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COLLISION-INDUCED $^2P_{1/2}$ —— $^2P_{3/2}$ MIXING

IN SODIUM

by

Brother John Pitre, F.S.C.

A Thesis
Submitted to the Faculty of Graduate Studies through the Department
of Physics in Partial Fulfillment of the Requirements for
the Degree of Doctor of Philosophy at the
University of Windsor

Windsor, Ontario

1967

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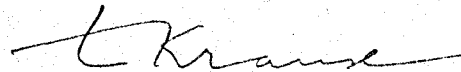
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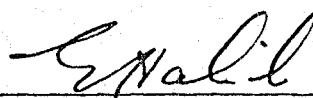789 E. Eisenhower Parkway
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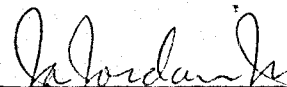Ann Arbor, MI 48106-1346

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ABSTRACT

The total cross sections for inelastic collisions between excited sodium and noble gas atoms in their ground states, leading to $3\,^2P_{1/2}$ —— $3\,^2P_{3/2}$ excitation transfer in sodium, have been determined in a series of sensitized fluorescence experiments with pure sodium vapour and its mixtures with noble gases. In the experiments with pure sodium, the sodium vapour pressure was varied over the range $0 - 1.6 \times 10^{-5}$ mm Hg, where radiation trapping effects were negligible and the cross section $Q_2(3\,^2P_{1/2} \leftarrow 3\,^2P_{3/2})$, for Na - Na collisions, was found to be 283 $\mathring{A}^2 \pm 10$ per cent. In the sodium-noble gas experiments the sodium vapour was maintained at a pressure of 5×10^{-7} mm Hg and the noble gas pressures ranged from 0 to 1 mm Hg. The cross sections $Q_1(3\,^2P_{1/2} \rightarrow 3\,^2P_{3/2})$ and $Q_2(3\,^2P_{1/2} \leftarrow 3\,^2P_{3/2})$ are as follows: Na - He, 86.0 and 44.8 $\mathring{A}^2$; Na - Ne, 67.0 and 35.4 $\mathring{A}^2$; Na - Ar, 110 and 55.9 $\mathring{A}^2$; Na - Kr, 85.0 and 43.6 $\mathring{A}^2$; Na - Xe, 89.8 and 45.6 $\mathring{A}^2$, all quoted with an accuracy of $\pm$ 5 per cent. The ratios Q_1/Q_2 are in agreement with the value predicted from the principle of detailed balancing and the cross sections also agree, as to order of magnitude, with some theoretically predicted values.

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I am indebted to the National Research Council for financial support in the form of a graduate studentship, and, finally, I would like to thank the Christian Brothers of the Toronto District who granted me the opportunity to pursue graduate studies.

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I. INTRODUCTION

In the past decade there has been a rapid resurgence of interest in the area of atomic interaction physics which had been relatively dormant since the war era. Initial impetus was received from the need in other fields, such as astrophysics, thermonuclear research and the diagnostics of hot gases, for collision cross sections and basic data of interaction mechanisms. Later, with the advent of rocketry, upper atmospheric studies with emphasis on thermal collisions directed much interest toward low temperature processes. Still greater stimulus was received in the sixties with the discovery of the laser, especially the gas laser of the helium-neon type, where population inversion is accomplished by collision induced transitions between excited states. The $3\,^2P_{1/2}$ and $3\,^2P_{3/2}$ resonance levels in sodium lend themselves very well to experimental studies of collision induced excitation transfer as they are close to one another and well removed from other levels which might perturb the processes.

If a cell containing pure sodium vapour at low pressure or a sodium vapour-noble gas mixture is illuminated with one component of the sodium resonance doublet then both fine structure components will appear in fluorescence. The component which has the same wavelength as is used for excitation is known as resonance fluorescence and the other component whose presence is due to the collisional excitation transfer is called sensitized fluorescence. The inelastic

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collisions proceed according to the equation,

$$Na(3^2P_{1/2}) + X + \Delta E \rightleftharpoons Na(3^2P_{3/2}) + X \ , \tag{1}$$
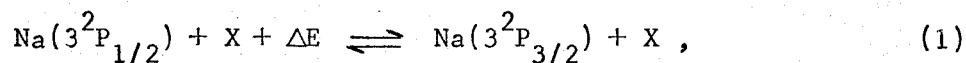
where X is a sodium or noble gas atom in the ground state and ΔE

represents a change in kinetic energy of relative motion. The term

diagram for sodium given in Fig. 1 shows the processes that ensue when

sodium vapour is irradiated with the D_1 component of the resonance

doublet.
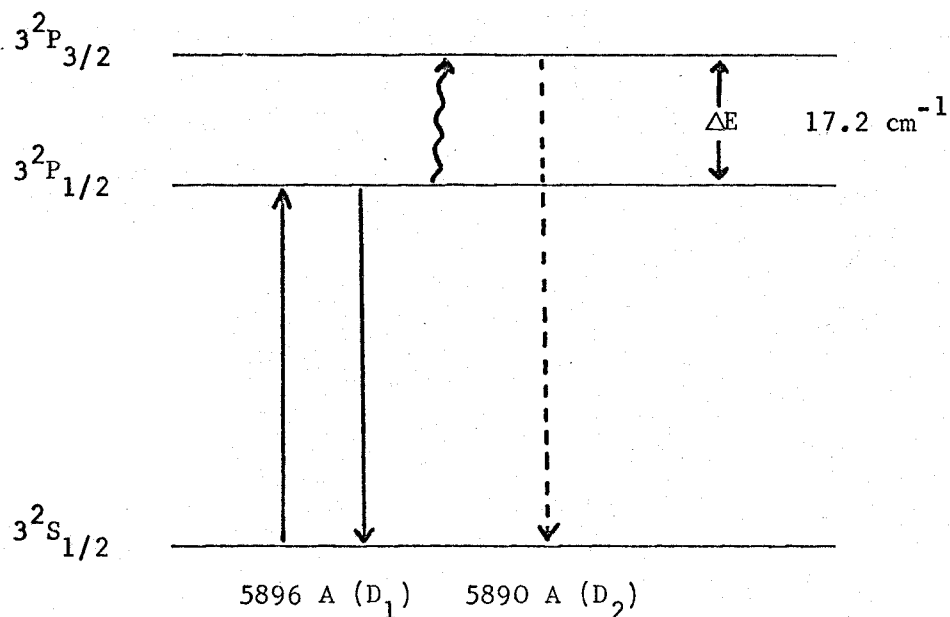
Fig. 1. The Sodium Term Diagram

The first studies of sensitized fluorescence in alkali metals

were made with sodium by Wood (1912, 1914) and Wood and Mohler (1918).

Later, collisions in pure sodium vapour as well as in its mixture with

argon were investigated by Lochte-Holtgreven (1928). Seiwert (1956)

determined collision cross sections for sodium-sodium collisions and

also interpreted Lochte-Holtgreven's work with argon, obtaining the

appropriate collision cross section.

The most recent work with sodium was carried out by Jordan and Franken (1966) who determined the cross sections for collisions between sodium and all the noble gas atoms. However, because of the small separation between the fine structure levels, there was difficulty in efficiently resolving the NaD lines without loss of intensity. Thus they found it necessary to illuminate the fluorescing vapour with both lines simultaneously and to monitor the degree of mixing as a function of inert gas pressure by scanning the fluorescent spectrum with a spectrometer and recording the D_2/D_1 intensity ratio in the fluorescent light. This method assumes that the D_2/D_1 intensity ratio in the exciting light remains constant throughout each experiment and that increasing noble gas pressures influence the absorption profiles of both D lines in the same manner.

In the present investigation the use of interference filters developed by Dobrowolski (1959) allowed efficient separation of the D lines with sufficient intensity in the exciting beam, and obviated the necessity of making either of the above assumptions.

Sodium-sodium collisions were investigated by Seiwert (1956) at high vapour pressures where large corrections for imprisonment of radiation must be made. Imprisonment of radiation occurs when an emitted photon is reabsorbed before escaping from the resonance cell and results in an apparent increase in the lifetimes of the excited states. Seiwert's corrections for trapping were made on the basis of a modified Holstein (1947) theory. No such corrections were necessary in the present

investigation since vapour pressures were always so low that radiation

trapping effects were negligible (Kibble, Copley and Krause, 1967a).

It is of interest to determine whether Na-Na collisions fit

into the scheme of Franck's rule (1929) which predicts that cross sections

should vary inversely with the energy defect ΔE. The agreement of K-K,

Rb-Rb and Cs-Cs cross sections with this rule has been used by Krause

(1966) to predict a cross section of 800 $\overset{\circ}{A}^2$ for the Na-Na collisions.

A detailed theoretical treatment for alkali-noble gas

collisions has been available only for the case of sodium where the

small energy defect allows for the assumption of semiclassical approxi-

mations. Accurate experimental data are thus necessary to verify the

existing theoretical calculations and to permit further refinements of

the theory.

II. THEORY

A. The Microscopic Theory

The detailed treatment of the problem of collisional excitation transfer presents difficulties since there is no adequate knowledge of interatomic interaction potentials, especially for excited states where wave functions are known at best approximately. There is also the additional difficulty of solving in closed form the dynamic problem. Such a solution has not as yet been achieved. The magnitude of ΔE is very important in that it divides the collisions into the two limiting cases, adiabatic and non-adiabatic, according to whether ΔE is greater or smaller than hv/a, where h is Planck's constant, v is the relative velocity of approach and a is an effective interaction distance of atomic dimensions. A detailed semiclassical explanation of this division, known as Massey's criterion, has been given by Thorson (1961). Of all the alkali metals, only lithium and sodium have ΔE small enough, so that the collisions may be considered non-adiabatic. In this case the theory is simplified since a semiclassical approach may be employed, which considers motion to follow the classical path where changes in velocity due to the interconversion of ΔE are ignored. It is for this reason that no detailed theoretical treatment is available for the heavier alkali atoms.

Moskowitz and Thorson (1963) studied non-adiabatic collisions by using, within the scope of perturbation theory, a modified "distorted-wave" approximation. They took, as a zero order approximation, the wave

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functions corresponding to elastic scattering of two rigid spheres. The distorted-wave method takes into account the distortion of the incident and outgoing waves by the scattering field and it is employed when the off-diagonal elements of the interaction matrix are small. In this treatment the sodium P orbitals distinguished by J and m_J have an orientation fixed in space, are not strongly coupled to the axial field of the colliding pair and do not follow the internuclear axis as it rotates throughout the collision. The nuclear motion occurs on an effective elastic scattering surface determined by the interaction energy of the filled shells of the sodium and noble gas atoms, and it is the axial perturbation which introduces transitions among the zero-order eigenfunctions of J^2 and m_J. Cross sections calculated in this approximation are extremely sensitive to the location of the classical turning points and a decrease of ten per cent in the hard-sphere diameter caused the cross section to increase by a factor of six. Comparison with the present investigation may be summed up by quoting Moskowitz and Thorson (1963), " Numerical results agree qualitatively with experiment but this agreement is fortuitous and is not the immediate aim of the calculation".

Nikitin (1965 a,b) disagreed with the above approach and held that transitions take place only in the restricted region where spin-orbit decoupling of the excited sodium atom commences and before the momentum of the optical electron is coupled with the axis of the quasi-molecule, which results in molecular terms between which there are no allowed transitions. Thus the cross sections cannot correlate with

the gas kinetic cross section of the colliding pair. In Nikitin's approach the problem reduced to the solution of three simultaneous equations for the probability amplitude. He discussed two models based on a choice of the axis of quantization used in the calculation of the interaction Hamiltonian. For the molecular model the axis of quantization was the molecular axis and this model was used when the angular velocity of rotation of the molecular axis was smaller than the fine structure frequency. The atomic model had the quantization axis fixed in space in such a manner that it coincided with the momentum vector of the relative motion of the colliding pair; this model was used for fast rotation.

Jordan (1964) employed time-dependent perturbation theory and used, as an approximation for the sodium atomic wave functions, modified hydrogenic wave functions. He further suggested that the electrostatic interaction arose from both van der Waals and quadrupole-induced dipole forces. The cross sections were found by determining the region beyond which the transition probability for a mixing collision dropped to zero and using the impact parameter corresponding to that region as the cross section radius. The greatest sources of error in his calculation arose from uncertainties in the choice of wave functions used for the excited ^{2}P state and in the value of the critical impact parameter.

