

INELASTIC SCATTERING OF Ne^+ BY H_2 AND D_2

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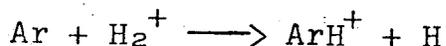
Inorganic Materials Research Division of the
Lawrence Berkeley Laboratory and Department of
Chemistry, University of California, Berkeley.

ABSTRACT

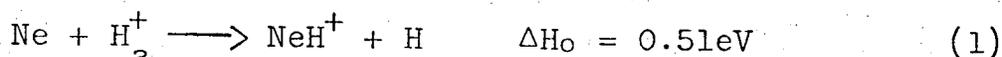
The reaction $\text{Ne}^+(\text{H}_2, \text{H})\text{NeH}^+$ is notable as a simple exoergic ion-molecule reaction which has a very small rate constant, less than 8×10^{-12} cc/sec. We have studied the velocity vector distributions of Ne^+ scattered non-reactively from H_2 and D_2 at a number of energies. There is no evidence for the occurrence of reactive or charge transfer collisions. The small angle ($<90^\circ$) scattering is elastic, but there is large angle inelastic scattering of three types: vibrational and rotational excitation of hydrogen, collisional dissociation to $\text{Ne}^+ + \text{H} + \text{H}$, and electronic excitation of hydrogen to $\text{B}^1\Sigma_u^+$ and perhaps other states. The results are consistent with conclusions drawn from a molecular orbital correlation diagram for the $(\text{Ne}-\text{H}_2)^+$ system.

The phenomena which occur in partially ionized mixtures of the various noble gases with hydrogen have played a central role in the development of ion-molecule reaction kinetics¹.

The reactions



and their isotopic variants have been studied extensively by a number of workers²⁻⁵ who have employed a variety of techniques. Both reactions are known to be rapid and exoergic, with thermal rate constants close to the Gioumousis-Stevenson value. The reaction

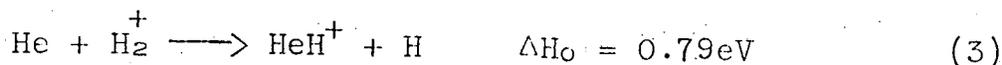


is endoergic, but possesses an appreciable reaction cross section once the necessary energy is supplied either as vibration of H_2^+ or relative translation⁴. In contrast, the reaction

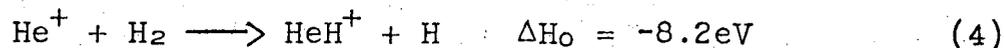


is exoergic as written, but has never been shown to occur. An upper limit for the room temperature rate constant has been set⁶ at 8×10^{-12} cc/sec, two orders of magnitude smaller than the Gioumousis-Stevenson value expected for such a simple exoergic reaction.

A similar situation prevails in the $(\text{He}-\text{H}_2)^+$ system. The endoergic reaction

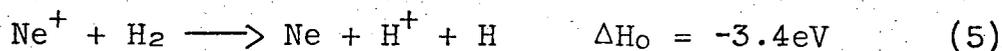


occurs with an appreciable cross section once the necessary energy is supplied either as vibration or translation⁴. On the other hand, the exoergic reaction



has never been observed with certainty.

Failure to observe reactions (2) and (4) has at various times been attributed^{7,8} to the fact that in each case their exoergicity greatly exceeds the bond energy of the product molecule, which might make it difficult to form product molecular ions which are stable with respect to dissociation. This argument leads to the expectation that dissociative charge transfer



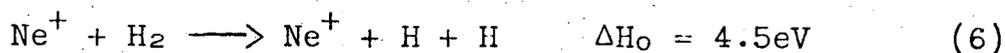
should be the dominant collisional process, since any incipient, energized NeH^+ in its electronic ground state should dissociate to Ne and H^+ . While some dissociative charge transfer has evidently been observed in the $\text{Ne}^+\text{-HD}$ system⁹, the estimated cross section of $2.3 \pm 1\text{\AA}^2$ is far smaller than the Gioumoussis-Stevenson value. Thus one is left with the conclusion that most $\text{Ne}^+\text{-H}_2$ collisions are non-reactive. The same conclusion applies to the $\text{He}^+\text{-H}_2$ system⁹, where the thermal dissociative charge transfer cross section is even smaller, 0.6\AA^2 or less.

An explanation based on molecular orbital correlation diagrams for the lack of reactivity of He^+ with H_2 has been given by Mahan¹⁰. Figure 1 shows a similar qualitative

correlation diagram which applies to the linear Ne^+-H_2 system. Because the lowest three orbitals of the reactants and products are well separated in energy, a facile and unique correlation may be made between them, and the fate of a particular reactant electron configuration can be predicted with some confidence. The $2p\sigma$ orbital of Ne combined in-phase with the $1\sigma_g$ orbital of H_2 forms the lowest bonding orbital of the system, and correlates with the in-phase combination of $\text{H}(1s)$ and the 3σ bonding orbital of NeH^+ . The out-of-phase combination of these reactant orbitals yields an orbital of initially increasing energy correlating diabatically with the excited $4\sigma^*$ orbital of NeH^+ , which has antibonding character. The in-phase combination of the $1\sigma_u^*$ antibonding orbital of H_2 with Ne $2p\sigma$ produces a three center orbital of initially decreasing energy which correlates diabatically with $\text{H}(1s)$ and its out-of-phase combination with the σ bonding orbital of NeH^+ . Configuration interaction removes the crossing of these two upper orbitals, and yields the qualitative adiabatic curves of Figure 1.

