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DYNAMICS OF THE REACTION OF N^+ WITH H_2 .

II. REACTIVE SCATTERING AT RELATIVE ENERGIES BELOW 3 eV

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ABSTRACT

The reaction $N^+(H_2, H)NH^+$ has been investigated in the range of initial relative energies from 0.79 to 2.8 eV. Despite the deep potential energy well (6 eV) associated with the symmetric NH_2^+ molecule ion, the reaction proceeds predominately by a direct interaction mechanism at relative energies as low as 2 eV. Only when the initial relative energy is near or below 1 eV does a long-lived complex mechanism appear to predominate. This behavior can be rationalized in terms of the qualitative features of potential energy surfaces deduced for the system from an electronic state correlation diagram.

A major goal of chemical kinetics is to elucidate the relation of reactivity and reaction dynamics to the known electronic properties of reactants, products, and intermediates. This problem is particularly intriguing in the area of ion-molecule reactions, since such systems frequently possess several low-lying potential energy surfaces, often of quite different character, on which a reaction might proceed. In order to be able to compare predictions of the nature of reaction dynamics based on electronic state correlation diagrams and ab initio calculations with experimental results, our laboratory has undertaken a series of investigations of the detailed dynamics of the collisions of C^+ , N^+ , O^+ , and Ne^+ with hydrogen.¹⁻⁵

Previous work from this laboratory² on the $N^+(H_2,H)NH^+$ reaction was limited to initial relative translational energies above 2 eV. However, the electronic state correlation diagram for this system suggests that the nature of the reaction dynamics might undergo a substantial change as the initial relative energy was lowered. In this paper, we report experimental results that we have obtained in the low energy regime, and discuss them in terms of the electronic state correlation diagram for the system.

Experimental

This is the first work from this laboratory which has been performed with a new ion beam collision apparatus specifically intended for use in the regime of low ion energies.

As is the case for our older apparatus,⁶ the newer model employs an electron impact ion source followed by a magnetic momentum analyzer and lens system to produce, select, and focus an ion beam of known mass and energy. The neutral target molecules were in this work contained in a collision cell, and thus had an isotropic velocity distribution. The ion gun-collision cell combination is mounted on a rotatable table inside a large vacuum chamber. By rotating this table, the experimenter can direct ions scattered at a desired angle into the detector. The detection train in this apparatus is fixed to one wall of the vacuum chamber, and consists of a Wien filter velocity analyzer followed by a quadrupole mass filter and a magnetic electron multiplier operated in the pulse counting mode. Data is recorded at fixed laboratory scattering angles by stepping the velocity analyzer through an appropriate range. The data are reduced and usually displayed as plots of the specific product intensity⁶ (Cartesian flux) in a velocity coordinate system which has its origin at the center-of-mass of the system. In what follows, the individual components of the apparatus are described in more detail.

The electron impact ion source consists of a small cylindrical equipotential region surrounded by a tantalum grid and mounting plate. Electrons emitted by an external tungsten filament are accelerated radially through the grid into the cylindrical equipotential region where they ionize the ambient gas. The entire assembly is surrounded by a

cylindrical electron reflector, so that electrons can make more than one pass through the ion source region. Ions are drawn from the source axially through an aperture in the grid mounting plate, and directly enter the lens system of the momentum analyzer.

The magnetic momentum analyzer is of the 90° deflection type with circular pole pieces so that second order focusing may be achieved. The radius of curvature of ions in the field is 3 cm, and the overall flight path of ions through the analyzer and its entrance and exit lens systems to the collision center is approximately 22 cm. Magnetic fields of up to 4000 gauss can be produced by two coils of 250 turns each that are energized by a current regulated power supply. The return yoke of the magnetic analyzer is a cylindrical can which completely surrounds the pole pieces in much the same way as in the magnet described by Carette and Kerwin.⁷ As a result, there is virtually no magnetic field leakage from the momentum analyzer region into other areas of the apparatus.

Ions which leave the momentum analyzer are rendered into a parallel beam and decelerated to their kinetic energy at ground potential by a three element lens based on the designs outlined by Heddle.⁸ A set of vertical and horizontal deflection plates are used to position the beam immediately before it enters the collision region.

