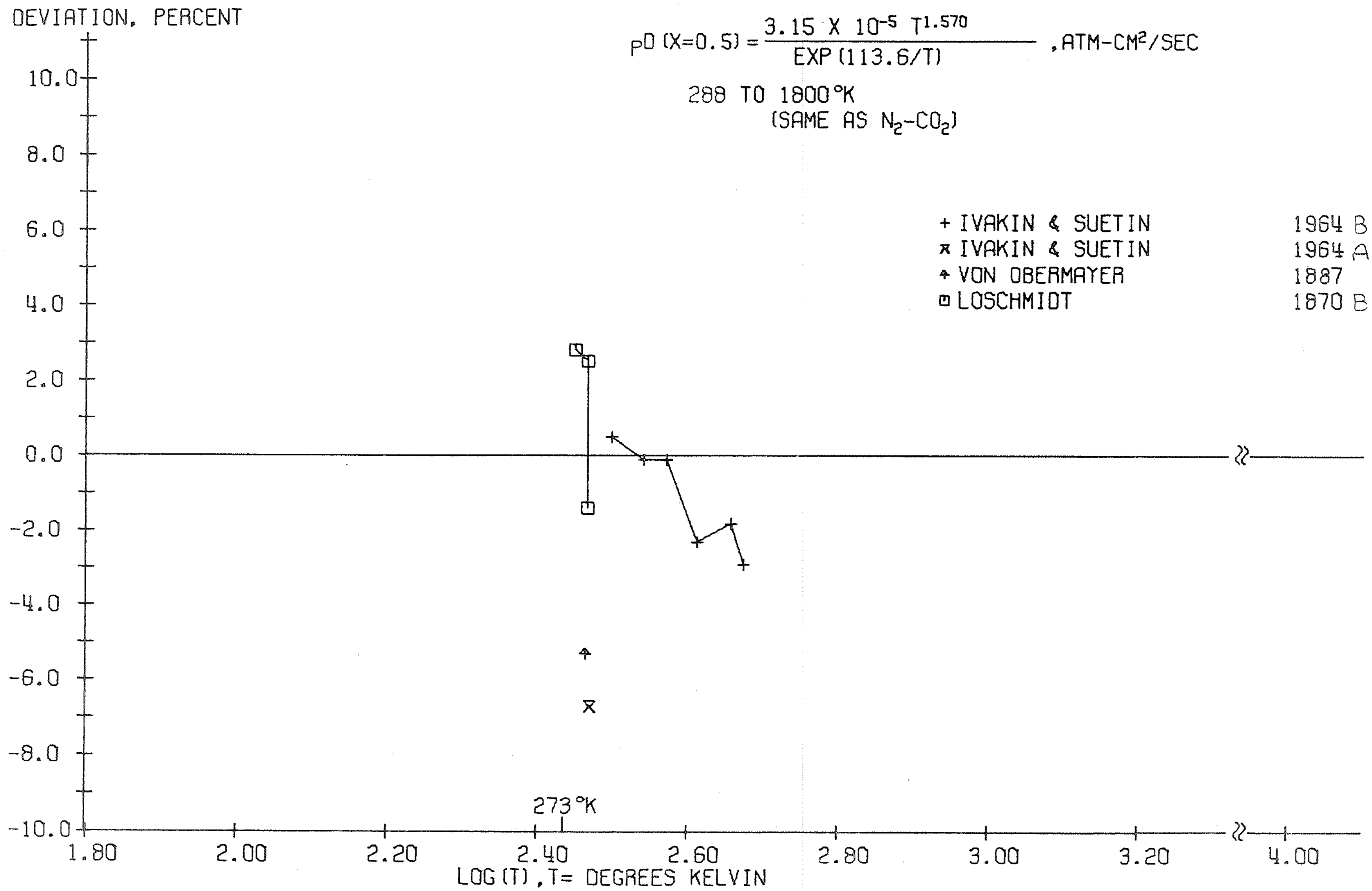


FIG. 68. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE - CO_2

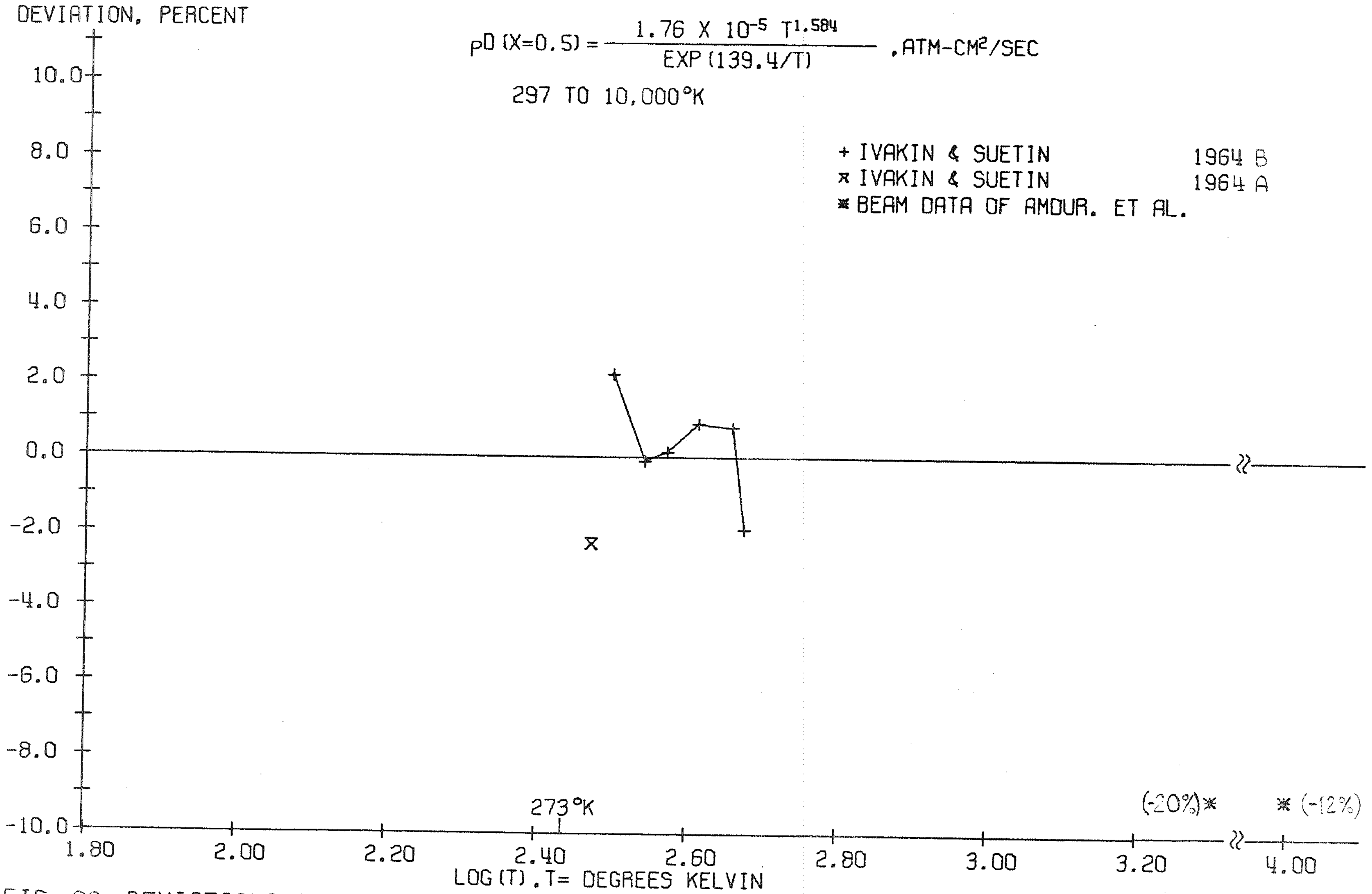


FIG. 69. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON MONOXIDE-SF_6

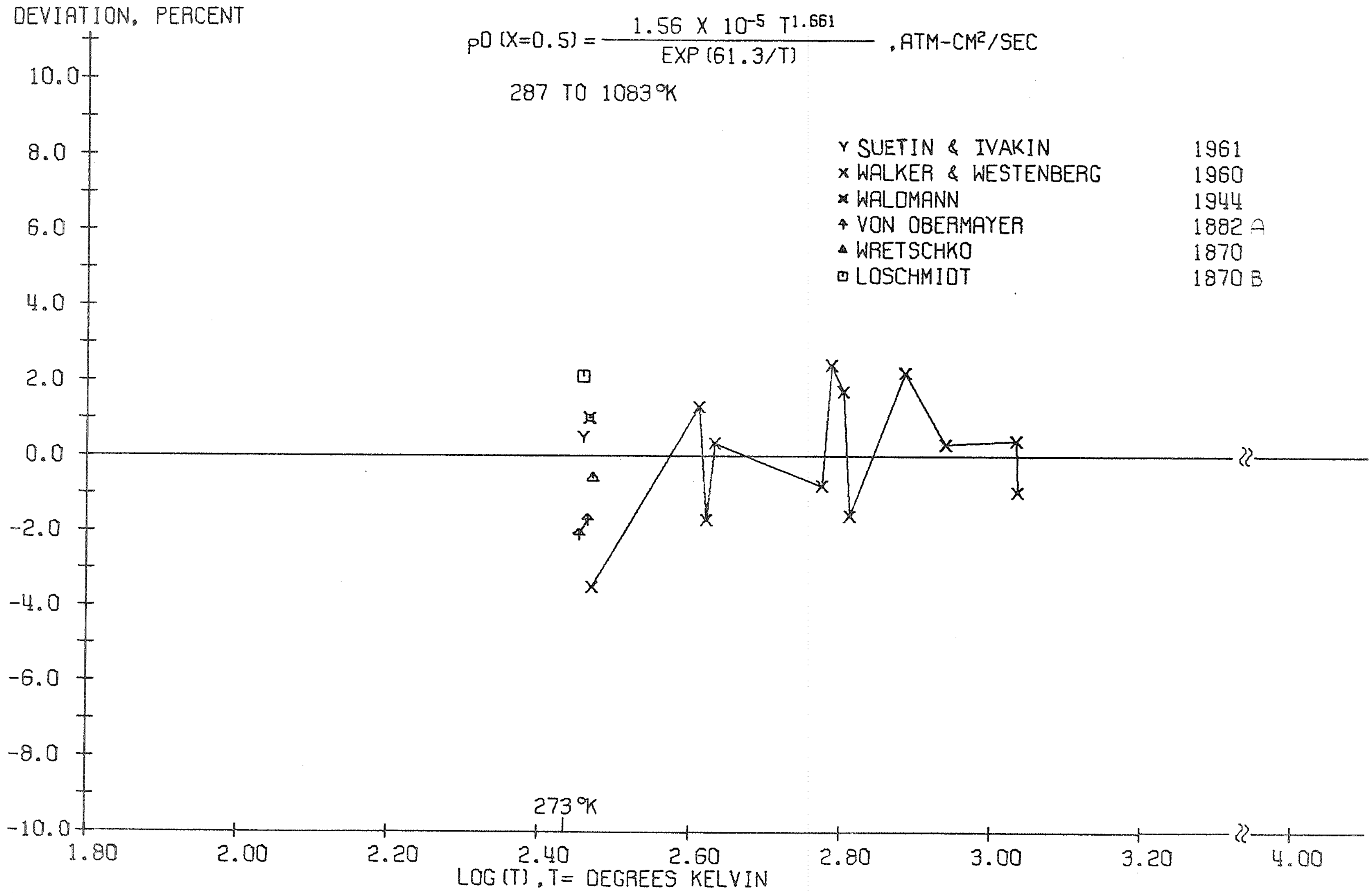


FIG. 70. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, OXYGEN - CARBON DIOXIDE

