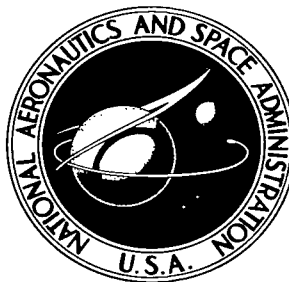


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# THERMAL CONDUCTIVITIES OF ORDINARY AND DEUTERATED GASEOUS HYDROGEN FLUORIDE AND THEIR EQUIMOLAR MIXTURE

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*Cleveland, Ohio*





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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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# THERMAL CONDUCTIVITIES OF ORDINARY AND DEUTERATED GASEOUS HYDROGEN FLUORIDE AND THEIR EQUIMOLAR MIXTURE\*

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## SUMMARY

Thermal-conductivity measurements were carried out at 373.8° and 422.3° K with an eight-cell hot-wire thermal-conductivity apparatus constructed of Monel. Lower temperatures were precluded because of the complication of the polymerization reaction. The thermal conductivity of hydrogen fluoride is higher than that of deuterium fluoride at both temperatures; the conductivity of the equimolar mixture is slightly greater than that of either pure component. The experimental results are analyzed to test further a recent hypothesis that a resonant exchange of rotational quanta causes the thermal conductivity of a polar gas to seem anomalously low in relation to its viscosity. These results corroborate the findings of our previous investigation on polar gases; that is, they are consistent with this hypothesis, the only qualification being that near-resonant exchange is also important.

## INTRODUCTION

The thermal conductivities of highly polar gases such as hydrogen fluoride, water, and ammonia seem anomalously low in relation to their viscosities. Mason and Monchick (ref. 1) have suggested that this effect is largely a result of a resonant exchange of rotational energy, presumed probable on grazing collisions of like polar molecules. Hence, a grazing collision with exchange is equivalent to a head-on collision without exchange, insofar as the transport of the rotational quantum is concerned. Baker and Brokaw (refs. 2 and 3) recently measured the thermal conductivities of several polar gases and their deuterated counterparts, as well as the equimolar mixture for each pair,

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\* A condensed version entitled "Thermal Conductivities of Gaseous HF, DF, and the Equimolar HF-DF Mixture" was published in the Journal of Chemical Physics, vol. 46, April 1967.

in an effort to test experimentally the theory of Mason and Monchick. A polar gas and its deuterated counterpart should be a good test of the theory (ref. 2) because the substitution of a deuterium atom for a hydrogen atom increases the moment of inertia by a factor of approximately two but changes the mass only slightly. Since the resonant correction depends inversely on the  $3/2$  power of the moment of inertia (ref. 1), the resonant correction for the deuterated gas is approximately one-third that of the ordinary gas.

The experimental results previously reported by Baker and Brokaw (refs. 2 and 3) are consistent with the notion that a resonant, or near-resonant exchange of rotational quanta is important in determining the thermal conductivity of polar gases. However, in each of these systems analysis was complicated either by exchange reactions which resulted in multicomponent systems (e. g. , the water system contains HDO as well as  $H_2O$  and  $D_2O$ ) or because the difference between the resonant effect for the gas pairs ( $N^{15}H_3 - NH_3$ ) was small. The hydrogen fluoride - deuterium fluoride (HF-DF) system overcomes both these problems. Because there is only a single natural isotope for the fluorine atom  $F^{19}$ , there is no possibility of a multicomponent system. In addition, hydrogen fluoride should exhibit the largest resonant effect of any polar gas because of its large dipole moment and small moment of inertia (ref. 1). Finally, hydrogen fluoride is a simple linear dipole, and resonant theory is on firmer ground for this class of molecules than for more complicated polar gases (such as asymmetric-top molecules).

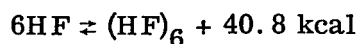
Because of these reasons, the experiments reported herein were undertaken as a more stringent test of the Mason and Monchick equations. Conductivities of HF, DF, and their equimolar mixture were measured at  $373.8^{\circ}$  and  $422.3^{\circ}$  K.

## EXPERIMENTAL

### Apparatus and Procedure

The hot-wire thermal-conductivity apparatus used for these conductivity measurements is essentially the same as the apparatus used previously and described in reference 3. However, because of the corrosive nature of HF, the platinum-iridium filaments were mounted in a Monel block. The gas-handling system was also constructed of Monel.