The scattering cell is 3.2 cm in diameter, 6.5 cm high, and has an exit slot 0.15 x 1.2 cm. The gas flow to the cell was controlled by a Granville-Phillips needle valve, and the

pressure was determined by using a capacitance manometer, making appropriate allowance for the conductance of the tubing between the manometer and the cell. In the experiments reported here, the scattering gas pressure was approximately 1 mtorr.

Velocity analysis of the product ions was performed by using a crossed electric and magnetic fields device. The magnetic field was produced by two 225 turn coils wound on rectangular pole pieces of 4.5 cm length. The return yoke of the electro-magnet was a mild steel box which completely enclosed the pole pieces, much in the manner of the design of the ion source momentum analyzer. Fields of up to 1000 gauss could be produced without causing detectable stray fields immediately outside the velocity filter. Ions were protected from the return and fringing fields of the pole pieces by mild steel tubes.

A conventional Wien velocity filter employs a uniform electric field perpendicular to the magnetic field, and is a stigmatic device which focuses ions only in planes perpendicular to the magnetic field. Our device uses the curved electric field plates described by Legler⁹ to produce a parabolic electrostatic potential, and simultaneous focusing in the two directions perpendicular to the axis of the filter. In operation, fixed values of the electric and magnetic fields were employed, so that the filter passed ions of one velocity only. The ion velocity spectrum was scanned by varying the potential of the filter with respect to ground. The resolution was determined by the virtual apertures formed at the entrance

and exit of the filter by the lens systems, and generally amounted to 4% of the energy at which the ions passed through the filter. The angular resolution of the system was determined by the same lens apertures, and was approximately 1.8° FWHM.

Results

Thirteen experiments were performed in which full product velocity vector distributions were determined. The results of six of these are displayed in Fig. 1. At 2.8 eV, the highest initial relative energy experiment shown, the product distribution is clearly asymmetric, and the highest intensity of NH^+ occurs very close to the spectator stripping velocity. This result is consistent with asymmetric distributions found in earlier work² with a different apparatus, and in the other experiments of comparable relative energy performed with the present apparatus.

As the rest of the intensity maps in Fig. 1 show, when the initial relative energy is decreased, the intensity peak broadens and moves to smaller barycentric velocities. The relative intensity at the center-of-mass velocity and at the larger angles increases steadily. At the lowest energy, 0.79 eV, the distribution is quite symmetric, with the intensity peak occurring at the center-of-mass velocity.

Discussion

As is well-known, a product velocity vector distribution which is asymmetric about the $\pm 90^\circ$ axis in the center-of-mass

system indicates that the reaction proceeds by a direct, short-lived interaction in which the collision partners are close for a time comparable to or less than a molecular rotational period. It is clear from Fig. 1 that at relative energies above 2 eV, the $N^+(H_2,H)NH^+$ reactions proceeds by a direct interaction mechanism.

The situation at lower collision energies is potentially more complicated, due to the increasing symmetry of the observed product distributions. A product distribution which is symmetric about the $\pm 90^\circ$ axis is consistent with the occurrence of a "long-lived" collision complex in which the collision partners are close to each other for a time which is long compared to the characteristic rotational period. The symmetry of the distribution results from the fact that the strongly interacting collision complex may decay to products at a rotational phase which is apparently randomly distributed with respect to the phase at the beginning of the interaction. While a symmetric product distribution is consistent with the occurrence of a long-lived complex, it does not necessarily indicate that such an extended interaction occurs. It is conceivable that a system may possess a potential energy surface which produces a highly symmetric product distribution, even though all the reactive trajectories are of the direct interaction type. In addition, slight asymmetries in the true product distribution can be masked by such effects as detector resolution and finite velocity distributions of the reactants. These factors can be

particularly important for endoergic reactions where the products may be confined by energy conservation to regions very close to the center-of-mass velocity.

The distributions of Fig. 1 show an increasing degree of symmetry as the initial relative energy is lowered. Not only does the intensity peak move toward the center-of-mass velocity, the intensity contours at the intermediate and higher velocities become more symmetric. This behavior is quite different from that ascertained for the $O^+(H_2,H)OH^+$ reaction,^{3,4} and is very similar to that observed in a study of the $C^+(H_2,H)CH^+$ reaction.¹ This suggests that at the lower energies, the $N^+(H_2,H)NH^+$ reaction proceeds to an increasing extent through a long-lived collision complex. Because of the considerations expressed in the previous paragraph, this conclusion must be tempered with caution. However, the marked departure of the intensity peak from the spectator stripping location clearly indicates that a stronger coupling and energy exchange between all atoms occurs in many of the collisions at lower energy.