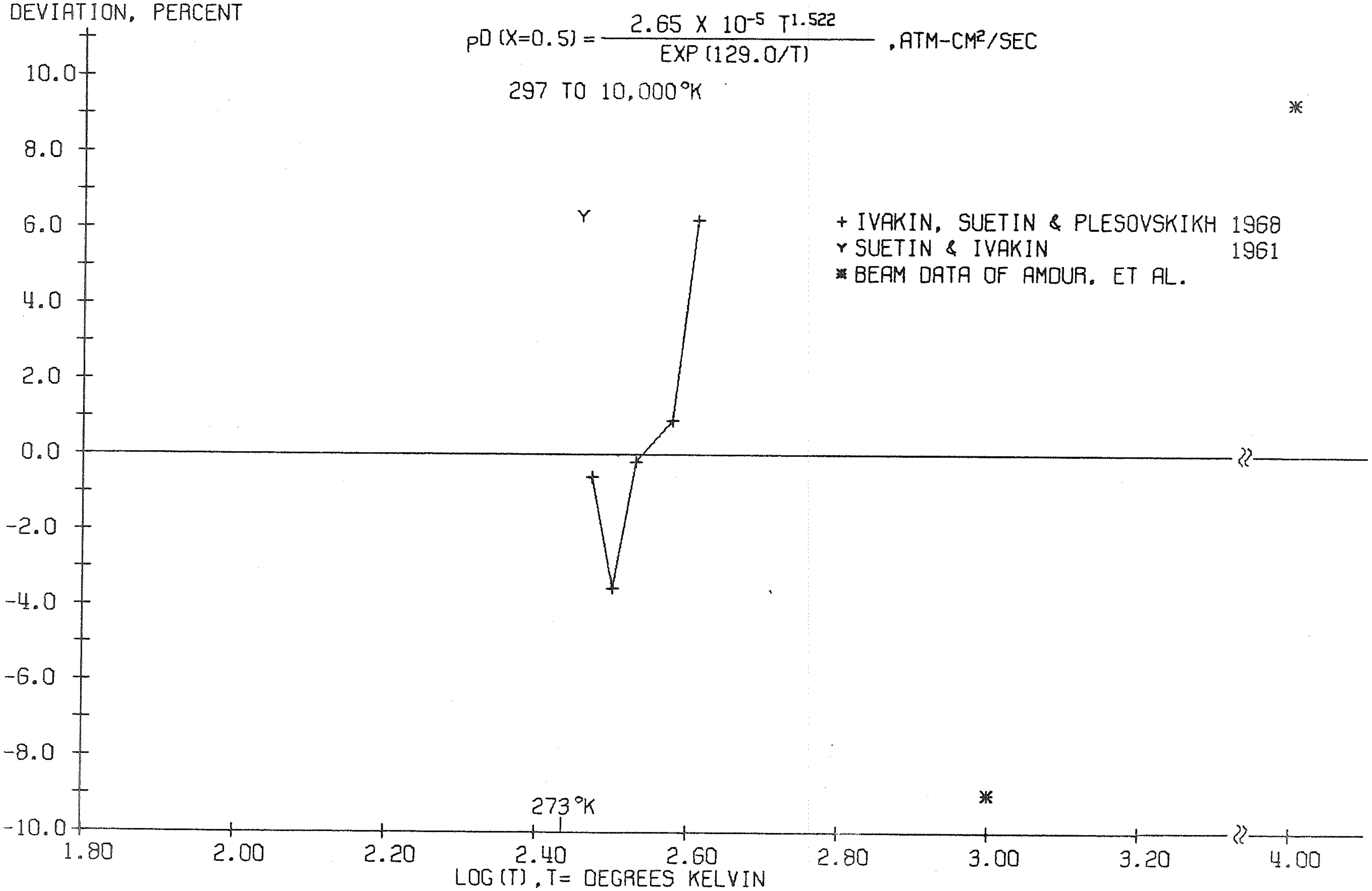


FIG. 71. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. OXYGEN - SF_6

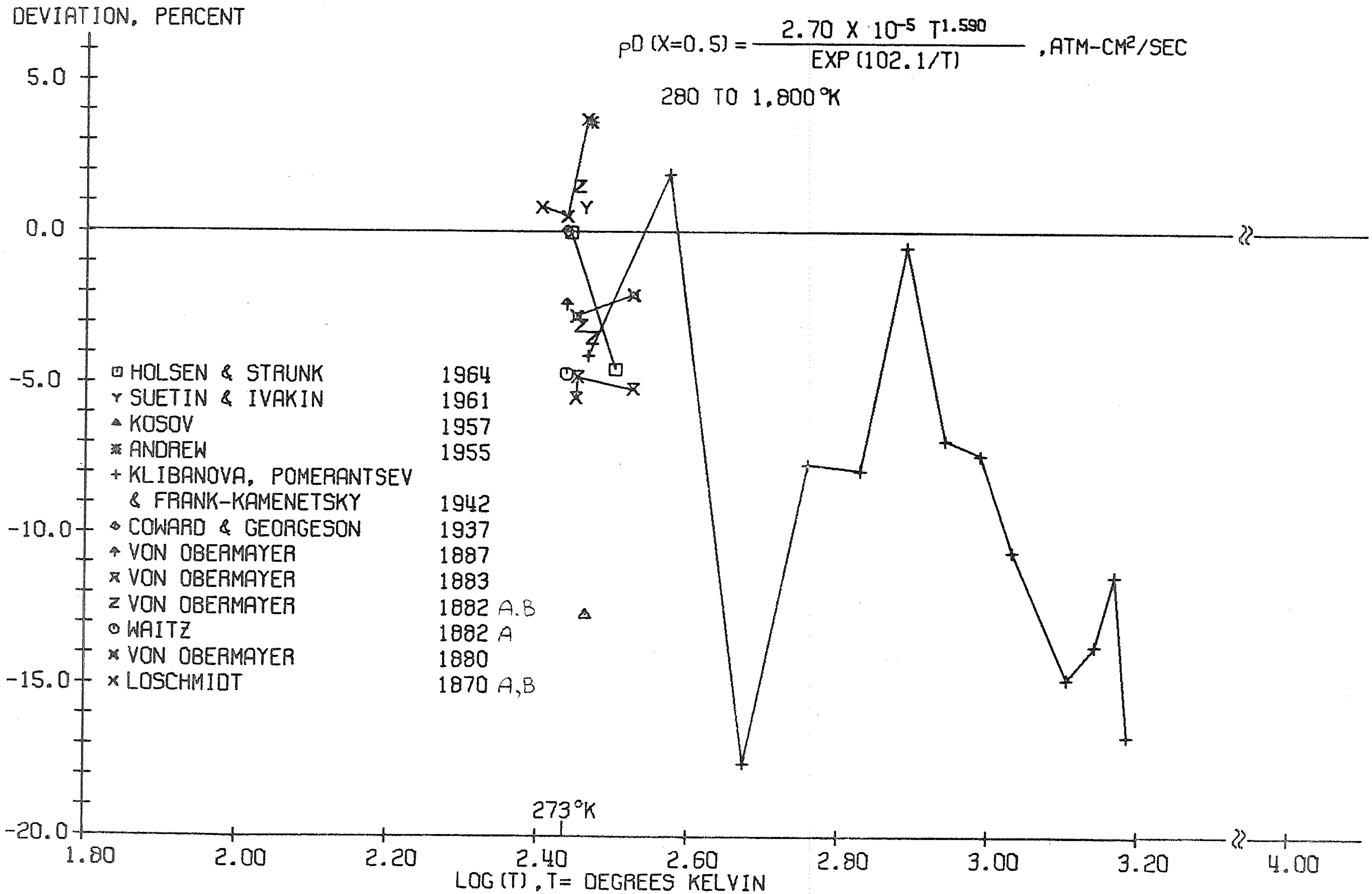


FIG. 72. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, AIR - CARBON DIOXIDE

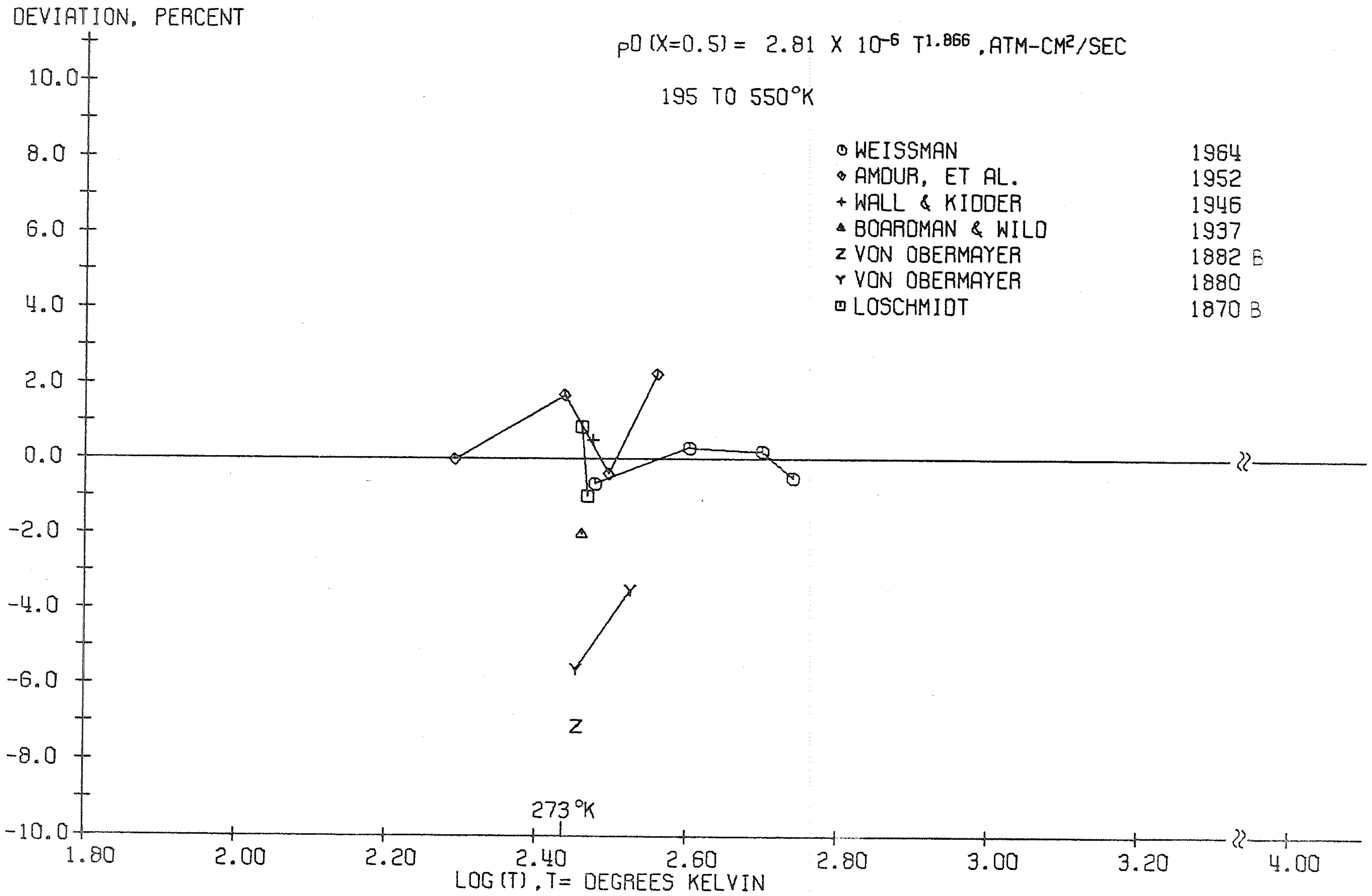


FIG. 73. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. CARBON DIOXIDE - N_2O

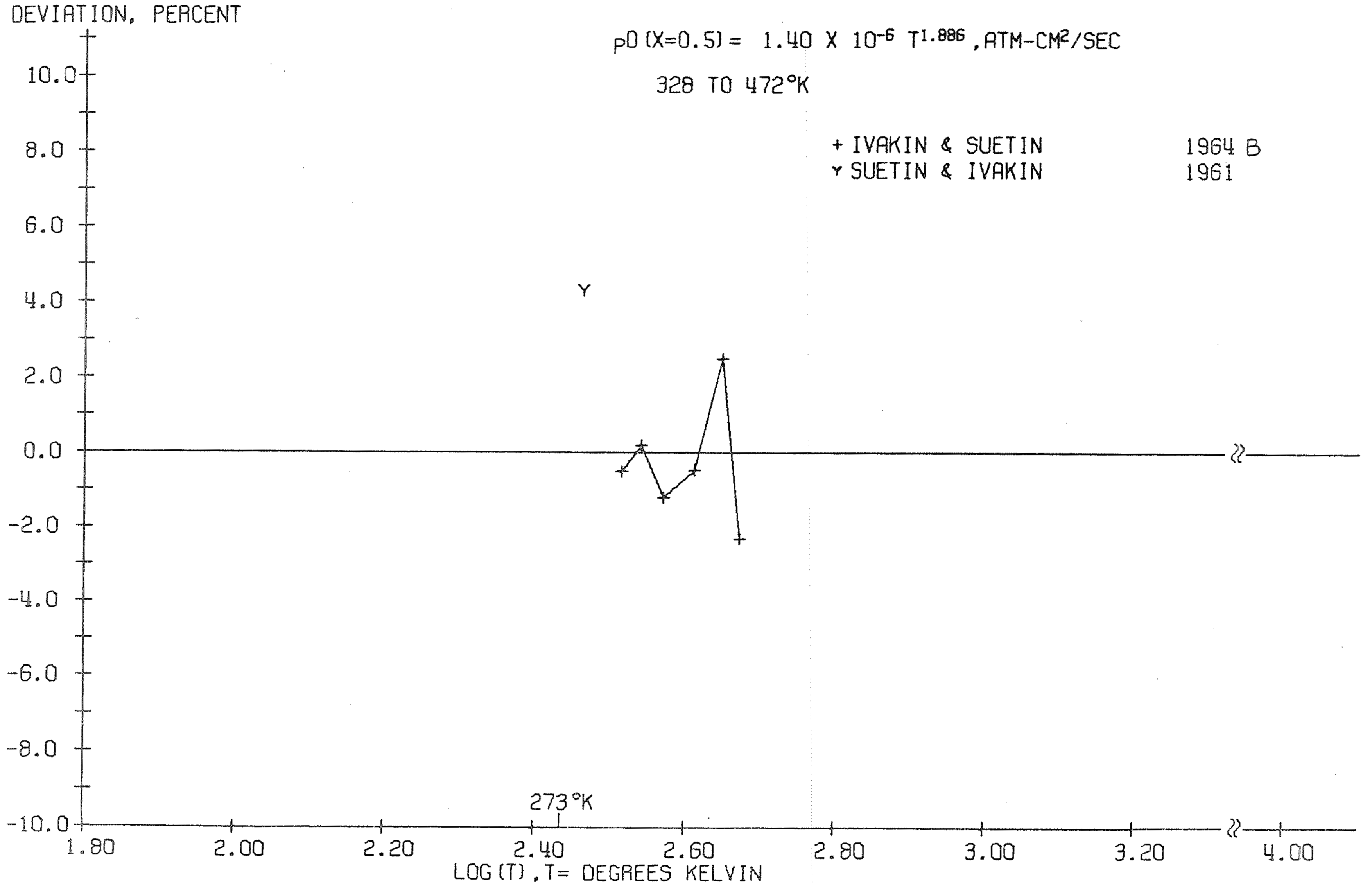


FIG. 74. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, CARBON DIOXIDE - SF_6