Reactants initially in the Ne^+-H_2 configuration correlate adiabatically with NeH^{++} and H^- . Even if NeH^{++} were to be stable against dissociation these products would lie at a very high energy, and would not be observed. Another possible product configuration which can be reached diabatically is $\text{H}(1s)$ and $\text{NeH}^+ (3\sigma)^1(4\sigma^*)^1$. The states of NeH^+ arising from this configuration are very probably unbound or weakly bound¹¹, since in the united atom limit they correlate with the high energy configuration $\text{Na}^+(2p)^5(3s)^1$. In the separated atom limit, these states correlate to $\text{Ne}^+(2p)^5$ and $\text{H}(1s)$.

Thus the process



should be of some importance at relative energies sufficient to effect the dissociation of hydrogen.

Another product configuration accessible diabatically from the $\text{Ne}^+ - \text{H}_2$ reactants is $\text{Ne}^+(2p)^5$ and $\text{H}_2(1\sigma_g)^1(1\sigma_u)^1$. The states of hydrogen derived from this configuration are the lowest triplet state $b^3\Sigma_u^+$ which is repulsive and dissociates to two ground state hydrogen atoms^{1,2}, and the $B^1\Sigma_u^+$ state, which separates to $\text{H}(1s)$ and $\text{H}(2s)$. Excitations of the $B^1\Sigma_u^+$ state involving approximately 11.2eV or greater inelasticity may therefore be expected at higher relative collision energies. Excitations to the $b^3\Sigma_u^+$ state may also be possible for initial relative energies above 4.5eV, but may be difficult to identify due to the wide range of excitation energies possible for non-vertical transitions to this repulsive state.

The correlation diagram makes it clear that the $\text{Ne}^+ - \text{H}_2$ configuration does not correlate adiabatically or diabatically with the ground state of NeH^+ , and thus the failure to observe these reaction products is understandable. Furthermore, the same argument (basically, the energy isolation of the $\text{Ne}^+ 2p$ orbital) suggests that the charge transfer channels should not be important, and the small (and relatively uncertain) cross section found for dissociative charge transfer is consistent with this expectation. The principal results of low energy (<4.5eV) collisions in the $\text{Ne}^+ - \text{H}_2$ system

should therefore be elastic and vibrationally inelastic scattering. At higher energies we expect dissociative excitation, and excitation to the B \sum_u^+ state of H₂. The purpose of the work reported here was to test these predictions experimentally.

EXPERIMENTAL

The instrument used in this work consists of a magnetic mass spectrometer for preparation of a collimated beam of primary ions of known mass and energy, a scattering cell to contain the target gas, an ion detection train made up of an electrostatic energy analyzer, a quadruple mass spectrometer, and an ion counter. The detector components and the exit aperture of the scattering cell are mounted on a rotatable lid which permits simultaneous angular and energy measurements of the ion products. The major components of the apparatus have been described previously¹³. In all important respects, the constitution and operation of the apparatus and the data acquisition and reduction techniques were the same as we have used in earlier work.

The primary ions were extracted from a microwave discharge through neon gas which contained small amounts of helium. The excited configurations of Ne⁺ lie 36eV above the ground state, so no such species will be found in the momentum analyzed ion beam, due to the low electron temperature of the microwave discharge. The two states $^2P_{3/2}$, $^2P_{1/2}$ of the lowest

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configuration of Ne^+ differ in energy by only 0.097eV, and are probably both populated.

RESULTS AND DISCUSSION

Figures 2-4 show contour maps of the scattering of Ne^+ from H_2 at three different initial relative energies. At the lowest relative energy, 4.75eV, (Figure 2) the Ne^+ intensity maximizes at or near the $Q = 0$ circle at all angles, so the scattering appears to be elastic, with perhaps some slight inelasticity appearing at angles near 180° . Moreover, the angular variation of intensity is of the general type that we have found for several non-reactive systems at moderate energies^{14,15}. There are no intensity variations with angle which would suggest the abrupt onset of dissociative charge transfer or reaction at the smaller impact parameters. The general appearance of the low energy non-reactive scattering is therefore consistent with the small cross sections that have been estimated for these reactive and charge transfer processes.

In Figure 3, the scattering appears to be perfectly elastic at angles smaller than 90° . Near 180° , however, the locus of maximum intensity clearly lies inside the $Q = 0$ circle. Thus head-on collisions do lead to noticeable inelasticity at this higher energy. This large angle inelastic scattering becomes particularly obvious in the map of Figure 4, where the initial relative energy is rather high, 18.1eV. Even at this high relative energy, the small angle scattering is quite elastic.

The contour maps of the scattered intensity give a general picture of the nature of the collision process. However, in constructing this type of map, some of the details of the intensity variations are suppressed by smoothing and interpolation processes. In order to reveal more of the detailed nature of the inelastic scattering, intensity profiles of Ne^+ scattered at 180° were obtained as functions of Q , the difference between final and initial relative energies. These intensity profiles in general resemble distorted and substructured gaussians, but because of the finite angular and energy resolution of the apparatus and the isotropic motion of the target gas molecules, the inelastic structure is rarely clearly resolved. Consequently, we have attempted to obtain a better approximation to the true scattering function from the experimental data by deconvolution procedures which make use of our knowledge of the ion beam profiles and detector bandpass function.