The thermal conductivity of hydrogen fluoride vapor is markedly dependent on the pressure (ref. 4). Long, Hildebrand, and Morell (ref. 5) studied pressure-volume-temperature data on hydrogen fluoride vapor and concluded that the experimental results could be well described by assuming the vapor to be an equilibrium mixture of monomer and hexamer:



Franck and Spalthoff (ref. 4) suggested that this reaction is responsible for the high thermal conductivities which they observed. Butler and Brokaw (ref. 6) have treated this problem theoretically and have shown that the aforementioned polymerization reaction does indeed account for the observed conductivities. To avoid the complications of this polymerization reaction, all measurements were made at elevated temperatures ( $>373^\circ \text{K}$ ) and at a reduced pressure (100 mm Hg). Under these conditions the polymerization increases the thermal conductivity of HF by less than 2 percent.

A slight drift in the electromotive force readings was noted during the experimental measurements. This amounted to an increase in thermal conductivity of about 0.1 percent during a 20-minute period. This steady drift was apparently due to a very slight reaction of the hydrogen fluoride with the walls of the cell which liberated hydrogen; hence, the thermal conductivity increased. To correct for this drift the electromotive force data were taken as a function of time and then extrapolated back to the time that gas was introduced into the cells. However, at  $473^\circ \text{K}$  the drift had increased to such an extent that the measurements were not considered precise enough for an investigation which involves small differences in thermal conductivity. Thus, no measurements are reported above  $422.3^\circ \text{K}$ .

As was the case in the earlier work (refs. 2 and 3), nitrogen was generally used as the reference gas, and helium and argon were used as the calibrating gases. However, DF and the equimolar HF-DF mixture were measured against HF. Thus, these small electromotive forces were measured with enhanced precision.

Potentiometer readings relative to argon are recorded in table I. These experimental measurements were reproducible to better than  $\pm 0.04$  millivolt. An additional significant figure is retained in these readings for use in the thermal conductivity calculations. Except for the calibrating gases, the values listed in table I are the average of at least four measurements. Because helium and argon are monatomic gases, it is possible to compute conductivity from viscosity through a rigorous, experimentally verified, theoretical relation to less than a few parts per thousand. Thus, thermal conductivities for the calibrating gases were assigned after considering both experimental viscosity and thermal-conductivity data. The details of this method have been fully described previously (ref. 2). The conductivities selected for calibration are given in table II.

## Experimental Results

The thermal conductivities derived from tables I and II by the method of reference 2

TABLE I. - EXPERIMENTAL ELECTRO-  
MOTIVE FORCE MEASUREMENTS  
RELATIVE TO ARGON

Gas	Temperature, °K	
	373.8	422.3
	Electromotive force, mV	
Helium	44.879	41.163
Nitrogen	15.306	13.957
Hydrogen fluoride	8.699	9.771
Deuterium fluoride	8.512	9.560
Hydrogen fluoride mixture (0.502 HF - 0.498 DF)	8.806	9.795

TABLE II. - THERMAL CONDUCTIVITIES OF HELIUM AND ARGON USED FOR CALIBRATION PURPOSES<sup>a</sup>

Temperature, °K	Thermal conductivity, $\mu\text{cal}/(\text{cm})(\text{sec})(^\circ\text{K})$	
	Helium	Argon
373.8	431.4	50.41
422.3	470.8	55.44

<sup>a</sup>Ref. 2.

TABLE III. - EXPERIMENTAL THERMAL CONDUCTIVITIES

Gas	Temperature, °K	
	373.8	422.3
	Thermal conductivity, $\mu\text{cal}/(\text{cm})(\text{sec})(^\circ\text{K})$	
Hydrogen fluoride	60.82	70.13
Deuterium fluoride	60.56	69.73
Hydrogen fluoride mixture (0.502 HF - 0.498 DF)	60.98	70.17
Nitrogen	72.14	79.10

are presented in table III. Although the primary purpose of this work is to measure accurately the difference in thermal conductivity of hydrogen fluoride and deuterium fluoride (and of HF and the HF-DF mixture), the absolute values obtained are in satisfactory agreement with values reported in the literature. For example, the nitrogen conductivities agree with values interpolated from the data of Vargaftik and Oleshchuk (ref. 7) to within 1 percent and are in even better agreement with the more recent results of Grier and Schäfer (ref. 8). The measurements of Franck and Spalthoff already mentioned (ref. 4) are the only conductivities for hydrogen fluoride vapor that have been previously reported; there do not appear to be any conductivity data for deuterium fluoride. The results listed in table III are about 4 percent higher than those given in reference 4. This degree of discrepancy is not uncommon for conductivity measurements, even among data for inert gases. In view of the special problems connected with hydrogen fluoride, this agreement seems satisfactory.