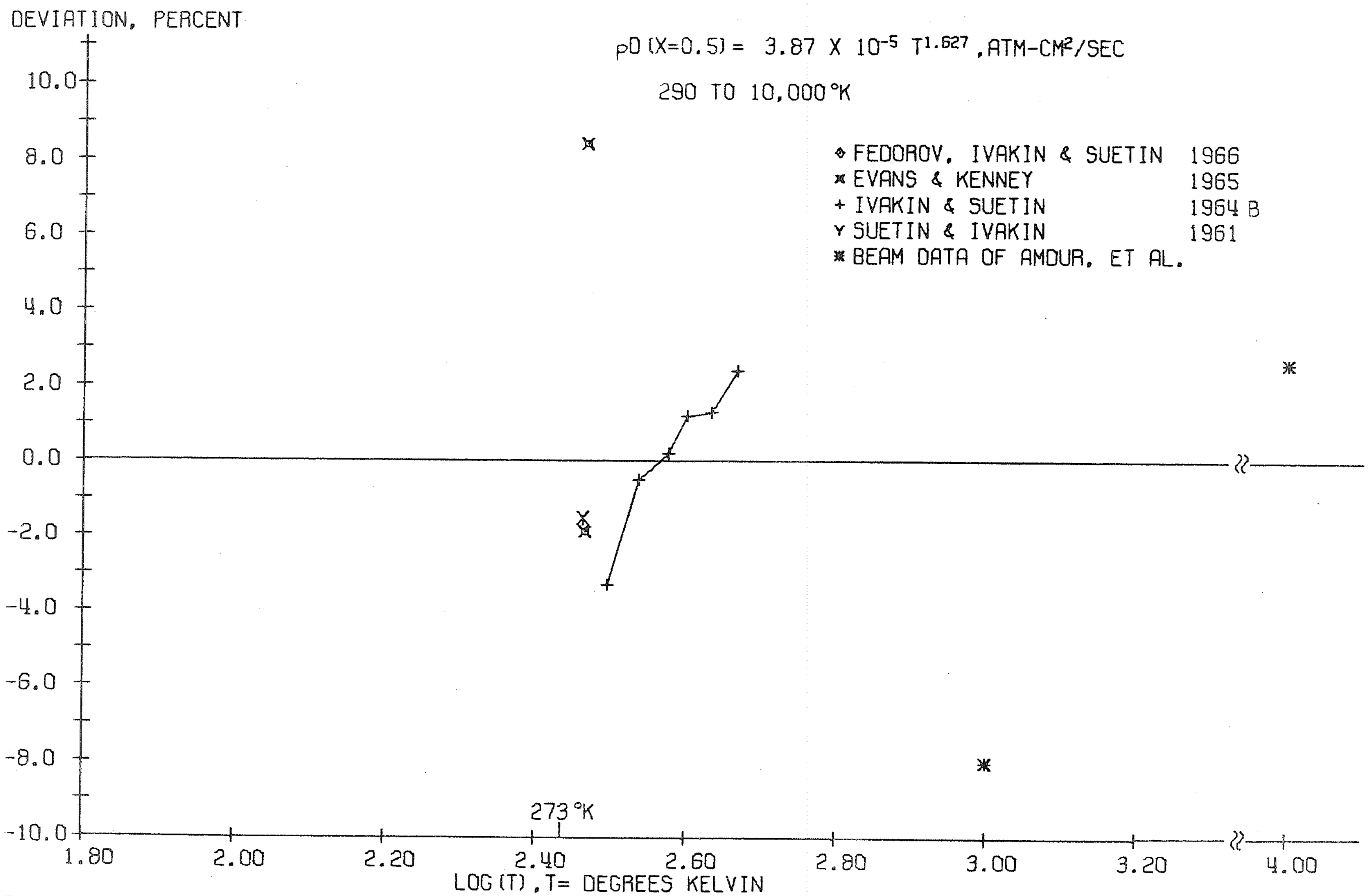


FIG. 75. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, HELIUM - SF_6

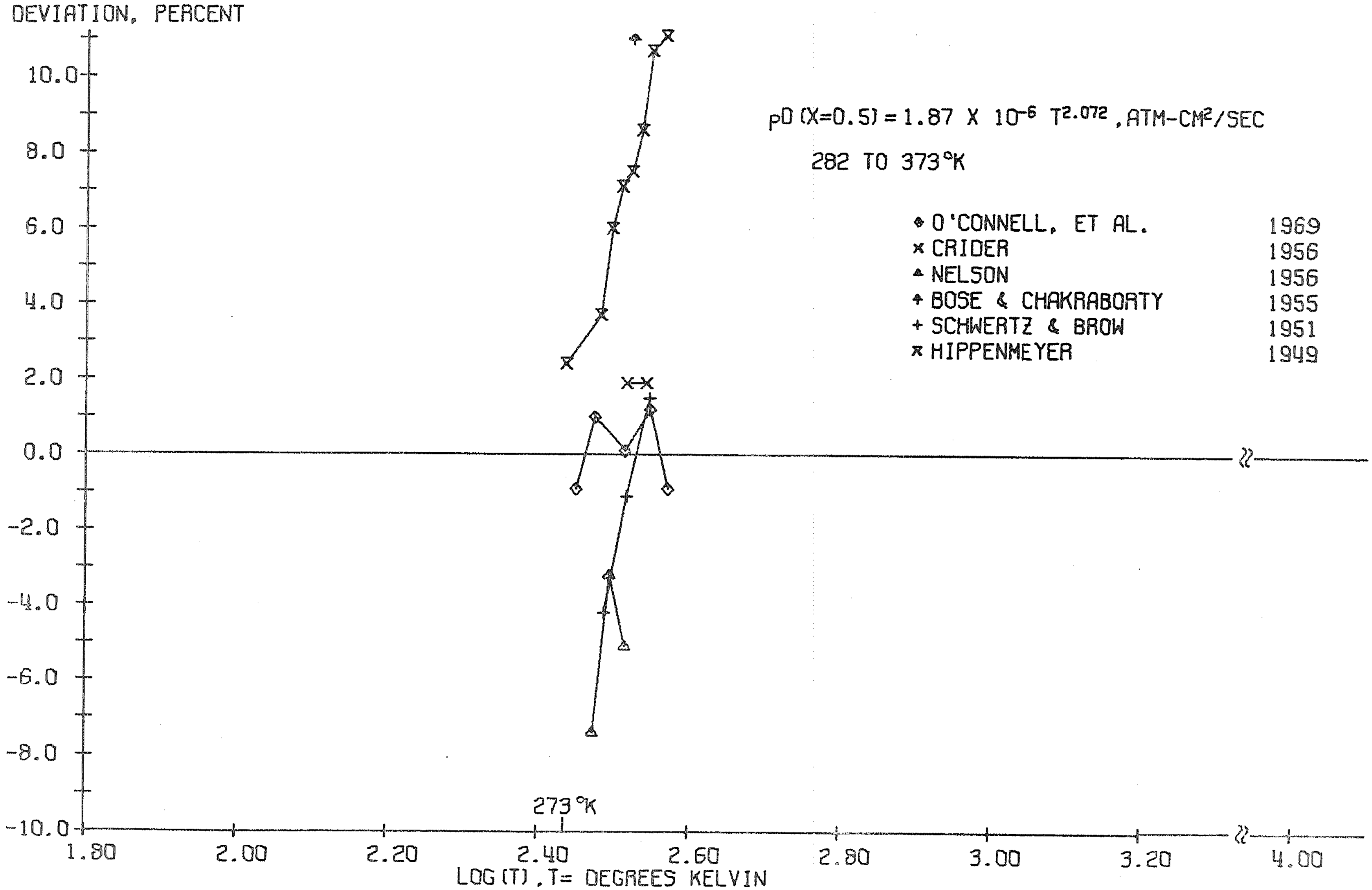


FIG. 76. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. WATER - NITROGEN

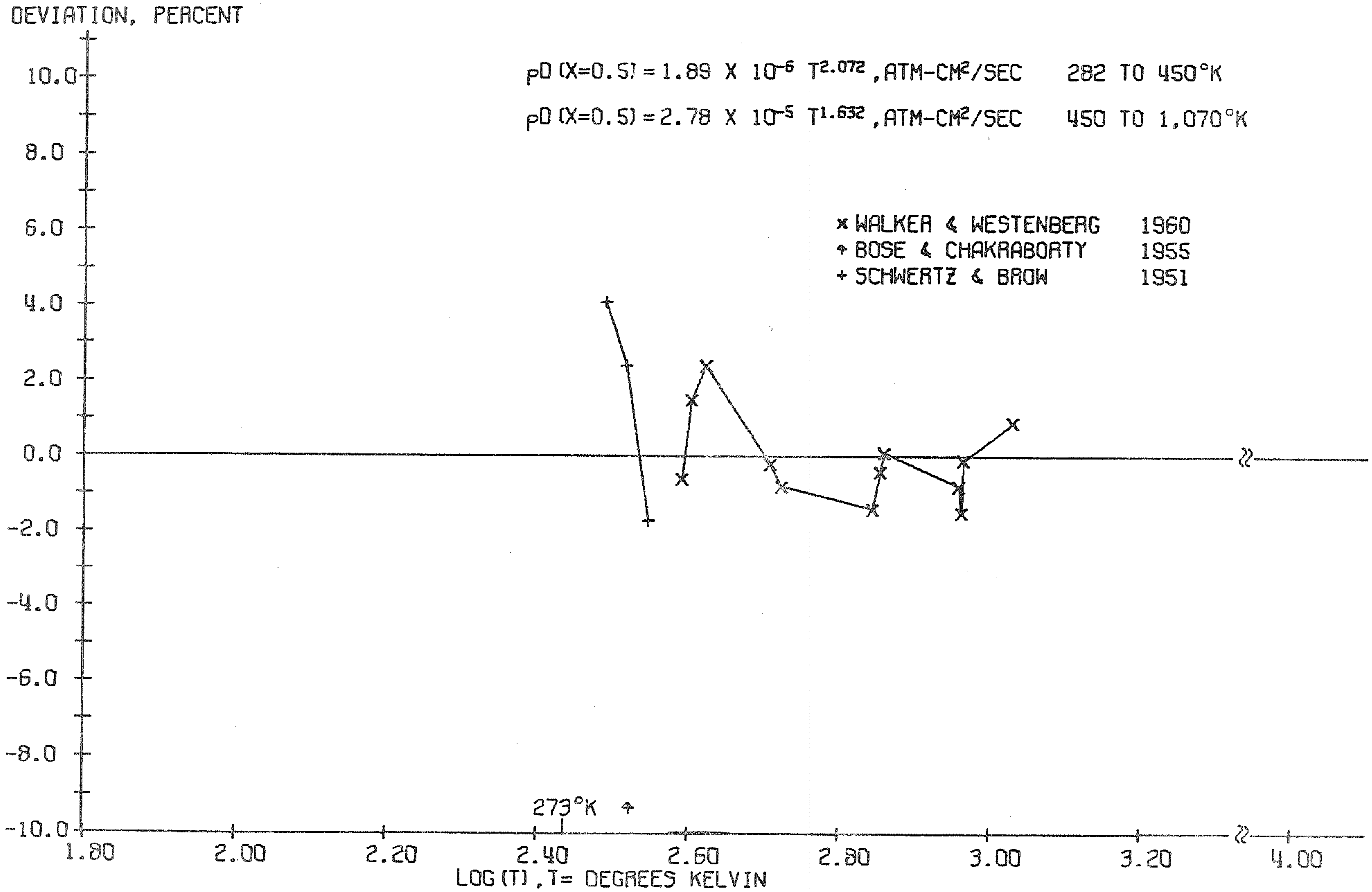


FIG. 77. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION. WATER - OXYGEN

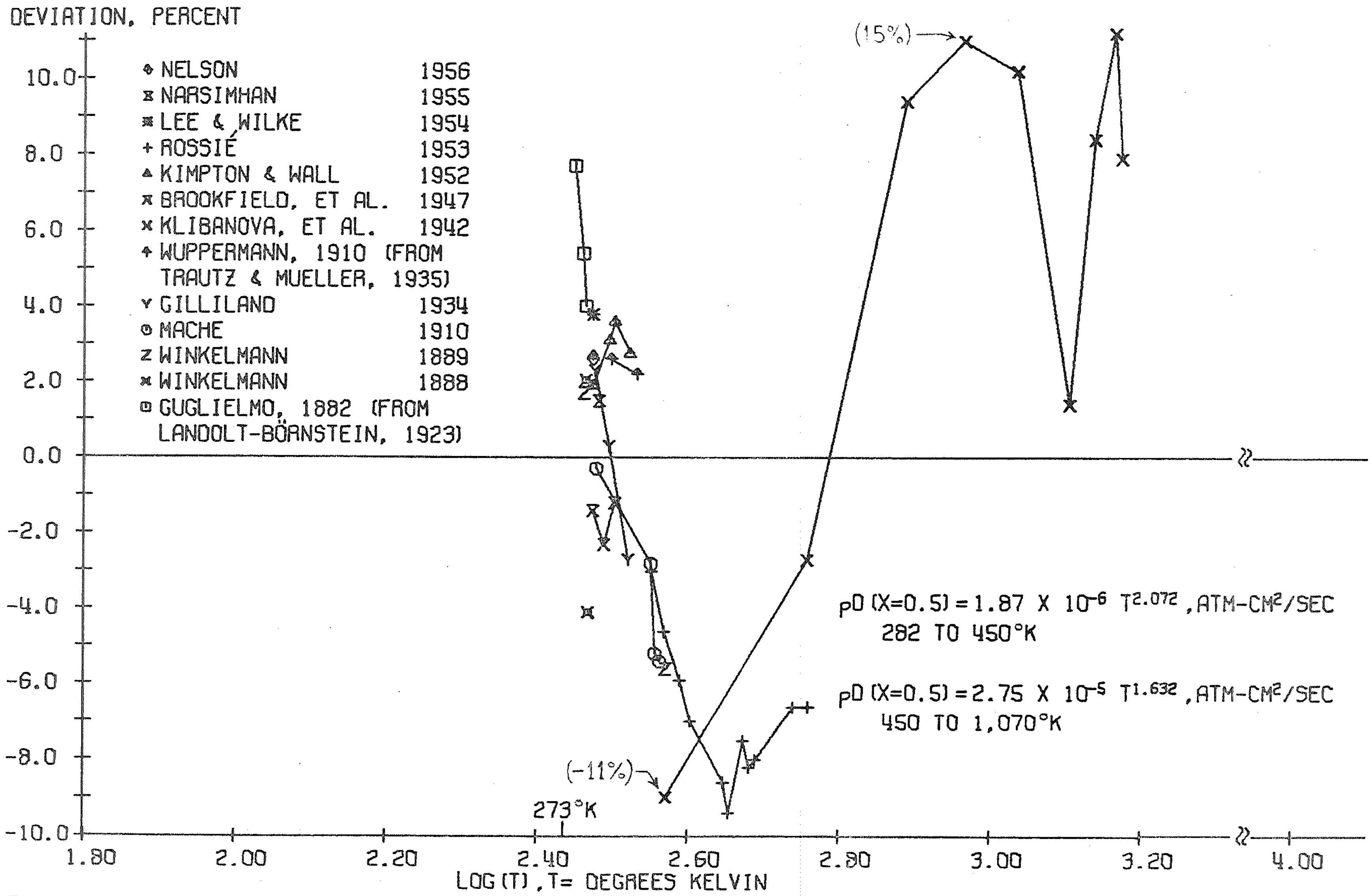


FIG. 78. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, WATER - AIR

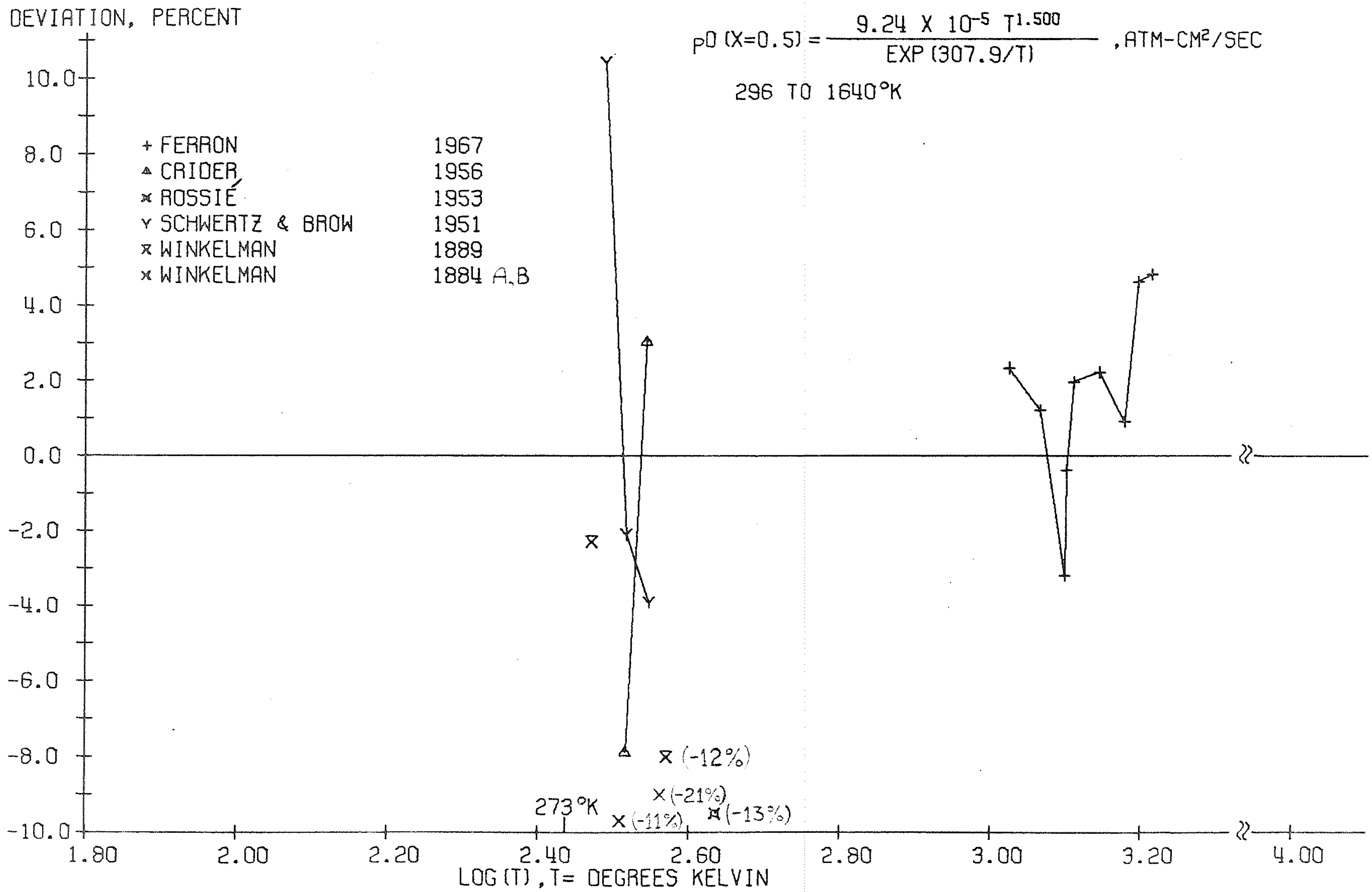


FIG. 79. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, WATER - CARBON DIOXIDE

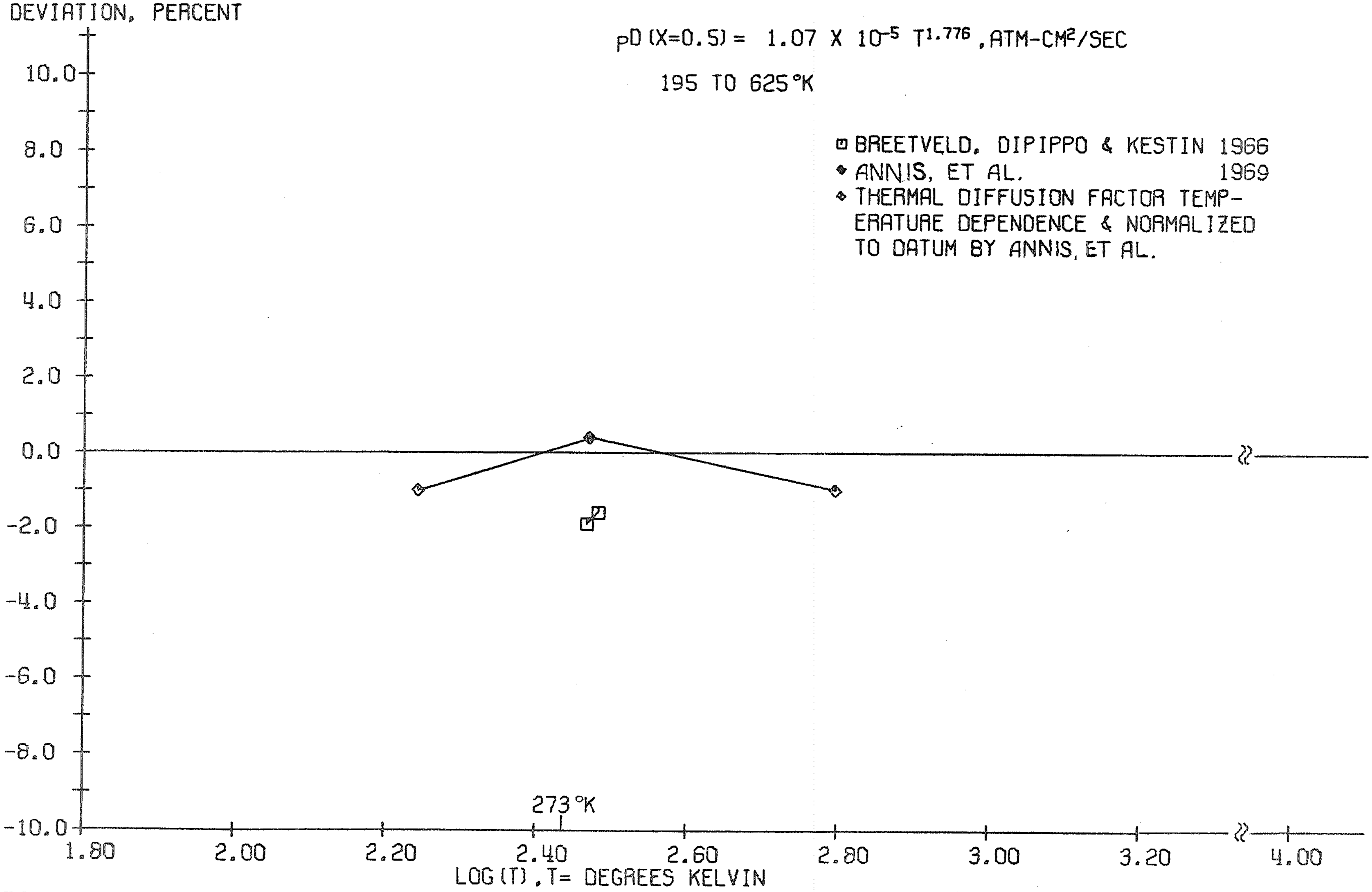


FIG. 80. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, NEON - CARBON DIOXIDE

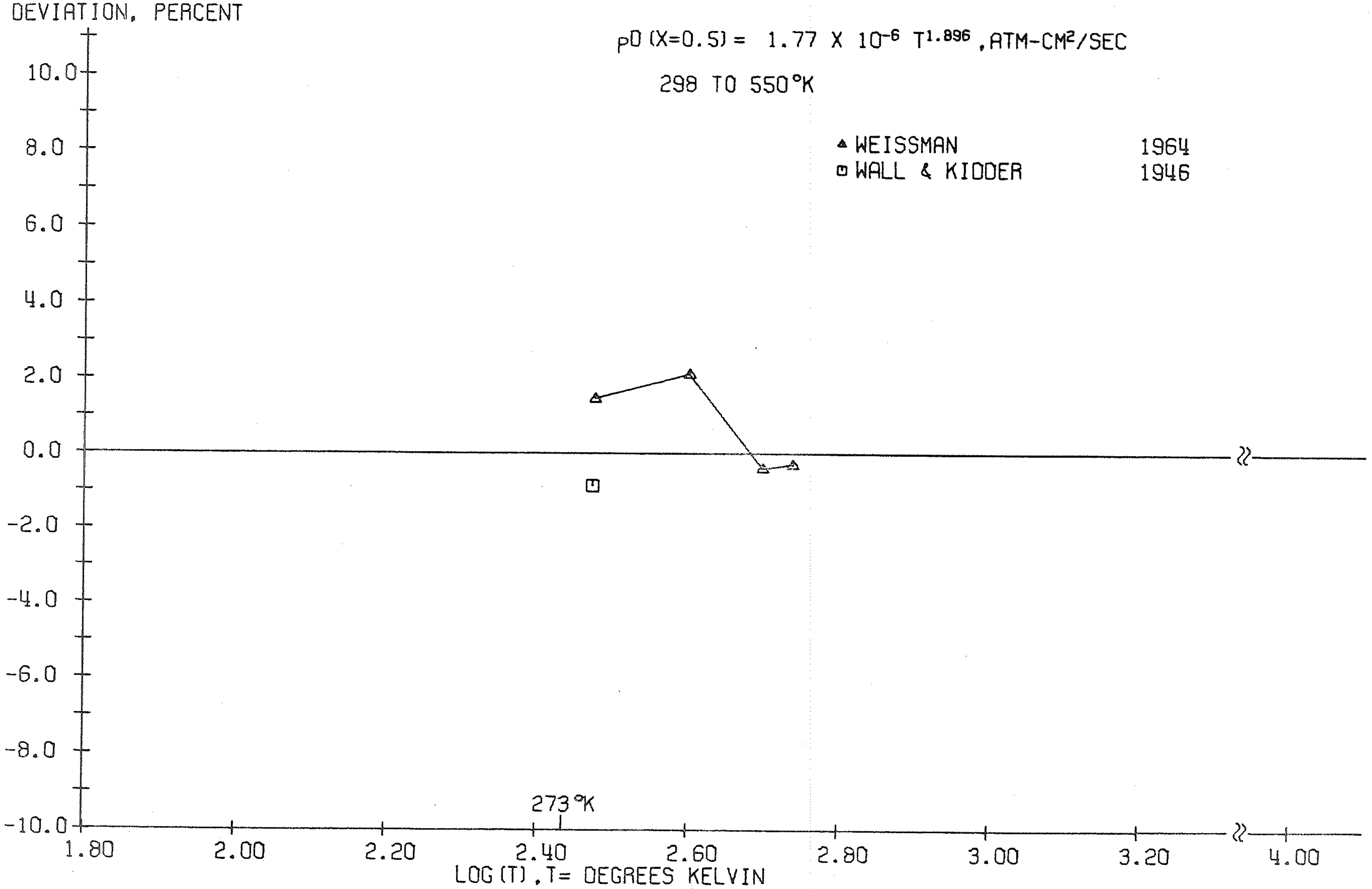


FIG. 81. DEVIATIONS OF DIFFUSION COEFFICIENTS FROM REFERENCE EQUATION, PROPANE - CARBON DIOXIDE

APPENDIX

General Correlation for Diffusion Coefficients, $\mathcal{D}_{12}$, by Corresponding-States

This appendix presents a new two-parameter corresponding-states equation for the temperature dependence of diffusion coefficients, $\mathcal{D}_{12}$. The derived equation is

