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Brief Reports

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Charge transfer in $N^{2+} + H$ collisions at slow to intermediate velocities

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We have calculated charge-transfer cross sections for collisions of ground-state N^{2+} ions with atomic hydrogen for barycentric collision energies up to 5.4 keV/amu. The calculations were done using a combination of close-coupled and unitarized distorted-wave techniques after an expansion in NH^{2+} molecular states. In addition to the entrance channel, we included a single product channel dissociating to $N^+(2s2p^3, {}^3D^o) + H^+$, both within the $^3\Pi$ symmetry. Our results are in good agreement with recent experimental measurements of state-to-state charge-transfer cross sections except at higher energies, where increasing population of the $N^+(2s2p^3, {}^3P^o)$ level was observed. The charge-transfer excitation process leading to the $^3P^o$ N^+ state is not well described by the Landau-Zener approach.

I. INTRODUCTION

We have recently calculated charge-transfer cross sections for low to intermediate velocity collisions of hydrogen or helium ions with a number of doubly or trebly charged first row ions. We report here results for the reaction

$$N^{2+}(\cdots 2s^2 2p, {}^2P^o) + H \rightarrow N^+(\cdots 2s2p^3, {}^3D^o) + H^+ ,$$

for barycentric collision energies up to 5.4 keV/amu. At the lowest collision energies, where a fully quantal treatment of the process is most appropriate, we used the close-coupled technique of Heil, Butler, and Dalgarno.[1] In this approach, one expands in electronic eigenstates of the aggregate collisional system and then numerically solves the resulting nuclear scattering equations by using a diabatic transformation to reduce them to standard potential coupling form. The calculation was done without the use of electron translation factors.

Our molecular basis set was comprised of the two states within the $^3\Pi$ symmetry of the NH^{2+} molecule, which dissociate to the products and reactants shown above. The potential curves and couplings for this case were published in Heil, Butler, and Dalgarno[1] with cross sections up to 8 eV/amu. This basis set was chosen to represent collision energies typical of low-temperature astrophysical plasmas.[2] As the collision energy increases, transitions to other final states become more probable; electron motions should tend from molecular to atomic orbitals, and electron translation effects can become larger. In addition, the close-coupled solution of the scattering equations requires progressively more computation. In order to extend efficiently these results into the keV energy range, we used the unitarized distorted-wave method of Bienstock, Heil, and Dalgarno,[3] where accurate to supplement close-coupled results.

Important new experimental data on the $N^{2+} + H$ charge transfer process have been reported recently by Wilkie *et al.*[4] In addition to measuring the total charge-transfer cross sections over the 0.043–13 keV/amu range, they utilized translational energy spectroscopy to study state-selective cross sections from 43–570 eV/amu. Of particular interest was their observation of the increased population of the $N^+(-2s2p^3, {}^3P^o)$ final state, in addition to other less important levels, with increasing collision energy; an increase such that the $^3P^o$ level may dominate over the $^3D^o$ final state at intermediate energies.

The population of both these final states involves the transfer of an electron from hydrogen and the excitation of an electron from the $2s$ shell of N^{2+}. With an increasing body of both experimental and theoretical data extending up to intermediate energies, studies of $N^{2+} + H$ charge transfer may contribute to understanding the dynamics of this important class of minimally four-body transfer/excitation processes.

Earlier experimental measurements of the total $N^{2+} + H$ charge-transfer cross section have been done by Phaneuf, Meyer, and McKnight[5] and by Seim *et al.*[6] Phaneuf, Meyer, and McKnight[5] measured cross sections for this reaction down to 2.9 keV/amu while the results of Seim *et al.*[6] extended down to 570 eV/amu.

II. RESULTS AND DISCUSSION

The calculated cross sections for charge transfer leading to the population of $^3D^o$ N^+ are given in Table I. Over the

TABLE I. Charge-transfer cross sections vs barycentric collision energy for the reaction of ground state $N^{2+}+H$ leading to $^3D^o$ product ions.

Energy (eV/amu)	Cross sections (10^{-16} cm^2)
0.0054	32.1
0.0107	36.9
0.107	16.6
0.298	10.1
1.61	5.10
4.19	4.17
8.68	3.03
26.8	4.62
53.6	3.39
80.4	3.05
134.0	4.01
268.0	5.65
536.0	5.15
1070.0	3.51
2140.0	2.11
5360.0	0.93

range 0.0054–5360 eV/amu the cross section shows an oscillatory structure with peaks near 0.01, 27, and 270 eV/amu. In Fig. 1 certain of these results are plotted for comparison with the experimental $N^{2+}+H$ total cross sections of Wilkie *et al.*,[4] Phaneuf, Meyer, and McKnight,[5] and Seim *et al.*[6] The calculated results are in good agreement with experiment below 800 eV/amu. Above this energy, our calculations predict that the cross section leading to $^3D^o$ N^+ ions drops off, whereas all experiments show the total cross section rises.

