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MOLECULAR BEAM STUDIES OF REACTION DYNAMICS
OF $F + H_2, D_2$

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ABSTRACT

Reactions of $F + H_2, D_2$ have attracted extensive theoretical and experimental attention in recent years and will undoubtedly become prototype chemical reactions in the future for the detailed understanding of reaction dynamics through meaningful comparison between theoretical calculations and experimental observations.

In this paper, recent state resolved, high resolution crossed molecular beams studies of these reactions are discussed. The experimental results reveal such detailed information as the dependence of the angular distribution of products on initial collision energies and vibrational quantum states of products and give indication of recently predicted quantum mechanical resonance phenomena.

INTRODUCTION

Recent advances in quantum chemistry have made it possible to understand some important features of the reaction dynamics of such simple systems as $H + H_2 \rightarrow H_2 + H$ entirely based on quantum mechanical calculations.¹ The extension of this remarkable achievement to other simple hydrogen containing triatomic systems, especially the calculation of accurate ab initio potential energy surfaces, is progressing rapidly, although full three dimensional quantal calculations still await the development of more efficient methods. For the understanding of reaction dynamics of hydrogen containing systems, quantal scattering calculations are very important, since the significant quantum effects of tunneling and resonance phenomena which appear in these systems preclude the usage of classical trajectory calculations as a means of obtaining reliable information on the reaction dynamics. In the development of efficient approximation methods for carrying out three dimensional quantal calculations, comparison with accurate experimental results is necessary when the results of exact theoretical calculations are not available.

One of the hydrogen-containing three atom systems which has already attracted extensive experimental and theoretical attention and will undoubtedly become the prototype reaction in the detailed understanding of reaction dynamics is the reaction of fluorine atoms with hydrogen molecules. The calculation of a limited *ab initio* potential energy surface of chemical accuracy has been carried out.² The basic features of this calculation were adopted in the determination of more complete semiempirical potential energy surfaces which have been utilized in extensive classical trajectory calculations.^{3,4} In exact one dimensional scattering calculations,^{5,6} interesting quantum resonance phenomena were predicted at certain specific collision energies while a very recent three dimensional calculation using an angular momentum decoupling approximation⁷ has provided some important information regarding resonance phenomena in three dimensional reactive scattering. The advances in recent years of various experimental approaches, such as chemical lasers,⁸ chemiluminescence methods⁹ and crossed molecular beams experiments,¹⁰ have not only revealed detailed information on product state distributions, but also have provided angular distributions of products in various quantum states at well defined collision energies. These results and expected improvements in the near future will certainly provide an unprecedented opportunity for making very meaningful comparisons between theoretical calculations and experimental observations.

In particular, the experimental observation of resonance phenomena via their effect on the state and angular distributions of the products under well-defined initial conditions could provide a very stringent test of the accuracy of theoretical calculations. Since the resonance phenomena in chemical reactions are very sensitive to the potential energy hypersurface at the critical configuration of the reaction intermediate, the observation of resonance phenomena at specific collision energies in the laboratory provides an opportunity to evaluate the reliability of the potential energy surface in the close contact region.

In one dimensional quantum mechanical calculations, a sharp resonance has been predicted for the production of $\text{HF}(v=2)$ at a collision energy of ~ 1 kcal/mole.^{5,6} This sharp enhancement of the production of $\text{HF}(v=2)$ at a specific collision energy was not observed in recent three dimensional calculations,⁷ although a similar resonance phenomenon in the production of $\text{HF}(v=2)$ predicted in the one dimensional calculation is still evident as collision energies are varied at a fixed orbital angular momentum of reactants. Because of both the shift of the resonance to higher collision energies when the orbital angular momentum is increased and the averaging of the distribution of orbital angular momenta in three dimensional reactive scattering, no pronounced variation of the state distribution as a function of collision energies is predicted in spite of the presence of resonance phenomena. This means that the measurement of state distributions as a function of collision energy will not be the appropriate method for the search for resonance phenomena. On the other hand, in the energy range where the resonance

is occurring, there is a substantial variation in the reaction probability as a function of orbital angular momentum, or, equivalently, of impact parameter, for collisions at a given initial relative velocity. Since the angular distribution of product molecules to a large extent reflects the dependence of reaction probability on impact parameter, especially for a system with the lowest entrance barrier for the collinear approach, the search for unusual features in the angular distributions of various quantum states of HF measured in a high resolution cross molecular beams experiment is probably the best approach for finding reactive resonance phenomena in the laboratory.