The 'general approximate' method of Callaway and Bauer (1965) was based on the possibility of obtaining, with the use of the impact parameter method, an exact expression for the transition amplitude. The

interaction of the atoms was described by means of an effective potential which acted to produce transitions in the sodium atom. The latter was regarded as a core with one electron whose wave function was represented by a Slater determinant. Thus the matrix element reduced to an integral involving a single electron's coordinates, and the equation giving the time dependence of the transition amplitude could be regarded as applying to a one-electron problem. It may be of interest to note that the cross section quoted by Callaway and Bauer for sodium argon collisions is larger than the value set as an upper limit by Nikitin.

A survey of the theoretical results shows that of all sets of calculations those of Jordan agree most closely with the cross sections obtained in this research. However, it is difficult to determine at this point which if any of these methods will prove the most satisfactory for the solution of the problem. By suitable adjustment of assumed parameters, or by the use of more exact wave functions for excited states et cetera, quite different results might be obtained in the future from similar approaches.

B. The Macroscopic Theory

In order to analyse the experimental data, it is necessary to examine the rate equations that apply to a three level atomic system which exists in a steady state involving continuous optical excitation by only one NaD frequency, spontaneous decay and binary inelastic collisions. Quenching effects due to noble gases as well as imprisonment of radiation under the conditions of this experiment have been

shown by Kibble, Copley and Krause (1967 a,b) to be negligible.

The rate equations which represent the rates of change in the densities of the collisionally populated levels are as follows:

$$dn_2/dt \; = \; 0 \; = \; Z_1 N_1 - n_2/\tau - Z_2 n_2 \quad , \tag{2}$$

$$dn_1/dt \; = \; 0 \; = \; Z_2 N_2 - n_1/\tau - Z_1 n_1 \quad . \tag{3}$$

The subscripts 1 and 2 refer to the $^2P_{1/2}$ and $^2P_{3/2}$ states respectively. N_1 and N_2 refer to the densities of atoms in the states being optically excited and n_1 and n_2 refer to the states being collisionally excited. τ is the average lifetime of the ^2P states, and Z_1 and Z_2 are the numbers of collisions per excited atom per second resulting in the transitions $^2P_{1/2} \longrightarrow {}^2P_{3/2}$ and $^2P_{3/2} \longrightarrow {}^2P_{1/2}$, respectively.

Dividing (2) and (3) by N_1 and N_2 respectively yields,

$$Z_1 \; = \; (1/\tau + Z_2) \, \eta_2 \quad , \tag{4}$$

and

$$Z_2 \; = \; (1/\tau + Z_1) \, \eta_1 \quad , \tag{5}$$

where

$$\eta_1 \; = \; I(D_1)/I(D_2) \; = \; n_1/N_2 \quad , \tag{6}$$

and

$$\eta_2 \; = \; I(D_2)/I(D_1) \; = \; n_2/N_1 \quad , \tag{7}$$

and where I refers to fluorescent intensity and the wavelengths in the denominators are in each case, the same as for the exciting light. In Equations (6) and (7) it is assumed $\tau_1 = \tau_2 = \tau$.

Solving (4) and (5) for Z_1 and Z_2 gives,

$$Z_1 \; = \; \frac{1}{\tau} \frac{\eta_2 + \eta_1 \eta_2}{1 - \eta_1 \eta_2} \tag{8}$$

and

$$Z_2 = \frac{1}{T} \frac{\eta_1 + \eta_1 \eta_2}{1 - \eta_1 \eta_2} ,$$ (9)

which in the case of small η reduce to

$$Z_1 = \eta_2/T ,$$ (10)

and

$$Z_2 = \eta_1/T .$$ (11)

The collision numbers Z are functions of temperature according to

$$Z(T) = N \int q(v) \, v \, f_T (v) \, dv ,$$ (12)

where N is the density of ground state atoms, $q(v)$ is the differential cross section, v is the velocity of relative motion and $f_T(v)$ is the Maxwell-Boltzmann probability that at temperature T degrees Kelvin the colliding partners have relative velocity between v and $v + dv$.

By analogy with the gas kinetic cross section we may define an effective total cross section Q by,

$$Z_1 = N Q_1 v_r \quad ; \quad Z_2 = N Q_2 v_r ,$$ (13)

where v_r is the mean relative velocity given by

$$v_r = (8kT/\pi \mu)^{1/2} ,$$ (14)

and where k is Boltzman's constant and μ is the reduced mass of the colliding atoms.

The relationship between the effective total cross section and the differential cross section is given from (12) and (13) as,

$$Q(T) = 1/v_r \int q(v) \, v \, f_T (v) \, dv .$$ (15)

According to the principle of detailed balancing the ratio

of the cross sections should be,

$$Q_1/Q_2 = g_2/g_1 \exp(-\Delta E/kT) \quad , \tag{16}$$

where $g_1 = 2$ and $g_2 = 4$ are the statistical weights of the $^2P_{1/2}$ and $^2P_{3/2}$ states respectively, and the Boltzmann factor, $\exp(-\Delta E/kT)$, is the probability that in a collision the kinetic energy of relative motion will be greater than ΔE.

III. DESCRIPTION OF THE APPARATUS

A. General Description

The arrangement of the apparatus is shown in Fig. 2. Light emitted from a sodium spectral lamp, after passing through a pin hole, was rendered parallel and one of the two D components was separated by interference filters. The monochromatic beam was then condensed into the fluorescence cell containing sodium vapour at a controlled pressure. Fluorescent light emitted normally to the incident beam was collimated, resolved into its two D components by interference filters and brought to a focus on the S_{20} photocathode of a photomultiplier which was enclosed in a liquid air-cooled vacuum cryostat. Intensities were recorded by means of a pulse counting train. An electromechanical sequencing system integrated with the counting train operated motor driven filter holders in the incident and fluorescent beams and allowed for an automatically programmed sequence of operations. (Pitre, 1965). The whole apparatus was enclosed in a light tight box to protect it from stray light.

B. The Spectral Lamp

A commercial Osram spectral lamp operated at 1.4 A was used for the sodium-noble gas collision experiments. However, for the sodium-sodium experiment, the intensity ratios η were much smaller and it was necessary to develop a light source which was more efficient in exciting

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A - sodium lamp
B - fluorescence tube and oven
C - greaseless stopcock
D - photomultiplier and cryostat
M - amplifier-discriminator
N - scaler
P - pinhole
L - lenses
S - stops
F - Spectrolab filters
F_1 - Kodak 301 filter
F_2 - neutral density filter
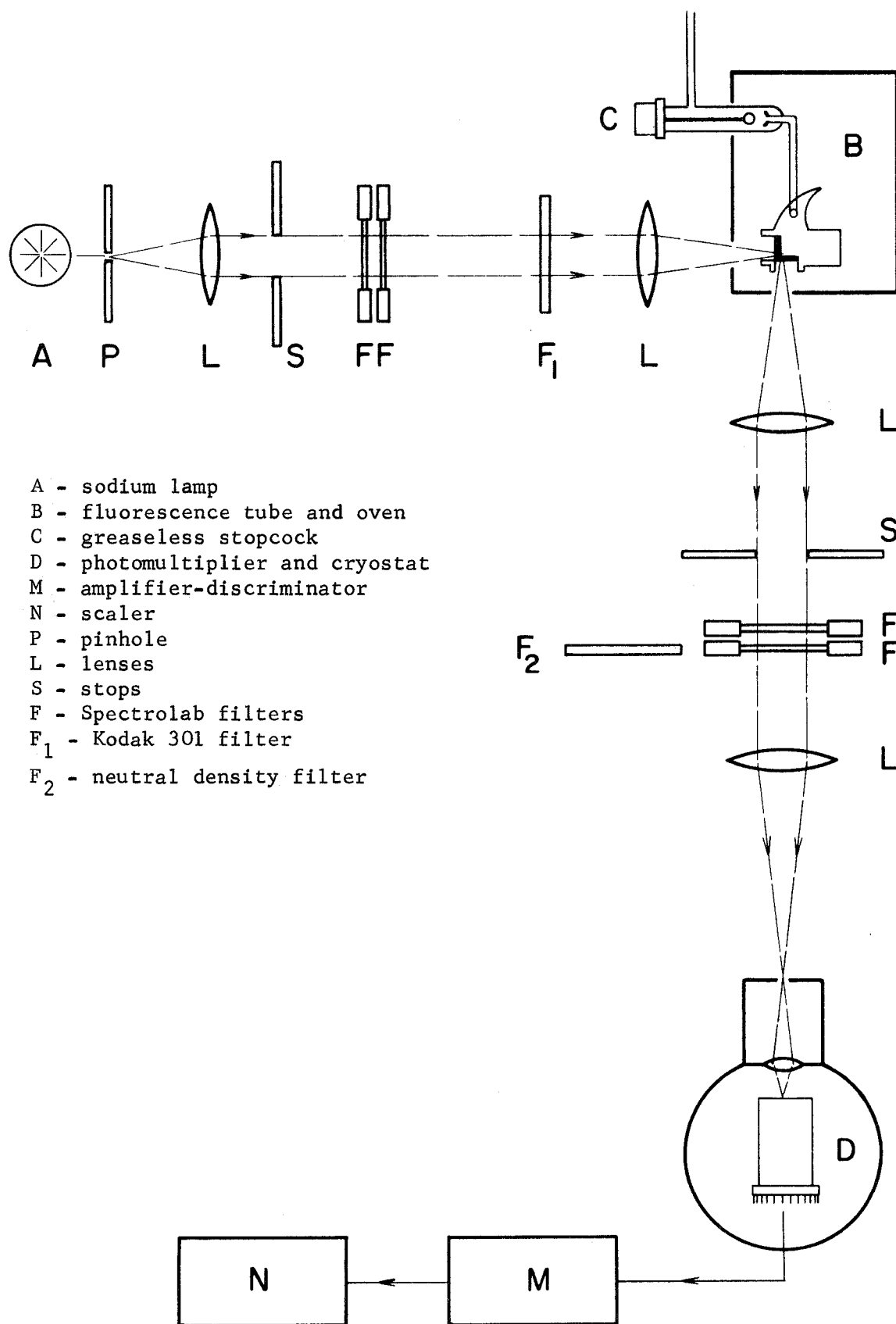
Fig. 2.   Schematic diagram of apparatus.

resonance fluorescence. This spectral lamp was a modification of the radiofrequency sources described by Berdowski, Shiner and Krause (1967) and Burling and Krause (1967).

It had been suggested by Bell, Bloom and Lynch (1961) that excitation by frequencies higher than 30 Mc/s should produce an increased lamp intensity since reduction in skin depth would reduce the thickness of the ring discharge and result in less self absorption. The only basic modifications necessary to achieve higher frequencies were the reduction of the inductance in the tuned circuit and the use of 4-65 A tetrodes. The tuned circuit and lamp excitation coils both consisted of 5 turns of 12-gauge wire and were 2 cm long and 2 cm in diameter. The latter coil was in parallel with a small variable capacitor which would be adjusted for maximum power transfer from the oscillator to the discharge tube. The main difficulty with sodium discharge lamps is caused by the rapid discoloration and crystallization of pyrex glass and, to a lesser degree, of Corning 1720 'alkali resistant' glass under the attack of hot sodium. The problem was resolved by using the inner discharge tube of a Philips spectral lamp which showed no signs of discoloration or deterioration after continuous operation during a period of over 200 hours. Typical power requirements of the oscillator were 220 ma at 500 V and care was taken not to overdrive it or else the glass of the discharge tube would melt. Because of the small dimensions of the excitation coil it was necessary to employ heaters at both ends of the discharge tube. For maximum intensity, the end which contained the sodium and controlled its vapour pressure was

held constant at 208°C while the other end was maintained at about 20°C higher.

This lamp operated at 146 Mc/s and was 3 times more efficient in exciting resonance fluorescence than new commercial Osram lamps but showed no definite increase over lamps operated at 40 Mc/s. However, its extreme stability as shown by a less than $\pm$ 4 per cent variation in fluorescence excited by this lamp over a nine hour period resulted in its adoption for the experiment.

