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**Vibrational Relaxation of HF($v = 1$ and 3) in H_2 , N_2 , and D_2
at 200 and 295 K**

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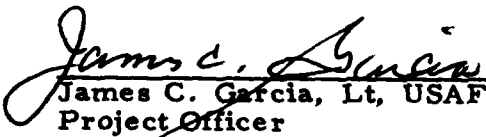
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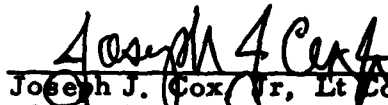
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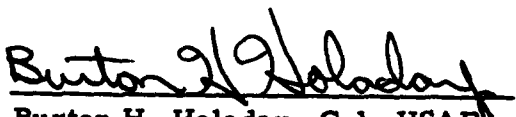
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I. INTRODUCTION

In recent studies,¹⁻⁴ the rate coefficients for the deactivation of the upper vibrational levels of HF and DF were found to scale more strongly with v than the $k_v \propto v$ scaling predicted for harmonic oscillators. The present studies have been performed at reduced temperatures to determine if the v dependence of the HF rate coefficients is sensitive to temperature in the range of 200 to 295 K.

The pumping reaction



directly populates $v = 1, 2$, and 3 , so that the scaling of the $\text{HF}(v)$ deactivation rates with v is important for understanding the detailed performance of the HF chemical laser. In previous studies,^{2,3} the rate coefficients at 295 K were found to scale as v^n with $n = 2.7 \pm 0.2$ for $\text{HF}(v = 1, 2, 3)$ in N_2 , O_2 , and HCl , and $n = 1.9 \pm 0.1$ for $\text{DF}(v = 1-4)$ in H_2 , HF , and N_2 . For comparison, we have measured deactivation rates of $\text{HF}(v = 1 \text{ and } 3)$ in H_2 , D_2 , and N_2 at 200 K.

II. EXPERIMENT

The experiments were performed in the fluorescence cell that was used for a study of H-atom deactivation of $\text{HF}(v = 3)$ and $\text{HF}(v = 2)$.^{5,6} The reports of those studies contain the details of the time-resolved infrared signal processing, the TEA HF pulsed laser, and the cooled fluorescence cell. An RCA C-31034 (GaAs) photomultiplier and an InSb detector were used to monitor the fluorescence of $\text{HF}(v = 3)$ at $0.89 \mu\text{m}$ and that of $\text{HF}(v = 1)$ at 2.5 to $3.0 \mu\text{m}$, respectively. Narrow band filters restricted the fluorescence to a $0.02\text{-}\mu\text{m}$ spectral band centered at $0.89 \mu\text{m}$ for $v = 3$ and to a $0.5\text{-}\mu\text{m}$ spectral band centered at $2.75 \mu\text{m}$ for $v = 1$.

The gases included hydrogen (Matheson 99.95%), nitrogen (Air Products 99.998%), deuterium (Precision Gas Products 98%), helium (Air Products 99.995%), and HF (Matheson 99.9%). The HF was purified by pumping at 77 K to remove the noncondensibles before distillation into a passivated stainless steel cylinder. The HF, diluted 1:10 with helium, was metered into the flow tube-fluorescence cell with a calibrated needle valve. The needle valve and Matheson rotating ball flowmeters used to meter the other gases were calibrated by pressure-rise measurements in a standard volume.

Experiments were performed at various flow rates of the collision partners with the HF-helium flow adjusted to give HF partial pressures of $\sim 1.5 \times 10^{-3}$ Torr. At this partial pressure, the self-deactivation rate of $\text{HF}(v = 1)$ is $1 \times 10^{-4} \mu\text{sec}^{-1}$ at 295 K and $2 \times 10^{-4} \mu\text{sec}^{-1}$ at 200 K.⁷ These rates are small contributions to the measured decay rates of $\text{HF}(v = 1)$. The deactivation rate of $\text{HF}(v = 3)$ by HF is ~ 20 times larger^{1,4} than that of $\text{HF}(v = 1)$ ($\sim 2 \times 10^{-3} \mu\text{sec}^{-1}$ at 295 K and $\sim 4 \times 10^{-3} \mu\text{sec}^{-1}$ at 200 K). The measurements of $\text{HF}(v = 3)$ deactivation rates were obtained for a wide range of quenching pressures in order to minimize the contributions of self-deactivation and diffusion.

III. RESULTS

Single exponential decay rates were determined from the fluorescence traces. These data for HF($v = 1$) and HF($v = 3$) are plotted versus the pressures of H_2 , N_2 , and D_2 in Figs. 1 through 6. The deactivation rate coefficients determined from the slopes of the lines through the data are listed in Table 1. For a discussion of the equations governing the deactivation of the several vibrational levels following laser excitation, see Ref. 2.

A. DEACTIVATION by H_2

The deactivation rates for HF($v = 3$) in H_2 have a small but measurable contribution from diffusion at 1 Torr. When 10 Torr of helium was added to a hydrogen partial pressure of 1.05 Torr, the deactivation rate decreased from 0.023 to $0.016 \mu\text{sec}^{-1}$, a value in agreement with an extrapolation of the higher pressure data of Fig. 2.

The present value of $(1.69 \pm 0.17) \times 10^{-2}$ for the rate coefficient for HF($v = 1$) deactivation by H_2 at 295 K compares favorably with values of $(1.43 \pm 0.15) \times 10^{-2}$ and $(1.7 \pm 0.1) \times 10^{-2} (\mu\text{sec Torr})^{-1}$ obtained previously in this laboratory.^{2,8} Several other literature values are listed in Ref. 8. The present value of $(1.13 \pm 0.12) \times 10^{-2} (\mu\text{sec Torr})^{-1}$ for the HF($v = 3$)- H_2 deactivation rate coefficient at $T = 295$ K is identical to that reported in Ref. 2 and is larger than the value of $(0.90 \pm 0.10) \times 10^{-2} (\mu\text{sec Torr})^{-1}$ obtained at 200 K. For the purpose of comparing rates measured at different temperatures, the probability of collisional deactivation has been calculated using collision diameters of 2.55 Å for HF and 2.9 Å for H_2 . In Fig. 7, these probabilities, P , are plotted versus $1/T$ for comparison with data obtained from laser-induced fluorescence experiments performed behind reflected shock waves at 295 to 1000 K.⁸ The data for HF($v = 1$) may be fitted by the expression $P = 1.57 \times 10^{-3} \exp(-84/T)$. It should be noted that this process is endothermic by 201 cm^{-1} (289 K) so that the probability of the reverse, exothermic process is given by $P = 1.57 \times 10^{-3} \exp(205/T)$.