In this paper, recent state resolved high resolution crossed molecular beams studies on $F + H_2 \rightarrow HF + H$ and $F + D_2 \rightarrow DF + D$ carried out at several collision energies will be discussed. The results reveal such detailed information as the dependence of the angular distribution of product molecules on both collision energy and product vibrational quantum state. These measurements illustrate the current state of the art of crossed molecular beams studies of reactive scattering and reflect predicted quantum mechanical resonances as well as other interesting features in the experimental observations of reactive scattering.

EXPERIMENTAL

The experiment is performed by crossing two supersonic molecular beams, one of atomic fluorine seeded in N_2 , and one of H_2 or D_2 , in a newly designed high resolution universal crossed molecular beams machine. The two beams cross at 90° in a well-defined interaction region. The detector, a triply-differentially pumped electron-impact ionization mass spectrometer, is rotated about the interaction region in order to obtain the product angular distributions. The product velocity distributions are determined by measuring time-of-flight for molecules passing through a rapidly spinning slotted disc to the ionizer.

The design of the new apparatus closely follows that of the original universal crossed molecular beam machine described by Lee et al.,¹¹ but several modifications extend its capabilities beyond those of the original design. The detector is suspended from a 35" diameter rotating chamber which allows one to obtain a 30 cm flight path in contrast to the 17 cm flight path in the original 25" design. This nearly doubles the resolution of the time-of-flight spectra and consequently allows the product state distribution to be more accurately determined than was previously possible. In addition, the main chamber has been enlarged in order to increase the laboratory angular scan range from 115° to 170° .

Other modifications include a set of removable precision-machined apertures for the detector slits to allow the detector angular

resolution to be varied simply by inserting a different set of apertures. Extreme care was taken in the choice and processing of all materials used in the three ultra-high vacuum detector regions, which has resulted in a background count rate at most masses that is nearly a factor of ten lower than in any of our previous machines. Also, a newly designed 1 μ s per channel multi-channel scaler compatible with the CAMAC standard has been utilized for the time-of-flight analysis in conjunction with a new on-line computer system.

The fluorine atom beam was produced by thermal dissociation of a 3% mixture of F₂ in N₂ at 700 torr in a nickel oven heated by thermo-coax¹² heating elements to a temperature of 706°C. The heated gases expand through a 0.15 mm nozzle at the tip of the oven. A 0.66 mm diameter skimmer was used with a nozzle-skimmer distance of 6 mm. The beam was collimated in a second chamber to an angular spread of 2.2°. The nozzle to interaction center distance was 5.5 cm. Nitrogen was used as a seed gas to avoid the increased background at mass 20 (HF) that would have resulted from the use of Ar or Ne. Using He as a seed gas resulted in such high F atom velocities that the orientation of the resulting Newton diagram was unfavorable for observing many important center-of-mass angles. The F atom beam obtained had a peak velocity of 13.7×10^4 cm and a FWHM velocity spread of approximately 25%.