Some preliminary refinement of certain of the data was obtained with an iterative deconvolution technique¹⁶. At its best, this method revealed the most prominent structure of the inelastic scattering function, but its reliability depended sensitively on the quality and spacing of the experimental data points. Noisy or limited data often produced spurious structure in the unfolded scattering function. In contrast, the integration fitting method¹⁷ of

deconvolution proved more generally applicable and reliable, and therefore was used to obtain approximations to the true scattering functions for all the experiments reported in this paper. In the integration fitting method, a functionality for the true differential scattering cross section is assumed, and then convoluted with the primary ion beam profile and detector band pass function to obtain a calculated laboratory intensity profile. The calculated laboratory intensities are then compared with experimental data, the parameters of the scattering function adjusted accordingly, and the convolution repeated until a satisfactory fit to the experimental data is obtained. The iterative unfolding and integration fitting methods of deconvolution have been discussed and compared by Gillen and Mahan¹⁸, who also gave examples of the application of the integration fitting method to elastic and inelastic large angle scattering.

In the present work, the scattering function for a barycentric angle of 180° was assumed to be of the form

$$F(Q) = \sum_{i=1}^n h_i \exp \left[-(Q - Q_i)^2 / 2\sigma_i^2 \right]$$

where n , the number of peaks, ranges from one to four, and h_i , Q_i , and σ_i are the height, location, and width of the i 'th peak. The number of peaks and the three parameters of each were taken as totally disposable, but in choosing initial values of the number of peaks and their location, we were guided by the known electronic energy levels of hydrogen.

Figure 5 shows a comparison of the fit to the experimental

data of two scattering functions which differ principally by the inclusion in one of them of an inelastic feature located at $Q = -4.6\text{eV}$. It is clear that the fit of the calculated intensity to the experiment is considerably improved when the inelastic feature is added to the scattering function. Moreover, it must be emphasized that the improvement displayed in Figure 5 is the least dramatic of all our results, since the amplitude of the inelastic feature is the second smallest of any we found necessary to add to the scattering function. Table 1, which gives the parameters of the best scattering functions found by deconvolution, shows that in the higher energy experiments the amplitudes of the various inelastic features were more nearly comparable, and thus their presence and placement in the experimental data was more obvious.

It will be noted in Figure 5 that the fit of the calculated intensity to the experimental data is poor at the extreme wing of the intensity profile in the region of positive Q . The principal reason for any intensity in this region is the failure of the electrostatic deflection energy analyzer to reject the unscattered primary beam perfectly, because of the scattering of a small fraction of the strong primary beam from the electrode surfaces in the analyzer. The high background at 0° laboratory angle in non-reactive scattering experiments has been a characteristic feature of this apparatus and of another in our laboratory. In the latter case, the background has been substantially

reduced by cutting a hole in the electric deflection energy analyzer which allows the largely undeflected primary beam to pass out of the analyzer without striking an electrode surface. This alteration is not possible in the present apparatus, and thus the extended wing of the intensity profile consists principally of insignificant background counts.

The experimental data represented in Figure 5 were smoothed using a third degree polynomial fit to five adjacent data points. Some smoothing procedure is essential to the iterative unfolding method if spurious features are to be avoided. Smoothing will, at worst, cause a slight bulging of a peak in the experimental data, but will not shift or otherwise distort it. Smoothing is not necessary in the integration fitting deconvolution method used in this paper, and in the majority of cases, we fit our calculated intensity directly to the unsmoothed experimental data.

Figure 6 gives as a function of initial relative energy the locations of the peaks in the deconvoluted scattering functions. At low relative energies ($<8\text{eV}$) only one peak appears, and corresponds to elastic or slightly inelastic scattering. As the energy increases, this first feature becomes more inelastic, with Q_1 values ranging to -3.5eV at the highest relative energy, 22.5eV . Values of Q_1 in this range correspond to vibrational and rotational excitation of H_2 and D_2 , and the fraction of the initial relative energy converted to internal excitation is qualitatively in agreement with Secrest's calculation¹⁹ of Q/E_{rel} based on hard sphere

collinear collisions of a mass 23 atom with H₂ and D₂.

At an energy of 8eV, a second peak appears, initially with a Q₂ value of -4.5eV. As the initial relative energy increases, the Q₂ of this peak also becomes more negative, eventually reaching -6.9eV. Table 1 shows that σ₂, the width parameter for this peak, increases markedly as the relative energy increases, in contrast to the width parameters of the other peaks. The location and broadening of the second peak suggests that it corresponds to a dissociation of hydrogen, either through a non-vertical excitation of the repulsive b ³Σ_u⁺ state, or by formation of one of the states of the NeH⁺ (3σ)¹(4σ*)¹ configuration, which dissociate to Ne⁺ and H. Excitation of the triplet repulsive state of hydrogen by the doublet neon ion is spin-allowed, and in a head-on collision which can involve considerable nuclear motion in the hydrogen molecule, the non-vertical nature of the transition may not be unreasonable. A vertical excitation of hydrogen to the b ³Σ_u⁺ state would require Q = -10.2eV, and no such feature is found in the experimental energy spectrum. In the absence of any detailed information about the excited states of NeH⁺ or the potential energy surface for Ne⁺ and H₂, it seems at least equally reasonable that the second peak involves transient formation of excited NeH⁺, as suggested by the correlation diagram, followed by dissociation to Ne⁺ and H.

In the relative energy range from 18 to 22eV, the third and fourth inelastic peaks become prominent. The third peak has $Q_3 = -11.5\text{eV}$, independent of initial relative energy over this rather short range. The minimum excitation energy of the $B \ ^1\Sigma_u^+$ state is 11.3eV, while vertical excitation of this state requires 12eV. According to the correlation diagram, the $B \ ^1\Sigma_u^+$ state should be easily accessible from the Ne^+-H_2 configuration, and therefore this assignment seems to be the most reasonable for the third inelastic peak.