## ANALYSIS AND DISCUSSION

The expression that Mason and Monchick (ref. 1) derived for the thermal conductivity of a polyatomic gas may be written as follows:

$$\lambda = \frac{\eta}{M} \frac{5}{2} \left[ C_{v,trans} + \frac{\rho D_{int}}{\eta} C_{int} - \frac{2}{\pi} \left( \frac{5}{2} - \frac{\rho D_{int}}{\eta} \right)^2 \sum \frac{C_k}{Z_k} \right] \quad (2)$$

where  $\lambda$  is thermal conductivity,  $\eta$  is viscosity,  $M$  is molecular weight,  $\rho$  is density,  $D_{int}$  is the average coefficient for the diffusion of internal energy,  $C_{v,trans}$  and  $C_{int}$  are the translational and the internal contributions to the molar heat capacity,  $Z_k$  is the number of collisions for relaxation of the  $k^{th}$  internal mode, and  $C_k$  is the heat capacity associated with that mode. In small rigid polyatomic molecules, the only collision numbers small enough to affect thermal conductivity significantly are those associated with rotational relaxation. For nonpolar gases,  $D_{int}$  is commonly assumed to be equal to the self-diffusion coefficient  $D_{11}$ . For polar gases, however, Mason and Monchick (ref. 1) suggest that

$$\eta_{int} = \frac{D_{11}}{1 + \delta} \quad (3)$$

where  $\delta$  is a correction term calculated from the theory of resonant collisions. Mason and Monchick (ref. 1) have developed expressions from which the resonant correction

for linear dipoles and symmetric tops can be computed. The expression used in the analysis of the linear dipoles HF and DF is as follows:

$$\delta = \langle a_4 \rangle \left( \frac{3}{16} \frac{\mu h}{\sigma_D kT} \right)^2 \frac{m^{1/2}}{\left( \frac{C_{\text{int}}}{R} \right)^{3/2}} \quad (4)$$

Here  $\langle a_4 \rangle$ , taken to be 0.44, is the mean value of a dimensionless quantity involving the rotational quantum numbers,  $\mu$  is the dipole moment,  $h$  is Planck's constant,  $\sigma_D$  is the kinetic-theory diameter characteristic of diffusion ( $\sigma_D \equiv \sigma [\Omega(1,1)^*]^{1/2}$  in the notation of ref. 9),  $k$  is the Boltzmann constant,  $m$  is the molecular mass,  $R$  is the universal gas constant, and  $I$  is the molecular moment of inertia.

### Pure Gases

The experimental data on the pure gases were analyzed as follows: The necessary resonant corrections were calculated from equation (4) and the results were used to compute  $D_{\text{int}}$  from equation (3). The collision number  $Z_{\text{rot}}$  was then calculated from equation (2) after  $\lambda$  had been replaced by the experimentally determined thermal conductivity. The ratio of the collision number of HF to that of DF was then compared with the theoretical results of Sather and Dahler for a rough sphere (ref. 10) as modified in reference 11:

$$Z_{\text{rot}}^{-1} \propto \frac{\frac{4I}{m\sigma_\eta^2}}{\left( 1 + \frac{4I}{m\sigma^2} \right)^2} \quad (6)$$

Here,  $m$  and  $I$  are the same as in equation (4), while  $\sigma_\eta$  is the viscosity collision diameter ( $\sigma_\eta \equiv \sigma [\Omega(2,2)^*]^{1/2}$  in the notation of ref. 9), and  $\sigma$  is the diameter of the rough sphere core. The rough sphere model was chosen because the experimental results of O'Neal and Brokaw (ref. 11) for a variety of near-spherical molecules (including methane and even ethane) suggest that this model is a reasonable one for substances that do not differ markedly from spherical symmetry.

Some temperature-independent properties necessary for the data analysis are listed in table IV. The values listed for the dipole moments deserve some comment. Until



TABLE IV. - MOLECULAR PROPERTIES OF HYDROGEN  
FLUORIDE AND DEUTERIUM FLUORIDE

Property	Hydrogen fluoride	Deuterium fluoride
<sup>a</sup> Moment of inertia, g-cm <sup>2</sup>	1.3356×10 <sup>-40</sup>	2.5414×10 <sup>-40</sup>
Molecular weight	20.0080	21.0142
Dipole moment, debyes	<sup>b</sup> 1.83	<sup>c</sup> 1.837

<sup>a</sup>Refs. 18 and 19.