Hydrogen and deuterium beams were produced by a very high pressure expansion of the gases through a 0.030 mm diameter nozzle. The nozzle-skimmer region was pumped by a Varian 10" NHS diffusion pump backed by a Leybold Heraeus WS-500 Roots blower package. The nozzle skimmer distance used was approximately 10 mm and was externally adjusted to allow maximization of beam intensity. The skimmer itself was constructed of electro-formed nickel¹³ and has the advantages of very sharp edges and very small included angles. The beam was collimated to a width of 2.2° and the nozzle was located at a distance of 6.5 cm from the scattering center. The collision energy of the experiment was varied by changing the stagnation temperature of the D₂ or H₂ beam. The beam source was heated with a short section of thermocoax wire, powered by a regulated D.C. power supply. At the higher temperatures, the stagnation pressures behind the nozzle were increased in order to maintain number density, and thus intensity and high quality of expansion. The operating conditions of the H₂/D₂ beam source for the four different sets of data to be presented here are shown in Table I along with the most probable collision energy for each condition.

Product velocity analysis for the F + D₂ experiments was performed by single shot time-of-flight. The single shot time-of-flight wheel consisted of a 0.13 mm thick stainless steel disk of 17.5 cm diameter. Eight slots, each 1.5 mm wide, were machined on its perimeter. A reducing slit, also 1.5 mm wide, was mounted to the front of the detector. The wheel was rotated at a speed of 200 Hz. Data was collected for 600 μ s following each wheel opening using a multichannel scalar channel width of 5 μ s (120 channels). Typical counting times

Table I. H_2/D_2 Operating Conditions

	Collision Energy kcal/mole	Temperature °C	Stagnation Pressure (Atm)	Peak Velocity cm/sec
H_2	2.00	32°C	27	27.2×10^4
	3.17	219°C	40	35.7×10^4
D_2	2.34	32°C	27	20.9×10^4
	4.5	421°C	54	39.9×10^4

were two hours at each angle.

Velocity distributions for the $F + H_2$ experiment were obtained by cross-correlation time-of-flight. Our particular implementation of this method has been described before.¹⁴ Cross-correlation was necessary for $H + HF$ due to the much higher background at mass 20 (HF) than at mass 21 (DF). However, it is not an appropriate technique for a system such as $F + D_2$, since, in that case, the signal to background ratio is sufficiently high such that cross-correlation would actually reduce the quality of the data.¹⁵ Additionally, we are presently limited by wheel design and wheel speed to a channel width of 10 μ s with cross-correlation. Data for $F + H_2$ was taken with this lower limit channel time of 10 μ s corresponding to a wheel speed of 392 Hz with 255 channels on the wheel. Typical counting times were approximately 70 minutes at each angle.

Angular distributions were obtained by modulation of the H_2/D_2 beam with a 150 Hz tuning fork chopper. Data was collected for equal times during the open and closed portion of the tuning fork cycle in a dual channel NIM scalar. The difference of the counts in the two channels constitutes the angular scan signal. Typical counting times for the angular scans were 2 minutes at each angle. Typical signal near the maximum in the angular distribution for $F + H_2$ would be about 6000 counts with a background of 19,000 counts. For $F + D_2$ we usually achieved 1700 counts of signal with about 120 counts of background. These numbers represent about one sixth of the actual count rate due to the duty cycle of the tuning fork chopper. Detector aperture slits were set to achieve a laboratory angular resolution of 1° for both angular scans and the TOF data.

RESULTS AND ANALYSIS

The measured laboratory angular distributions for $F + H_2$ at 2.00 and 3.17 kcal/mole collision energy are shown in Figs. 1 and 2 along with the calculated best fits. The nominal Newton diagrams for each system are also shown. The corresponding data and Newton diagrams for $F + D_2$ at collision energies of 2.34 and 4.51 kcal/mole are shown in Figs. 3 and 4.

Fig. 1. Experimental LAB angular distribution of the HF product from the $F + H_2$ reaction at 2.00 kcal/mole.

Fig. 2. Experimental LAB angular distribution of the HF product from the $F + H_2$ reaction at 3.17 kcal/mole.

Fig. 3. Experimental LAB angular distribution of the DF product from the $F + D_2$ reaction at 2.34 kcal/mole.

Fig. 4. Experimental LAB angular distribution of the DF product from the $F + D_2$ reaction at 4.51 kcal/mole.