C. The Interference Filters

The sodium resonance doublet was resolved by Spectrolab Type 'U' interference filters which typically transmitted about 60 per cent of the wanted resonance line and one per cent of the unwanted component. Two such filters in series were used in the exciting beam for the sodium-noble gas experiment, and three for the investigation with pure sodium vapour. In the former experiment exciting light was also passed through a Kodak '301' short wave pass filter which removed all other alkali resonance radiation which would have caused fluorescence in traces of alkali impurities that might be present in the sodium. An additional Kodak '301' filter plus a Spectrolab Type 'P' interference filter with a 60 Å transmission band width centred at the NaD lines were inserted into the incident beam for the pure sodium investigation where much greater spectral purity was required. Knowledge of the transmissions of the filters in the incident beam to the NaD lines was unnecessary since they did not enter into the calculation of cross sections. Type 'U' filters were also

used to separate the D lines in fluorescence and a neutral density filter was employed in measurements of resonance fluorescence to prevent the scaler from becoming overloaded. Considerable effort was made to ensure that, in both the exciting and fluorescent beams, the light passing through the interference filters should be strictly parallel as it must be for optimal resolution. This was achieved, at some expense to the intensity of the signal, by means of a series of pinholes and stops. The latter also confined the light beam to the central regions of the filters whose edges appeared to reject less efficiently the undesirable spectral components than their central portions. The filters themselves were mounted in holders which permitted them to be precisely oriented with respect to each other and to the light beam, and to be rotated about the light beam axis. These holders, in turn, were fixed to motor driven slides which were capable of remote operation.

D. The Fluorescence Cell

The fluorescence cell which has been described by Rae and Krause (1965) was made of pyrex glass and consisted of two parts: the main body, where fluorescence took place, and a side-arm containing the liquid sodium. A blackened horn, which was connected to the vacuum and gas handling systems by a greaseless stopcock, was situated behind the cell windows. The exciting beam was focussed just inside the entrance window of the cell and as near as possible to the side window without causing any reflections from it. The cell was so designed that the centre of the side window was immediately behind the centre of the front window and the exciting and fluorescent light had to pass

through only a very thin layer of unexcited sodium atoms. This design minimizes both unwanted absorption of the incident beam and diffusion of radiation.

E. The Ovens

The oven containing the fluorescence cell consisted of a double-walled copper box, thermally insulated from the outside by glass wool. The exciting and fluorescent beams passed through glass windows set in annular teflon plugs. The heater, a 56 ohm coil non-inductively wound on a ceramic cylinder, was situated in one corner behind a copper shield and heating took place by convection. The A.C. current for the heater was controlled by a powerstat and a saturable core reactor. Control of the D.C. current for the latter was provided by a transistorized D.C. amplifier employing a suitably placed Veco 41A2 thermistor as a sensing element. The circuit diagrams for all the heater control units were given by Pitre (1965). A similar circuit was used to control the current for the heaters of the r.f. discharge lamp.

The side arm of the fluorescence cell which contained the sodium sample* was always kept at a lower temperature than the main body of the cell in order to prevent condensation of sodium on the cell windows and also to provide precise control of the sodium vapour pressure. This was accomplished by passing low viscosity paraffin oil from a Wobser Ultrathermostat through a closely fitting 3/16 inch copper coil surrounding the side arm. With this arrangement the main oven temperature

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* Natural sodium of nominal purity 99.9 per cent was obtained from the
A. D. MacKay Company of New York.

T' could be stabilized to within $\pm$ 0.1°C over a range extending from 25°C to 300°C and the side arm temperature T could be maintained to within .05°C over a range extending from 100°C to 200°C. All temperatures were measured by means of copper-constantan thermocouples and a Leeds and Northrup 8662 potentiometer.

F. Vacuum and Gas Handling System

A vacuum of better than 1 x 10^{-7} mm Hg was obtained using a Speedivac E02 diffusion pump operated with Dow Corning 704 fluid and backed by an ES35 rotary pump. The diffusion pump was protected from cooling water failures by an FSM-1 Flowtrol unit.

The admission of small quantities of Linde M.S.C. noble gases from one litre flasks to the system was monitored by a Speedivac Model 8/1 Pirani type gauge. Final pressures were measured with a C.V.C. GM-100A McLeod gauge which was isolated from the system by a liquid air trap and stopcock. To prevent possible contamination of the system by mercury vapour the stopcock to the gauge was only opened when there was a positive pressure from the system to the gauge.

G. The Detection and Recording System

The fluorescent beam was detected by an I.T.T. type FW-130X, 16 dynode, end-on photomultiplier which was cooled in a liquid air cryostat. The Sb - K - Na - Cs photocathode had an S20 spectral response, was 0.1 inch in diameter and had a quantum efficiency of about 7.5 per cent for the NaD lines. Pulses from the photomultiplier were passed through the amplifier-discriminator unit PW 4270 and were recorded on the scaler PW 4230. The scaler readings were registered by means of a

printer control PW 4200 and a Victor printer. It was not necessary to preamplify the pulses at the photomultiplier and thus direct connection could be made between the photomultiplier and the amplifier-discriminator unit. The dark noise of the photomultiplier was also determined, which, with an overall tube-gain of 3×10^5 and an effective quantum efficiency[*] of 3 per cent, amounted to 30 counts/min (equivalent to a D.C. dark current of 2×10^{-14} amp) and remained constant for many months.

The dead time of the counting system which depends on the height and decay time of the photomultiplier pulses and also on the discriminator setting, was obtained using the following formula which is applicable to a non-paralyzable counter.

$$a = A/1 - A\tau_o , \qquad (17)$$

where a is the true count rate, A is the observed count rate and τ_o is the dead time.

The true count rate a was determined by inserting an electrometer between the anode of the photomultiplier and ground, and measuring the photomultiplier current g. The relation,

$$g = k a , \qquad (18)$$

where k is a constant, assumes only that the photomultiplier output is linear with input intensity. Combining equations (17) and (18) gives

$$1/A = k/g + \tau_o . \qquad (19)$$

A plot of 1/A versus 1/g yielded the dead time τ_o as the intercept of this linear plot. The dead time was found to be 1.4 ± 0.2 μ sec. and this gave rise to corrections, never exceeding 1.4 per cent, to the observed counting rates.

Footnote for page 19.

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* The effective quantum efficiency is the product of the photocathode quantum efficiency (7.5 per cent at 5890 Å for an S20 surface) and a factor arising from the use of pulse discriminating techniques.

IV. EXPERIMENTAL PROCEDURE

A. Experiments with Sodium-Noble Gas Mixtures

At the beginning of the experiment, the fluorescent cell was evacuated and baked at a pressure of 1×10^{-7} torr, and a drop of sodium was distilled into the side-arm. The Pyrex bottles containing the noble gases (Linde M.S.C. grade), which had been connected to the system, were opened under vacuum by glass-enclosed magnetic breakers. The temperature of the sodium in the side-arm, which controlled the vapour pressure in the fluorescence cell, was $115^{\circ}C$ and corresponded to a vapour pressure of 5×10^{-7} mm Hg. The higher temperature of the main cell, $124^{\circ}C$, prevented condensation of sodium on the windows. The transmissions of the filter sets were measured in situ and were remeasured several times during the experiments.

The transmission of a Type 'U' Spectrolab filter to the appropriate NaD component depends on the polarization of the incident light (Dobrowolski, 1959). Thus the D_2 component in the fluorescent light could not be directly isolated by means of the appropriate filter because of its partial polarization which amounted to about 20 per cent in the absence of inert gases and which decreased rapidly with increasing gas pressure. Instead, it was found practicable to determine first the total fluorescent intensity and then, with the D_1 filters in the fluorescent light beam, the intensity of the D_1 component. The subsequent subtraction of the two intensities yielded the intensity of

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the D_2 component. These procedures were followed at each chosen gas pressure, starting with the lowest. A complete sequence of measurements was carried out with the D_1 component used for excitation, followed by a similar sequence with D_2 excitation. Before each experimental run the fluorescence cell was evacuated and baked overnight. Each η value was determined in a series of 21 alternating one-minute photon count accumulations carried out with and without interference filters in the fluorescent light beam. After correcting the counts for filter transmissions, the readings were taken in pairs to give ten values of η whose arithmetic mean was accepted as the η_1 or η_2 value corresponding to the particular gas pressure. It should be noted that the determination of η_2 involved the subtraction of nearly equal intensities of total and resonance fluorescence from one another, to obtain the very low sensitized fluorescent intensity. However, even this difficult measurement produced quite satisfactory results.

The dark noise of the photomultiplier, which amounted to 30 counts/min resulted, in the case of the very faint sensitized fluorescence measurements, in a signal-to-noise ratio ranging from 10 to 100. With the much more intense resonance fluorescence, dark noise was negligible but corrections for light reflected in the cell, which amounted to about 0.5 per cent, were made to the measured intensities.

Due to the optical activity of the mica in the interference filters, the incident beam was partially linearly polarized with the axis of polarization at an angle of 70° to the plane defined by the incident and fluorescent beams. A simple calculation (Feofilov, 1961)

showed that under these conditions the effects due to anisotropy of

the D_2 radiation on the calculation of cross sections were negligible.

B. The Experiment with Pure Sodium

All traces of inert gases were removed from the cell by

baking it at $185^\circ C$ and then pumping on it for an extended time period

at lower temperatures. At all times when data was being collected,

the greaseless stopcock remained open and the cell which was connected

to the vacuum system by 8 mm tubing had a residual gas pressure never

exceeding 1×10^{-7} mm Hg.

In order to minimize imprisonment of radiation, the image

of the slit, which was 2.7 mm in width, was positioned so that its

edge was about 0.5 mm from the side window of the cell. In this

configuration the fluorescent light had only to pass through a very

thin layer of unexcited atoms before escaping from the cell and yet

the light reflected in the cell was only 0.5 per cent of the resonance

fluorescent intensity.

After passing through three interference filters which

contained optically active mica, the exciting beam was almost completely

linearly polarized with the plane of polarization close to the plane

defined by the incident and fluorescent beams. Under these circumstances

the D_2 resonance was only slightly polarized (Feofilov, 1961), and

corrections to the cross section for radiation anisotropy were

negligible.

At times when no data were being collected, the main cell

was maintained at more than $10^\circ C$ above the temperature of the side arm.

However, during the experimental runs, the side arm temperature was raised to within $2^{\circ}C$ of the main cell to avoid any significant temperature gradients in the fluorescent cell. The absence of condensation on the cell windows during these periods was verified by periodic cooling of the entire fluorescence cell and checking for scattered light. The vapour pressure of the sodium as determined by these temperatures was calculated from the empirical equation of Nesmeyanov (1963). However, in the pressure range of this investigation Nesmeyanov's equation is an extrapolation from existing vapour pressure data and as such may be regarded as a small source of systematic error.

Since the transmissions of the appropriate NaD components depend on the polarization of the incident light and since the polarization of the sensitized fluorescence is different from that of the resonance fluorescence (Gough, 1967), it was not possible to determine the transmission of the interference filters to D_2 sensitized fluorescence. Thus, although η_1 could be measured directly, η_2 could not; nor could η_2 be obtained indirectly by the method outlined in the procedure for experiments with noble gases since all η_2 values would be smaller than 1×10^{-4} in the experimental pressure range.

In the determination of η_1, four D_1 interference filters were used in series to separate the D_1 component in the fluorescent beam. The transmission of this filter set was measured in situ by employing two additional D_1 filters in the exciting beam. Each experimental η_1 value was a result of approximately 100 one-minute counts of signal alternating with 100 similar counts of background noise which were obtained by opening and closing a shutter in the exciting

beam. Every nineteen minutes a one-minute accumulation of resonance fluorescence was registered, and thus it was only necessary for the lamp to remain constant for a nineteen minute interval, but, in fact, as was pointed out earlier, the lamp remained constant to within a few per cent during each 215 minute period. The background noise which was due almost entirely of photomultiplier dark noise amounted to 60 counts per minute. This increase in dark noise over that quoted for the sodium-noble gas experiments was a result of a minor electronic alteration which increased the effective quantum efficiency of the detection system.