<sup>b</sup>Refs. 14 to 16.

<sup>c</sup>Calculated from HF data on basis of reduced mass.

TABLE V. - DATA ANALYSIS FOR PURE HYDROGEN  
FLUORIDE AND DEUTERIUM FLUORIDE

	Temperature, °K	
	373.8	422.3
Viscosity of hydrogen fluoride, micropoises	158.2	180.0
$C_{int}/R$		
Hydrogen fluoride	1.00548	1.00634
Deuterium fluoride	1.00698	1.01091
$\rho D_{11}/\eta (= 6/5 \langle A^* \rangle)$	1.3277	1.3328
Collision number, $Z_{rot}$		
Hydrogen fluoride	9.5	9.1
Deuterium fluoride	4.1	3.7
$Z_{rot, DF}/Z_{rot, HF}$	0.44	0.41
<sup>a</sup> $(Z_{rot, DF}/Z_{rot, HF})_{cal}$	0.58	0.58
Resonant correction, $\delta$		
Hydrogen fluoride	2.22	1.88
Deuterium fluoride	0.91	0.76
<sup>b</sup> Hydrogen chloride	0.34	0.28

<sup>a</sup>Calculated from eq. (6).

<sup>b</sup>From ref. 3.

recently a value of 1.91 debyes was generally accepted as the dipole moment for hydrogen fluoride. This value was obtained by Hannay and Smyth (ref. 12) and Oriani and Smyth (ref. 13) about two decades ago by measuring the dielectric constant of the hydrogen fluoride vapor. However, recent determinations of the dipole moment from isotopic variations of the rotational magnetic moment (ref. 14) and by the molecular beam electric resonance method (ref. 15), as well as by a molecular Hartree-Fock calculation (ref. 16), indicate a value of about 1.83 debyes. The dipole moment for deuterium fluoride was calculated from this value for hydrogen fluoride by assuming that the ratio of  $\mu_{DF}/\mu_{HF}$  to the ratio of reduced masses of DF and HF is equal to this same ratio for the HCl-DCl system.

Certain temperature-dependent quantities also needed for the data analysis are shown in table V. The viscosity values for hydrogen fluoride were obtained from a short linear extrapolation of the data of Posey (ref. 17). Unfortunately, these data appear to be the only viscosity measurements for hydrogen fluoride that have been reported. Viscosities of DF were calculated from the HF viscosities. Since there were no data available that compared the viscosities of the isotopic pairs, the viscosity collision cross sections for HF and DF were assumed to be the same. (This assumption was also made in the earlier work with HCl-DCl and  $NH_3 - ND_3$ , ref. 3.)

The heat-capacity data given in table V were calculated from spectroscopic constants (refs. 18 and 19) by a computer program described by McBride, Heimerl, Ehlers, and Gordon (ref. 20). The dimensionless quantity  $\rho D_{11}/\eta$  that involves the self-diffusion coefficient was computed as  $(6/5)\langle A^* \rangle$  (ref. 9, p. 540). The quantity  $\langle A^* \rangle$  is a ratio of collision integrals (essentially the ratio of the viscosity to the diffusion cross sections) and is insensitive to temperature and the details of the intermolecular force law, at least for spherically symmetric potentials. Since there is no experimental information on the self-diffusion of hydrogen fluoride,  $\langle A^* \rangle$  was taken from calculations for a modified (angle independent) Stockmayer potential tabulated in reference 21. The required potential parameters were obtained by fitting the viscosity data ( $\sigma = 2.394$ ,  $\epsilon/k = 400$ , and  $\delta_{max} = 2.0$ ). The determination of  $\langle A^* \rangle$  is the only place in the analysis where it is necessary to make any assumption about the intermolecular force law.