$$(\Omega_\circ)^{-1} = \frac{\mathcal{D}_{12}(X_1 \approx 0.0)\sqrt{\frac{2M_1M_2}{M_1+M_2}}\ \omega}{T^{3/2}}$$

$$= 0.05702 \Big/ \left\{\ln\left[(T/\Theta)/4.10 \times 10^7\right]\right\}^2 \exp\left[16.61/(T/\Theta)\right] \times \exp\left[35.87/(T/\Theta)^2\right], \quad \text{(A-1)}$$

for $T/\Theta \geq 36.6$, and

$$(\Omega_\circ)^{-1} = 5.46 \times 10^{-5} (T/\Theta)^{1/3}, \quad \text{(A-2)}$$

for $T/\Theta < 36.6$,

where Θ and ω are adjustable parameters determined by corresponding-states analysis. The quantum effects evident in measurements of $\mathcal{D}_{12}$ at very low temperatures and for light gases were approximated by the hyperbola

$$\Delta\Omega = 10^4\left\{0.2415 - \left[0.05832 + 48.92\, X^2\right]^{1/2}\right\}, \quad \text{(A-3)}$$

$$X = (\Theta/T)(\mu_\circ/\omega),$$

where $\mu_\circ$ denotes a relative reduced mass. Equations (A-1), (A-2), and (A-3) appear to serve better than any other known method for the correlation and prediction of diffusion coefficients. These equations were developed as follows.