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V. RESULTS AND DISCUSSION

A. Experiments with Sodium-Noble Gas Mixtures

The experimental values of η_1 and η_2 for the Na - He mixture are listed in Table I and are plotted against helium pressures in Fig. 3; the resulting curve is representative of the remaining mixtures with noble gases. Because of the small energy defect between the ^2P resonance levels in sodium, complete mixing takes place at relatively low noble gas pressures. Above 80 torr $\eta_1 = 0.51$ and $\eta_2 = 1.9$ which is in good agreement with the theoretically predicted values of 0.53 and 1.88. The most rapid changes of η with gas pressure occur at pressures below 1 torr and all precise measurements were carried out in this pressure range. Figures 4, 5, 6, 7 and 8 are the graphical representations of Tables II, III, IV, V and VI and show plots of the η-values against pressures of helium, neon, argon, krypton and xenon, respectively, in the range 0 - 1 torr. Each curve represents at least two experimental runs. Values of η_1 and η_2 were read off the appropriate curves at 0.05 torr intervals and were used in a computer program to obtain the collision numbers Z_1 and Z_2 from Eqs. (8) and (9) and hence, from Eq. (13), the corresponding values for the cross sections Q_1 and Q_2 which were then averaged and yielded the final cross sections for the particular sodium-inert gas collisional process. The divergences between the individual Q_1 or Q_2 values, before averaging, were gratifyingly small as may be seen in the plots of the collision numbers Z_1 and Z_2

TABLE I

Fluorescent Intensity Ratios for High Helium Pressures

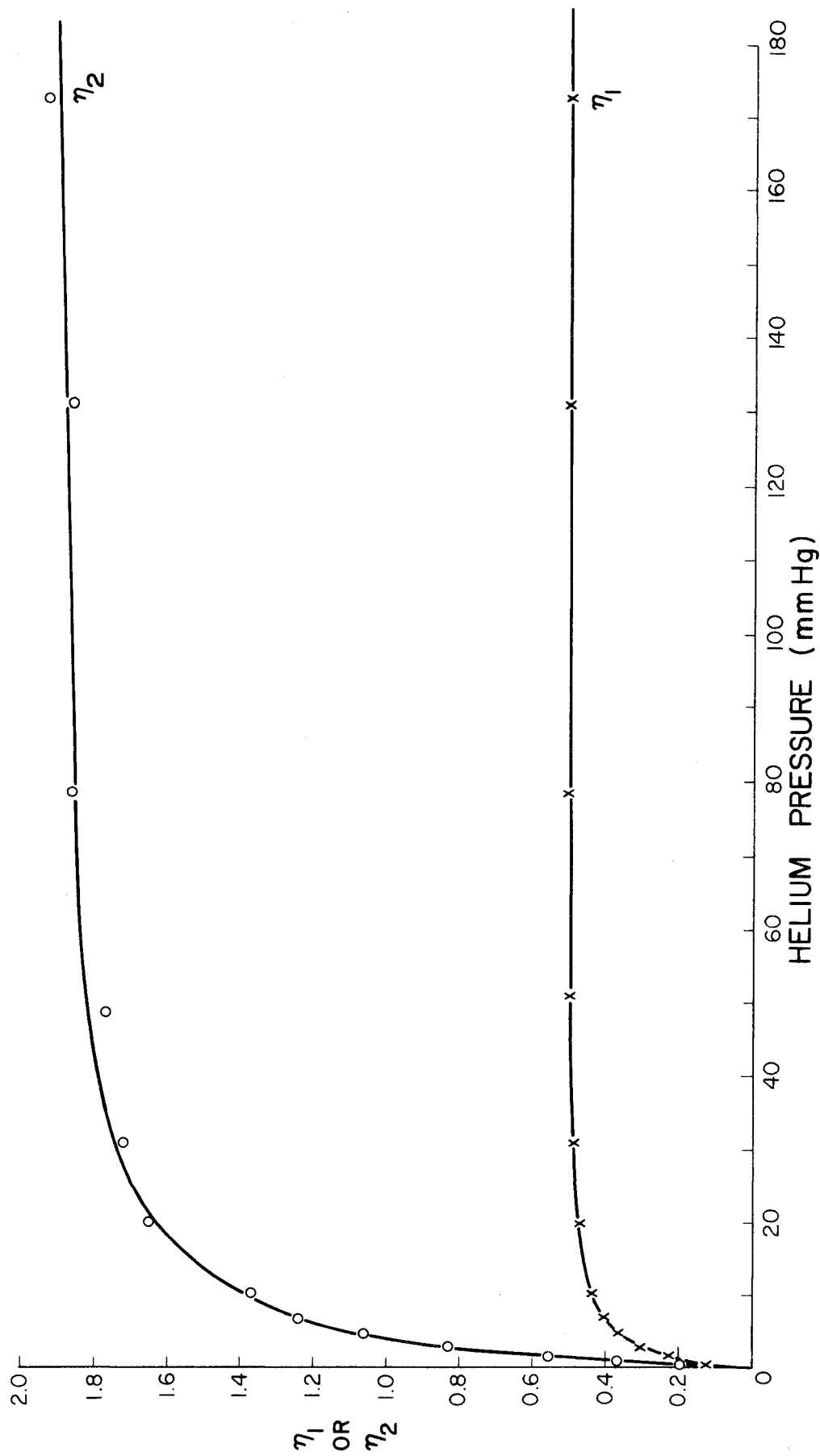| Helium Pressure (mm Hg)	η_1 D_1/D_2	Helium Pressure (mm Hg)	η_2 D_2/D_1
.601	.126	.41	.195
1.48	.227	.86	.366
2.81	.306	1.48	.556
4.74	.364	2.81	.831
6.79	.404	4.74	1.06
10.29	.438	6.79	1.24
20.0	.472	10.29	1.37
31.0	.487	20.0	1.65
51.2	.500	31.0	1.72
78.4	.507	51.2	1.77
131.	.511	78.4	1.86
173.	.505	131.	1.86
		173.	1.93

Fig. 3. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and Helium

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TABLE II

Fluorescent Intensity Ratios for Sodium-Helium Collisions

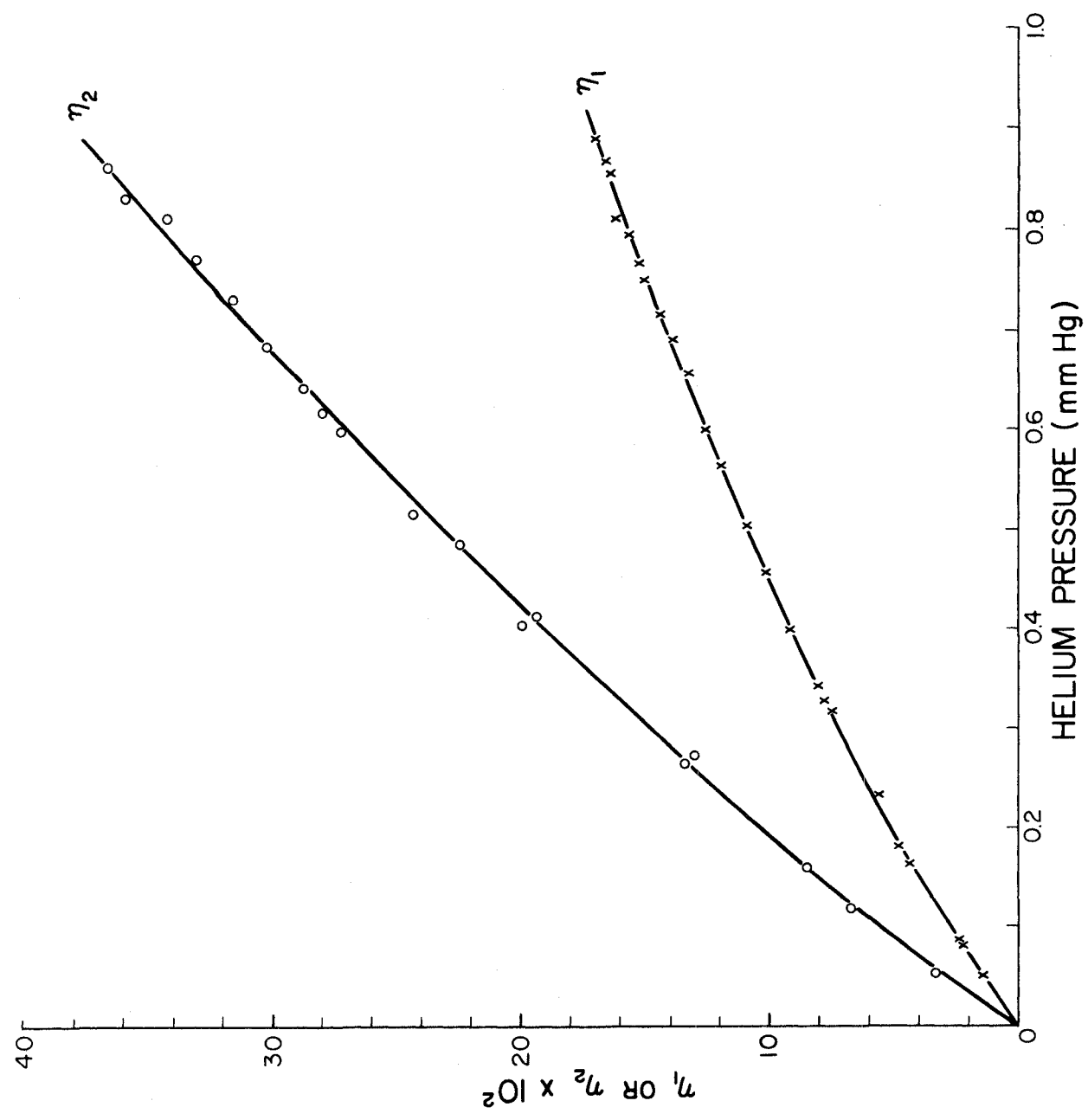| Helium Pressure (mm Hg)	$\eta_1 \, (\times \, 10^{-2})$ D_1/D_2	Helium Pressure (mm Hg)	$\eta_2 \, (\times \, 10^{-2})$ D_2/D_1
.050	1.46	.053	3.28
.081	2.27	.119	6.72
.087	2.41	.160	8.47
.163	4.30	.265	13.36
.181	4.76	.274	12.95
.232	5.57	.404	19.88
.318	7.45	.412	19.47
.328	7.81	.485	22.38
.342	8.03	.515	24.29
.400	9.11	.598	27.23
.458	10.10	.617	27.94
.504	10.90	.641	28.74
.563	11.94	.683	30.12
.601	12.63	.732	31.52
.657	13.31	.769	32.99
.691	13.89	.810	34.17
.716	14.38	.829	35.84
.750	15.02	.860	36.55
.768	15.22		
.796	15.67		
.811	16.15		
.855	16.34		
.868	16.58		
.891	17.00		

Fig. 4. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and
Helium at Low Pressures

