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Generation of H^-, $H_2(v'')$, and H Atoms by H_2^+ and H_3^+ Ions Incident Upon Barium Surfaces

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I. Introduction

The generation of vibrationally excited molecules by electron excitation collisions and the subsequent generation of negative ions by dissociative attachment to these molecules has become a standard model for volume source operation.[1] These processes have been supplemented recently by the demonstration of atom-surface recombination to form vibrationally excited molecules[2,3], and enhanced negative ion formation by protons incident upon barium electrodes.[4] In this paper we consider the additional processes of molecular vibrational excitation generated by recombination of molecular ions on the electrode surfaces, and negative ion formation by vibrationally excited molecules rebounding from low work-function electrodes.

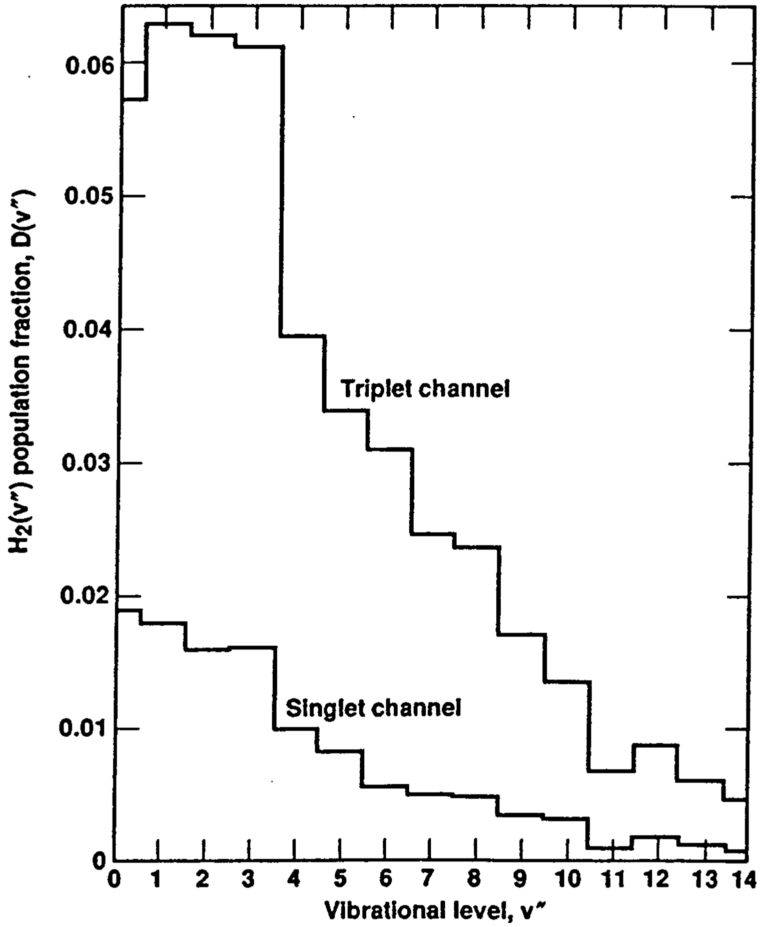

Figure 1: Population distribution per incident H_2^+ ion

II. Recombination and Dissociation of H_2^+ and H_3^+ on Surfaces

A four-step model has been developed for the formation of excited molecules, $H_2(v'')$, generated either by H_2^+ ions or H_3^+ ions incident upon metal surfaces.[5,6,7] The final vibrational distribution per incident H_2^+ ion is shown in Fig. 1 for the case of neutralized ions impinging on the surface with 4eV of translational energy, and an internal excitation distribution appropriate to H_2^+ neutralization. The distinguishing feature of this figure is the broad vibrational distributions extending across the entire spectrum of bound vibrational levels. The singlet channel refers to H_2^+ neutralization directly into the singlet ground state of the H_2 molecule, the triplet channel corresponds to neutralization proceeding through the lowest excited triplet state prior to ground state formation. The total production is the sum of these two distributions.

In Fig. 2 is shown the population distribution per incident ion for incident H_3^+ neutralized to form 6eV H_3, which in turn spontaneously dissociate into a 4eV H_2 molecule and a two eV H atom immediately prior to the "hard" collision with the surface. Here again the final population distribution spans the bound spectrum. A comparison of the distributions of Figs. 1,2 shows that the final population for $v'' \geq 5$ is larger in the case of H_3^+recombination than for H_2^+recombination. For this reason the H_3^+ ion appears to be a more useful ion leading to H^- formation than does the H_2^+ ion.