Rotational relaxation collision numbers obtained from the experimental conductivities and resonant corrections calculated from equation (4)<sup>1</sup> are given in table V. (To emphasize the magnitude of these resonant corrections, the values for hydrogen chloride are also listed.) There are no collision numbers for HF obtained by other

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<sup>1</sup>The derivation of equation (4) involves, among other approximations, the replacement of summations by integrations. Resonant corrections obtained from this expression are about 8 percent too large for HF and 4 percent too large for DF. The  $\delta$  values shown in table V have been corrected for this effect.

experimental techniques with which to compare these values. Nonetheless, these collision numbers appear somewhat high when compared with the 3.6 and 3.2 collisions obtained for HCl at these temperatures (ref. 3). The larger dipole moment for HF should result in a smaller  $Z_{\text{rot}}$ ; however, the effect of moment of inertia is in the other direction. On the other hand, if collision numbers are calculated with the assumption that the resonant correction is zero, unrealistic values of less than one collision are obtained. Since the collision number is sensitive to the absolute value of both viscosity and thermal conductivity, errors in these quantities might possibly account for the seemingly high value of  $Z_{\text{rot}}$ . However, the viscosity would have to be more than 10 percent larger or the thermal conductivity more than 10 percent smaller to reduce  $Z_{\text{rot}}$  to the value obtained for HCl. Experimental errors of this magnitude certainly seem unlikely. Ratios of the collision numbers of HF and DF are also given in table V along with the theoretical ratios calculated from equation (6). The agreement is about the same as for the other gas pairs studied (ref. 3) and seems satisfactory in consideration of the sensitivity of this ratio to the ratio of the viscosity collision cross sections. For example, if the viscosity collision cross section for DF were actually 3.5 percent larger than for HF (the value of  $\text{D}_2\text{O}$  is 3 percent larger than for  $\text{H}_2\text{O}$ ), then the agreement would be exact. Moreover, the ratio of the collision numbers calculated with the assumption that the resonant correction is zero is slightly greater than unity at each temperature, clearly in disagreement with all current theories on rotational relaxation.

## Equimolar Mixture

In this section the experimental conductivities are compared with values calculated with the use of the first-order linearized form of the equations of Monchick, Pereira, and Mason (ref. 22) for the thermal conductivity of polyatomic and polar gases. This form of their results can be regarded as equivalent to the Hirschfelder-Eucken formula for the thermal conductivity of polyatomic gas mixtures (refs. 23 and 24) plus a number of first-order correction terms. All the mixture calculations (as well as most of the pure gas calculations mentioned earlier) were performed on a high-speed computer.

In calculating the mixture conductivity it is necessary to know the value of the resonant correction for the various interactions in order to compute the coefficient of diffusion for internal energy (see eq. (3)). For the like interactions, these corrections will be the same as for the pure gases; for the unlike interactions, the situation is more complicated. Of course, if the resonance effect actually required an exact matching of rotational energy differences, the correction for the unlike interactions would be zero. However, previous results with the HCl-DCl equimolar mixture (ref. 3) strongly indicate that near-resonant exchange of rotational quanta does occur. The situation in an

HF-DF mixture should be quite similar. In fact, the same arguments involving rotational energy level differences (discussed in detail for HCl and DCl in ref. 3) were used for HF and DF. Again it was evident that, as a consequence of the moment of inertia of DF being nearly a factor of two greater than that of HF, near-resonant collisions are always possible for an HF molecule interacting with a DF molecule, whereas for DF interacting with HF, near-resonant collisions are possible only half the time. On this basis, the following relations were assumed:

$$\delta_{\text{HF-DF}} = \delta_{\text{HF-HF}} \quad (7)$$

and

$$\delta_{\text{DF-HF}} = \frac{\delta_{\text{DF-DF}}}{2} \quad (8)$$

It is also necessary to know the rotational collision numbers for the unlike interactions in order to make the mixture calculations. Ratios of the collision number for like interaction to that for unlike interaction were calculated on the basis of equation (6). In using equation (6),  $2\gamma_{\text{HF-DF}}$ , where  $\gamma$  is the reduced mass, was substituted for both  $m_{\text{HF}}$  and  $m_{\text{DF}}$ . The following results were obtained:

$$\frac{Z_{\text{HF-HF}}}{Z_{\text{HF-DF}}} = 0.976 \quad (9)$$

and

$$\frac{Z_{\text{DF-DF}}}{Z_{\text{DF-HF}}} = 1.025 \quad (10)$$

The results of the mixture calculations are presented in table VI and are shown graphically in figure 1. The method referred to as "inelastic" uses the equations of reference 22. The Hirschfelder-Eucken calculations, which involve no inelastic effects, are also given in order to show the magnitude of the first-order correction terms. In addition, both these calculations were carried out with the assumption that there was no resonant exchange during unlike collisions, that is,  $\delta_{\text{HF-DF}} = \delta_{\text{DF-HF}} = 0$ . The experimentally determined conductivities are also listed in table VI along with the conductivities calculated with the assumption of a linear mixing rule.