The fourth peak has $Q_4 = -14.8\text{eV}$, again relatively independently of the initial collision energy. There are several states of hydrogen which have repulsive walls lying 14 to 16eV above the ground state at internuclear distances approximately equal or smaller than the equilibrium bond distance. The ones which are known (including the $B \ ^1\Sigma_u^+$ state) either dissociate directly to, or cross, the asymptote of $\text{H}(1s) + \text{H}(2\ell)$ for energies greater than 14.5eV above the ground state molecule. It seems likely, therefore, that the fourth peak at $Q_4 = -14.8\text{eV}$ corresponds to excitation of hydrogen to one or more of the several states which dissociate to one normal and one excited atom.

In summary, we can conclude that the predictions based on the molecular orbital correlation diagram for Ne^+-H_2 are largely substantiated by the non-reactive scattering experiments. Most of the Ne^+-H_2 scattering is non-reactive and does not involve charge transfer. There is inelastic scattering of

three types: vibrational and rotational excitation of hydrogen, collisional dissociation either through the repulsive triplet state of hydrogen or by formation of unstable, electronically excited NeH^+ , and electronic excitation of hydrogen to $B^1\Sigma_u^+$ and perhaps other states. The notable lack of reactivity of the Ne^+-H_2 system even in relatively high energy collisions is a consequence of the large ionization energy of Ne, which keeps the excited Ne^+-H_2 surfaces well separated from the ground state $\text{Ne}-\text{H}_2^+$ surface, apparently the only one that can lead to stable NeH^+ .

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Figure Captions

- Fig. 1 A correlation diagram for the lowest σ valence orbitals of the $(\text{Ne-H}_2)^+$ system.
- Fig. 2 A contour map of the specific intensity of Ne^+ scattered from H_2 at 4.75eV relative energy. A barycentric coordinate system is used, with angles measured with respect to the primary Ne^+ beam direction, and speed measured relative to the center of mass velocity. The quantity Q is the difference between final and initial relative energies.
- Fig. 3 A contour map of the specific intensity of Ne^+ scattered from H_2 at 13.7eV relative energy. Note that the intensity maxima near 180° lie inside the elastic $Q = 0$ circle, thereby indicating inelastic backscattering.
- Fig. 4 A contour map of the specific intensity of Ne^+ scattered from H_2 at 18.1eV relative energy. The inelastic feature at 180° is a composite of the four processes discussed in the text.
- Fig. 5 Intensity profiles of Ne^+ scattered at 180° from D_2 at 8.1eV relative energy. Comparison of the upper and lower panels shows the improvement of the fit of the calculated intensity profile (squares) to the experimental data (solid line) by adding a small inelastic feature to the scattering function $F(Q)$.
- Fig. 6 A plot of the positions of the maxima of the inelastic features in the scattering function as a function of initial relative collision energy.

Table I
Scattering Function Parameters^a

E_{rel}	h_1	$-Q_1$	σ_1	h_2	$-Q_2$	σ_2	h_3	$-Q_3$	σ_3	h_4	$-Q_4$	σ_4
4.7	1.0	0.3	1.1									
6.7	1.0	0.3	1.0									
8.2	1.0	0.22	1.0	0.15	4.6	0.20						
10.0	1.0	0.65	1.0	0.40	4.6	0.20						
10.4 ^b	1.0	1.2	2.2	0.10	4.6	0.60						
11.3	1.0	1.1	1.0	0.50	4.6	0.30						
12.2	1.0	1.2	1.2	0.50	4.6	0.40						
12.6 ^b	1.0	1.0	1.8	0.25	4.7	0.70						
15.0 ^b	1.0	1.5	1.8	0.40	5.2	0.80						
17.7 ^b	0.40	2.4	1.5	1.0	6.2	1.7	0.30	11.5	0.80	0.07	14.6	1.0
20.7 ^b	0.50	3.3	1.1	1.0	6.5	1.7	0.40	11.5	0.80	0.15	14.8	0.90
22.5 ^b	0.60	3.5	0.9	1.0	6.9	1.7	0.50	11.6	0.70	0.25	14.8	1.0

a) E_{rel} , Q , σ in eV.

b) Runs done with D_2 .