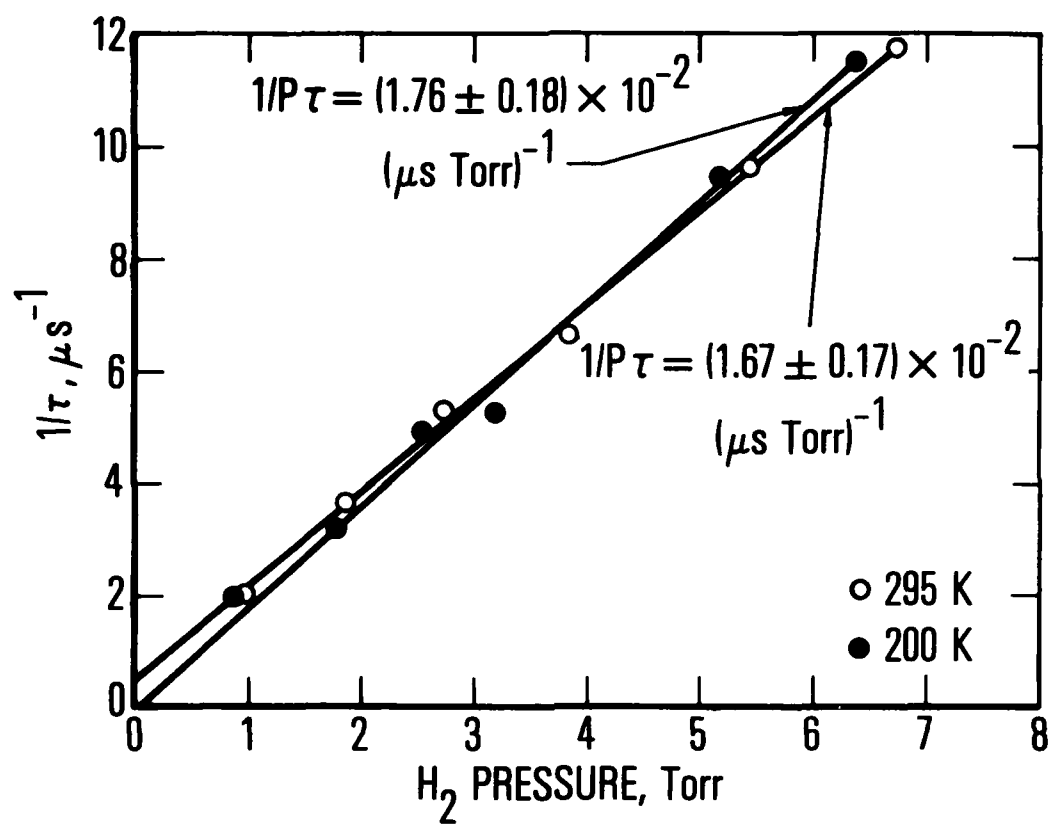


Figure 1. Relaxation Rates of $HF(v = 1)$ versus H_2 Pressure at 200 and 295 K

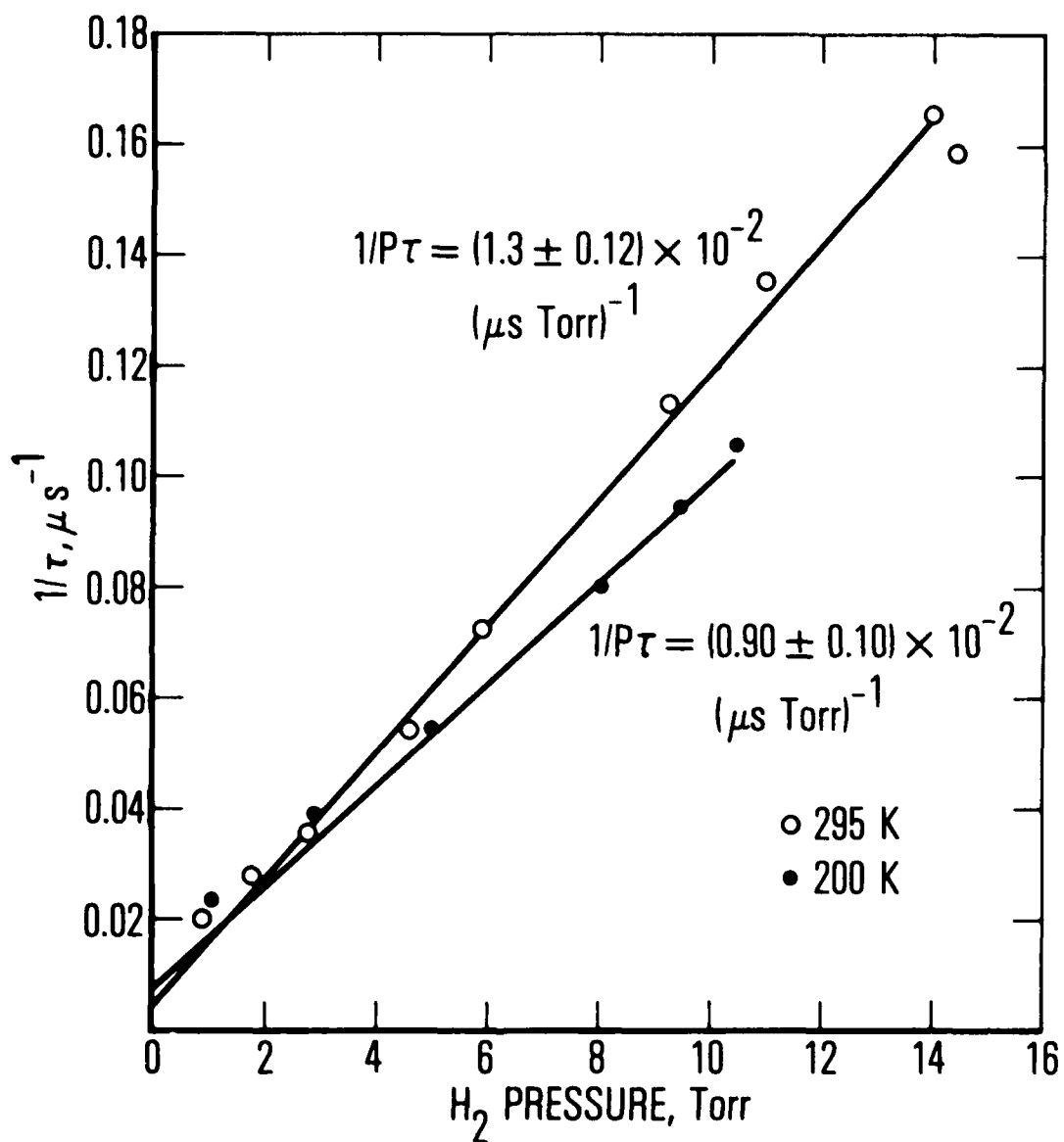


Figure 2. Relaxation Rates of HF($v = 3$) versus H_2 Pressure at 200 and 295 K

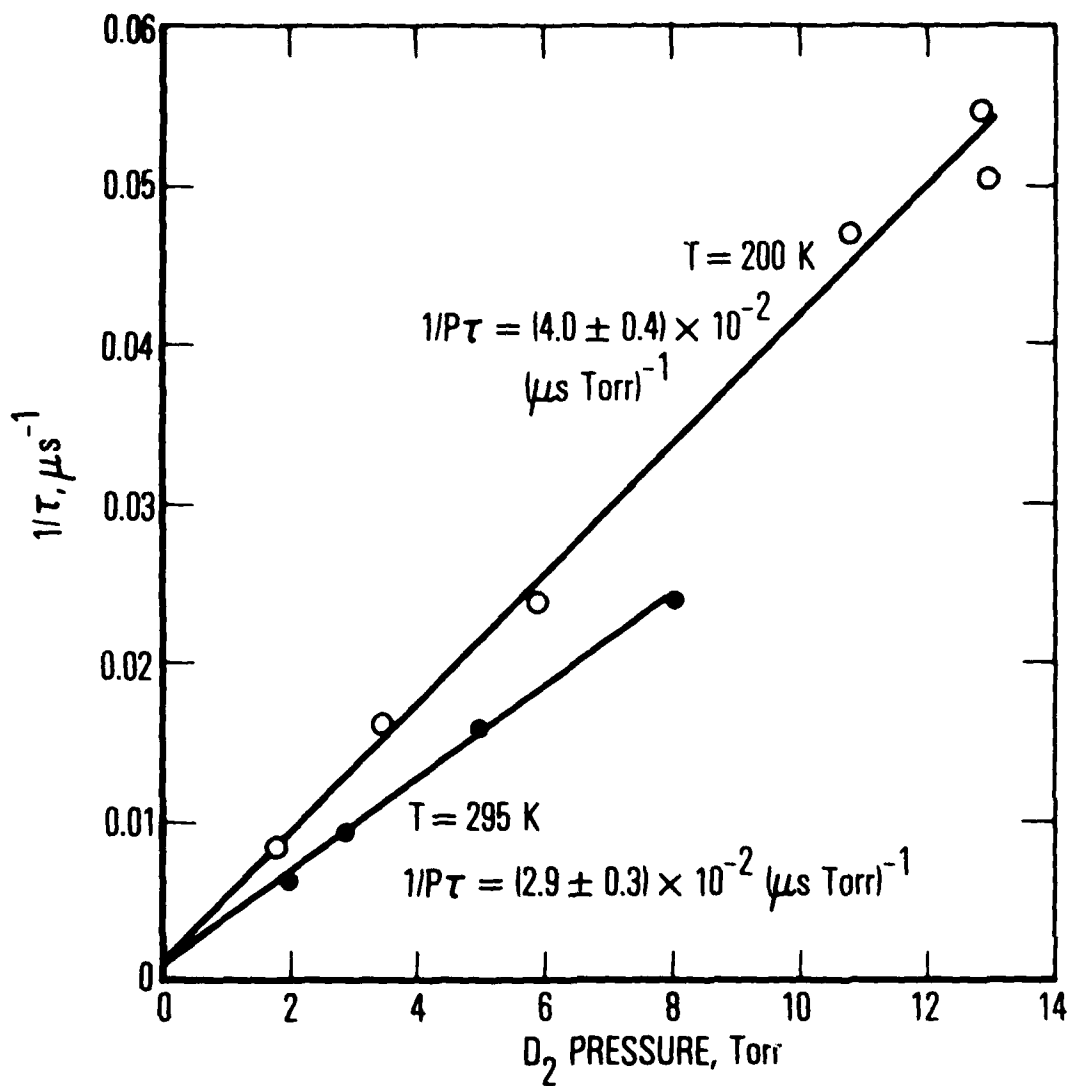


Figure 3. Relaxation Rates of $HF(v = 1)$ versus D_2 Pressure at 200 and 295 K

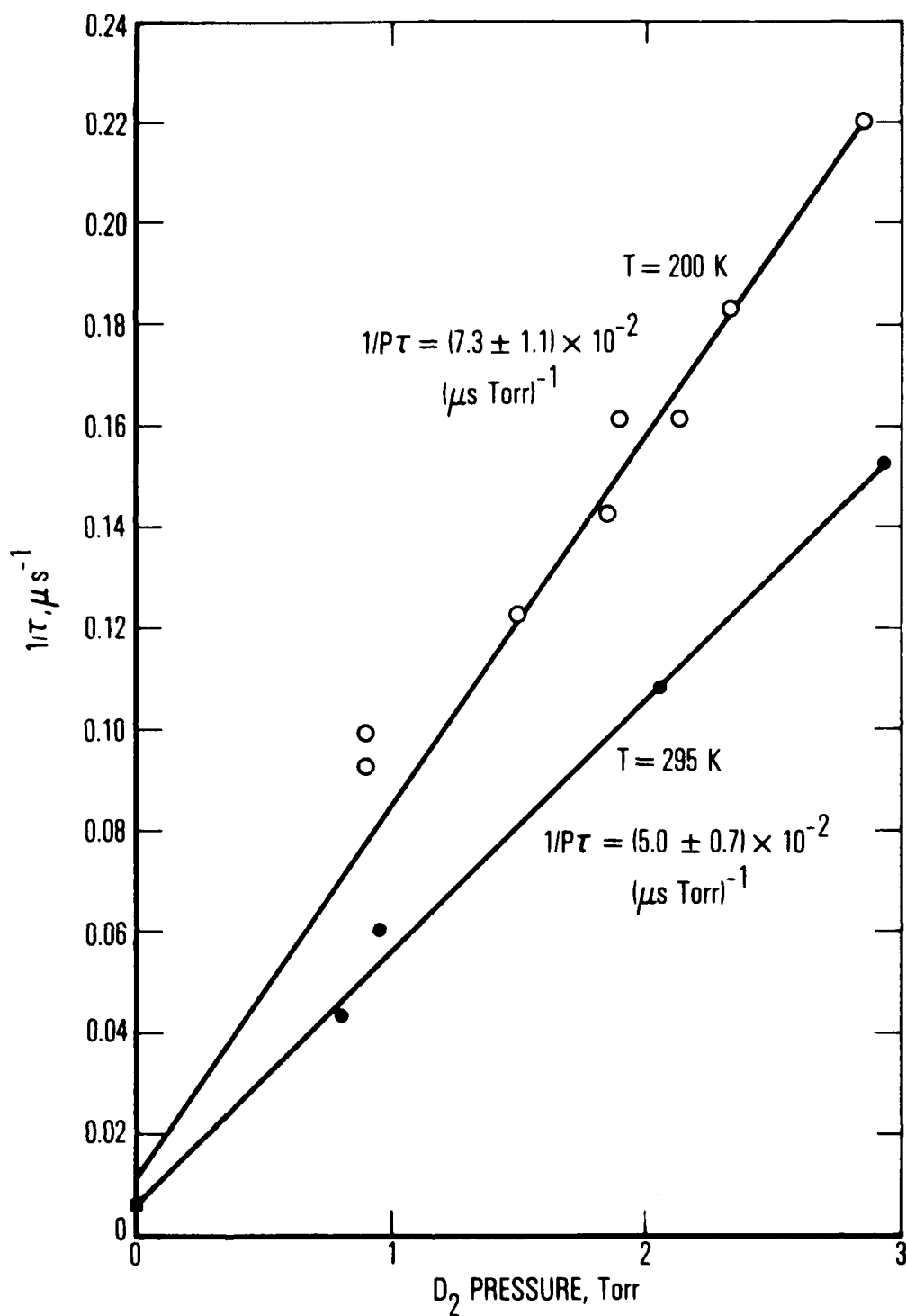


Figure 4. Relaxation Rates of HF($v = 3$) versus D_2 Pressure at 200 and 295 K

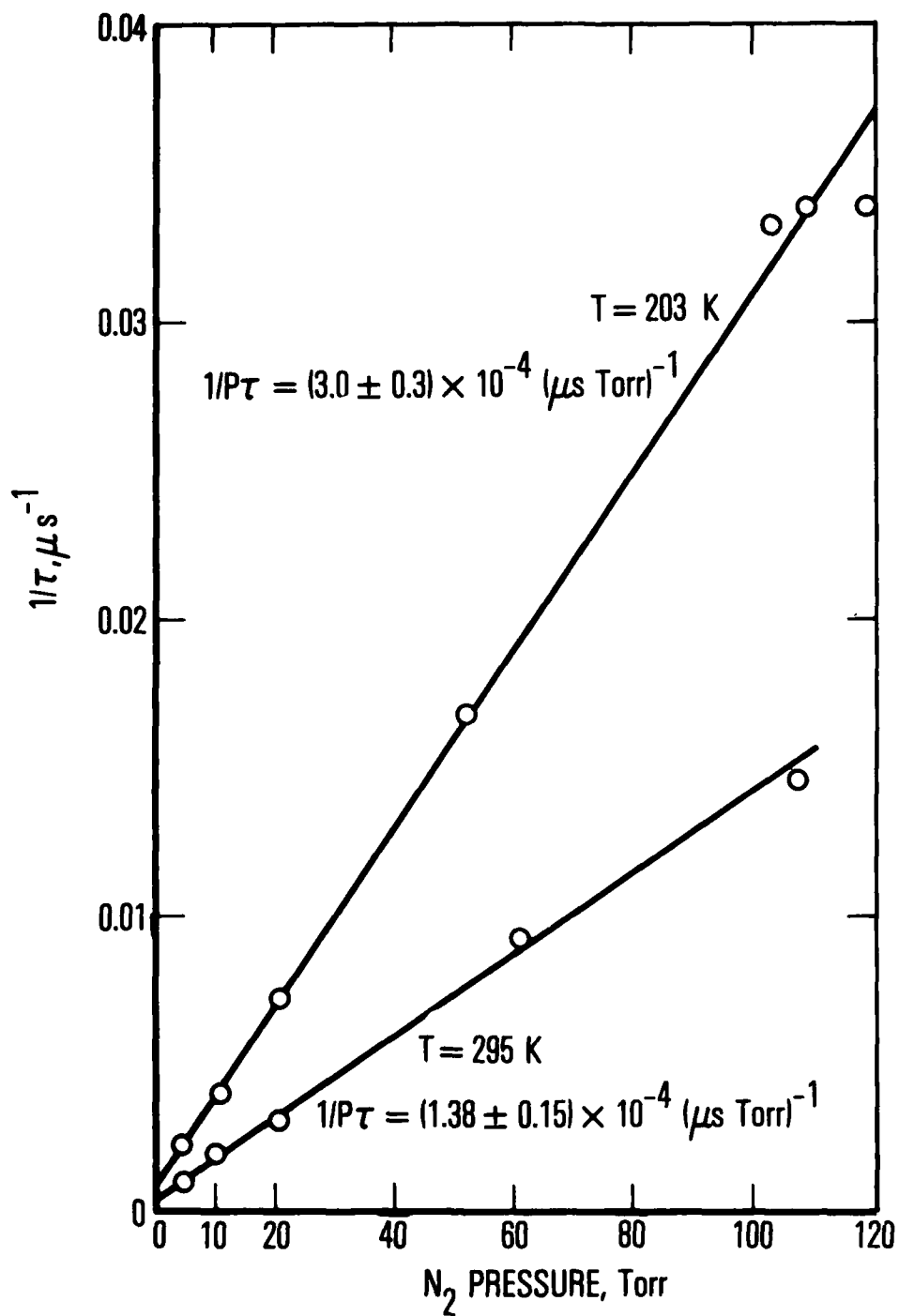


Figure 5. Relaxation Rates of HF($v = 1$) versus N_2 Pressure at 203 and 295 K

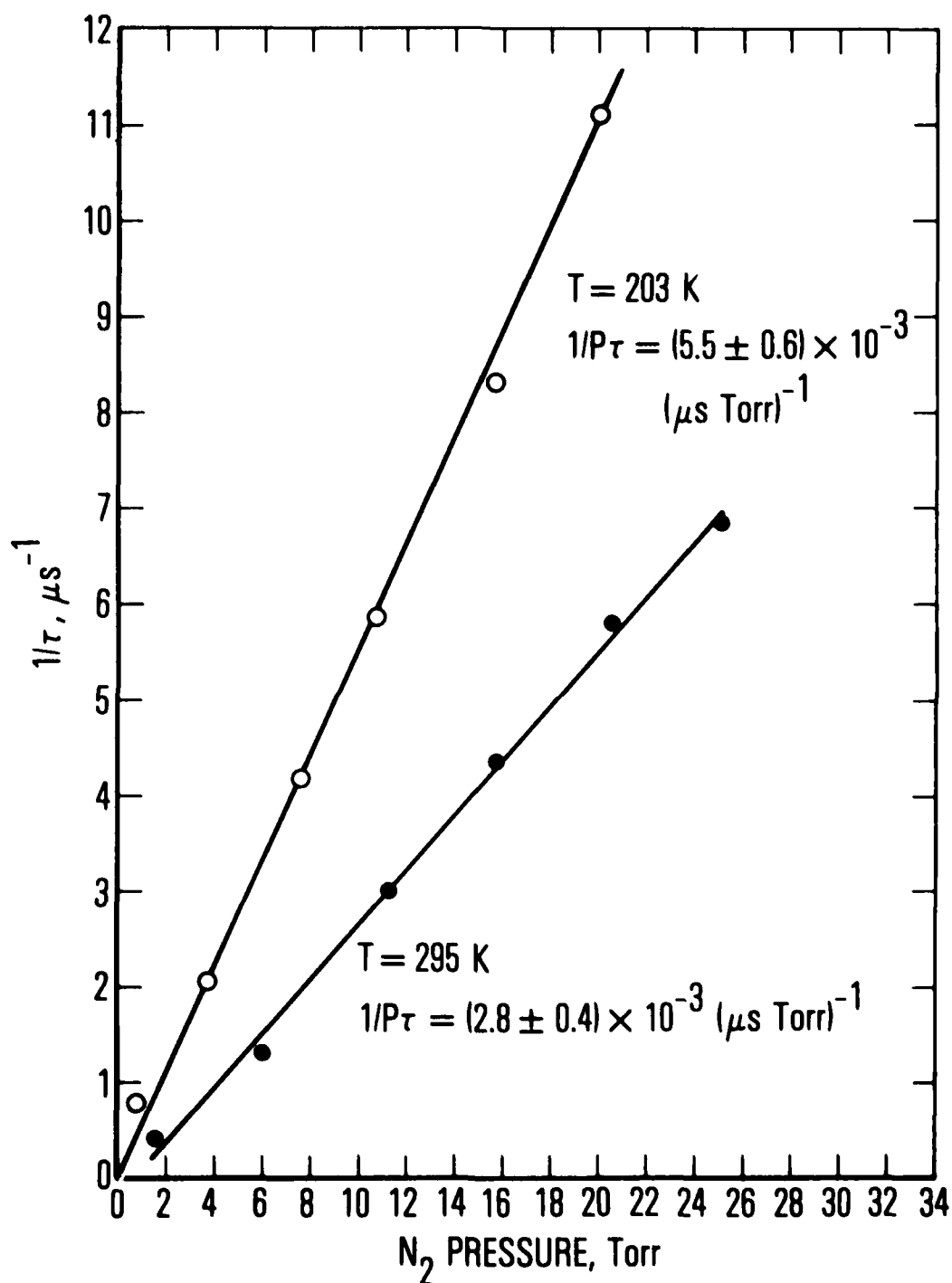