In view of the fact that the ground state of symmetric NH_2^+ lies approximately 6 eV below the minimum energy of reactants and products,¹⁰ it is surprising that there is no evidence that the $N^+(H_2,H)NH^+$ reaction proceeds by a long-lived complex mechanism at relative energies above 2 eV. This is particularly true when one considers that the product distributions from the $C^+(H_2,H)CH^+$ reaction¹ show considerable

symmetry at relative energies as high as 3.5 eV, even though this system has a potential energy well which is only 4.4 eV deep. Thus our interpretation of the data for the $\text{N}^+(\text{H}_2, \text{H})\text{NH}^+$ reaction is that while there is a substantial indication of the occurrence of a long-lived collision complex mechanism at relative energies near and below 1 eV, it remains to be explained why this persistent complex is not in evidence at still higher energies.

The answer to this question lies in the details of the potential energy surface which controls the reaction dynamics. The salient features of potential energy surfaces can often be deduced by consideration of correlation diagrams which connect the electronic states of reactants, intermediates, and products. Construction of a correlation diagram for the $\text{N}^+(\text{H}_2, \text{H})\text{NH}^+$ reaction is facilitated by the results of ab initio calculations¹¹⁻¹³ on NH , NH^+ , and NH_2^+ , as well as the standard thermodynamic and spectroscopic information¹⁰ that is available. In addition, the existence of a correlation diagram¹ for the C^+-H_2 system which is consistent with experimental data and with ab initio calculations¹⁴ of the potential energy surfaces is very helpful, since the properties of some of the states of the N^+-H_2 system should be closely related to those of the C^+-H_2 system.

Figure 2 shows the correlation diagram for the N^+-H_2 system. Correlations are shown explicitly for collinear ($\text{C}_{\infty\text{v}}$) and isosceles ($\text{C}_{2\text{v}}$) approaches of N^+ to H_2 . The behavior of the system for the more general C_s conformations can be

inferred by making appropriate alterations to the surfaces of higher symmetry.

The collinear approach of $N^+(^3P_g)$ to $H_2(^1\Sigma_g^+)$ gives rise to $^3\Sigma^-$ and $^3\Pi$ states of the collinear complex. The former of these has, aside from the 1s and 2s electrons of nitrogen, two non-bonding atomic 2p electrons with parallel spins, and two electrons in a bonding orbital which covers all three nuclei. The latter has only one 2p non-bonding electron, and three σ electrons, one of which is in an orbital which is antibonding between the terminal atoms. Thus the $^3\Sigma^-$ state is expected to lie lower than the $^3\Pi$ states, and must correlate to the products $NH^+(^4\Sigma^-) + H$. The $^3\Pi$ states must correlate to $NH^+(^2\Pi)$ and H . On the basis of their electronic configurations, neither of these states is expected to have a deep potential energy well or a high potential barrier between reactants and products. In fact, the $^3\Sigma^-$ and $^3\Pi$ states of N^+-H_2 are related respectively to the $^2\Pi$ and $^2\Sigma$ states of the C^+-H_2 system by the addition of a non-bonding $2p\pi$ electron. Ab initio calculations on the C^+-H_2 system show¹⁴ only a small (~ 0.5 eV) potential energy well for the $^2\Pi$ state, and a small (~ 0.5 eV) activation energy barrier for the $^2\Sigma$ state. Since these features can be expected to be largely preserved in the corresponding states of N^+-H_2 , we can conclude that collinear or near collinear approaches of N^+ to H_2 offer the possibility of product formation at any relative energy by a direct interaction mechanism.