TABLE III

Fluorescent Intensity Ratios for Sodium-Neon Collisions

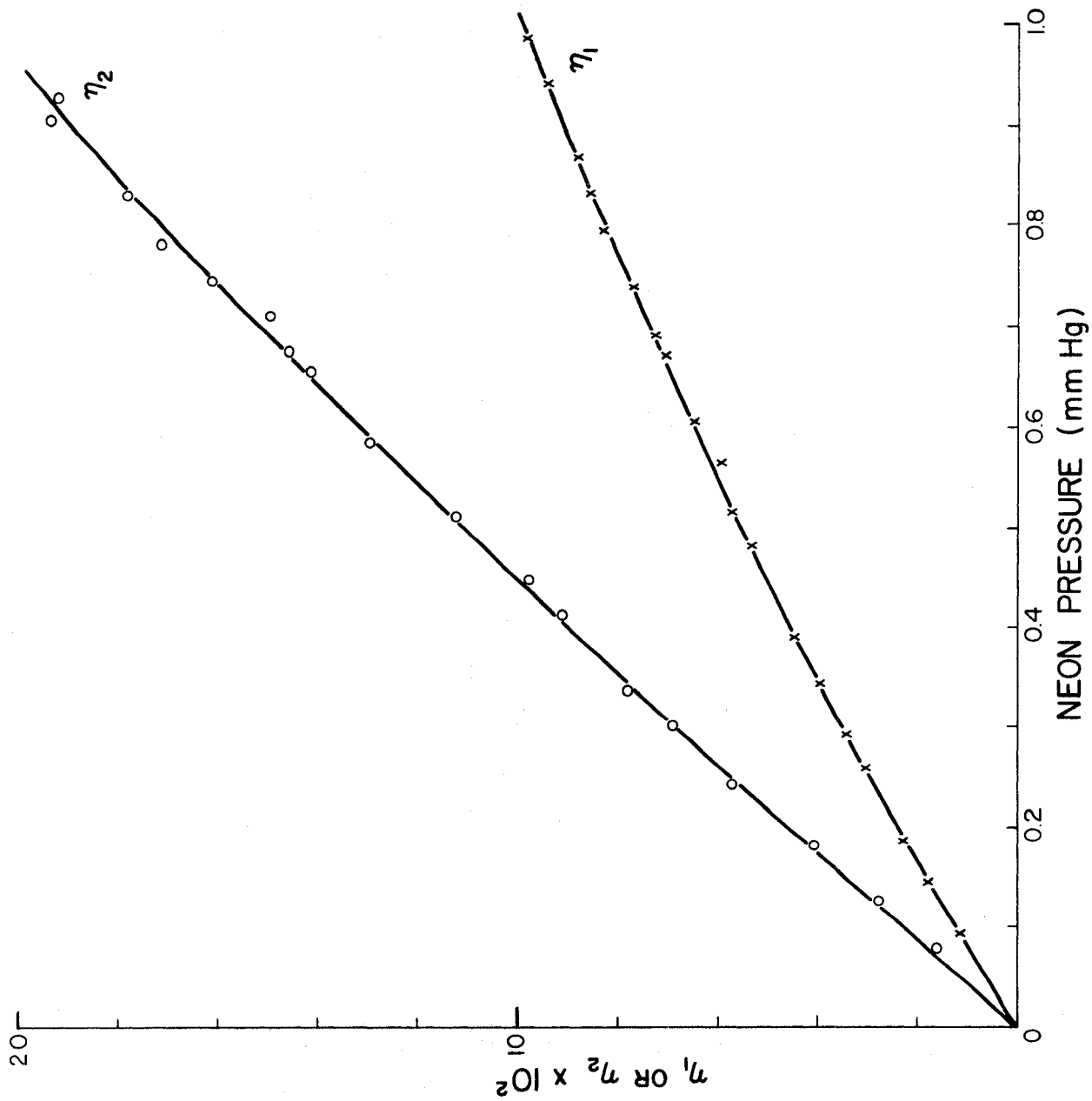| Neon Pressure (mm Hg)	η_1 (x 10^{-2}) D_1/D_2	Neon Pressure (mm Hg)	η_2 (x 10^{-2}) D_2/D_1
.093	1.12	.080	1.60
.147	1.78	.128	2.77
.186	2.26	.182	4.04
.259	3.02	.242	5.69
.293	3.41	.301	6.89
.342	3.93	.338	7.79
.391	4.44	.413	9.11
.482	5.27	.448	9.78
.517	5.68	.512	11.21
.566	5.91	.585	12.94
.606	6.44	.653	14.12
.671	7.01	.673	14.58
.690	7.23	.710	14.94
.738	7.65	.743	16.11
.795	8.26	.781	17.09
.832	8.53	.830	17.82
.868	8.78	.904	19.33
.941	9.38	.927	19.18
.985	9.81		

Fig. 5. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and Neon at Low Pressures

TABLE IV

Fluorescent Intensity Ratios for Sodium-Argon Collisions

Argon Pressure (mm Hg)	η_1 (x 10^{-2}) D_1/D_2	Argon Pressure (mm Hg)	η_2 (x 10^{-2}) D_2/D_1
.027	.49	.133	4.45
.062	1.05	.197	6.24
.135	2.17	.257	8.30
.210	3.32	.317	10.39
.329	5.06	.339	10.35
.340	5.18	.419	13.38
.411	6.08	.442	13.87
.458	6.61	.483	15.23
.562	8.10	.569	17.11
.578	8.19	.632	18.63
.700	9.39	.672	19.69
.765	10.14	.699	20.42
.841	10.83	.744	21.78
.960	12.00	.831	24.04
.992	12.60	.869	25.02
1.030	13.15	.933	26.01
		1.002	29.20

η_2

η_1

ARGON PRESSURE (mm Hg)

η_1 OR η_2 x 10^2

40

30

20
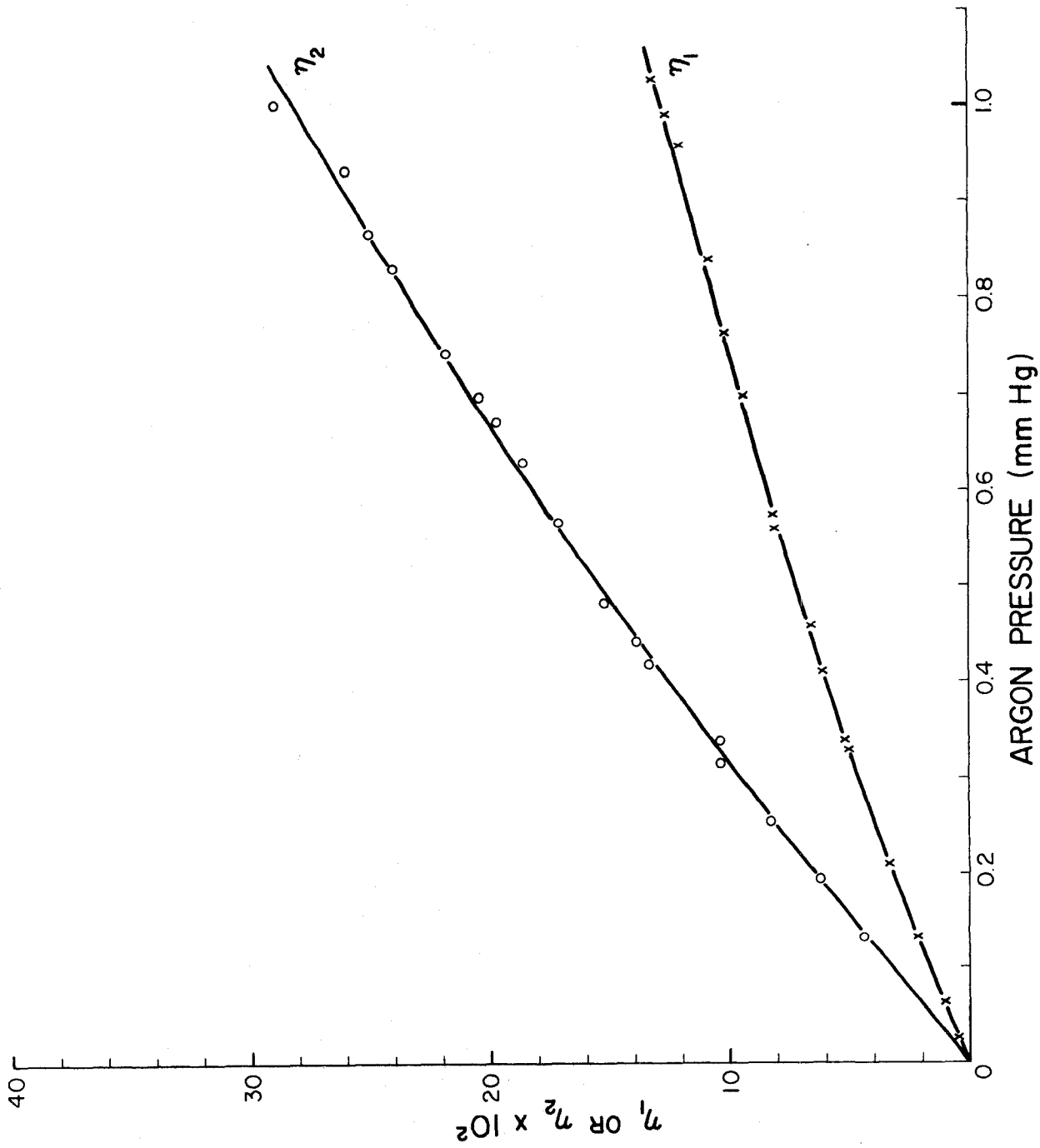
10

0

0.2 0.4 0.6 0.8 1.0

Fig. 6. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and Argon at Low Pressures

TABLE V

Fluorescent Intensity Ratios for Sodium-Krypton Collisions

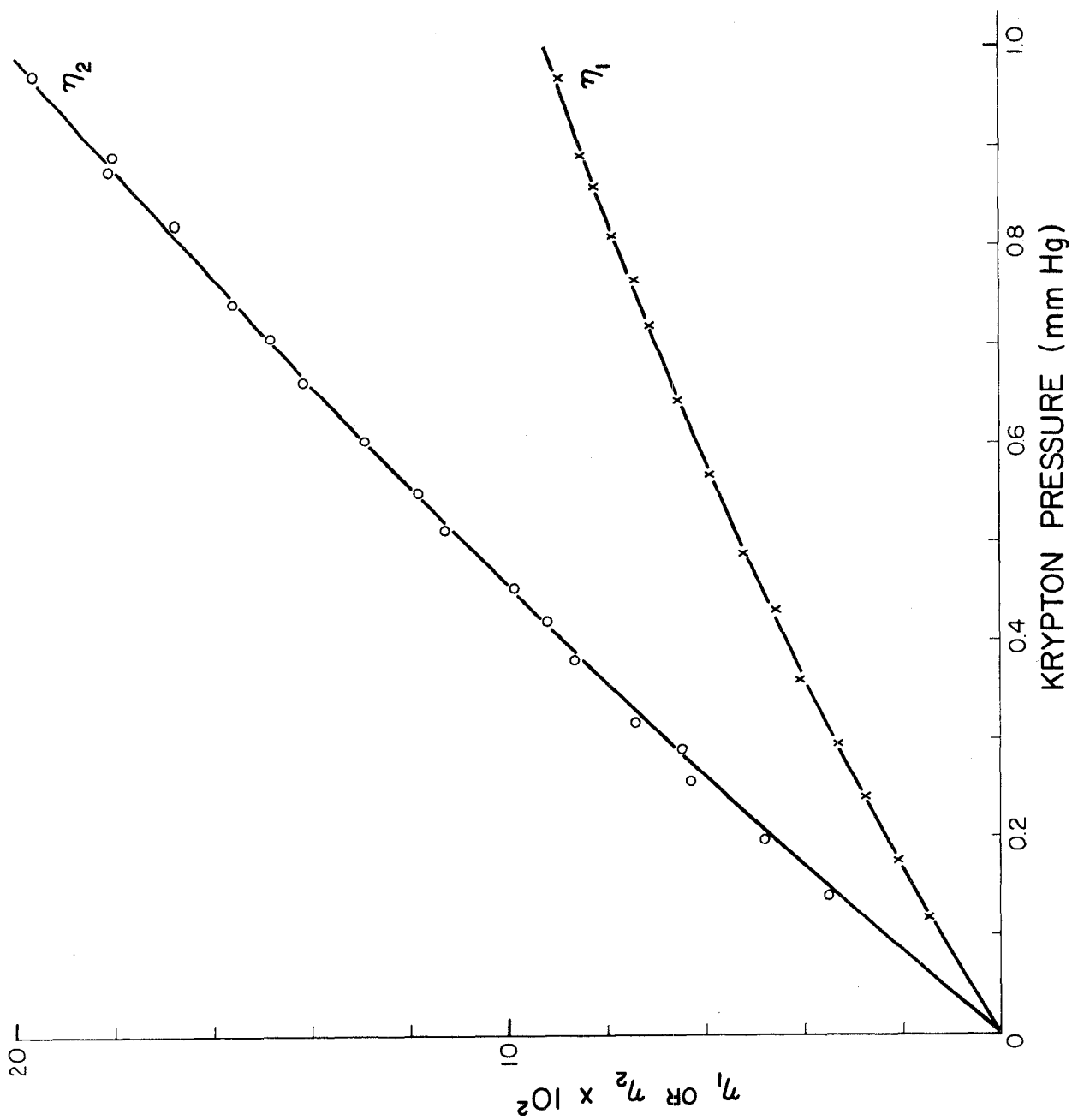| Krypton Pressure (mm Hg)	η_1 ($\times 10^{-2}$) D_1/D_2	Helium Pressure (mm Hg)	η_2 ($\times 10^{-2}$) D_2/D_1
.118	1.43	.140	3.51
.177	2.06	.198	4.79
.241	2.76	.258	6.32
.296	3.29	.291	6.47
.361	4.06	.318	7.43
.431	4.56	.381	8.63
.488	5.23	.420	9.18
.568	5.91	.453	9.84
.642	6.53	.513	11.26
.720	7.10	.549	11.79
.766	7.41	.603	12.89
.811	7.85	.664	14.17
.861	8.22	.708	14.81
.889	8.51	.741	15.58
.968	8.96	.821	16.75
		.875	18.06
		.890	17.98
		.971	19.61