Figure 6. Relaxation Rates of $\text{HF}(v = 3)$ versus N_2 Pressure at 203 and 295 K

Table 1. Deactivation Rate Coefficients, ($\mu\text{sec-Torr}$)⁻¹

	295 K	200 K
HF(v = 1)-H ₂	$(1.67 \pm 0.17) \times 10^{-2}$	$(1.76 \pm 0.18) \times 10^{-2}$
HF(v = 3)-H ₂	$(1.13 \pm 0.12) \times 10^{-2}$	$(0.90 \pm 0.10) \times 10^{-2}$
HF(v = 1)-N ₂	$(1.38 \pm 0.15) \times 10^{-4}$	$(3.0 \pm 0.3) \times 10^{-4}{}^a$
HF(v = 3)-N ₂	$(2.85 \pm 0.4) \times 10^{-3}$	$(5.5 \pm 0.6) \times 10^{-3}{}^a$
HF(v = 1)-D ₂	$(2.9 \pm 0.3) \times 10^{-3}$	$(4.0 \pm 0.4) \times 10^{-3}$
HF(v = 3)-D ₂	$(5.0 \pm 0.7) \times 10^{-2}$	$(7.3 \pm 1.1) \times 10^{-2}$

^aT = 203 $\pm$ 4 K.