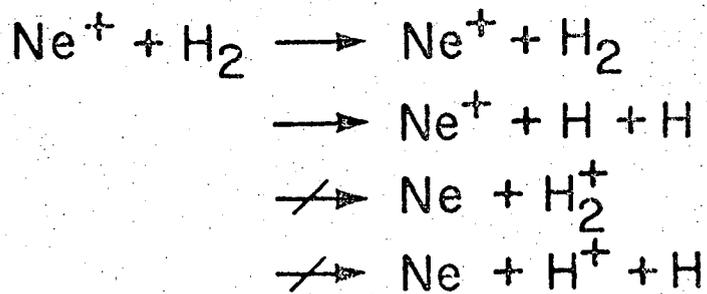
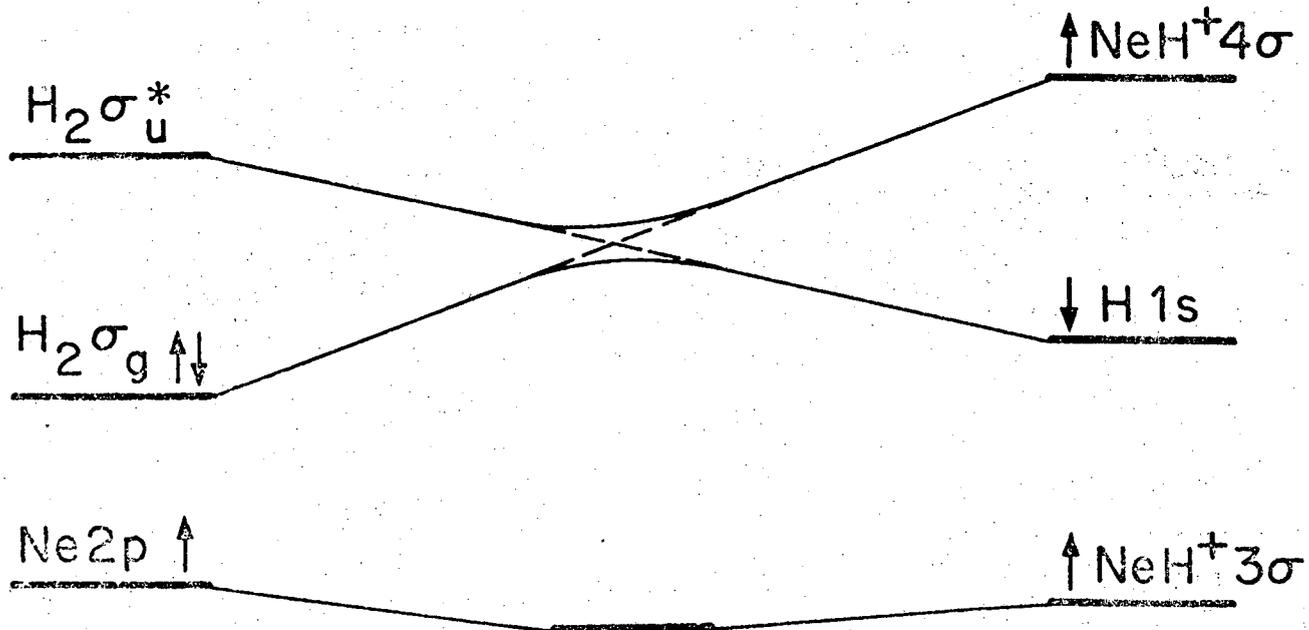
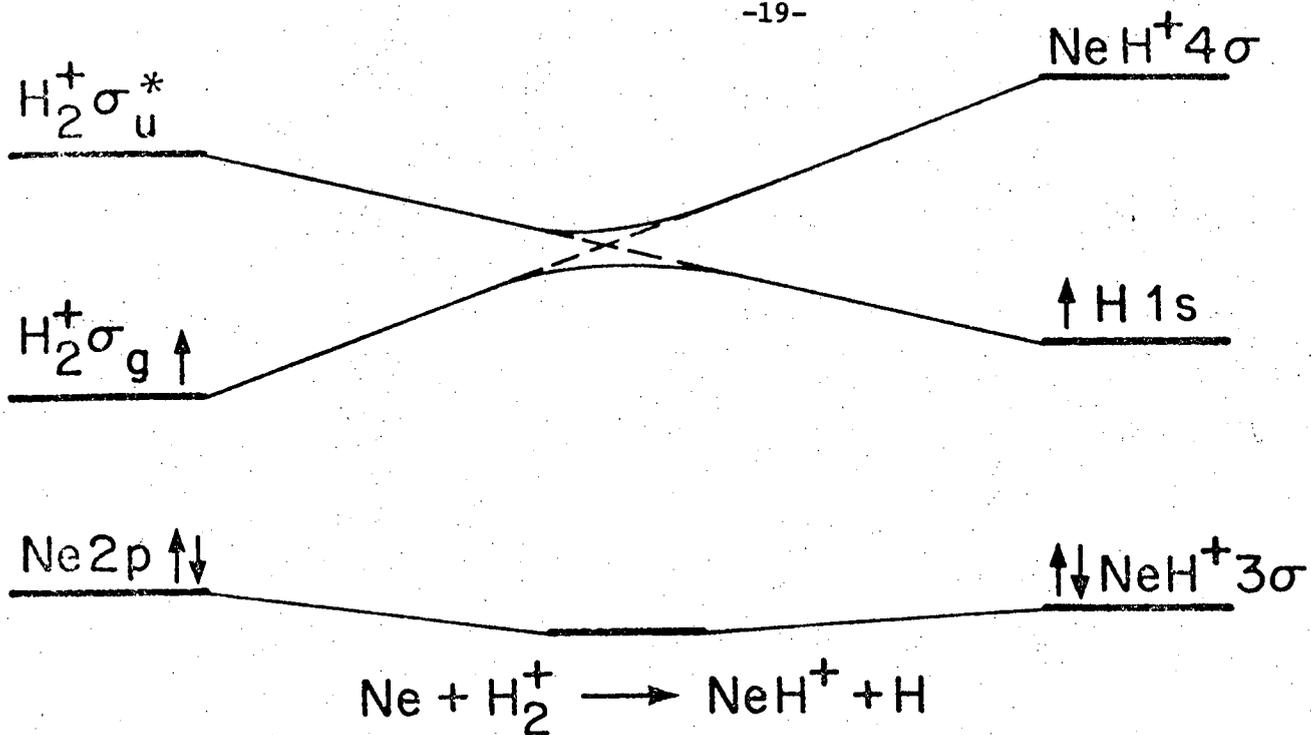