Approach of $N^+(^3P_g)$ to $H_2(^1\Sigma_g^+)$ in the symmetric triangular (C_{2v}) geometry produces potential energy surfaces of 3B_1 , 3B_2 , and 3A_2 symmetry. It is profitable to compare the electron configuration of these asymptotic reactant surfaces with that of the 3B_1 electronic ground state of NH_2^+ . Apart from the 1s electrons of nitrogen, the configuration of this latter state is $(2a_1)^2 (1b_2)^2 (3a_1)^1 (1b_1)^1$. The $2a_1$ and $1b_2$ orbitals have strong N-H bonding properties, the $1b_1$ (π) orbital is of non-bonding character, while the $3a_1$ orbital is somewhat bonding or largely non-bonding, depending on the HNH angle. On the asymptotic 3B_1 surface, the reactant configuration corresponds to $(2a_1)^2 (3a_1)^2 (1b_1)^1 (4a_1^*)^1$, in which the $1b_2$ bonding orbital of NH_2^+ is empty, and one electron is in a strongly antibonding $4a_1^*$ orbital. We expect, therefore, that as N^+ approaches H_2 on the 3B_1 surface, the potential energy must first rise rather steeply. This asymptotic 3B_1 surface will eventually attempt to cross the 3B_1 surface which has the electron configuration of the equilibrium ground state of NH_2^+ , and which correlates in lowest order to the electron configuration of $N(^2D)$ and $H_2^+(^2\Sigma_g^+)$. Because these states are of the same symmetry, the crossing is avoided. Thus, after having risen, the lowest adiabatic 3B_1 surface abruptly drops as the equilibrium geometry of symmetric NH_2^+ is approached.

The 3B_1 surface of N^+-H_2 is related to the 2A_1 surface of C^+-H_2 by addition of a $1b_1$ non-bonding electron. The ab initio calculations of Liskow, Bender, and Schaefer¹⁴

on the C^+-H_2 system show that the lowest 2A_1 surface in fact has a barrier of approximately 3 eV before it drops to the potential well which corresponds to the equilibrium ground state of CH_2^+ . The behavior of the 3B_1 surface of the N^+-H_2 system should be very similar, and thus this state does not offer a low energy path from the reactants to the deep potential energy well of NH_2^+ .

When $N^+(^3P_g)$ approaches H_2 on the 3B_2 surface, the valence electron configuration is $(2a_1)^2 (1b_2)^1 (3a_1)^2 (4a_1^*)^1$. Despite the presence of three bonding electrons, this still corresponds to a highly excited configuration of NH_2^+ near the geometry of the equilibrium ground state. Thus it is unlikely that on this 3B_2 surface there are any deep wells of the type that can give rise to a long-lived collision complex.

In the reactant region, the electron configuration of the 3A_2 state of N^+-H_2 is $(2a_1)^2 (1b_2)^1 (3a_1)^2 (1b_1)^1$. Since there are no antibonding electrons present, and only one of the lowest lying bonding electrons is absent, it is likely that this state has, at least initially, a rather constant potential energy as N^+ approaches H_2 . Support for this conclusion comes from the observation that the 3A_2 state of N^+-H_2 is related to the 2B_2 state of C^+-H_2 by the addition of a non-bonding $1b_1$ electron, and calculations¹⁴ show that this 2B_2 surface is flat at large C^+-H_2 separations, and rises as the equilibrium ground state geometry of CH_2^+ is approached. There is no reason to believe that the 3A_2 state of N^+-H_2 will differ appreciably in its behavior.

It is evident that none of the three surfaces generated by $N^+(^3P_g)$ and $H_2(^1\Sigma_g^+)$ offers a low energy path from reactants to the deep ground state potential energy well of symmetric NH_2^+ when the system is constrained to C_{2v} geometry. However, an important change occurs when distortions to the more general scalene triangular geometry (C_s) are considered. The crossing of the 3A_2 and 3B_1 surfaces which occurs for C_{2v} geometry becomes avoided in the less symmetric conformations, since in C_s symmetry, both these states become $^3A''$. Consequently, a system initially on the 3A_2 - $^3A''$ reactant surface has available to it an adiabatic low energy path to the 3B_1 - $^3A''$ ground state potential energy well of NH_2^+ . Since the 3A_2 - $^3A''$ part of the surface is flat, and the 3B_1 - $^3A''$ part drops in energy as reactants approach, it is very unlikely that there are any substantial potential barriers between reactants and the NH_2^+ intermediate. A very similar situation exists in the C^+-H_2 system,¹ where the crossing of the 2B_2 and 2A_1 states becomes avoided in C_s symmetry, and opens a low energy path to the CH_2^+ intermediate. As noted earlier, the 3A_2 and 3B_1 states of N^+-H_2 are related to the 2B_2 and 2A_1 states of C^+-H_2 by the addition of a non-bonding $1b_1$ electron, so the similarities between these sets of surfaces in the two systems should be quite marked.