From the figure it is clear that for each method of calculation the conductivities

TABLE VI. - COMPARISON OF EXPERIMENTAL AND CALCULATED VALUES FOR THERMAL CONDUCTIVITY OF EQUIMOLAR HYDROGEN FLUORIDE - DEUTERIUM FLUORIDE MIXTURES

Method	Temperature, °K	
	373.8	422.3
Thermal conductivity, $\mu\text{cal}/(\text{cm})(\text{sec})(^\circ\text{K})$		
Inelastic		
Unlike interaction	61.82	71.21
No unlike interaction	65.42	75.36
Hirschfelder-Eucken		
Unlike interaction	60.89	70.18
No unlike interaction	61.67	71.27
Linear mixing rule, $0.502 \lambda_{\text{exp HF}} + 0.498 \lambda_{\text{exp DF}}$	60.69	69.93
Experimental	60.98	70.17

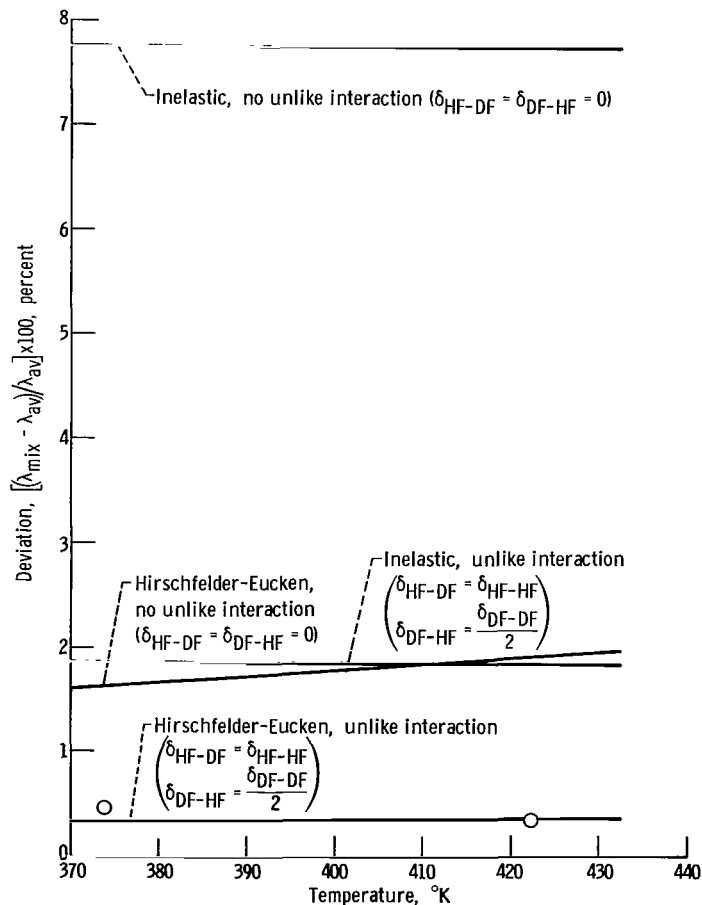


Figure 1. - Deviation of thermal conductivities of equimolar hydrogen fluoride - hydrogen chloride mixtures from linear average. Open circles are experimental results.

obtained when unlike interaction is considered agree much better with experiment than when unlike interaction is neglected. Also, it is obvious that the Hirschfelder-Eucken calculation is superior to the inelastic calculation for this particular mixture. This suggests that the first-order correction terms calculated from the equations of reference 4 are too large. Finally, these results support the conclusion previously reached (ref. 3) that the Hirschfelder-Eucken formula is satisfactory for calculating the thermal conductivities of mixtures of isotopic polar gases provided that inelastic and resonant effects are included in the pure component calculations.

## CONCLUDING REMARKS

The experimental results with both the pure gases and the equimolar mixture are consistent with the notion that a resonant or near-resonant exchange of rotational quanta is important in determining the thermal conductivity of polar gases. This same conclusion was reached in previously reported work with other polar gas systems (HCl-DCl, NH<sub>3</sub>-N<sup>15</sup>H<sub>3</sub>). However, the HF-DF system is a more stringent test of the theory because the resonant effect predicted is so much greater than in the other systems (three times greater than in the HCl-DCl system, for example). Thus, the current results strengthen the concept that resonant or near-resonant exchange of rotational energy plays an important role in determining the thermal conductivity of polar gases.

Lewis Research Center,  
National Aeronautics and Space Administration,  
Cleveland, Ohio, March 30, 1967,  
129-01-02-02-22.

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