Fig. 7. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and Krypton at Low Pressures

TABLE VI

Fluorescent Intensity Ratios for Sodium-Xenon Collisions

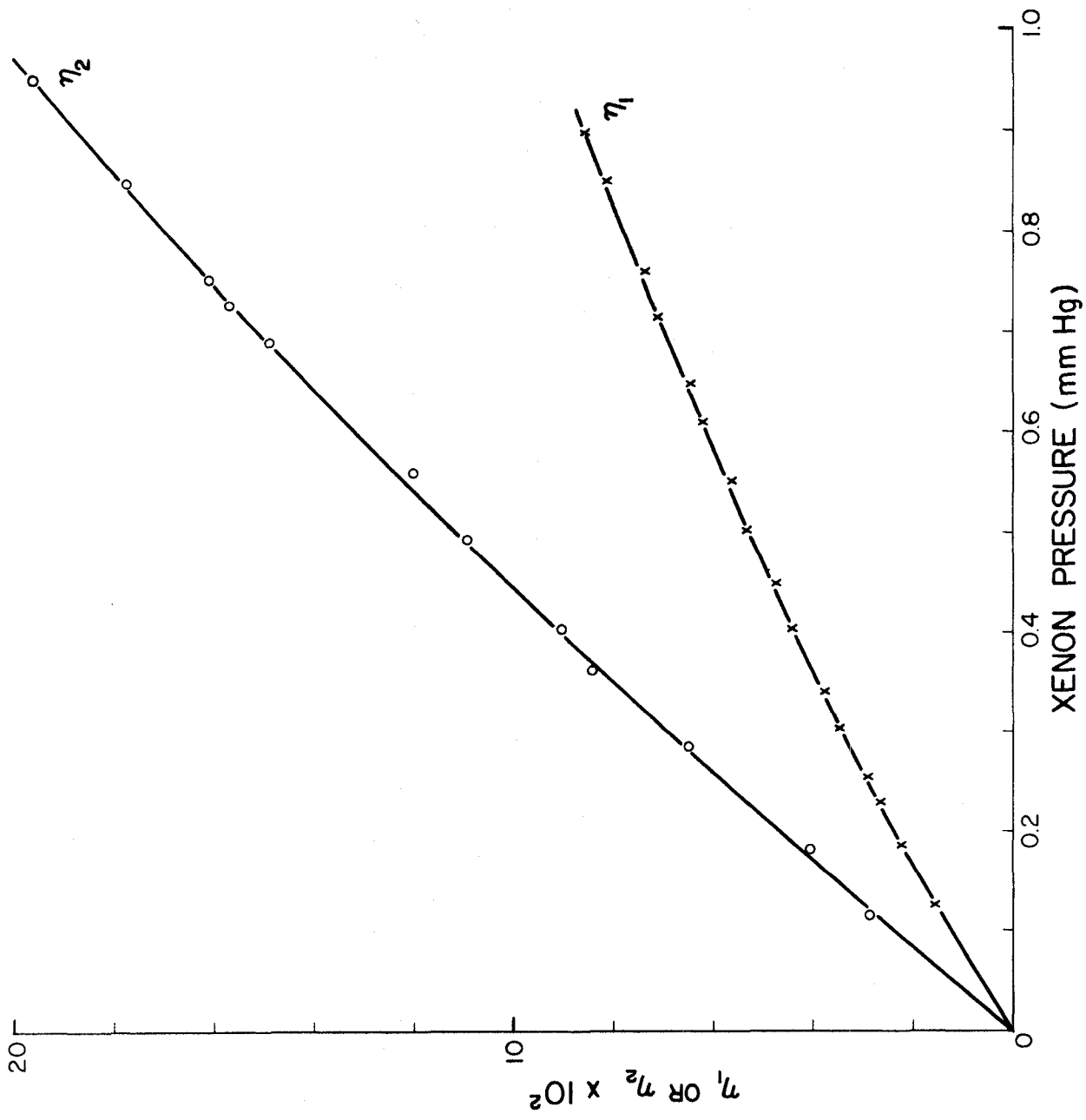| Xenon Pressure (mm Hg)	η_1 (x 10^{-2}) D_1/D_2	Xenon Pressure (mm Hg)	η_2 (x 10^{-2}) D_2/D_1
.127	1.56	.116	2.87
.187	2.21	.182	4.03
.230	2.62	.288	6.50
.257	2.90	.362	8.41
.303	3.45	.404	9.02
.341	3.76	.493	10.95
.405	4.41	.559	11.98
.449	4.74	.689	14.87
.503	5.33	.727	15.68
.552	5.62	.752	16.09
.610	6.22	.848	17.74
.648	6.42	.950	19.60
.715	7.12		
.760	7.35		
.851	8.09		
.899	8.54		

Fig. 8. Fluorescent Intensity Ratios in Mixtures of Sodium Vapour and Xenon at Low Pressures

against pressures of helium, neon, argon, krypton and xenon which appear

in Figs. 9, 10, 11, 12 and 13, respectively. These graphs show pairs

of straight lines whose slopes are proportional to the cross sections

Q_1 and Q_2 and the constancy of the results is indicated by the virtually

complete collinearity of the points.

Table VII shows the collision cross sections Q_1 and Q_2

together with the experimental and theoretical values quoted by other

authors. The figure of 5 per cent quoted for the accuracy of the cross

sections is supported by the agreement within 5 per cent between all

the ratios Q_1/Q_2 and the theoretically predicted value. In the calcula-

tion of the cross section τ was taken as 1.63×10^{-8} sec, (Kibble,

Copley and Krause, 1967) and the temperature used in the expression for

v_r was the main cell temperature, $124^{\circ}C$. The Na - A cross sections

calculated by Seiwert (1956) from Lochte-Holtgreven's experimental

data are in adequate agreement with the results of this investigation,

as are Jordan and Franken's (1966) results, with the exception of the

Na - Kr and Na - Xe cross sections.

The cross sections $Q_2 (^2P_{1/2} \longleftarrow {}^2P_{3/2})$ whose magnitudes,

unlike those of the cross sections $Q_1 (^2P_{1/2} \longrightarrow {}^2P_{3/2})$ are not influenced

by the thermal energy distribution, do not exhibit any predictable

variation over the range of the noble gases. The collisional processes

in sodium are non-adiabatic (Massey and Burhop, 1952) and thus differ

in character from the corresponding processes in K; Rb and Cs. This

might well account for the absence of the correlation between the cross

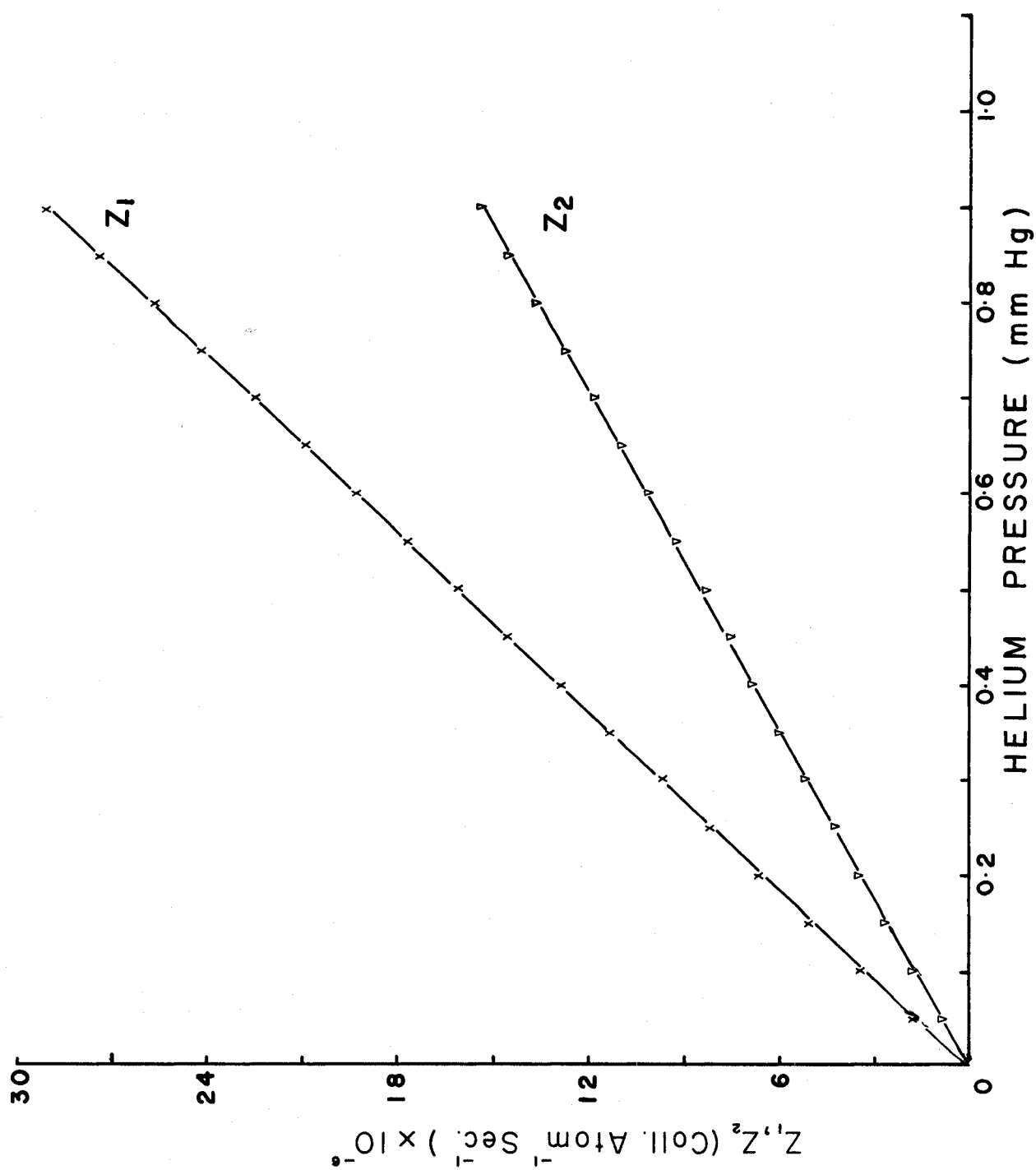section Q_2 and the elastic cross sections for collisions between electrons