If the distance between the two hydrogen nuclei is regarded as fixed, the potential energy of the N^+-H_2 system can be represented in three dimensions as a function of two coordinates. These can be taken to be R , the distance from the center-of-mass

of H_2 to N^+ , and θ , the angle between this direction and the internuclear axis of H_2 . When θ is equal to $\pi/2$, the system has C_{2v} symmetry, and the 3A_2 and 3B_1 surfaces can intersect and cross each other. For $\theta < \pi/2$, the intersection is avoided, and there are upper and lower $^3A''$ surfaces. The latter connects the $^3A_2 - ^3A''$ reactants to the $^3B_1 - ^3A''$ ground state potential well of NH_2^+ . Thus for a fixed separation of the hydrogen nuclei, the intersection of the $^3B_1 - ^3A''$ surfaces is of the conical type.

Conical intersections of potential energy surfaces have been discussed by several authors.¹⁵⁻¹⁹ In general, one does not expect that such cones will be circular, but rather, the variation of the potential energy in the directions of the two orthogonal coordinates will be different. In the case at hand, if the potential energy of the system is not particularly sensitive to the value of θ , the splitting between the upper and lower $^3A''$ surfaces will not increase rapidly as θ departs from $\pi/2$, and the interior angle of the cones in this direction will be large. However, if the slopes of the 3B_1 and 3A_2 surfaces with respect to variations in R are quite different, the interior angle of the cone in this direction will be rather small.

The analysis of Teller¹⁵ demonstrates how the properties of a conical intersection can affect the behavior of a dynamical system. In the limit of low initial relative velocity, the system moving on the lower surface will in general avoid the region of the apex of the cone, where the separation of the

two surfaces is small and vanishing. As a consequence, it will most often behave adiabatically and remain on the lower of the two surfaces. A system with higher initial relative energy can visit the region near the conical intersection. As a consequence of the small separation between the surfaces and the finite nuclear velocity, there may be a considerable probability that the system will make a transition to the upper branch of the cone. The probability of a transition will be large if the conical intersection is such that the two surfaces are close for a substantial range of the coordinate θ , but separate rapidly as R is changed. In the N^+-H_2 system, such diabatic behavior at higher velocity will result in product formation by a direct interaction mechanism, just as is observed experimentally. In the limit of low velocities, almost all systems initially on the $^3A_2-^3A''$ surface will behave adiabatically, visit the $^3A''-^3B_1$ potential energy well, and display the characteristics of a long-lived collision complex. This too is consistent with experimental observations.

The potential energy surfaces of the N^+-H_2 system, as far as they can be deduced from electronic state correlation diagrams, offer an explanation of why the $N^+(H_2,H)NH^+$ reaction proceeds by a direct interaction mechanism at initial relative energies that are considerably less than the maximum potential energy well depth. The same features of the potential surfaces also provide an explanation of why indications of a long-lived complex mechanism are observed in experiments performed at

the lowest relative energies. The validity of these explanations can perhaps best be assessed by an accurate calculation of the surfaces in the vicinity of the 3B_1 - 3A_2 conical intersection, and an examination of the computed dynamics on these surfaces at several initial relative energies.

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Figure Captions

Figure 1. Intensity contour maps of the velocity vector distribution of NH^+ from the $\text{N}^+(\text{H}_2, \text{H})\text{NH}^+$ reaction at six initial relative energies. The quantity plotted is the specific intensity of product (Cartesian flux) on a coordinate system whose origin is the center-of-mass velocity of the system. The zero angle is taken as the original direction of the N^+ projectile. Note that as the initial relative energy is lowered, the maximum intensity moves from the spectator stripping velocity (marked by a small cross) toward the centroid velocity, and the symmetry of the distributions with respect to the $\pm 90^\circ$ axis increases.

Figure 2. An electronic state correlation diagram for the low lying surfaces of the $\text{N}^+ - \text{H}_2$ system. At the far right, a collinear approach of N^+ to H_2 is assumed. At the far left, N^+ approaches H_2 along the perpendicular bisector of the hydrogen molecule. Surface crossings that are avoided in C_{2v} symmetry are indicated by straight dashed lines. Surface crossings that become avoided in C_s symmetry are indicated by curved dashed lines.

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Fig. 1

Fig. 2

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