Theory. Most of the theory has been presented in Chapter 2, and the terms used in this appendix are consistent with the body of the report. According to corresponding-states the collision integral, $\bar{\Omega}_{12}^{(1,1)}$, can be expressed as a function of two parameters. One parameter that is related to the true well-depth of the intermolecular potential, denoted by Θ, and the second parameter that is a characteristic range-of-force distance, denoted by $\omega^{1/2}$. The relationship between the collision integral and the first approximation of $\mathcal{D}_{12}$ is

$$[\mathcal{D}_{12}]_1 = \frac{0.008258\ T^{3/2}}{p[2M_1M_2/(M_1+M_2)]^{1/2}\ \bar{\Omega}_{12}^{(1,1)}}\,. \tag{2.2-8}$$

Eq. (2.2-8) can be rearranged to

$$\frac{[\mathcal{D}_{12}]_1\,[2M_1M_2/(M_1+M_2)]^{1/2}}{T^{3/2}} \equiv \frac{1}{\bar{\Omega}}\,, \tag{A-4}$$

where p has been assumed constant, at one atmosphere. To simplify notation the subscripts and superscripts on the collision integral have been dropped because this appendix concerns only diffusion in binary mixtures. On the assumption of a two-parameter potential the collision integral can be expressed in terms of Θ and ω, that is,

$$\bar{\Omega} = \omega\,\Omega(T/\Theta). \tag{A-5}$$

The values of ω and Θ can be obtained from experimental measurements of $\mathcal{D}_{12}$ and an auxiliary theoretical formula. The values of $\mathcal{D}_{12}$ should be corrected to a mixture composition which corresponds to a trace concentration of the heavy component; then the measurements will closely approximate $[\mathcal{D}_{12}]_1$, see Section 2.4. The theoretical formula is an expression for the London dispersion constant, see Section 2.3. The London constant, C, is proportional to the potential parameters,

$$C \propto \Theta(\omega)^3. \tag{A-6}$$

Given the London constant the low-temperature classical asymptotes for $[\mathcal{D}_{12}]_1$ may be readily calculated, as was shown in Section 2.6-a),

$$\bar{\Omega} = 71.1\ (C/T)^{1/3}. \tag{A-7}$$

For mixtures free of quantum effects Eqs. (A-5), (A-6), and (A-7) can lead to quantitative estimates of Θ and ω, when reliable values of C and $[\mathscr{D}_{12}]_1$ as a function of temperature are available.

For mixtures with significant quantum effects theory indicates that the differences between the quantum- and classical-diffusion collision integrals may be simply related to Λ^*/T, that is, the ratio of the de Boer parameter to temperature. Since $\Lambda^* \equiv h/[\sigma(2\mu\epsilon_{12})^{1/2}]$, quantum effects may be approximated by

$$\Delta\Omega = (\Omega_o^* - \Omega_o) \propto \left(\frac{\Theta}{T}\right)\left(\frac{\mu_o}{\omega}\right), \qquad \text{(A-8)}$$

where the asterisk denotes the presence of quantum effects, the subscript $_o$ denotes values taken relative to a reference gas pair, and μ_o is the relative mass ratio of the reference mixture to a particular mixture.

Outline of Calculation Method. The information necessary for the calculation of the corresponding parameters is available from the critically evaluated diffusion coefficient data presented in the body of this report and from London constants in the literature.[1] The most reliable values of $\mathscr{D}_{12}$ are experimental results for the ten noble gas pairs, whose molecular characteristics are most consistent with the theoretical formulas. Of all the data available only the experimental results by van Heijningen *et al.*[2] were selected for this corresponding-states analysis; the temperature range of this data is between 65° and 400°K. The values of $\mathscr{D}_{12}$ calculated from molecular-beam potentials were used at 1000° and 10,000°K; and they have been listed in Table 17. The data from both sources were corrected to a mixture composition which corresponds to a trace of the heavy component by the method given in Chapter 4, Section 2. From this information values of $(1/\bar{\Omega})$ were calculated in accordance with Eq. (A-4), the results are given in Table 26. The values were plotted on precise graphs on a log-log basis, and appear as unconnected points. At low temperatures the London constants given in Table 14 were used to calculate values of $1/\bar{\Omega}$ in accordance with Eqs. (A-4) and (A-7). These low-temperature data appear as straight lines with slopes equal to 1/3 on the log-log plots constructed for each noble gas mixture. By graphical analysis the corresponding-states parameters Θ and ω were obtained. The gas pair He-Ar was considered a reasonable choice as a reference system to which the two parameters would be normalized. The procedure to determine Θ and ω was essentially as follows. On the graph for He-Ar, the graph for each noble gas pair was superimposed and shifted along the asymptote of He-Ar until the experimental points were most nearly coincident. Similarly, the data of other gas pairs were matched. A displacement of the abscissa from values for He-Ar gave the $\log_{10}\Theta$. With Θ known, the values of ω were calculated in accordance with Eq. (A-6). In Table 26 the fifth and sixth columns list the estimated values of Θ and ω for the ten noble gas pairs.

The determined Θ and ω were used to generate values of (T/Θ) and $[\mathcal{D}_{12}]_1 \times [2M_1M_2/(M_1 + M_2)]^{1/2} \omega / T^{3/2}$ which were plotted on a large-scale graph on a log-log basis. If the two-parameter assumption for the corresponding-states analysis is good, as well as the use of reliable data, then these points should fall on a single curve. This curve would correspond to the collision integral for the reference system, He-Ar. The results were quite consistent. Almost every one of the points, 61 in all, had deviations of less than 10% and the average absolute deviations for each noble gas pair ranged from about 1 to 3%. The data clearly indicated a complex nonlinear relationship, except, of course, at temperatures covered only by the classical asymptote. A simple mathematical form applicable over the entire temperature range was not obtained. An equation was derived as follows. Through the data points a smooth curve was drawn. Points were selected off the curve which are listed in Table 27, and fitted to the form of Eq. (4.3-1). The numerical values for the corresponding-states equation were calculated by a linear least-squares procedure. The parameter s of Eq. (4.3-1) was found to be almost zero (0.0210); to simplify the expression the parameter s was taken as exactly zero, and the final values for the other parameters were again obtained by least-squares calculations. These results are given by Eq. (A-1). The intersection of Eq. (A-1) and the classical asymptote, for He-Ar, established the upper temperature limit for the asymptote, Eq. (A-2). The value of the intersection point was read directly off the large-scale graph. The results are illustrated in Fig. 82.

The hyperbolic approximation for quantum effects was calculated based on differences between the experimental data and points on the drawn curve only between 65° and 400°K. These differences were plotted and found to be consistent with Eq. (A-8). In addition data[3] for H_2- N_2 were included for which the Θ and ω values were estimated from a relationship between Θ, C, and polarizabilities. This relationship is discussed below. Inspection of the graph indicated that a correlation for quantum effects had the form of a hyperbola. The determined expression is given by Eq. (A-3), and Fig. 83 shows that quantum effects may be well estimated by this approximation.

The experimental values of $\mathscr{D}_{12}$ were compared with predictions by Eqs. (A-1), (A-2), and (A-3); the deviations are listed in Table 28. The values of Θ and ω used in these calculations are listed in Table 26.

The experimental data of $\mathscr{D}_{12}$ were also compared with a previously published correlation for $\mathscr{D}_{12}$.[4] This correlation requires knowledge of atomic volumes which are empirical parameters. The correlation is illustrated for the He-Ar mixture on Fig. 82. The deviations presented in Table 29 indicate that it is a very good simple method for estimates of $\mathscr{D}_{12}$.

Discussion. The utility of the corresponding-states equation is in three areas: (1) for mixtures with much reliable data the applicable temperature range may be extended to higher or lower temperatures with considerable reliability, (2) for mixtures with meager data the extrapolations can be made with less reliability but these extrapolations are sensitive to the temperature dependence of $\mathcal{B}_{12}$, and (3) for mixtures with no measurements of $\mathcal{B}_{12}$, values may be predicted over several decades of temperature.

The application of the equation requires two parameters. Several calculations were made for different methods useful for the determination of Θ and ω; the results are presented in Table 30. First from the measurement of a single value of $\mathcal{B}_{12}$ and knowledge of the London constant values of Θ can be obtained by simple graphical analysis. The technique is similar to that used to obtain the original values of Θ and ω. If the London constant is not available from the literature, then C may be computed from the Slater-Kirkwood formula[5]

$$C = \frac{(3/2)\alpha_1\alpha_2}{\left(\frac{\alpha_1}{N_1}\right)^{1/2} + \left(\frac{\alpha_2}{N_2}\right)^{1/2}} , \qquad \text{(A-9)}$$

where α is the polarizability of the atom or molecule, and N is the number of valence electrons. The Slater-Kirkwood expression is a fairly reliable means to obtain the London dispersion constant. Second, if no measurement of $\mathcal{B}_{12}$ is available then values of $\mathcal{B}_{12}$ may still be predicted given the London constant or the well depth.

The London constant has been correlated to the values of Θ. The form for this correlation was derived by an argument which start~~e~~s with $C \propto \epsilon_{12}(\sigma_{12})^6$. The well-depth, ϵ_{12}, is assumed proportional to Θ. The polarizability between atoms or molecules may be considered as, $\alpha \propto \sigma^{3}$, where σ can be approximated as the arithmetic mean value of the individual gases. By these assumptions and a plot with the appropriate coordinates, the equation obtained for θ in terms of (C,α) is

$$\theta = \frac{40.3\ C^{1.516}}{(\alpha_1^{1/3} + \alpha_2^{1/3})^6} \ . \tag{A-10}$$

Equation (A-10) and the available values of θ are shown in Fig. 84. If no values of the London constants are available then θ may still be predicted given the well-depth of the potential. The values of θ were plotted against ϵ_{12}/k from three sources, see Fig. 85. The most reliable source of ϵ_{12} is molecular-beam measurements[6], less reliable values are obtained from viscosity data and the combination rules[7], and from diffusion coefficients.[2] Fig. 85 indicates that there is a direct proportionality between θ and ϵ_{12} but, because of the relatively large uncertainties in ϵ_{12}, a detail analysis is unwarranted. The data presented in Fig. 85 was simply approximated by

$$\theta = 0.021\ (\epsilon_{12}/k). \tag{A-11}$$

In Table 30 the results are presented for a number of test cases about the predictive abilities of Eqs. (A-1), (A-2), and (A-3). Two gas pairs were chosen, one a noble gas pair other than He-Ar, and a gas pair with diatomic molecules, H_2- N_2. The deviations indicate that with one experimental measurement of $\mathcal{D}_{12}$ reliable values of $\mathcal{D}_{12}$ can be predicted over more than three decades of temperature. The absence of a measurement of $\mathcal{D}_{12}$ increases the uncertainties in predicted values of $\mathcal{D}_{12}$ by a factor of about 1.5, or less.

<u>Concluding Remarks</u>. In Table 30 the deviations as a function of temperature indicate that general correlation does not have great enough curvature,

an upswing, at temperatures between 1000° and 10,000°K. This shortcoming is also evident in Fig. 82. The magnitudes of these systematic errors are less than the uncertainty limits for mixtures of Group III, see Chapter 5, Section 1. In Table 28 the deviations for Ne-Ar appear systematically high by a few percent. This suggests that the values of Θ and ω should be slightly adjusted to improve the curve-fit for Ne-Ar.

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Table 26. Experimental Data and Corresponding-States Parameters.[a]

T, °K	$10^4/\bar{\Omega}$	Gas Pair	Θ	$(\omega)^{1/2}$
65.35	4.032	He - Ne	0.78	0.86
77.35	4.210			
90.2	4.321			
169.3	4.846			
295.0	5.290			
1,000.	6.743			
10,000.	12.00			
90.2	2.949	He - Ar	1.00	1.00
169.3	3.394			
295.0	3.762			
400.0	3.991			
1,000.	(4.919)[b]			
10,000.	(8.747)[c]			
111.7	2.744	He - Kr	0.89	1.07
169.3	3.009			
295.0	3.359			
400.0	3.493			
1,000.	4.169			
10,000.	(7.587)[d]			
169.3	2.611	He - Xe	0.71	1.19
231.1	2.713			
295.0	2.877			
400.0	3.033			
1,000.	3.546			
10,000.	(6.601)[e]			

Table 26. (continued).