Time-of-flight data for all of these systems has been obtained. TOF spectra were generally taken for every other angle at which angular data was taken, however, over regions in which interesting effects were anticipated, spectra were taken for every angle at which angular scan data was taken. Angles with less than one fourth of the total intensity at the peak of the angular distribution were not considered profitable for TOF analysis. This procedure resulted in 11 to 18 TOF spectra for each system. Figure 5 shows the experimental TOF data for $F + H_2$ at 2.00 kcal/mole collision energy. The plots also show our calculated best fit to the data, including the separate contributions of the various vibrational states.

Data for both of the $F + H_2$ systems and for the lower energy $F + D_2$ have been analyzed to obtain the center-of-mass (C.M.) product distribution. We have used a forward convolution trial and error fitting technique in which the C.M. angular and energy distribution is input as a trial function. The corresponding laboratory angular and velocity distributions are then calculated from it and compared to the experimental data. The original trial function is adjusted and the process repeated until a satisfactory fit is obtained simultaneously to both the TOF spectra and the angular scan. The appropriate equations and basic methodology of forward convolution have been well discussed elsewhere.¹⁶ We, however, found it necessary to write an entirely new computer program for the actual calculation. Our very large quantity of data forced us to use direct graphical output of our calculated distributions. We considered completely different angular and energy distributions for each vibrational state, as well as having the energy distribution of each state being angular dependent.

The C.M. angle-recoil energy distribution that we have used is a generalized RRK form with separate parameterization for each vibrational state. The functional form used is:

$$I_{C.M.}(E, \theta) = \sum_{v=0}^n C_v(\theta) N_v(\theta) \cdot \begin{cases} (E - E_{\min}(\theta, v))^{\alpha(\theta, v)} (E_{\max}(\theta, v) - E)^{\beta(v)} & \text{if } E_{\min}(\theta, v) \leq E \leq E_{\max}(\theta, v) \\ 0 & \text{if } E < E_{\min}(\theta, v) \text{ or } E > E_{\max}(\theta, v) \end{cases} \quad (1)$$

where $C_v(\theta)$ is a relative angular intensity factor, which is input in point form every 10° and interpolated linearly by the program for points between 10° intervals. $N_v(\theta)$ is a normalization factor calculated by the program to normalize the rest of the function to an area of 1 at each angle. $\alpha(\theta, v)$ is input in point form and interpolated in the same manner as $C_v(\theta)$. $E_{\min}$ and $E_{\max}$ were defined as circles in (E, θ) space with a given radius in E and a given displacement along the 0° - 180° axis. Usually the radius and displacement for $E_{\min}$ were chosen as 0, while the displacement for $E_{\max}$ was usually chosen as zero with the radius being set at the sum of the collision energy and the exothermicity minus the vibrational energy of the particular state. The

Fig. 5. Time-of-flight analysis of the HF product from $F + H_2$ reaction at 2.00 kcal/mole. Plots show experimental LAB TOF spectra at indicated angles along with the best fits calculated from center-of-mass angular and velocity distributions for various vibrational states of HF. (●) Experimental. (—) Calculated best fit summed over contributions from $v=1, 2$, and 3. (· —) $v=1$. (— —) $v=2$. (·····) $v=3$.

parameter β was fixed for each vibrational state. The total distribution, then, represents the entire product flux recoil energy distribution while the contribution from each vibrational state, in principle, represents the flux as a function of rotational excitation of that vibrational state.

Thus the final result of our data fitting analysis is a center-of-mass flux distribution as a function of angle and product recoil energy. In order to represent this distribution in the usual form superimposed on a Newton diagram it must be converted from an energy space to a velocity space distribution. The transformation is straightforward and the results are shown in Figs. 6-8.

Fig. 6. Center-of-mass velocity-space contour plot of HF product distribution from $F + H_2$ reaction at 2.00 kcal/mole.