Fig. 1

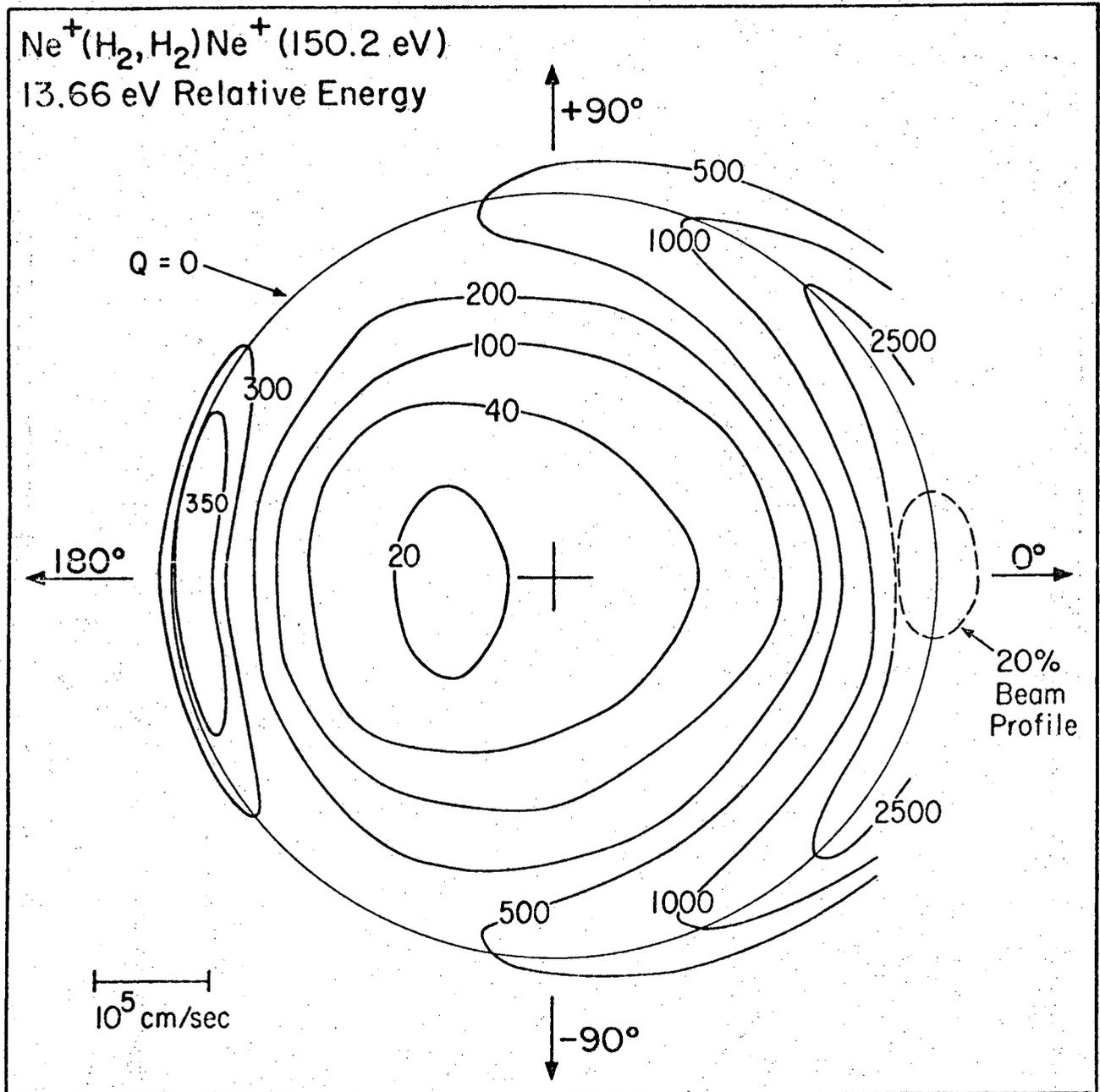


Fig. 3

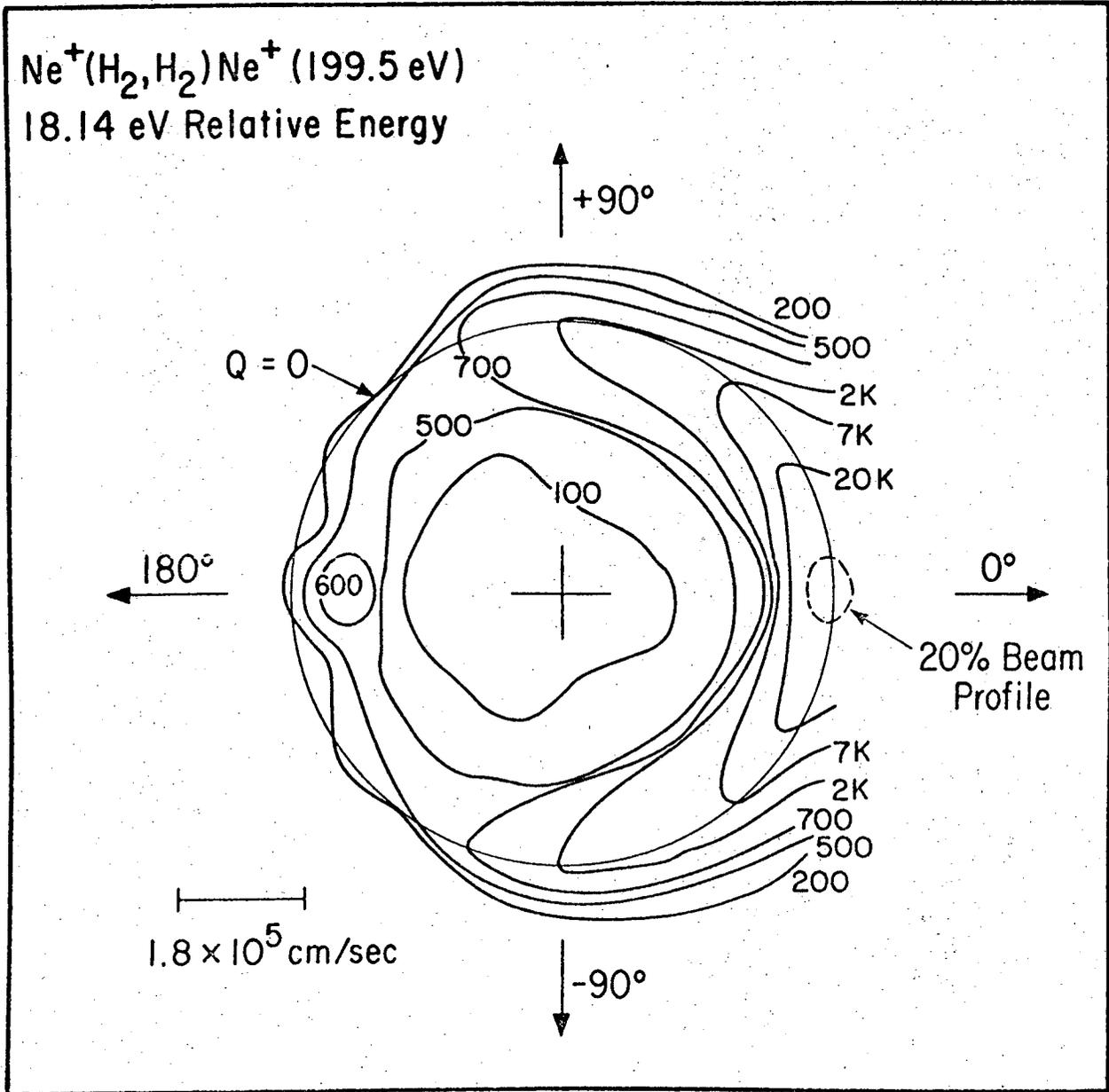