T, °K	$10^4/\bar{\Omega}$	Gas Pair	Θ	$(\omega)^{1/2}$
90.2	2.241	Ne - Ar	1.55	1.05
169.3	2.812			
295.0	3.201			
400.0	3.395			
1,000.	4.185			
10,000.	7.223			
111.7	2.136	Ne - Kr	1.35	1.13
169.3	2.483			
295.0	2.852			
400.0	3.075			
1,000.	3.696			
10,000	$(6.274)^f$			
169.3	2.170	Ne - Xe	1.29	1.21
231.1	2.348			
295.0	2.500			
400.0	2.658			
1,000.	3.231			
10,000.	5.741			
169.3	1.576	Ar - Kr	3.02	1.21
231.1	1.798			
295.0	1.965			
400.0	2.163			
1,000	2.700			
10,000	$(4.368)^g$			

Table 26. (Continued).

T, °K	$10^4/\bar{\Omega}$	Gas Pair	θ	$(\omega)^{1/2}$
169.3	1.336	Ar - Xe	3.72	1.24
231.1	1.538			
295.0	1.710			
400.0	1.898			
1,000.	2.479			
10,000.	4.089			
169.3	1.153	Kr - Xe	4.27	1.29
231.1	1.347			
295.0	1.482			
400.0	1.658			
1,000.	2.241			
10,000	3.969			

[a] $\bar{\Omega}$ is defined by Eq. (A-4).

Values in parenthesis are slightly low and more reliable values are as follows:

[b] 4.976×10^{-4}

[c] 9.265×10^{-4}

[d] 8.513×10^{-4}

[e] 7.155×10^{-4}

[f] 6.494×10^{-4}

[g] 4.680×10^{-4}

Table 27. Data for Curve-Fitting the Corresponding-States Equation.

$\log_{10}(T/\Theta)$	$-\log_{10}(\Omega_\circ)$	$\log_{10}(T/\Theta)$	$-\log_{10}(\Omega_\circ)$
1.600	-3.715	2.450	-3.424
1.700	-3.666	2.500	-3.415
1.800	-3.621	2.550	-3.406
1.900	-3.578	2.600	-3.397
1.950	-3.557	2.700	-3.377
2.000	-3.538	2.800	-3.356
2.050	-3.520	2.900	-3.334
2.100	-3.503	3.000	-3.311
2.150	-3.488	3.200	(-3.265)[a]
2.200	-3.474	3.400	(-3.218)[b]
2.250	-3.462	3.600	(-3.169)[c]
2.300	-3.452	3.800	(-3.118)[d]
2.350	-3.442	4.000	(-3.066)[e]
2.400	-3.433		

Values in parenthesis are slightly low and more reliable values are as follows:

[a] -3.262

[b] -3.210

[c] -3.150

[d] -3.094

[e] -3.037

Table 28. Deviations of Experimental Data from the Corresponding-States Equation.

Gas Pair	T, °K	Deviation, %	Gas Pair	T, °K	Deviation, %
He - Ne*	65.35	-1.89	Ne - Ar*	90.2	+5.20
	77.35	-0.63		169.3	+4.47
	90.2	-0.95		295.0	+1.89
	169.3	-2.01		400.0	+0.44
	295.0	-4.42		1,000.	+0.91
	1,000.	-4.75		10,000.	+7.37
	10,000.	+1.51	Ne - Kr	111.7	+1.07
He - Ar*	90.2	+2.38		169.3	+2.58
	169.3	-0.15		295.0	+1.67
	295.0	-2.52		400.0	+2.10
	400.0	-3.10		1,000.	+0.97
	1,000.	-1.05		10,000.	+4.60
	10,000.	+6.33	Ne - Xe	169.3	+1.44
He - Kr	111.7	+1.62		231.1	+0.85
	169.3	-0.47		295.0	+1.08
	295.0	-2.47		400.0	+0.17
	400.0	-5.05		1,000.	+0.26
	1,000.	-6.25		10,000.	+8.58
	10,000.	+2.64	Ar - Kr	169.3	+0.36
He - Xe	169.3	+1.09		231.1	+0.43
	231.1	-2.18		295.0	+0.64
	295.0	-1.64		400.0	+0.84
	400.0	-2.76		1,000.	+0.27
	1,000.	-5.85		10,000.	+0.11
	10,000.	+4.42			

Table 28. (Continued)

Gas Pair	T, °K	Deviation, %	Gas Pair	T, °K	Deviation, %
Ar - Xe	169.3	-0.74	Kr - Xe	169.3	+0.39
	231.1	-1.80		231.1	-0.87
	295.0	-1.03		295.0	-2.06
	400.0	-1.04		400.0	-2.12
	1,000.	-1.34		1,000.	+2.40
	10,000.	+2.94		10,000.	+11.36

*Deviations for these systems include corrections for quantum effects.

Table 29. Comparison of the Corresponding-States Equation and the Correlation by Fuller, Seager, and Giddings.[a]

Gas Pair	Average Absolute Deviation,% This Report	FSG	Temperature, °K
He - Ne	2.31	5.83	65 - 10,000
He - Ar	2.59	3.01	90 - 10,000
He - Kr	3.08	4.49	112 - 10,000
He - Xe	2.99	10.02	169 - 10,000
Ne - Ar	3.38	2.91	90 - 10,000
Ne - Kr	2.17	3.07	112 - 10,000
Ne - Xe	2.06	10.26	169 - 10,000
Ar - Kr	0.44	6.09	169 - 10,000
Ar - Xe	1.48	4.28	169 - 10,000
Kr - Xe	3.20	6.49	169 - 10,000

[a] Reference 4.

Table 30. Test Calculations for Corresponding-States Equation.

Gas Pair	T,°K	$\mathscr{D}_{12}(x_1 20.0)$, cm^2/sec	C,a.u.	ϵ_{12}/k,°K	Θ	ω	Avg. Abs. Deviation,[a] %
Ar - Kr	295.0	0.1354	91.8[b]	-	3.11[c]	1.45	0.41*
	-	-	91[d]	145	3.05[e]	1.46	0.45
	-	-	91.8[b]	-	3.17[f]	1.45	0.97
	-	-	-	-	-	-	6.09*[g]
H_2 - N_2	294.8	0.7521	30[d]	-	2.20[c]	1.12	4.22*
	294.8	0.7521	31.3[b]	-	2.10[c]	1.16	5.39
	-	-	30[d]	62.9	1.32[e]	1.33	6.51
	-	-	30[d]	-	1.64[f]	1.24	5.07
	-	-	31.3[b]	-	1.74[f]	1.23	7.42
	-	-	-	-	-	-	4.63*[g]

*The deviations calculated are as follows:

		Deviation, %				Deviation, %	
Gas Pair	T,°K	This Report	FSG[g]	Gas Pair	T,°K	This Report	FSG[g]
H_2 - N_2	65.25	-9.29	-11.5	Ar - Kr	169.3	+0.45	-11.2
	77.35	-0.10	-6.18		231.1	+0.39	-6.41
	90.2	+0.64	-3.94		295.0	+0.65	-3.62
	169.3	+1.24	+1.74		400.0	+0.72	-1.70
	294.8	-0.04	$+1.15_5$		1,000.	-0.04	-2.40
	1,000.	-2.36	-4.79		10,000.	-0.22	-11.2
	10,000.	+15.9	+3.13				

[a] Average absolute deviations are for data over a temperature range of 169° to 10,000°K for Ar-Kr (6 points) and 65° to 10,000°K for H_2-N_2 (7 points).

[b] Estimated from Slater-Kirkwood formula, Eq. (A-9).

[c] Estimated by graphical analyses.

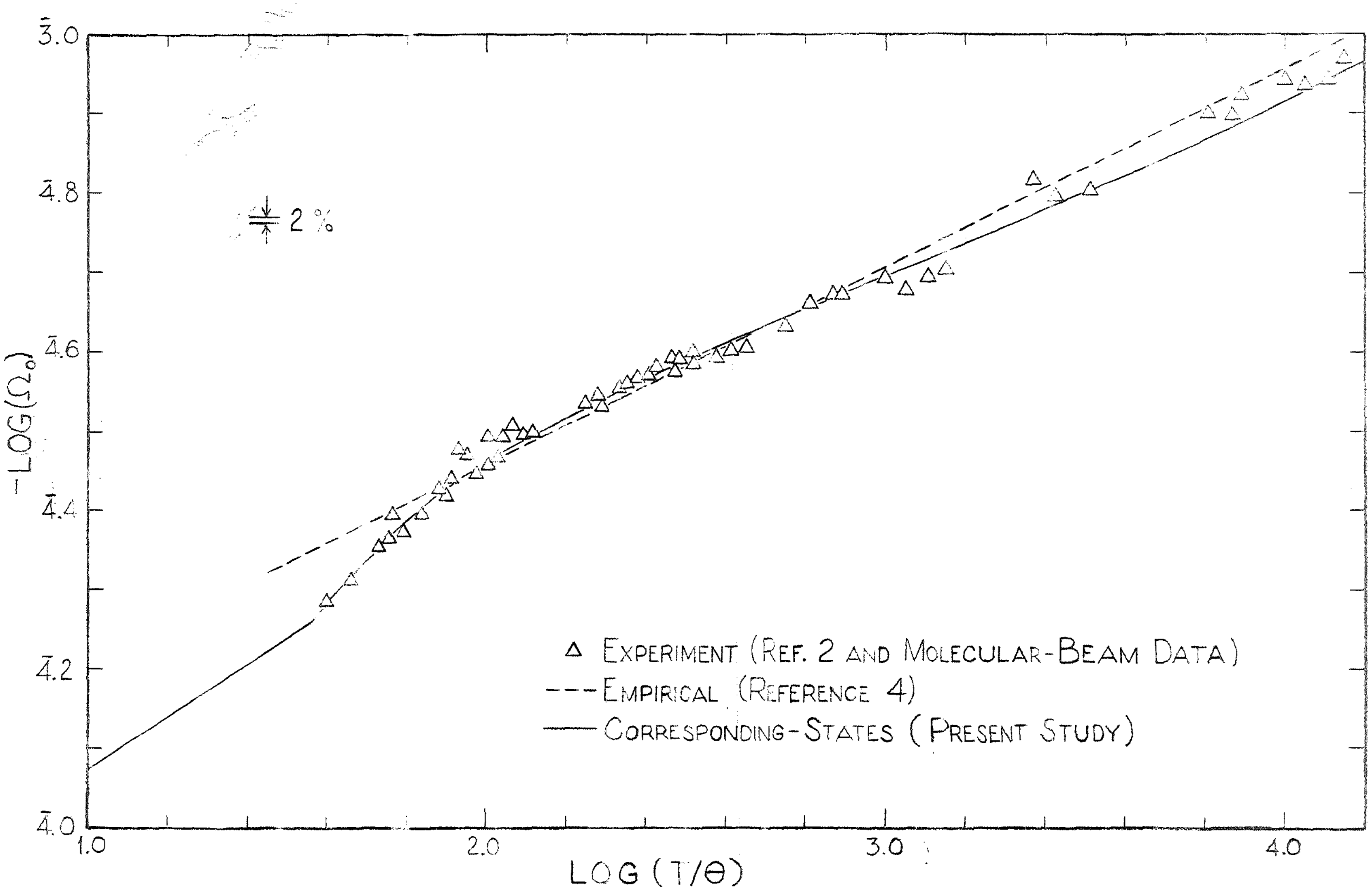
[d] Obtained from literature.[1]

[e] Calculated from Eq. (A-11).

[f] Calculated from Eq. (A-10).