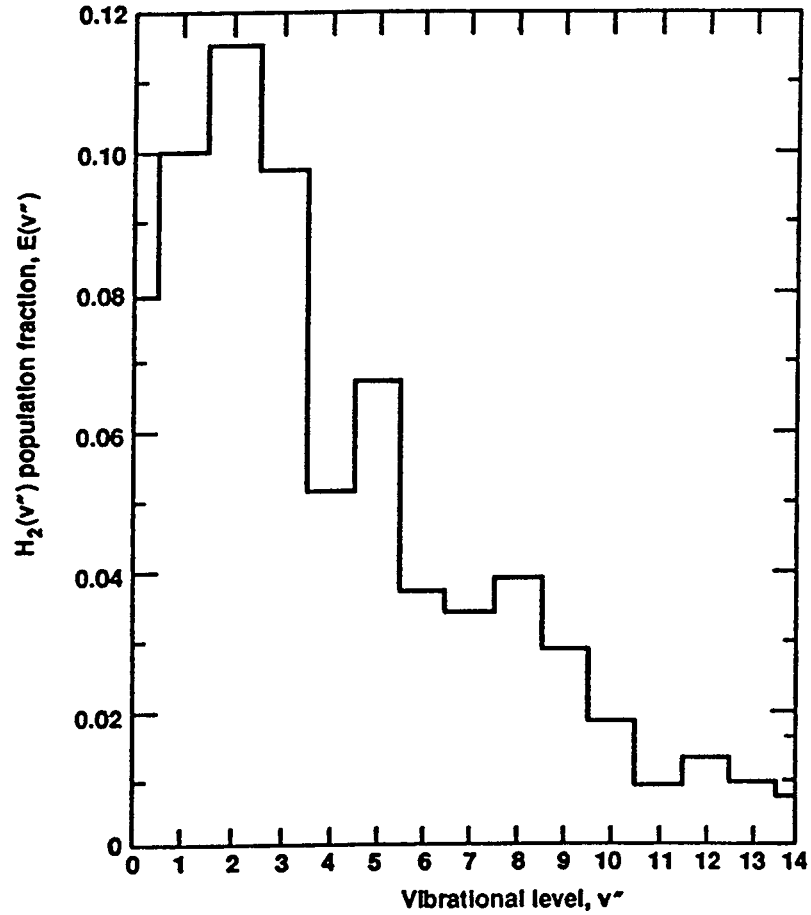

Figure 2: Population Distribution per Incident H_3^+ Ion

III. H_3^+ Ions Incident Upon Barium Surfaces

The material of this section is discussed in more detail in Refs. 6,7. We present here only a summary.

Molecular ion collisions with low work-function surfaces will allow for resonant captures into higher electronic states. These captures in turn will be Auger relaxed to provide an additional contribution to the ground state vibrational distribution. Low work-function collisions also allow for two additional mechanism for negative ion generation: Direct H^- production by electron capture to recombination product H atoms backscattering from the surface, and H^- production by dissociation of H_2^- ions formed from rebounding $H_2(v'')$ moving outward through the surface selvage.

The generation of H^- ions by H^+, H incident upon barium surfaces has been characterized experimentally by van Os et al.[4] In Fig. 3 their data is re-plotted for comparison with the product function

$$N(-) = a(H)\left[1 - \exp - v_\perp / v_\perp^\circ\right] , \qquad (1)$$

plotted against the outgoing perpendicular velocity component, $v_\perp$.

From this figure the asymptotic dependence on $v_\perp$ indicates a formation probability, a (H), equal to 0.3; the survival probability, given by the bracketed factor, remains as large as 0.33 for perpendicular energy components as low as 2.0 eV.