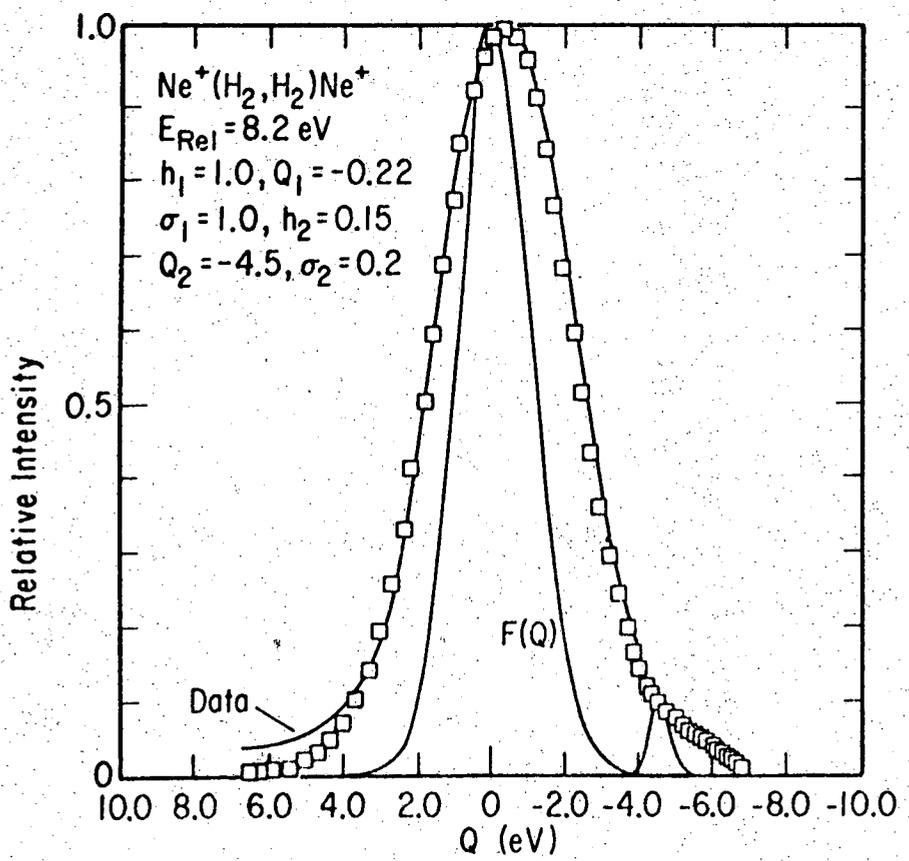
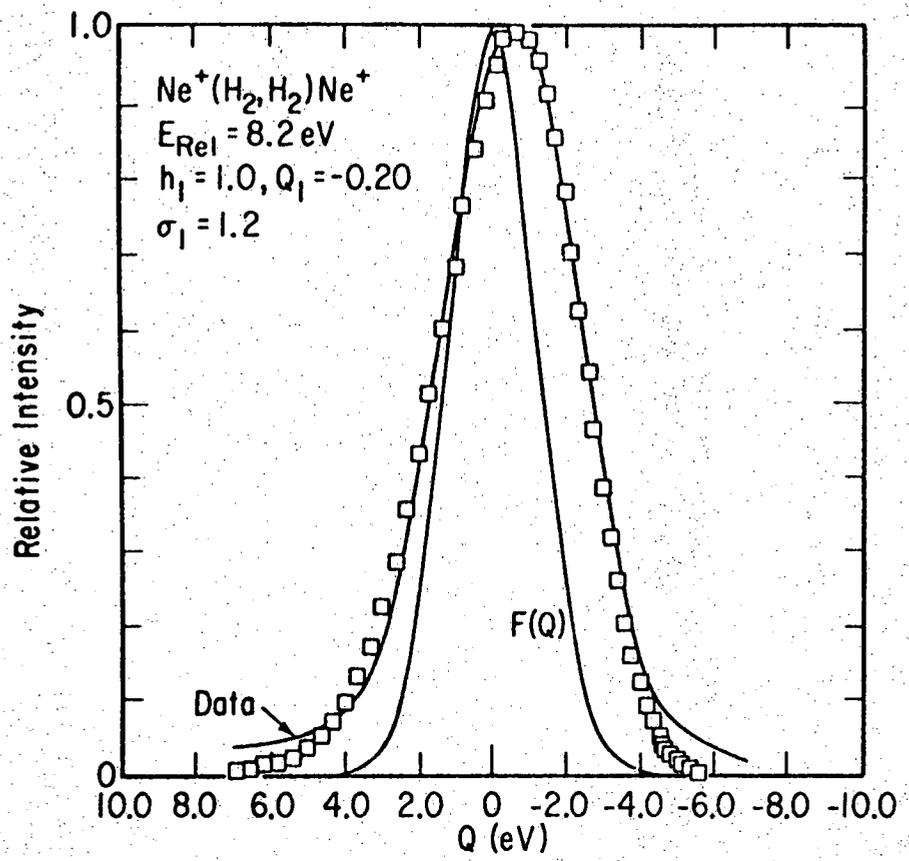