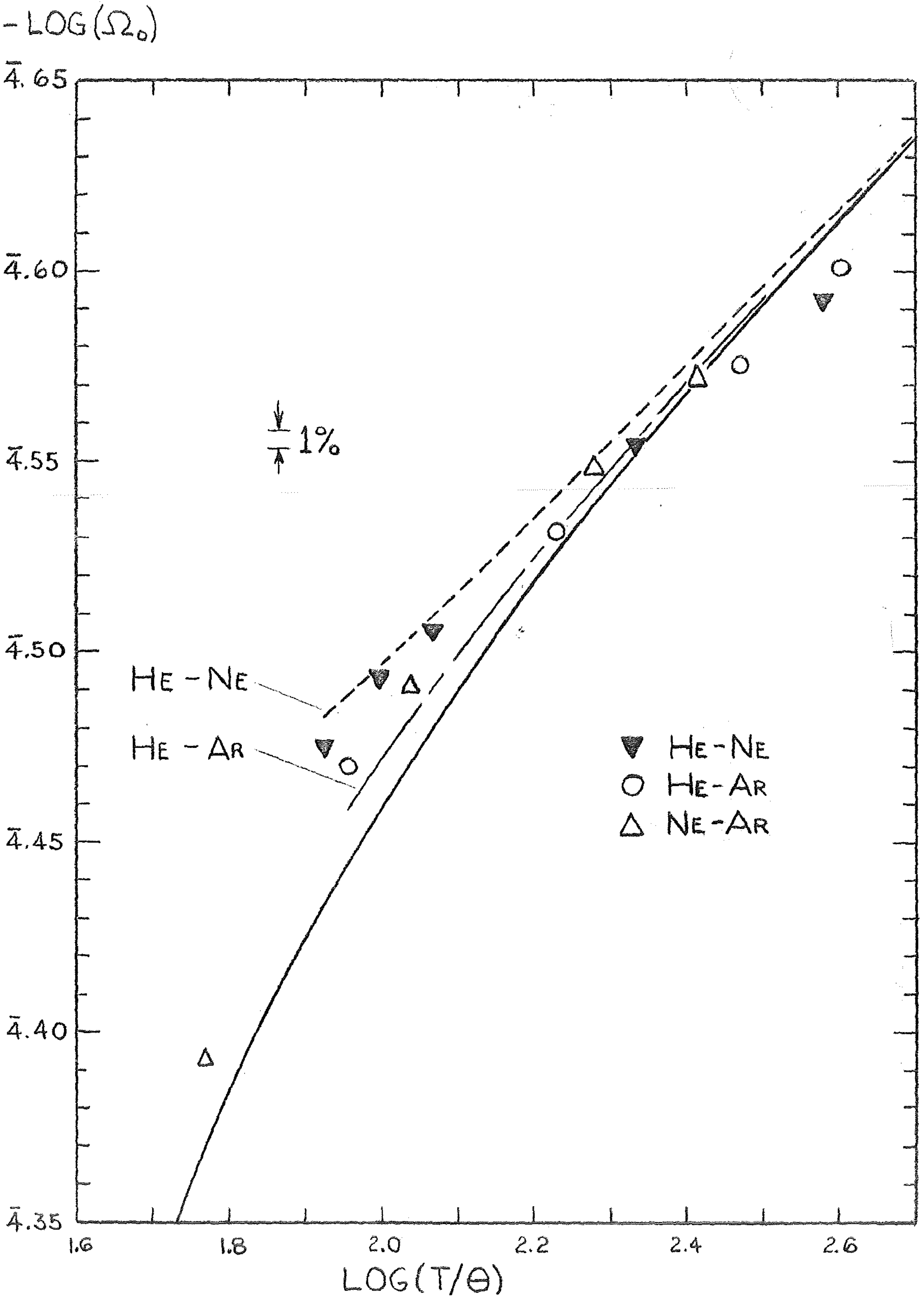
[g] Based on correlation by Fuller *et al*. (1966).[4]

Fig. 82. Corresponding-States Correlation for Gaseous Diffusion Coefficients.



3.0
4.8
4.6
4.4
4.2
4.0
-LOG(Ω_o)
1.0
2.0
3.0
4.0
LOG (T/Θ)
2 %
△ EXPERIMENT (REF. 2 AND MOLECULAR-BEAM DATA)
--- EMPIRICAL (REFERENCE 4)
— CORRESPONDING-STATES (PRESENT STUDY)

Fig. 83. Corresponding-States Correlation for Mixtures with Quantum Effects.



- LOG(Ω₀)
4̄.65
4̄.60
4̄.55
4̄.50
4̄.45
4̄.40
4̄.35
1%
He - Ne
He - Ar
▼ He-Ne
○ He-Ar
△ Ne-Ar
1.6
1.8
2.0
2.2
2.4
2.6
LOG(T/Θ)

Fig. 84. Correlation of θ as a Function of the London Dispersion Constant.

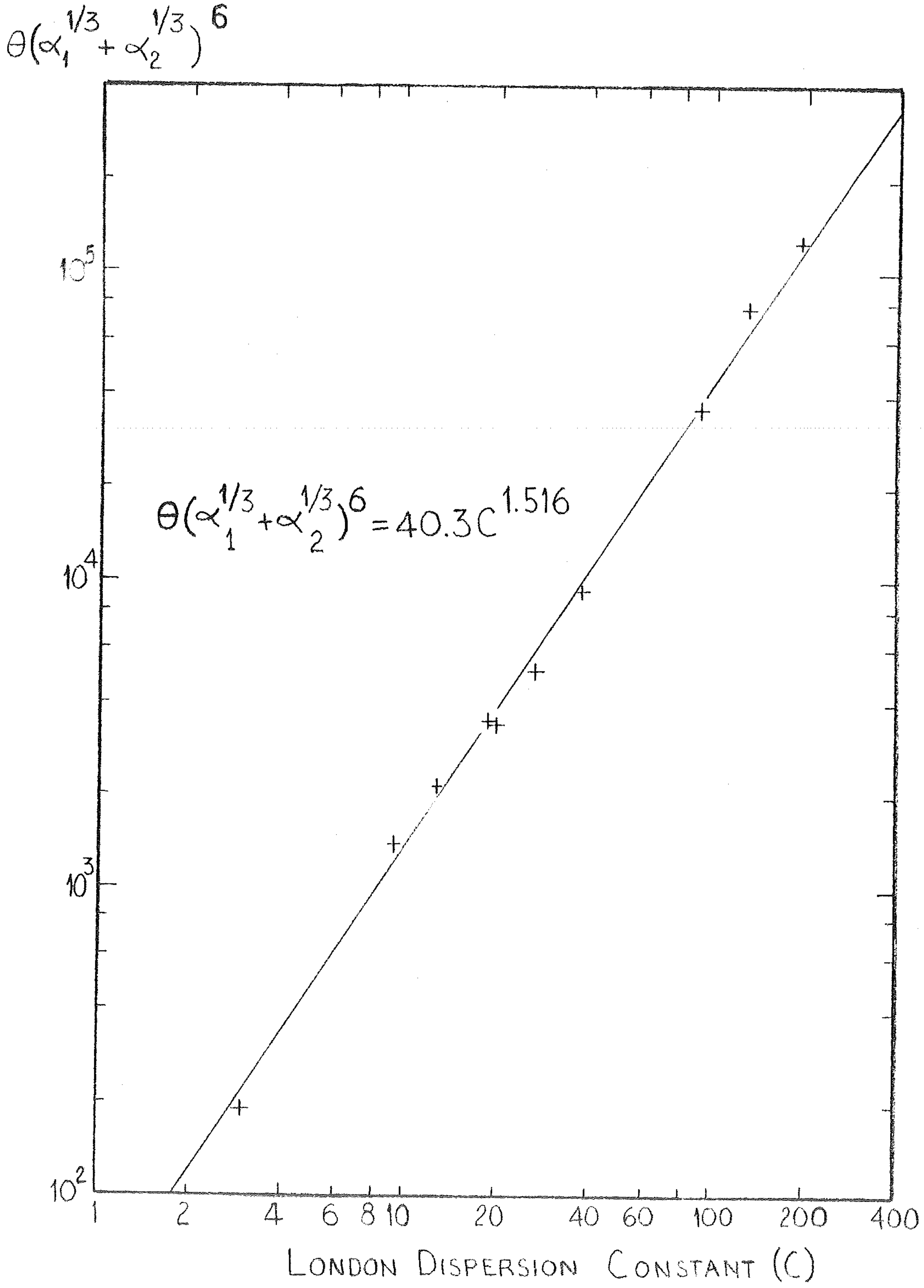
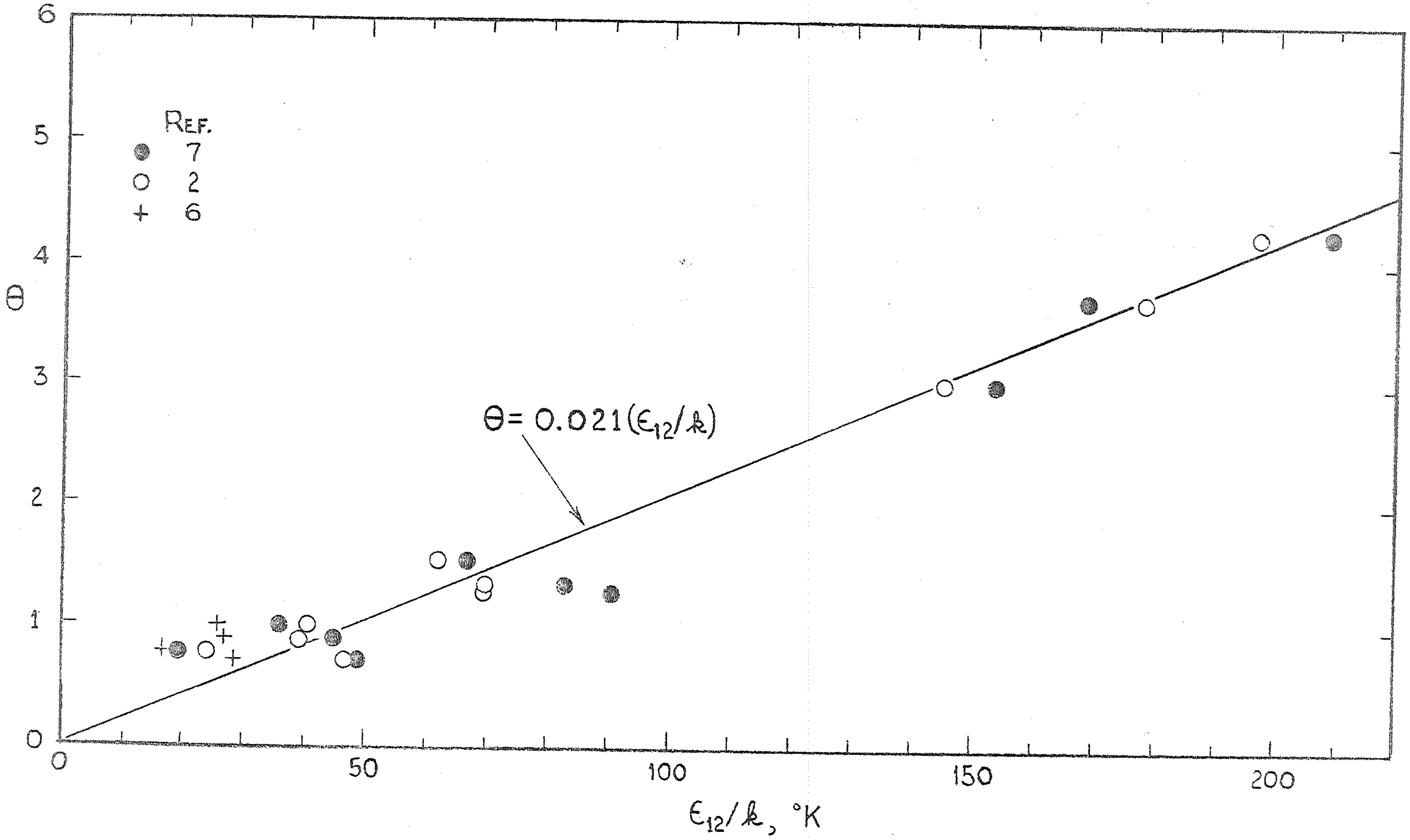

$\theta(\alpha_1^{1/3}+\alpha_2^{1/3})^6$
10^5
10^4
10^3
10^2
$\theta(\alpha_1^{1/3}+\alpha_2^{1/3})^6=40.3C^{1.516}$
1
2
4
6
8
10
20
40
60
100
200
400
LONDON DISPERSION CONSTANT (C)

Fig. 85. Correlation of θ as a Function of the Potential Well-Depth, ϵ_{12}/k.



REF.
● 7
○ 2
+ 6
Θ
0
1
2
3
4
5
6
$\Theta = 0.021(\epsilon_{12}/k)$
0
50
100
150
200
ϵ_{12}/k, °K

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von Obermayer, A. (1882 b). Sitzber. Akad. Wiss. Wien 85, 748. O_2-(air, N_2), air-CO_2, N_2O-CO_2. closed tube.

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Weissman, S. (1968 a). Proceedings of the Fourth Symposium on Thermophysical Properties, edited by J. R. Moszynski, (ASME, New York), pp. 360-5. H_2O-(methanol, ethanol, H_2O_2), methanol-1-butanol, ethanol-1-butanol, NO_2-N_2O_4, methyl ether-(SO_2, CH_3Cl), CH_3Cl-SO_2, propanol-butanol. mixture viscosity.

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Weissman, S. (1969). Private Communication. Ne-Xe, CH_4-CO_2. two-bulb apparatus.

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Weissman, S. and E. A. Mason (1962 b). J. Chem. Phys. 37, 1289. ^3He-^4He, He-(Ne, Ar, Kr, Xe), Ne-(Ar, Kr, Xe), Ar-(Kr, Xe), Kr-Xe, H_2-(He, Ne, Ar, Xe, HD, D_2, CH_4, N_2, CO, O_2), D_2-(HD, Ne), N_2-(CO, O_2), CO-O_2. mixture viscosity.

Weissman, S., S. C. Saxena, and E. A. Mason (1961). Phys. Fluids 4, 643. Ne-CO_2. two-bulb apparatus and thermal separation rate.

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Westenberg, A. A. and G. Frazier (1962). J. Chem. Phys. 36, 3499. Ar-H_2. point source.

Wicke, E. and P. Hugo (1961). Z. Physik. Chem. (Frankfurt). 28, 401. H_2-CO_2, N_2-CO. diffusion bridge (porous septum).

Wicke, E. and R. Kallenbach (1941). Kolloid Z. 97, 135. N_2-CO_2. diffusion bridge (porous septum).

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Winkelmann, A. (1884 b). Ann. Physik 22, 152. H_2O-(H_2, air, CO_2). evaporation tube.

Winkelmann, A. (1884 c). Ann. Physik 23, 203. H_2, air, and CO_2(each with the esters as follows)-(ethyl formate, propyl formate, methyl acetate, ethyl acetate, i-butyl acetate, methyl propionate, ethyl propionate, propyl propionate, amyl propionate, methyl butyrate, propyl butyrate, i-butyl butyrate, methyl-i-butyrate, ethyl-i-butyrate, i-butyl propionate, ethyl butyrate, propyl-i-butyrate, i-butyl-i-butyrate, amyl-i-butyrate, ethyl valerate, propyl valerate, i-butyl valerate. evaporation tube.