Fig. 9.   The Variation of the Sodium-Helium Collision Numbers with Helium Pressure
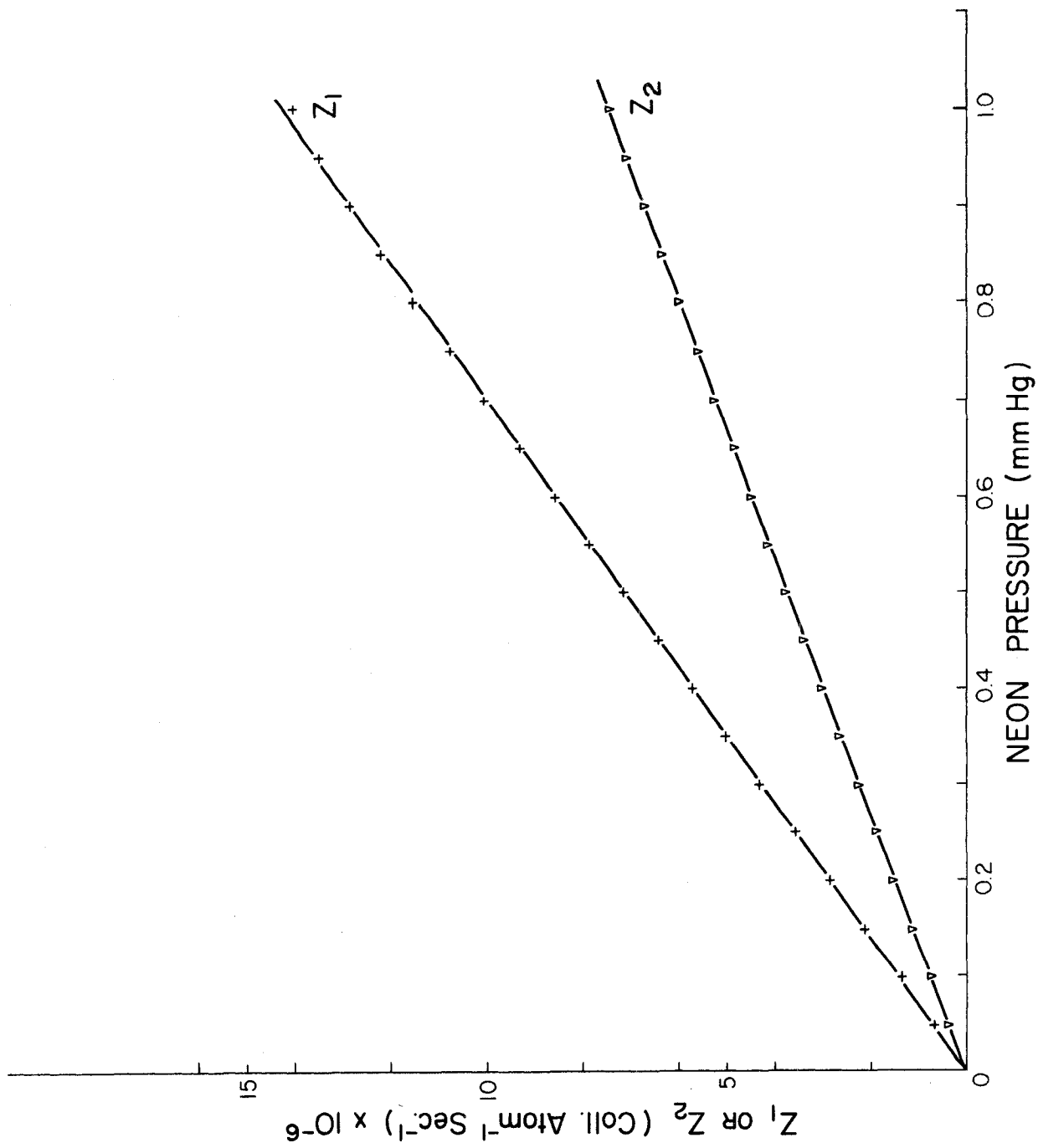
Fig. 10. The Variation of the Sodium-Neon Collision Numbers with Neon Pressure
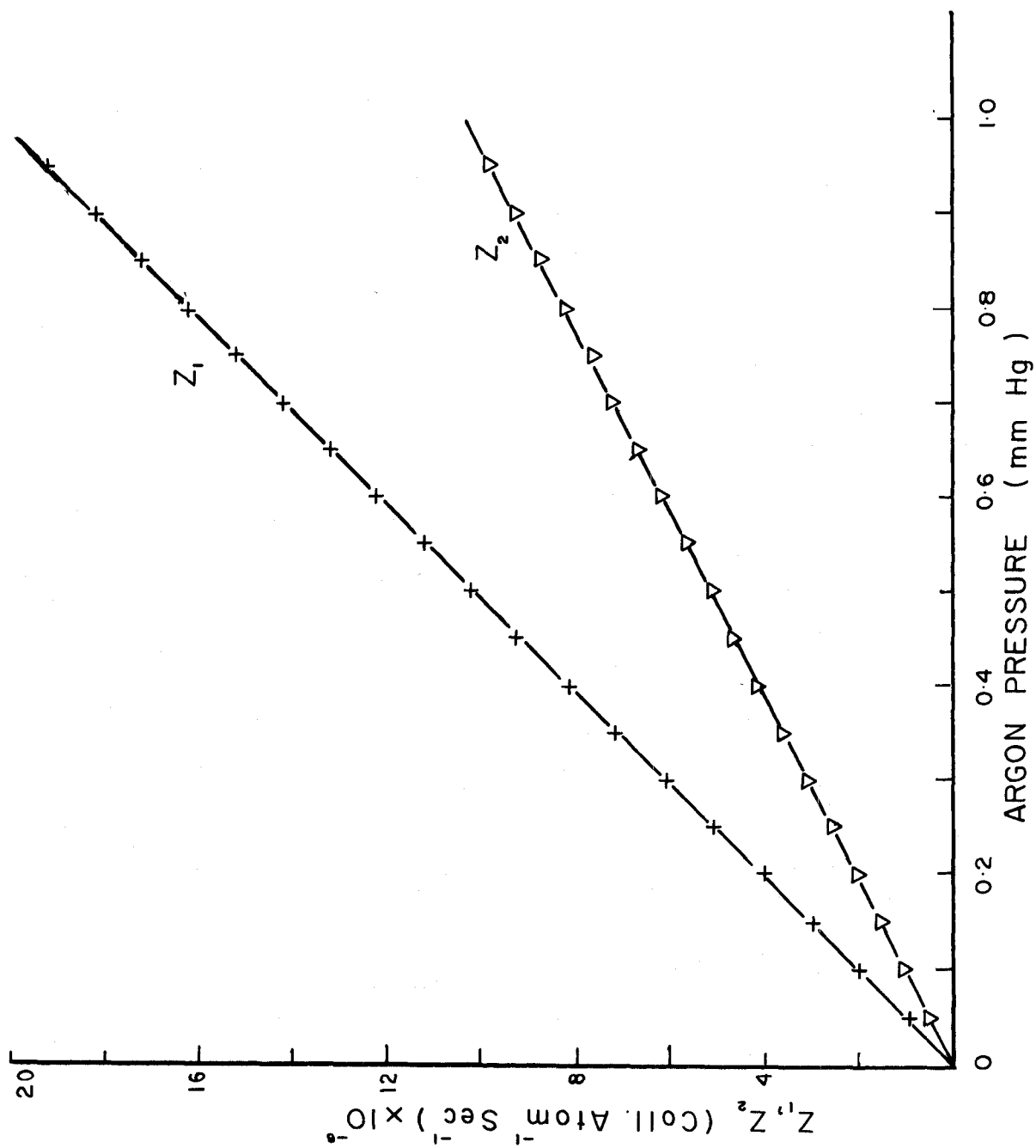
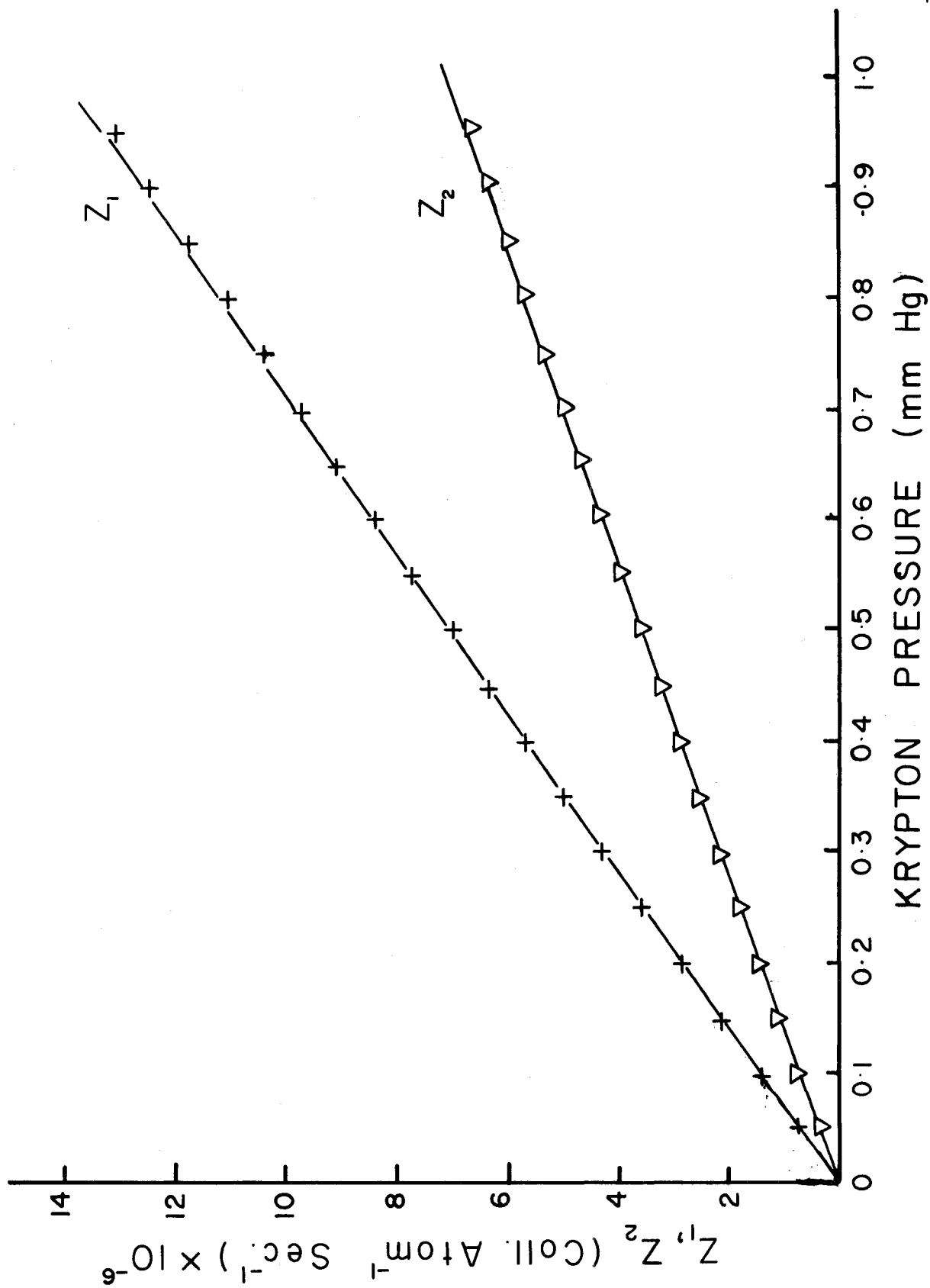Fig. 11. The Variation of the Sodium-Argon Collision Numbers with Argon Pressure

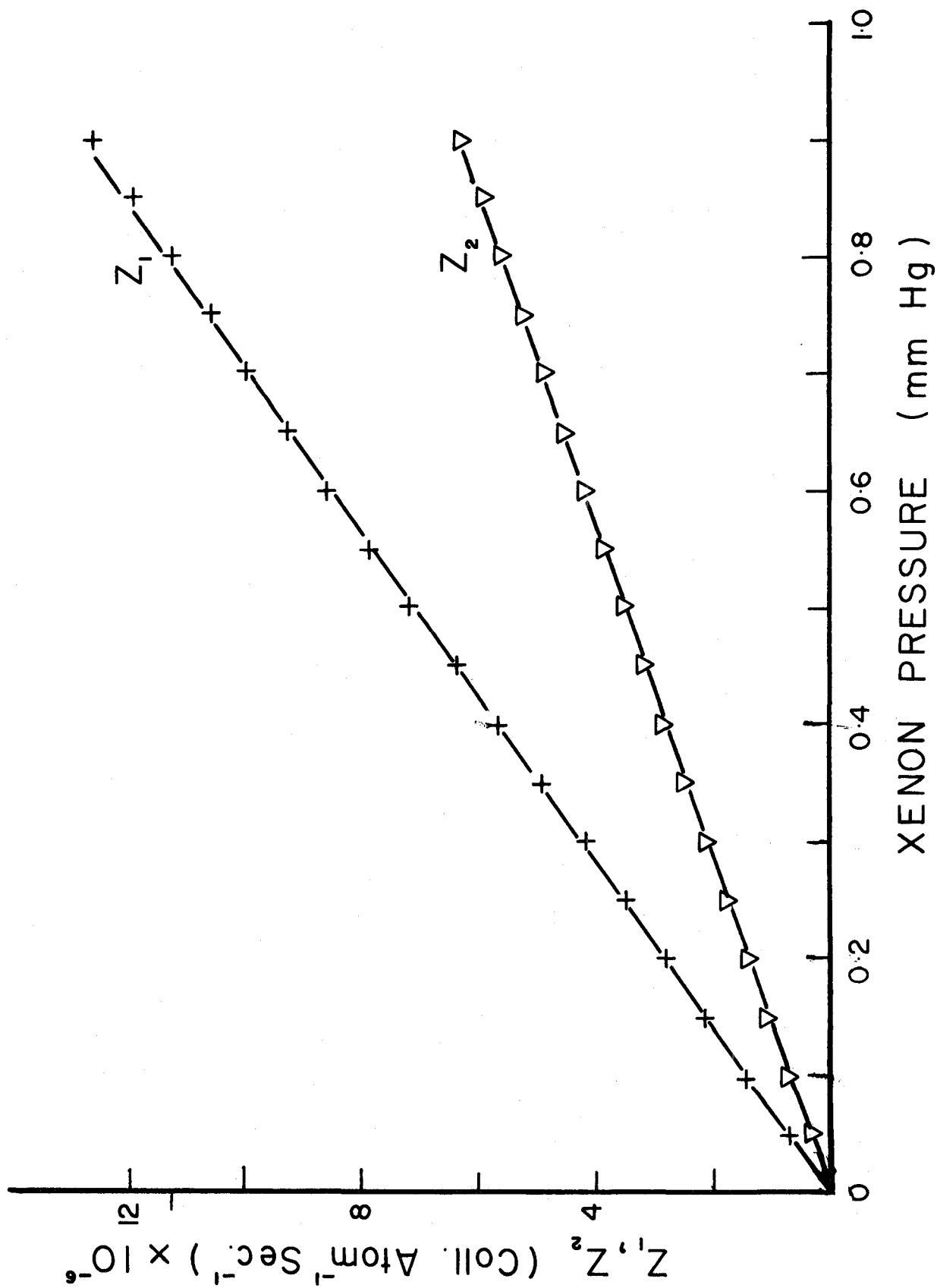Fig. 12. The Variation of the Sodium–Krypton Collision Numbers with Krypton Pressure