Fig. 4



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Fig. 5

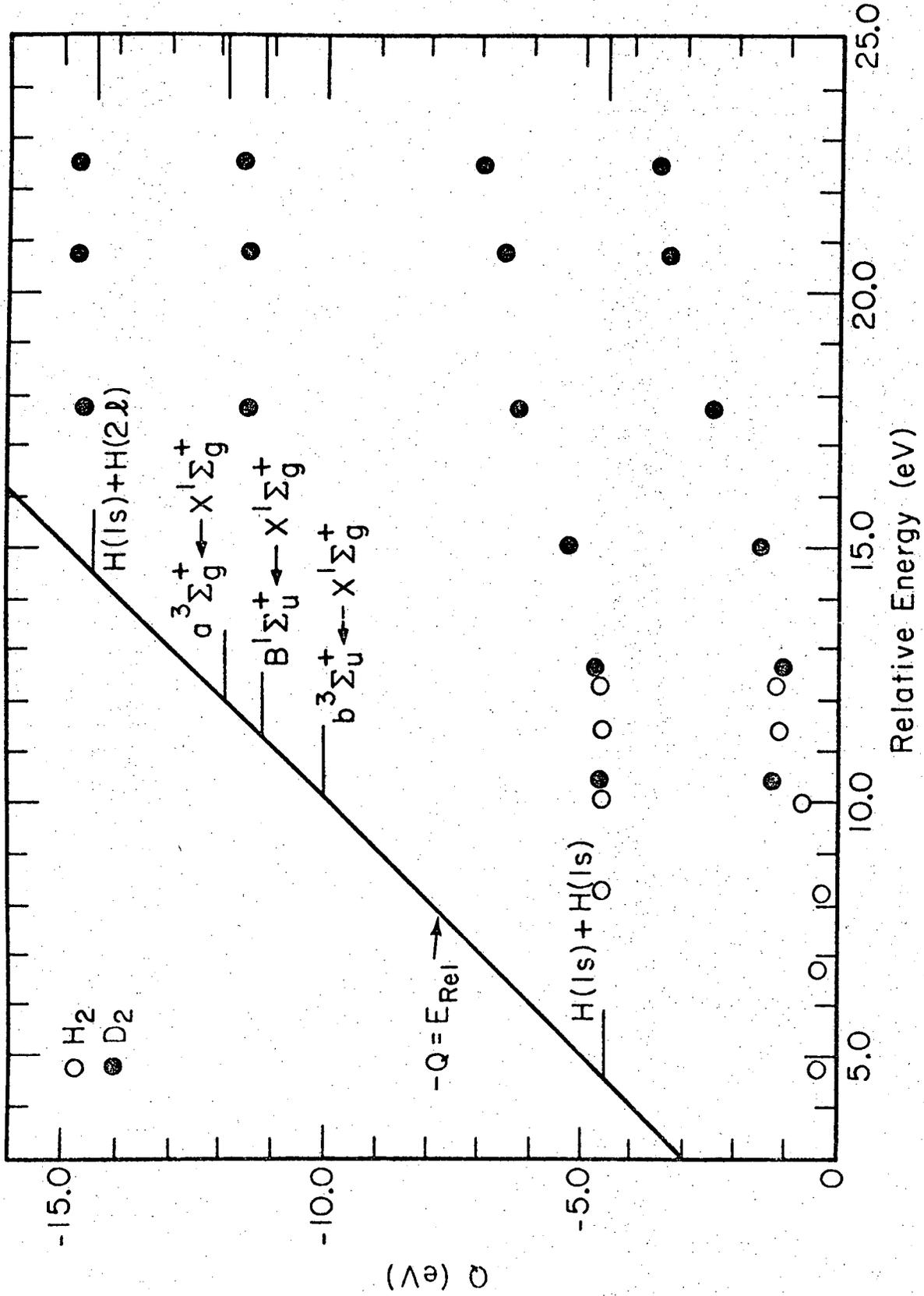


Fig. 6

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