The state-selective cross sections given by Wilkie *et al.*[4] suggest the resolution of this discrepancy. Their results show an increase in the production of $^3P^o$ N^+ above 250 eV/amu. In addition, the measurements show some population of the endothermic $N^+(\cdots 2s2p^3, {}^1D^o)$ and $N^+(\cdots 2s^22p3p, {}^3P)$ transfer/excitation channels, channels generally excluded by slow collision molecular models. In Fig. 2 our results are plotted with the $^3D^o$ final-state data

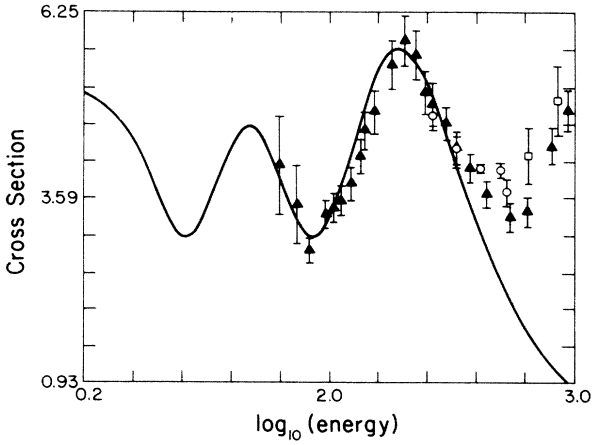

FIG. 1. Cross sections (10^{-16} cm^2) vs logarithm of (eV/amu) for $N^{2+}+H \rightarrow N^+ + H^+$. Theory (curve) and experiment: ▲, Wilkie *et al.* (Ref. 4); □, Phaneuf *et al.* (Ref. 5); ○, Seim *et al.* (Ref. 6).

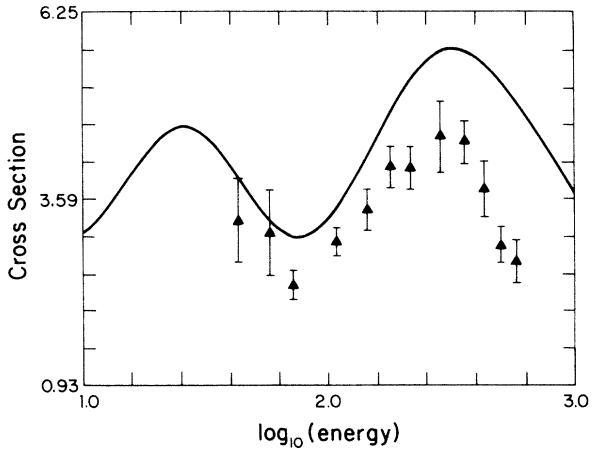

FIG. 2. $N^{2+}+H$ cross sections (10^{-16} cm^2) leading to $^3D^o$ N^+ vs logarithm of (eV/amu). Theory (curve) and experiment: ▲, Wilkie *et al.* (Ref. 4).

of Wilkie *et al.*[4] The calculated values are generally above the experimental data at the higher energies considered, where increasing transitions to the three other levels are sufficient to produce the apparently fortuitous agreement seen in Fig. 1.

The $^3P^o$ channel is exothermic by 2.45 eV so that $^3\Pi$ states correlating to this level will exhibit an avoided crossing with the neutral channel near $11.1a_0$. Since the molecular states are of the same symmetry, the dominant coupling between them should be radial.

In the absence of accurate potential curves and couplings, one can use the Landau-Zener approximation, as given by Butler and Dalgarno,[7] to estimate the contribution of this channel. Their technique assigns the Landau-Zener parameters based upon asymptotic splittings and a parametrization of the potential curve separation, $U_{11}-U_{22}$, at the avoided crossing depending upon the number of electrons involved. For this two-electron process, their method gives $U_{11}-U_{22}=3.4\times10^{-4}$ eV, but there results a cross section with a maximum well below 43 eV/amu, the lowest data point of Wilkie *et al.*[4] The $^3P^o$ cross section measured by Wilkie *et al.*[4] levels off and may be near a maximum at their highest-energy point, 570 eV/amu, with a value of 1.5×10^{-16} cm^2. The Landau-Zener approximation, with the above energy splitting, gives a cross section of 8.2×10^{-21} cm^2.

An alternative parametrization of Boyd and Moiseiwitsch[8] can be used to align the maximum of the Landau-Zener cross section with that of experiment. If we assume that the $^3P^o$ cross section does indeed reach a maximum near 570 eV/amu, the procedure of Boyd and Moiseiwitsch[8] gives an energy splitting of 0.5 eV at the avoided crossing, but a cross section of 4.8×10^{-16} cm^2 at 570 eV/amu.

III. CONCLUSIONS

It is evident that the Landau-Zener model is too simple to reliably model transfer/excitation processes. The oscillatory structure of the calculated cross sections for production of $^3D^o$ N^+ also supports this conclusion. The Landau-Zener model leads to cross sections with a single maximum. However, the experimental results in the $^3P^o$ level should be

strongly coupled to the $^3D^o$ level at intermediate energies making it difficult to estimate its effect if included in a larger close-coupled calculation.

An intuitive description of charge-transfer reactions says that they should be best characterized by multicenter molecular electron orbitals in the low velocity limit turning to atomic orbitals for fast collisions. The recent experimental data of Wilkie *et al.*[4] would appear to span the interesting transition region for $N^{2+}+H$. Further experiments and calculations are indicated for this system. An experimental confirmation of the theoretically predicted oscillations and predominance of the $^3D^o$ cross sections at low energies would be of great interest. In addition, theoretical and experimental measurements of the $^3P^o$ cross section should be extended over a greater energy range to see if it exhibits similar structure. This would also appear to be a good test for atomic orbital expansion methods, since the doubly excited atomic final states may be significantly influenced by correlation effects, requiring sophisticated atomic wave functions for accurate representation.

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