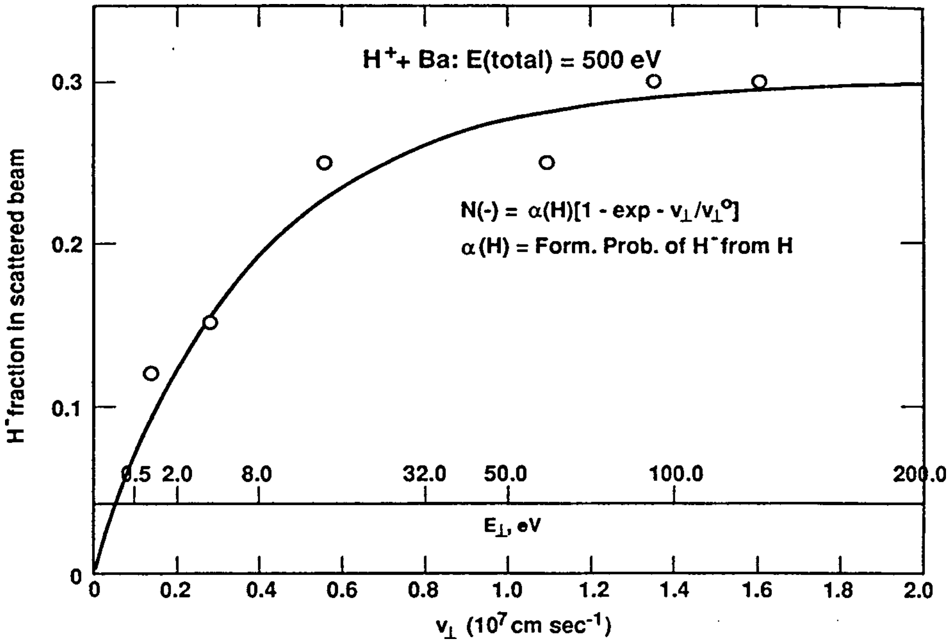

Figure 3: H^- generation vs outgoing normal velocity or energy, Eq.1.

In general one expects the energy level of a neutral atomic or molecular system to be raised as it experiences an increasing image field. Bruch and Ruijgrok[8] have shown however, that there is an insignificant level shift for image-plane separations $z - z_o > 3a_o$, the range where resonant electron capture would be expected to occur. For the downward shift of the negative ion level we use the expansion,

$$\Delta E = -\frac{1}{4}\frac{1}{z - z_o} - \frac{1}{2!}\alpha\frac{1}{16(z - z_o)^4} - \frac{1}{4!}\beta\frac{1}{256(z - z_o)^8} + \cdots \qquad (2)$$

The dipole and quadrupole polarizabilities, α, β for H^- are 215.5 and 7765.0, respectively.[9,10] No information is available for the polarizability of the H_2^- ion, but in a first approximation the H_2^- configuration is an H^- + H configuration from the level crossing outward, and for this discussion we shall approximate the H_2^- level shift by Eq. (2) and using the H^- polarizabilities.

The energetics of the barium H_2^-, $H_2(v'')$ system is illustrated isometrically in Fig. 4 where is shown the energy variations as a function of image-plane separation. The relative positions of the H_2^-, $H_2(v'')$ potentials are shown in the right hand plane for infinite surface-molecule separation. At this separation the H_2^- level lies too high to allow resonant capture from the barium (Ø = 2.7 eV) to $H_2(v'')$ to form H_2^-.