Fig. 13. The Variation of the Sodium-Xenon Collision Numbers with Xenon Pressure

TABLE VII

Cross Sections for $3^2P_{1/2} \rightarrow 3^2P_{3/2}$ Mixing Induced by Sodium-Noble Gas Atomic Collisions

Collision Partners	Designation Q_1 or Q_2	This Investigation			Seiwert (1956)	Jordan and Franken (1966)		Callaway and Bauer (1965)	Moskowitz and Thorson (1963)	Nikiti (1965)
		Q_1 or Q_2 ($Å^2$)	Q_1/Q_2	$2e^{-\Delta E/kT}$		Experimental	Theoretical			
Na - He	Q_1	86.0	1.92	1.88				44	3.1	
Na - He	Q_2	44.8				41.1	35.2	22	66	
Na - Ne	Q_1	67.0	1.91	1.88						
Na - Ne	Q_2	35.4				36.1	46.0			
Na - A	Q_1	109.9	1.97	1.88	111			132		45-120
Na - A	Q_2	55.9			66	54.6	69.9	66		
Na - Kr	Q_1	85.0	1.95	1.88						
Na - Kr	Q_2	43.6				63.2	74.0			
Na - Xe	Q_1	89.8	1.97	1.88						
Na - Xe	Q_2	45.6				61.8	82.6			

and noble gas atoms, which was observed in all the other cases (Krause, 1966). On the other hand, there is also no proportional relation, predicted by the various theories, between the cross sections Q_2 and the noble gas polarizabilities which increase monotonically from He to Xe. Such proportionality, which was also found lacking in the cases of the other alkali atoms (Krause, 1966), would be expected on the basis of an electrostatic van der Waals interaction or of a 'quadrupole-induced dipole' interaction (Jordan, 1964). The various theoretically calculated cross sections all agree as to order of magnitude with the experimental values except for Moskowitz and Thorson's (1963) Na - He results.

B. The Experiment with Pure Sodium

In the experiment with pure sodium the η values were found to be very small and thus Q_2 could be obtained directly from η_1 by the use of Eq. (11). From Eqs. (11) and (13) it would be expected that, in the absence of radiation trapping, a plot of η_1 against atomic density should be a straight line passing through the origin and having a slope proportional to the cross section $Q_2(^2P_{1/2} \longleftarrow {}^2P_{3/2})$. The experimental η_1 values, given in Table VIII and plotted in Fig. 14 against sodium density, show such a linear relationship although the straight line has an intercept of $\eta_1 = 3 \times 10^{-6}$. This intercept arises from an incomplete rejection, by the three interference filters in the incident beam, of the D_1 component emitted by the spectral lamp (Rae and Krause, 1965). Since the Spectrolab specifications give a transmission for the unwanted component of about one per cent of peak

TABLE VIII

Fluorescent Intensity Ratios for the Pure Sodium Investigation

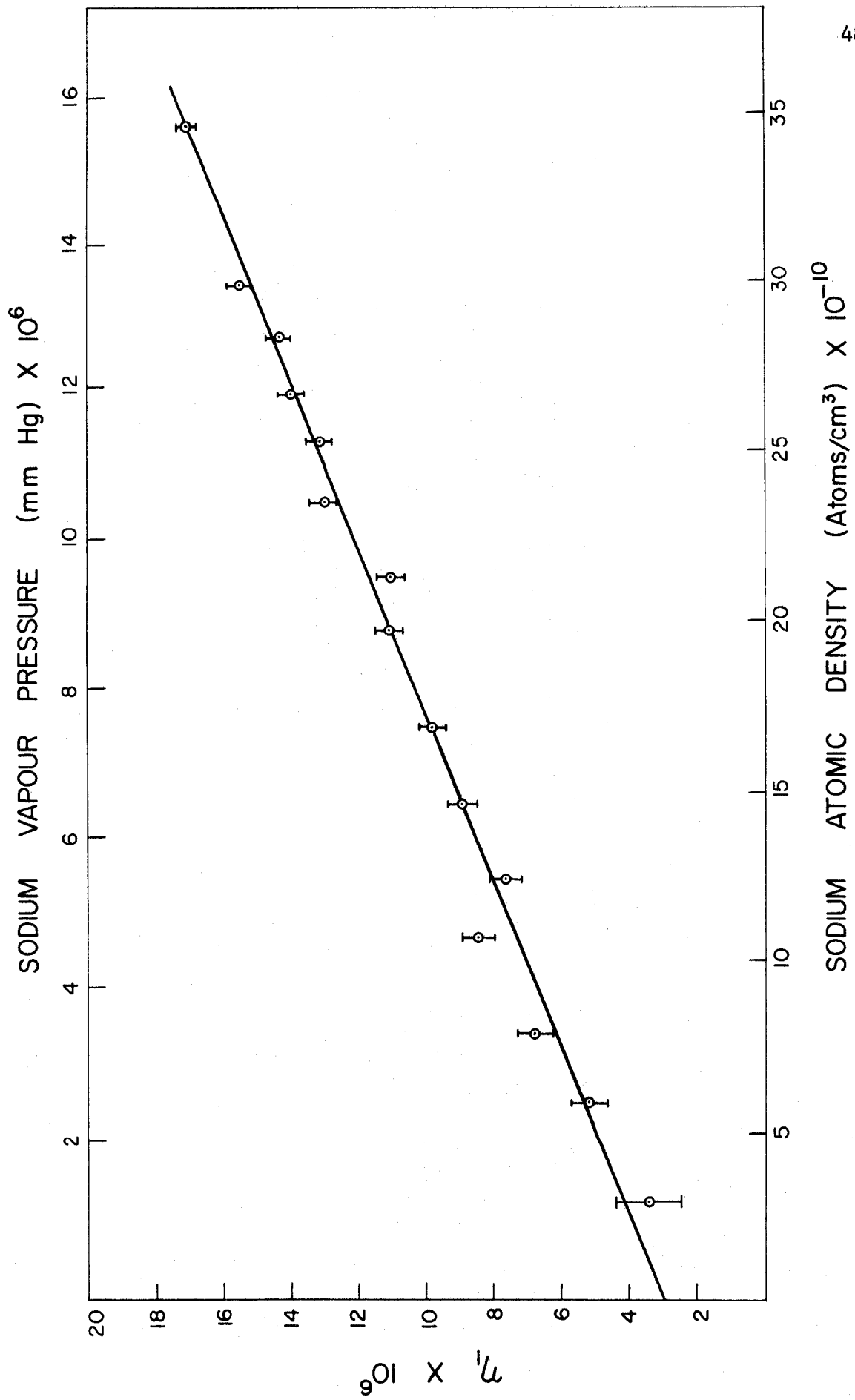| Side Oven Temperature ($^{\circ}$C)	Sodium Vapour Pressure ($\times 10^{-6}$ mm Hg)	Sodium Atomic Density ($\times 10^{10}$ Atoms/cm^3)	η_1 D_1/D_2 ($\times 10^{-6}$)
126.2	1.20	2.89	3.41
135.6	2.48	5.84	5.20
139.7	3.36	7.84	6.81
144.1	4.63	10.69	8.44
147.2	5.42	12.42	7.64
148.7	6.41	14.64	8.96
150.8	7.43	16.88	9.87
153.2	8.74	19.75	11.09
154.3	9.46	21.32	11.05
155.8	10.48	23.53	12.99
156.9	11.29	25.29	13.12
157.7	11.92	26.65	14.00
158.7	12.70	28.33	14.31
159.6	13.43	29.89	15.50
161.6	15.56	34.47	16.91

Fig. 14. The Variation of the Fluorescent Intensity Ratio η_1 with Sodium Atomic Density

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transmission it is to be expected that three filters in series will yield an intercept of the order of 1×10^{-6} which is in good agreement with the observed value. Although there is a contribution to the intercept arising from a small leakage of resonance fluorescence through the filters used in the fluorescent beam for measurements of sensitized fluorescence, this contribution is expected to be negligibly small, since, in this case, four filters in series were employed. In the absence or radiation trapping, both effects are independent of vapour density and do not, therefore, affect the slope of the straight line.

The onset of radiation trapping which depends on the geometry of the system and which would cause the η_1 curve to curve upwards, occurs at or just beyond the upper end of the range of atomic densities of this experiment (Rae and Krause, 1965; Czajkowski and Krause, 1965).

The experimental points in Fig. 14 were taken from three separate experimental runs, two with increasing and one with decreasing sodium vapour pressure. Between the two former runs, the main cell was baked while the side arm was held at a low temperature as a precaution against possible effects of condensation in the main cell. The reproducibility of the results showed such effects to be absent. The statistical uncertainties in the individual experimental points are shown by the error bars and equalled about $\pm$ 3 per cent over most of the experimental range.

A least squares analysis yielded a cross section of $Q_2 = 283 \ \overset{\circ}{A}^2$ and although the statistical scatter of the points is small, it is estimated that this value is correct to $\pm$ 10 per cent if possible sources

of systematic error in the determination of the vapour pressure are taken into consideration. In the calculation of the cross section the temperature employed in the expression for v_r was $151^\circ C$ which corresponded to the atomic density of the middle of the experimental range. Although the cross section Q_1 was not measured, it may be obtained from Eq. (16) since the ratios of the cross sections have been found to obey this relation for potassium, rubidium and cesium pure metal investigations, (Krause, 1966) and also for the sodium-noble gas experiments described here.

The value of the cross section Q_2 obtained in this investigation is about one third of the rough estimate of 800 $Å^2$ made by Krause (1966), who obtained this value from an extrapolation of a plot of the cross sections for K, Rb, and Cs against their fine structure splittings ΔE. However, since this estimate takes into account only the effect of ΔE on the cross sections and since for sodium ΔE is so small that the collisions are non-adiabatic, the cross section obtained in this investigation may not fit into a scheme valid for cross sections for the adiabatic collision processes of K, Rb and Cs.

The only other experimental result available for comparison is Seiwert's (1956) value of $Q_2 = 100$ $Å^2$ for a temperature of $287^\circ C$. It is not surprising that the agreement between the two cross sections is not good since Seiwert worked in a vapour pressure region where radiation trapping is pronounced and corrected his results for imprisonment of radiation using a modified Holstein (1947) theory. The merits of such corrections and the extent of the apparent increase in lifetime of the ^2P states is discussed by Kibble, Copley and Krause (1967a).

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VITA AUCTORIS

I was born on July 15, 1941, in Windsor, Ontario. In 1958 I entered the novitiate of the Brothers of the Christian Schools at Scarborough, Ontario, and two years later I completed my high school education at Christian Brothers College, Scarborough. In 1960 I enrolled at Assumption University of Windsor (became the University of Windsor in 1963) with an entrance scholarship which I maintained until my graduation in Honours Physics in 1964. I then entered the Faculty of Graduate Studies and obtained the Master of Science degree in Physics in 1965. I have been aided in my post graduate studies by a National Research Council graduate bursary and an N.R.C. studentship.

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