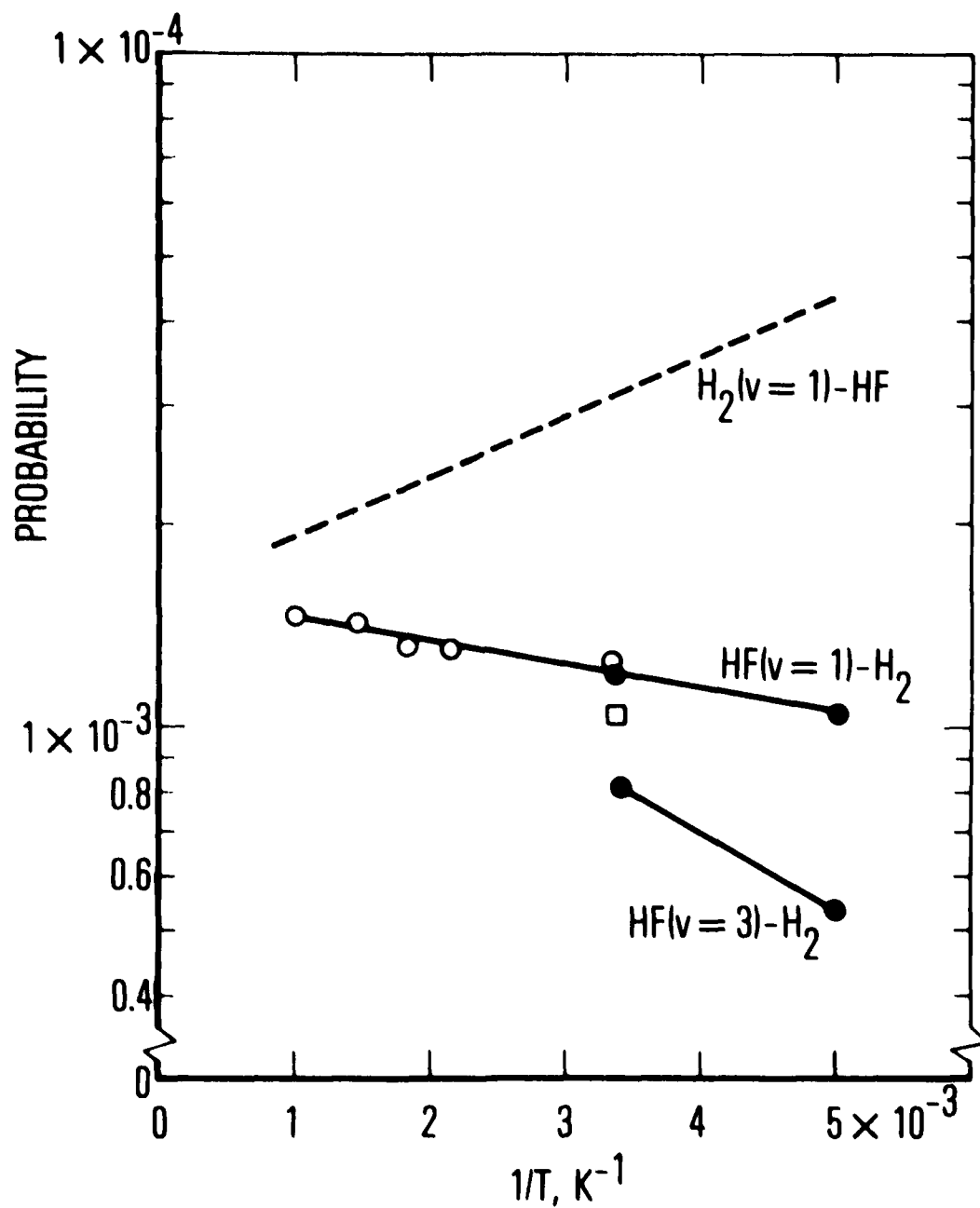


Figure 7. Deactivation Probabilities for HF($v = 1$) and HF($v = 3$) in H₂ versus Reciprocal Temperature. $\circ$; $\square$; $\bullet$, present data; —, exothermic exchange probability for H₂ ($v = 1$)-HF, see discussion

B. DEACTIVATION by D₂

The fit to the HF(v = 3)-D₂ data at T = 295 K (Fig. 4) shows no discernible effect of diffusion at the lowest pressure (~ 1 Torr) studied. The present value of $(2.9 \pm 0.3) \times 10^{-3} (\mu\text{sec Torr})^{-1}$ for the rate coefficient for HF(v = 1) deactivation by D₂ at 295 K compares with previously measured values of $(3.1 \pm 0.6) \times 10^{-3}$ (Ref. 8) and $(3.7 \pm 0.4) \times 10^{-3}$ (Ref. 9). The value of $(5.0 \pm 0.7) \times 10^{-2} (\mu\text{sec Torr})^{-1}$ for the rate coefficient for HF(v = 3) deactivation by D₂ is in good agreement with the value of $(4.4 \pm 0.8) \times 10^{-2}$ reported by Douglas and Moore.¹⁰ The probabilities of HF(v = 1) deactivation by D₂ plotted versus 1/T in Fig. 8 were calculated using a collision diameter of 2.9 Å for D₂. These probabilities show a complex temperature dependence with a minimum near 300 K. The probability of deactivation, P, can be described with $P = 1.74 \times 10^{-4} (T/295)^{1/2} \exp(152/T)$ for temperatures between 200 and 1000 K.