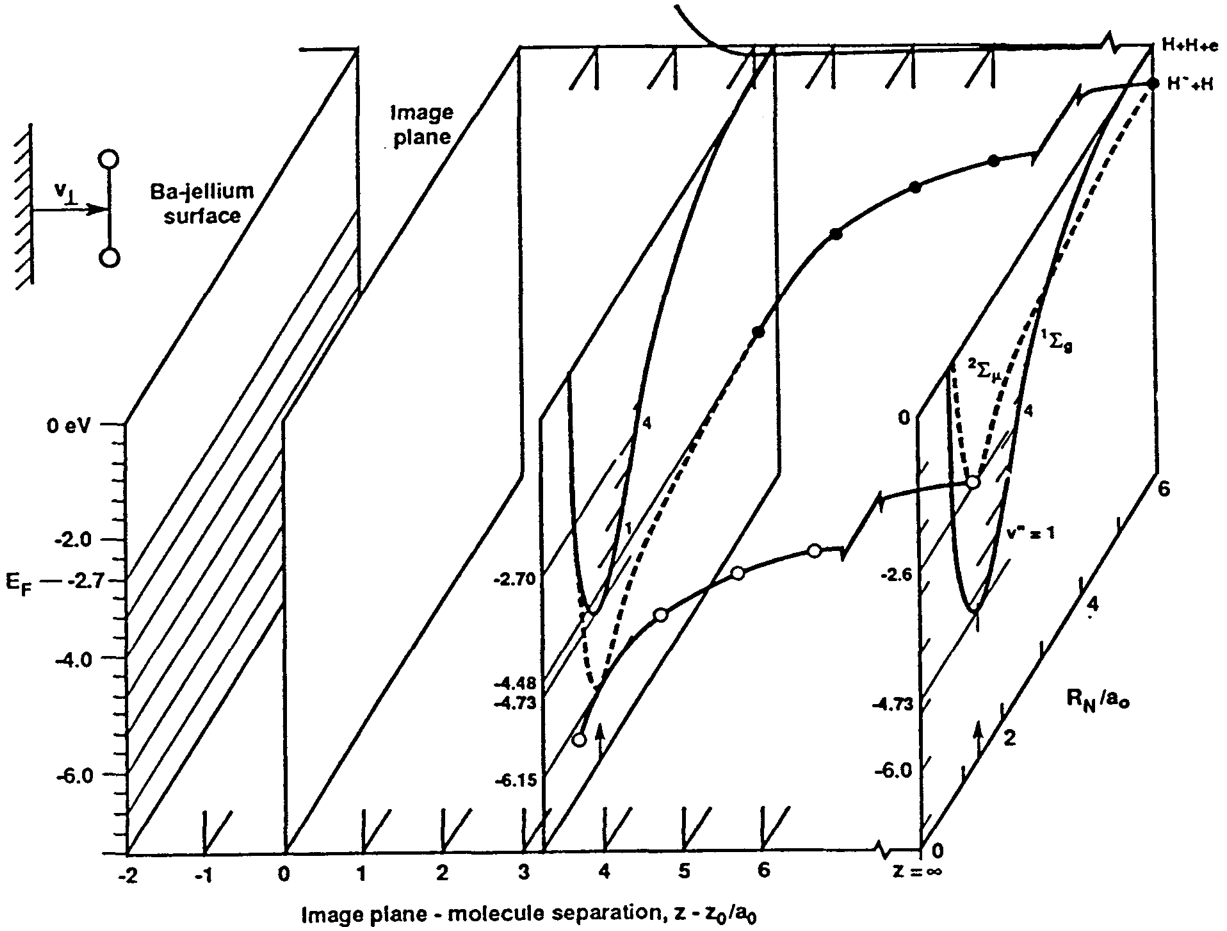

Figure 4: Energetics of the barium H_2^- , $H_2(v'')$ system.

At an image-plane molecule separation of 3.2 a_o the H_2^- configuration has shifted downward with respect to $H_2(v'')$ such that the asymptotic H_2^- potential lies as low as the $H_2(v'' = 0)$ level. At this separation resonant electron capture can occur from the barium to levels v" = 0, 1, 2, and 3 of H_2, resulting in H_2^- dissociation into $H^- + H$.

We can now summarize the resonant capture possibilities for a 6 eV incident H_3^+ ion rebounding as 4 eV $H_2(v'')$ and 2 eV H fragments. In the initial recombination process 20% of the H_3^+ leads to 3H dissociation and 80% to $H + H_2$ to give a yield $1.4H/H_3^+$. From the van Os data of Fig. 3 we conclude that 10% of these atoms will appear as H^- ions to give $0.14H^-/H_3^+$.

From Ref. 7 we have that 32% of the rebounding $H_2(v'')$ are in levels v" = 0, 1, 2, and 3. These molecules are subject to H_2^- formation and dissociation in their flight from $z - z_o = 2$ to 3.2 a_o. From Fig. 3 we find that 0.33 of these H^- survive to give an additional 0.10 H^-/H_3^+. The remaining flux is then 0.54 H_3^+. For those $H_2(v'' > 3)$, representing a fraction 0.48 H_2/H_3^+, no resonant capture to form H_2^- can occur.

One can now take inventory of the subsequent capture and dissociation processes for 6.0 eV incident H_3^+ on barium. The relative ratios $H : H_2 : H^-$ per incident H_3^+ ion are found to be 1.80:0.48:0.24. The new contribution offered by the low work-function barium surface is the 0.24 H^-/H_3^+. The H_2^- intermediary state has effectively converted the low-lying $H_2(v'')$ population into an additional source of H^- and H.

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