Figures 6 and 7 show the velocity space contour maps of our best fit distribution for $F + H_2$ at 2.00 and 3.17 kcal/mole respectively. Figure 8 shows the corresponding information for $F + D_2$ at 2.34

Fig. 7. Center-of-mass velocity space contour plot of HF product distribution from $F + H_2$ reaction at 3.17 kcal/mole.

kcal/mole. When transformed to laboratory angular and velocity space, these contour maps correspond to the total calculated distributions shown in the angular scan plots of Figs. 1-3 and the velocity distribution plots of Fig. 5.

DISCUSSION

The dynamics of the $F + H_2$ and $F + D_2$ reactions produce several features which lend themselves well to study by the crossed molecular beams method. The large product vibrational spacings in conjunction with narrow rotational energy distributions in many cases allow the vibrational states of the HF and DF products to be identified from distinct peaks which appear in the measurements of laboratory velocity. The potential energy barrier to reaction is lowest in the configuration

Fig. 8. Center-of-mass velocity-space contour plot of DF product distribution from F + D₂ reaction at 2.34 kcal/mole.

where the fluorine atom approaches collinear to the axis of the hydrogen molecule. At low collision energies this is expected to cause the products to scatter predominantly along the relative velocity vector of the reactants, and consequently peaks corresponding to individual vibrational states are observed in the laboratory angular distributions. Thus, both the laboratory velocity measurements and angular distributions provide detailed information on product state distributions and the two measurements effectively complement one another.

At low collision energies the angular distributions of HF and DF in the center of mass coordinate system shown in Fig. 6 and Fig. 8 are found to peak backward with respect to the direction of the incident fluorine atom. This is consistent with the nature of the potential energy surfaces derived from *ab initio* and semi-empirical calculations for F + H₂. The potential energy barrier for this reaction depends on the bending angle of H-H-F. The barrier is lowest for the collinear configuration, with a value of about 1 kcal/mole. Consequently, only those collisions involving a nearly collinear configuration of the two hydrogen atoms and the fluorine atom contribute to the formation of products, and these products are predominantly backward scattered.

The center-of-mass angular and velocity distribution for HF product from the $F + H_2 \rightarrow HF + H$ reaction at a collision energy of 3.17 kcal/mole is shown as a contour map in Fig. 7. At this collision energy the center-of-mass angular distribution differs markedly from that at the lower energy. The distributions for different vibrational states seem to be entirely decoupled. The products in $v=1$ and $v=3$ remain backward peaked as at the lower collision energy, but the angular distribution for product in $v=2$ is very unusual. The intensity remains high as the center-of-mass scattering angle decreases, and even shows a sideways peaked relative maximum. This is illustrated more clearly when the center-of-mass velocity distributions are plotted as a function of angle as in Fig. 9. The laboratory angular distribution also shows a significant change between the two energies. The structure due to strongly backward peaked HF($v=2$) product at 2.00 kcal/mole is absent when the collision energy is raised to 3.17 kcal/mole. The dramatic change in the center-of-mass angular distribution of the product formed in $v=2$ at 3.17 kcal/mole compared to that at 2.00 kcal/mole is consistent with the reactive resonance phenomenon predicted by the three dimensional calculations of Redmon and Wyatt. In that study it was shown that the effect of resonances in the $F + H_2$ reaction appears as a change in the dependence of the reaction probability on reactant orbital angular momentum. The reaction probability for forming HF($v=3$) is maximum at zero orbital angular momentum, independent of collision energy, while the probability for forming HF($v=2$) peaks at non-zero orbital angular momentum for a collision energy around 3 kcal/mole, and the orbital angular momentum corresponding to the peak reaction probability changes as a function of collision energy. Since the orbital angular momentum is determined by the impact parameter for the collision, the reaction probability for forming HF($v=2$) peaks at a non-zero impact parameter for a collision energy around 3 kcal/mole. Products formed from a larger impact parameter collision should scatter at smaller center-of-mass angles, so our experimentally observed results at 3.17 kcal/mole are in qualitative agreement with what one would expect from the reactive resonance in $F + H_2 \rightarrow HF(v=2) + H$ predicted in the calculation of Redmon and Wyatt.