C. DEACTIVATION by N₂

The present value of $(1.38 \pm 0.15) \times 10^{-4} (\mu\text{sec Torr})^{-1}$ for the rate coefficient for HF(v = 1) deactivation by N₂ at 295 K compares with previously measured values of $(1.25 \pm 0.6) \times 10^{-4}$ (Ref. 9), $(1.52 \pm 0.15) \times 10^{-4}$ (Ref. 8), and $(1.45 \pm 0.15) \times 10^{-4}$ (Ref. 2). The present value of $(2.85 \pm 0.40) \times 10^{-3} (\mu\text{sec Torr})^{-1}$ for the HF(v = 3) deactivation rate coefficient at 295 K is in good agreement with the previously reported² value of $(2.92 \pm 0.30) \times 10^{-3}$.

Probabilities for HF(v = 1) removal by N₂ were calculated from the data using a collision diameter of 3.7 Å for N₂; these values are plotted versus 1/T in Fig. 9. The trend displayed in the data for HF(v = 1) removal by D₂ is accentuated in Fig. 9 with the probability having a more pronounced minimum and rising more steeply with temperature above 1000 K. Empirically, these data may be described by the equation $P = (9.4 \times 10^{-2} T^{-3/2} + 18 \times 10^{-13} T^{2.5})$ for T = 200 to 1600 K. The removal probability passes through a minimum near 420 K. More importantly, this behavior, which is a common feature of hydrogen halide quenching,⁸ suggests that more than one mechanism is responsible for HF(v = 1, 3) deactivation by the molecules H₂, D₂, and N₂.

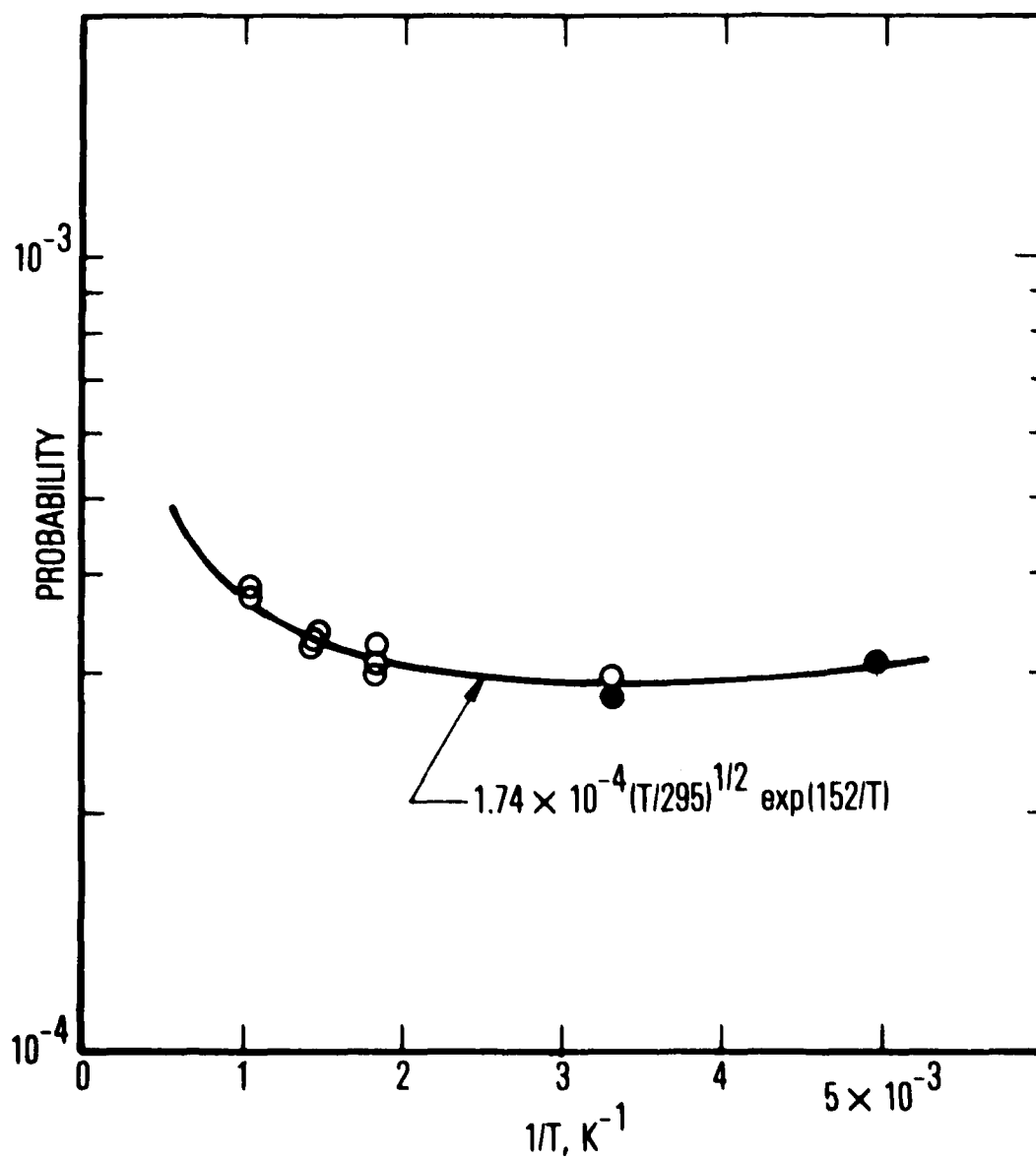


Figure 8. Deactivation Probabilities for HF($v = 1$) in D₂ versus Reciprocal Temperature. ○ - Ref. 8, ● - present data