Winkelmann, A. (1885). Ann. Physik 26, 105. H_2, air, and CO_2 (each with the compounds as follows)-(formic acid, acetic acid, propionic acid, i-butyric acid, n-butyric acid, i-valeric acid, methanol, ethanol, 1-propanol, 1-butanol, 2-butanol, 1-pentanol, active amyl alcohol, 1-hexanol). evaporation tube.

Winkelmann, A. (1888). Ann. Physik 33, 445. H_2O-air. evaporation tube.

Winkelmann, A. (1889). Ann. Physik 36, 93. H_2O-(H_2, air, CO_2). evaporation tube.

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Winn, E. B. (1950). Phys. Rev. 80, 1024. Ne-Ne, Ar-Ar, CH_4-CH_4, N_2-N_2, O_2-O_2, CO_2-CO_2. two-bulb apparatus.

Winn, E. B. and E. P. Ney (1947). Phys. Rev. 72, 77. CH_4-CH_4. two-bulb apparatus.

Winter, E. R. S. (1951). Trans. Faraday Soc. 47, 342. N_2-N_2, O_2-O_2, CO_2-CO_2. two-bulb apparatus.

Wintergerst, E. (1930). Ann. Physik 4, 323. NH_3-air. closed tube.

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Wretschko, A. (1870). Sitzber. Akad. Wiss. Wien 62, 575. H_2-(O_2, CO_2), O_2-CO_2. closed tube.

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Zhukhovitskiǐ, A. A., S. N. Kim, and M. O. Burova (1968). Zavod. Lab. 34, 144; in Chem. Abstr. 69, 13082 c (1968). He-N_2. open tube and gas chromatography.

Zmbov, K. F. and Ž. V. Knežević (1961). Bull. Inst. Nucl. Sci. "Boris Kidrich" (Belgrade) 11 (236), 141; in Chem. Abstr. 56, 10931 f (1962). BF_3-BF_3. two-bulb apparatus.

II. Molecular-Beam Measurements

The gas pairs investigated and the potential energy separation ranges are noted.

1. Massachusetts Institute of Technology (Amdur et al.)

Amdur, I. and R. R. Bertrand (1962). J. Chem. Phys. 36, 1078 He-He (0.55 - 1.0 Å).

Amdur, I. and A. L. Harkness (1954). J. Chem. Phys. 22, 664. He-He (1.27 - 1.59 Å).

Amdur, I. and E. A. Mason (1954). J. Chem. Phys. 22, 670. Ar-Ar (2.18 - 2.69 Å).

Amdur, I. and E. A. Mason (1955 a). J. Chem. Phys. 23, 415. Ne-Ne (1.76 - 2.13 Å).

Amdur, I. and E. A. Mason (1955 b). J. Chem. Phys. 23, 2268. Kr-Kr (2.42 - 3.14 Å).

Amdur, I. and E. A. Mason (1956 a). J. Chem. Phys. 25, 624. Xe-Xe (3.01 - 3.60 Å).

Amdur, I. and E. A. Mason (1956 b). J. Chem. Phys. 25, 630. He-H (1.16 - 1.71 Å).

Amdur, I. and E. A. Mason (1956 c). J. Chem. Phys. 25, 632. Ne-Ar (1.91 - 2.44 Å).

Amdur, I. and A. L. Smith (1968). J. Chem. Phys. 48, 565. He-H_2 (1.44 - 1.76 Å), He-D_2(1.45 - 1.79 Å).

Amdur, I., E. A. Mason, and A. L. Harkness (1954). J. Chem. Phys 22, 1071. He-Ar(1.64 - 2.27 Å).

Amdur, I., E. A. Mason, and J. E. Jordan (1957). J. Chem. Phys. 27, 527. He-N_2(1.79 - 2.29 Å), Ar-N_2(2.28 - 2.83 Å); derived potential, N_2-N_2(2.43 - 3.07 Å).

Amdur, I., J. E. Jordan, and S. O. Colgate (1961 a). J. Chem. Phys. 34, 1525. He-He(0.97 - 1.48 Å).

Amdur, I., M. S. Longmire, and E. A. Mason (1961 b). J. Chem. Phys. 35, 895. He-CH_4(1.92 - 2.37 Å), He-CF_4(2.43 - 2.74 Å); derived potentials, CH_4-CH_4(2.47 - 3.06 Å), CF_4-CF_4(3.43 - 3.77 Å).

Amdur, I., J. E. Jordan, and R. R. Bertrand (1964). in Atomic Collision Processes, edited by M. R. C. McDowell, (North-Holland Publishing Co., Amsterdam), pp. 934-43. He-He(0.52 - 0.98 Å), Ar-Ar(1.6 - 2.0 Å).

Colgate, S. O., J. E. Jordan, I. Amdur, and E. A. Mason (1969). J. Chem. Phys. 51, 968. He-Ar(1.40 - 1.81 Å), Ar-Ar(1.96 - 2.41 Å), Ar-H_2 (1.81 - 2.36 Å).

Jordan, J. E. and I. Amdur (1967). J. Chem. Phys. 46, 165. He-He (0.61 - 1.12 Å).

Jordan, J. E., S. O. Colgate, I. Amdur, and E. A. Mason (1970). Preprint, to be published in J. Chem. Phys. Ar-N_2(2.04 - 2.53 Å), Ar-CO (2.09 - 2.68 Å). Ar-O_2(2.01 - 2.50 Å); and derived potentials: N-N_2(2.07 - 2.61 Å), N-NO(2.06 - 2.59 Å), N-O_2(2.04 - 2.58 Å), O-N_2(2.03 - 2.57 Å), O-NO(2.02 - 2.55 Å), O-O_2(2.00 - 2.54 Å), N_2-N_2(2.12 - 2.65 Å), N_2-NO(2.11 - 2.63 Å), NO-NO(2.09 - 2.62 Å) O_2-N_2(2.09 - 2.62 Å), O_2-NO(2.08 - 2.60 Å), O_2-O_2(2.07 - 2.59 Å).

Mason, E. A. and I. Amdur (1964). J. Chem. Phys. 41, 2695. Ar-CH_4 (2.31 - 2.66 Å).

2. Moscow State University (Leonas *et al.*)

Belyaev, Yu. N. and V. B. Leonas (1966 a). Soviet Phys.-Tech. Phys. 11, 257 [Zh. Tekh. Fiz. 36, 353 (1966)]. Ar-N_2(2.26 - 2.86 Å), Ar-O_2 (2.18 - 2.94 Å), N_2-N_2(2.34 - 3.05 Å), N_2-O_2(2.34 - 3.05 Å), O_2-O_2 (2.34 - 3.15 Å). Errata in Soviet Phys.-Doklady 11, 866 (1967).

Belyaev, Yu. N. and V. B. Leonas (1966 b). Soviet Phys.-JETP Letters 4, 92 [Zh. Eksper. Teor. Fiz. Pis'ma 4, 134 (1966)]. H-N_2(1.46 - 1.81 Å), H-O_2(1.46 - 1.84 Å), N-N_2(1.76 - 2.54 Å), N-O_2(1.84 - 2.49 Å), O-N_2 (2.00 - 2.48 Å), O-O_2(2.05 - 2.46 Å).

Belyaev, Yu. N. and V. B. Leonas (1966 c). High Temp. (USSR) 4, 686 [Teplofiz. Vys. Temp 4, 732 (1966)]. Same results as reported by Belyaev and Leonas (1966 a).

Belyaev, Yu. N. and V. B. Leonas (1967 a). Soviet Phys.-Doklady 11, 866 [Dokl. Akad. Nauk SSSR 170, 1039 (1966)]. Ar-N_2(2.12 - 2.67 Å), Ar-O_2(2.15 - 2.63 Å), N_2-N_2(2.34 - 3.05 Å), N_2-O_2(2.34 - 3.05 Å), O_2-O_2(2.34 - 3.15 Å).

Belyaev, Yu. N. and V. B. Leonas (1967 b). Soviet Phys.-Doklady 12, 233 [Dokl. Akad. Nauk SSSR 173, 306 (1967)]. He-He(1.10 - 1.53 Å), He-H_2(1.15 - 1.89 Å), H_2-H_2(1.34 - 1.95 Å), H-He(0.79 - 1.35 Å), H-H_2(1.00 - 1.24 Å).

Belyaev, Yu, N. and V. B. Leonas (1967 c). High Temp. (USSR) 5, 1014 [Teplofiz. Vys. Temp. 5, 1123 (1967).]. Same results as reported by Belyaev and Leonas (1967 b).

Belyaev, Yu. N. and V. B. Leonas (1968). High Temp. (USSR) 6, 182 [Teplofiz. Vys. Temps 6, 188 (1968)]. N_2-N_2(2.18 - 2.72 Å), N_2-O_2(2.08 - 2.68 Å), O_2-O_2(2.05 - 2.75 Å); other results as reported by Belyaev and Leonas (1966 b).

Belyaev, Yu. N., V. B. Leonas, and A. V. Sermyaguin (1967). Private Communication from V. B. Leonas; in *Abstracts of Papers, Fifth International Conference on the Physics of Electronic and Atomic Collisions* (Publishing House "Nauka," Leningrad, USSR, 1967), p. 643. N_2-NO(2.28 - 2.85 Å), N_2-CO(2.22 - 2.77 Å), CO-CO(2.31 - 2.92 Å), N-NO(1.90 - 2.40 Å), O-Ar(1.78 - 2.40 Å), O-Kr(1.91 - 2.24 Å), O-Xe(2.08 - 2.37 Å), F-Ar(1.80 - 2.22 Å), F-Kr(1.93 - 2.28 Å), F-Xe(2.03 - 2.33 Å).

Kamnev, A. B. and V. B. Leonas (1965 a). Soviet Phys.-Doklady **10**, 529 [Dokl. Akad. Nauk SSSR **162**,798 (1965)]. He-He(0.87 - 1.27 Å), He-Ne(1.3 - 1.65 Å), He-Ar(1.63 - 2.06 Å), Ne-Ne(1.7 - 2.18 Å), Ne-Ar(1.93 - 2.49 Å), Ar-Ar(2.26 - 3.14 Å).

Kamnev, A. B. and V. B. Leonas (1965 b). High Temp. (USSR) **3**, 744 [Teplofiz. Vys. Temp. **3**, 804 (1965)]. Same results as reported by Kamnev and Leonas (1965 a).

Kamnev. A. B. and V. B. Leonas (1966 a). Soviet Phys.-Doklady **10**, 1202 [Dokl. Akad. Nauk SSSR **165**, 1273 (1965)]. He-Kr(1.67 - 2.04 Å), He-Xe(1.73 - 2.2 Å), Ne-Kr(2.15 - 2.52 Å), Ne-Xe(2 - 2.56 Å), Ar-Kr(2.4 - 3.1 Å), Ar-Xe(2.48 - 3.27 Å), Kr-Kr(2.4 - 3.02 Å), Kr-Xe(2.44 - 3 Å), Xe-Xe(2.48 - 3.09 Å).

Kamnev, A. B. and V. B. Leonas (1966 b). High Temp. (USSR) **4**, 283 Teplofiz. Vys. Temp. **4**,288 (1966)]. Same results as reported by Kamnev and Leonas (1966 a).

3. Other References

Fink, R. D., J. S. King, Jr., and J. H. Freeman (1969). J. Chem. Phys. **50**, 2773. He-CF_4(2.69 - 2.80 Å), He-CHF_3(2.57 - 2.70 Å), He-CH_2F_2 (2.47 - 2.60 Å).

Mason, E. A. and J. T. Vanderslice (1958). J. Chem. Phys. 28, 1070. H-Ne(1.7 - 2.5 Å), H-Ar(2.1 - 3.0 Å), H-H_2(0.4 - 1.2 Å).

4. Reviews

Amdur, I. (1961). Physical Chemistry in Aerodynamics and Space Flight (Pergamon Press, New York, 1961), pp. 228-35.

Amdur, I. (1962). AIChE J. 8, 521.

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Amdur, I. (1968). in Methods of Experimental Physics, edited by B. Bederson and W. L. Fite, (Academic Press, New York), Vol. 7A, pp. 341-360.

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Amdur, I. and E. A. Mason (1958). Phys. Fluids 1, 370.

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Leonas, V. B. (1964). Soviet Phys.-Uspekhi 7, 121 [Usp. Fiz. Nauk 82, 287 (1964)].

Leonas, V. B. and E. V. Samuilov (1966). High Temp. (USSR) 4, 664 [Teplofiz., Vys. Temp. 4, 710 (1966)].

Mason, E. A. and J. T. Vanderslice (1962). in Atomic and Molecular Processes, edited by D. R. Bates, (Academic Press, New York), pp. 663-94.

III. London Dispersion Constants

Dalgarno, A. (1967). Adv. Chem. Phys. 12, 143. This is a comprehensive review and summary.

IV. Related Viscosity Measurements

The gas pairs investigated and the temperature ranges are noted.

1. Primary References

Becker, E. W. and R. Misenta (1955). Z. Physik 140, 535. ^{3}He(14.18° to 20.33°K), ^{4}He(14.12° to 20.33°K), H_2(14.52° to 90.0°K), HD(14.12° to 90.0°K), H_2(14.26° to 90.0°K).

Coremans, J. M. J., A. van Itterbeek, J. J. M. Beenakker, H. F. P. Knaap, and P. Zandbergen (1958 a). Physica 24, 557. ^{4}He(20° to 80°K), Ne(20° to 80°K), H_2(20° to 80°K), D_2(20° to 80°K).

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