In the laboratory angular distribution of DF product from the reaction $F + D_2 \rightarrow DF + D$ at 4.51 kcal/mole (Fig. 4), there is another surprising feature: a sharp peak appears at the laboratory angle corresponding to forward scattering of DF($v=4$). Such a sharp forward product peak has not been observed previously in a reaction which does not form a long lived complex. But just as the extent of the polarization of product orbital angular momentum determines the sharpness of forward-backward peaks in angular distributions of products from a long lived complex, the strong forward peaking of DF($v=4$) indicates a strong correlation between the final orbital angular momentum of the product and the initial orbital angular momentum. The reliability of the angular momentum decoupling calculations carried out on $F + H_2 \rightarrow HF + H$ by Redmon and Wyatt depend on the extent of this correlation. For the $F + H_2$ and $F + D_2$ reactions, this approximation might appear to be valid. Once again, this strong correlation of final and initial angular

Fig. 9. Center-of-mass distributions at indicated angles of HF product from $F + H_2$ reaction at (a) 2.00 kcal/mole and (b) 3.17 kcal/mole.

momentum might be the consequence of the nature of the potential energy surface for these reactions, which has a bending angle dependent barrier which is minimum at the collinear configuration.

CONCLUSION

The experimental results presented in this paper are examples of what can be measured at present in the laboratory by the crossed beams method: The experimental resolution could be further improved in the future, but the incentive for future effort toward carrying out a better experiment certainly depends on the advancement of a theoretical calculations, making possible a more quantitative comparison

between theory and experimental results.

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REFERENCES

1. Kupperman, A., and Schatz, G.C.: 1975, J. Chem. Phys. 62, 2502.
2. Bender, C.F., Pearson, P.K., O'Neil, S.V., and Schaefer, III, H.F.: 1972, J. Chem. Phys. 56, 4626.
3. Muckerman, J.T.: 1971, J. Chem. Phys. 54, 1155.
4. Wilkins, R.L.: 1973, J. Chem. Phys. 58, 3038.
5. Schatz, G.C., Bowman, J.M., and Kupperman, A.: 1973, J. Chem. Phys. 58, 4023.
6. Latham, S.L., McNutt, J.F., Wyatt, R.E., and Redmon, M.J.: 1978, J. Chem. Phys. 69, 3746.
7. Redmon, M.J., and Wyatt, R.E.: 1979, Chem. Phys. Lett. 63, 209.
8. Parker, J.H., and Pimentel, G.C.: 1969, J. Chem. Phys. 51, 91; Coombe, R.D., and Pimentel, G.C.: 1973, J. Chem. Phys. 59, 251.
9. Polanyi, J.C., and Woodall, K.B.: 1972, J. Chem. Phys. 57, 1574.
10. Schafer, T.P., Siska, P.E., Parson, J.M., Tully, F.P., Wong, Y.C., and Lee, Y.T.: 1970, J. Chem. Phys. 53, 3385.
11. Lee, Y.T., McDonald, J.D., LeBreton, P.R., and Herschbach, D.R.: 1969, Rev. Sci. Instr. 40, 1402.
12. Semco Instruments Incorporated, 11505 Vanowen Street, North Hollywood, California 91605 USA
13. Beam Dynamics Corporation, 623 East 57th Steet, Minneapolis, Minnesota 55417 USA.
14. James Martin Farrar, Ph.D. dissertation, The University of Chicago, Chicago, Illinois, 1974.
15. Hans-Dieter Meyer, Diplomarbeit, Max-Planck-Institut für Strömungsforschung, Göttingen, West Germany, 1974.
16. Richard James Buss, Ph.D. dissertation, The University of California, Berkeley, California, 1979.

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