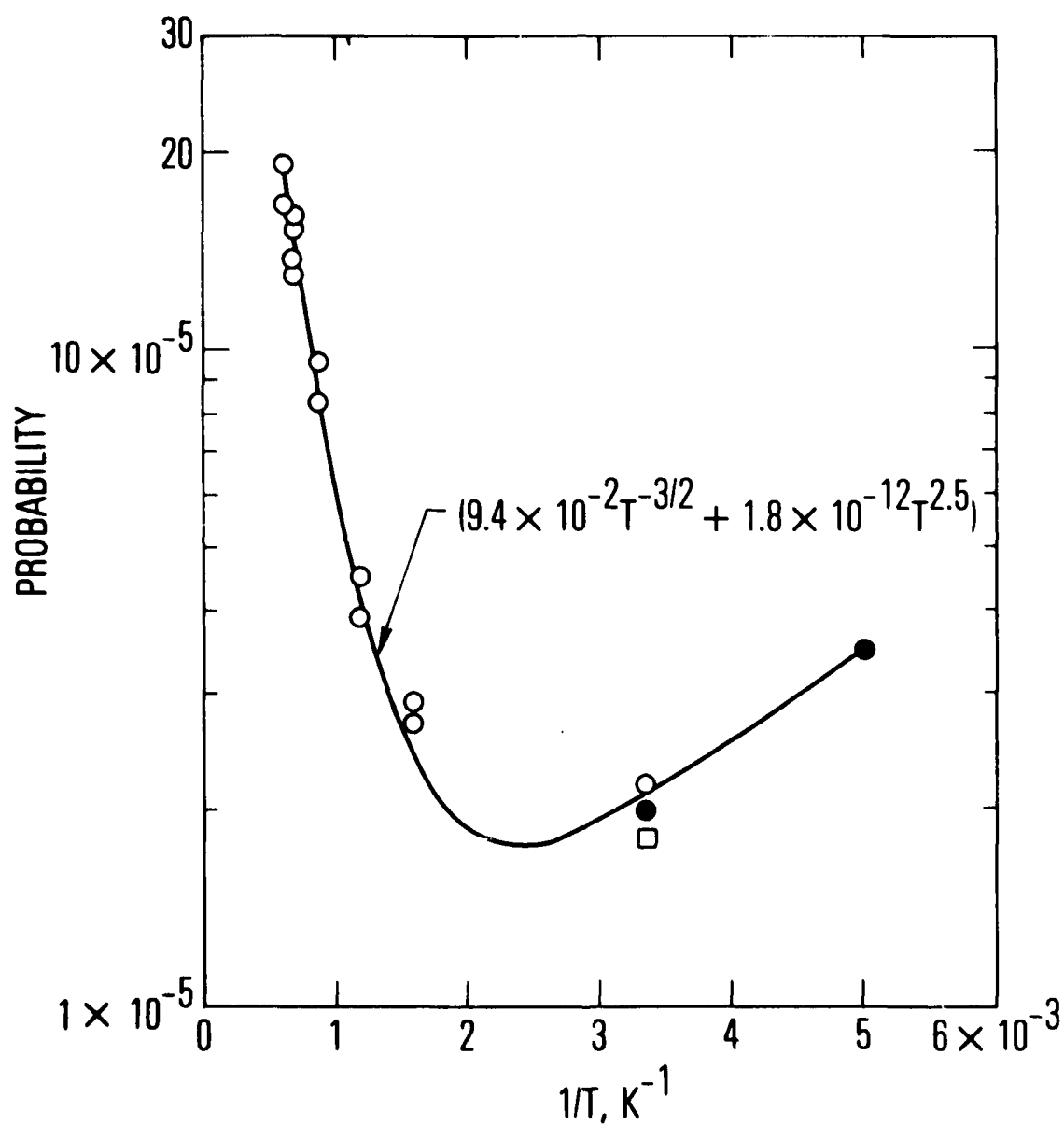


Figure 9. Deactivation Probabilities for HF($v = 1$) in N₂ versus Reciprocal Temperature. O :⁸ □ :⁹ ● , present data

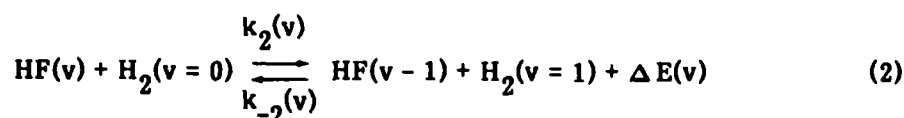
IV. DISCUSSION

A large body of high-temperature work^{11,12} suggests that HF($v = 1$) is relaxed with a rate coefficient that increases with increasing temperature. Several models have been used to describe these data, including the Landau-Teller formulation, which predicts that $(P\tau)^{-1}$ is proportional to $\exp(-AT^{-1/3})$. Although some success has been realized with this approach at temperatures above 1300 K, the relaxation data studied to date exhibit a more complex behavior at lower temperatures. The rates for HF($v = 1$) deactivation by H_2 , D_2 , DF, HCl, and N_2 (and for HF self-relaxation) show an inverse temperature dependence in certain temperature ranges.⁸ The probability of HF self-relaxation has a negative temperature dependence below about 1200 K, and HCl and HBr self-relaxation probabilities show a similar behavior below about 400 K.¹³ The temperatures at which these probability minima occur are approximately proportional to the binding energies for the respective dimers, and the large attractive interactions of the hydrogen halides have been used to explain qualitatively the low temperature inverse temperature dependence.

The attractive interactions can lead to long-lived bimolecular collisions or to the formation of excited complexes. Zittel and Moore¹¹ discussed the effect of attractive interactions on collisions with large impact parameters, where the rate of collisions surmounting any centrifugal barrier and experiencing a repulsive collision increases with decreasing temperature, relative to the rate of hard sphere collisions. Shin^{14,15} used a collision model to calculate HF and DF self-relaxation rates and attributed the low-temperature behavior of the relaxation rates to collisions in which a loosely bound nonrigid dimer is formed at low temperatures. The vibrational energy of the initially excited molecule is transferred partly to hindered rotational motion and partly to back and forth translational motion. Long-lived complexes have been observed in classical trajectory studies over surfaces with sufficient well depths. Billing and Poulsen¹⁶ found trajectories in which the molecules stick together and collide more than once. These "orbiting" collisions contributed significantly to the deactivation of HF($v = 1$) at low tempera-

tures and resulted in large changes of angular momentum. On the other hand, in the classical trajectory study by Wilkins,¹⁷ the vibrational energy was converted to rotational energy of the initially excited HF molecule by means of a V → R energy transfer process without orbiting collisions.

The deactivation of HF(v) by N₂, H₂, and D₂ can result in the vibrational excitation of the collision partner with the residual energy going into rotation of HF as well as relative translation. The potentials of interaction between polar HF and N₂, H₂, and D₂ are much smaller than between HF and HF, and the probabilities of HF(v = 1) deactivation by N₂, D₂, DF, and HCl have minima at lower temperatures than HF self-relaxation. For HF(v)-H₂ collisions, V → V exchange can occur by



Process (2) is endothermic by only 201 cm⁻¹ for v = 1 so that the vibrational energy exchange requires only a small conversion of rotational-translational energy. On the other hand, the V-R,T deactivation of HF(v = 1) by H₂ requires the conversion of ~3960 cm⁻¹ to rotational energy and has a rate coefficient of ≤ 3.8 × 10⁻⁴ (μsec Torr)⁻¹ at 295 K.¹⁸ Both the probability for Process (2) and the probability for the reverse, exothermic Process (-2) have been plotted in Fig. 7, neglecting the small V-R,T contribution. The exothermic probability is described by P = 1.57 × 10⁻³ exp(205/T) between 200 and 1000 K. Any minimum in the H₂-HF exchange probability must occur at a temperature greater than 1000 K.

If the entire deactivation rate for HF(v = 3) by H₂ is assigned to Process (2), then the reverse exothermic exchange also has an inverse temperature dependence. However, Poole and Smith¹ explained the v dependence of the total deactivation rate coefficient with a V-R,T contribution that increases with v and a V-V exchange contribution that decreases with v. If the rate coefficients for V-R,T deactivation of HF(v) by H₂ have the same v dependence as those found for other diatomic molecules, the V-R,T contribution to the deactivation of HF(v = 3) by H₂ at 295 K could be as much as 19 × 3.8 × 10⁻⁴ = 7.2 × 10⁻³ (μsec Torr)⁻¹ or 64% of the total rate coefficient.

When vibrational energy is transferred from $\text{HF}(v = 1)$ to $\text{D}_2(v = 1)$ or $\text{N}_2(v = 1)$, the excess energy of 968 cm^{-1} or 1627 cm^{-1} , respectively, must be accommodated by rotational and translational degrees of freedom. The $\text{HF}(v = 1)$ deactivation probabilities decrease with increasing energy mismatch (H_2 , D_2 , N_2 , O_2). The probability of $\text{HF}(v = 1)$ deactivation by D_2 is almost constant between 200 and 1000 K with a shallow minimum near 300 K (Fig. 8). The probability of $\text{HF}(v = 1)$ deactivation by N_2 has a much more pronounced minimum near 420 K (Fig. 9). Sentman and Solomon¹⁹ have compared theoretical V-V exchange rates at room temperature and above. The theoretical calculations gave good agreement with experimental data above $\sim 700 \text{ K}$ but did not contain long-range interactions that are the dominant contribution to the deactivation rates at low temperatures.

The rates for the deactivation of $\text{HF}(v = 1, 3)$ by D_2 and N_2 scale with v as $v^{2.7 \pm 0.1}$ at both 295 and 200 K (see Table 2). The rate for $\text{HF}(v = 4)$ deactivation in D_2 obtained by Douglas and Moore¹⁰ at 293 K is also consistent with this scaling. Although the nature of the collision partner does not affect this v dependence, it does affect the magnitude of the quenching probabilities. The same conclusion was reached in a study of $\text{DF}(v)$ deactivation³ although a $v^{1.9 \pm 0.1}$ dependence holds in the case of DF . The exceptions are the endothermic V-V exchanges $\text{HF}(v) \rightarrow \text{H}_2$ and $\text{DF}(v) \rightarrow \text{D}_2$. The same v dependence may not hold at higher temperatures where short-range interactions become more important and distributions over rotational levels shift toward higher states.

Table 2. Ratios of the Deactivation Rates $k(v = 3)/k(v = 1)$

	295 K	200 K
H ₂	0.68, 0.79 ^a	0.51
N ₂	20.7, 20 ^a	18
D ₂	17.2	18
O ₂	≥ 16.7	
HCl	23.5 ^b	
HF	17 ^c	

^aRef. 2.

^bHF(v = 3)-HCl data of Ref. 2, $1.7 \times 10^{-2} (\mu \text{ sec Torr})^{-1}$ for HF(v = 1)-HCl.

^cHF(v = 3)-HF data of Ref. 4, $5.8 \times 10^{-2} (\mu \text{ sec Torr})^{-1}$ for HF(v = 1)-HF.

V. CONCLUSIONS

The temperature dependence of $\text{HF}(v = 1)$ deactivation by N_2 and D_2 below room temperature differs markedly from that at high temperatures. At low temperatures, the deactivation probabilities are rather small with a zero or small inverse temperature dependence in contrast to the positive dependence at higher temperatures. $\text{HF}(v = 3)$ is quenched ~ 18 times more rapidly than $\text{HF}(v = 1)$ by both N_2 and D_2 at 200 and 295 K and by HCl and O_2 at 295 K. For the near-resonant exothermic exchange between $\text{H}_2(v = 1)$ and $\text{HF}(v = 0)$, Process (-2), the deactivation probabilities are much larger and show a small inverse temperature dependence. The detailed interpretation of the v dependence of $\text{HF}(v)$ quenching by H_2 is obscured by the increasing endothermicity with v and possible contributions of V-R,T deactivation of the higher vibrational levels. Further experiments are required to establish the vibrational scaling of $\text{HF}(v)$ deactivation at higher temperatures.

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LABORATORY OPERATIONS

The Laboratory Operations of The Aerospace Corporation is conducting experimental and theoretical investigations necessary for the evaluation and application of scientific advances to new military concepts and systems. Versatility and flexibility have been developed to a high degree by the laboratory personnel in dealing with the many problems encountered in the nation's rapidly developing space and missile systems. Expertise in the latest scientific developments is vital to the accomplishment of tasks related to these problems. The laboratories that contribute to this research are:

Aerophysics Laboratory: Launch and reentry aerodynamics, heat transfer, reentry physics, chemical kinetics, structural mechanics, flight dynamics, atmospheric pollution, and high-power gas lasers.

Chemistry and Physics Laboratory: Atmospheric reactions and atmospheric optics, chemical reactions in polluted atmospheres, chemical reactions of excited species in rocket plumes, chemical thermodynamics, plasma and laser-induced reactions, laser chemistry, propulsion chemistry, space vacuum and radiation effects on materials, lubrication and surface phenomena, photo-sensitive materials and sensors, high precision laser ranging, and the application of physics and chemistry to problems of law enforcement and biomedicine.

Electronics Research Laboratory: Electromagnetic theory, devices, and propagation phenomena, including plasma electromagnetics; quantum electronics, lasers, and electro-optics; communication sciences, applied electronics, semiconducting, superconducting, and crystal device physics, optical and acoustical imaging; atmospheric pollution; millimeter wave and far-infrared technology.

Materials Sciences Laboratory: Development of new materials; metal matrix composites and new forms of carbon; test and evaluation of graphite and ceramics in reentry; spacecraft materials and electronic components in nuclear weapons environment; application of fracture mechanics to stress corrosion and fatigue-induced fractures in structural metals.

Space Sciences Laboratory: Atmospheric and ionospheric physics, radiation from the atmosphere, density and composition of the atmosphere, aurorae and airglow; magnetospheric physics, cosmic rays, generation and propagation of plasma waves in the magnetosphere; solar physics, studies of solar magnetic fields; space astronomy, x-ray astronomy; the effects of nuclear explosions, magnetic storms, and solar activity on the earth's atmosphere, ionosphere, and magnetosphere; the effects of optical, electromagnetic, and particulate radiations in space on space systems.

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