Relative role of charge-exchange and electron-detachment processes in $H^- + Na$ collisions

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Results of a differential time-of-flight energy-loss study of $H^- + Na$ collisions in the 100—500-eV laboratory collision energy range are discussed. It is shown that the relative importance of electron-detachment processes in the total stripping cross section increases with collision energy. The relative magnitude of Na excitation processes is determined. As in negative-ion—molecule collisions, charge exchange to the $Na^-(^3P)$ shape resonance lying at 80 meV may be important.

I. INTRODUCTION

Electron stripping from hydrogen negative ions in collisions with alkali-metal atoms has long been of interest in view of production of intense hydrogen-atom beams for tokamak plasma heating. This has motivated a number of experimental studies concerned with the measurement of the total stripping cross section for H^- incident on various alkali-metal atom targets.[1,2] Electron stripping in these collisions occurs as a result of

(i) charge exchange (CE), e.g.,

$$H^- + Na \rightarrow H + Na^-$$

(ii) electron detachment

$$H^- + Na \rightarrow \begin{cases} H + Na + e^- \\ H + Na^* + e^- \end{cases},$$

where $H + Na + e^-$ is direct detachment (DD) and $H + Na^* + e^-$ is detachment accompanied by excitation processes (DE).

The total cross-section measurements reported up until now correspond to a sum over all these processes and do not give any information about the relative importance of the different channels. The only exception appears to be the recent work of Howald *et al.*[2] in which the $Na(^3P)$ excitation cross section has been reported for 1—25-keV $H^- + Na$ collisions. No information, however, exists about the relative role of charge exchange and detachment, which could give some insight into the intricate interrelation of these processes and hence provide sensitive tests for theory. In this paper we report results of such measurements for $H^- + Na$ collisions for energies below 1 keV.

A few introductory words about the $H^- + Na$ system are pertinent here. For energies below 1 keV one can attempt to describe detachment within a quasimolecular model. The $(HNa)^-$ and HNa molecular states have been calculated by several authors.[3] Figure 1 shows a schematic diagram of the states of the $(HNa)^-$ systems obtained in a configuration-interaction (CI) calculation by Olson and Liu.[3] One of the remarkable aspects of this system contrary to the H_2^- system, which we studied previously,[4] lies in the fact that the $X^2\Sigma$ $(HNa)^-$ ground state remains bound for all calculated internuclear distances. This characteristic of the $(HNa)^-$ system may be understood in simple terms if one remembers the high value of the dipole moment of the HNa molecule responsible for the strong dipole field in which the electron is bound.[5]

Electron detachment in this system may proceed either via the $A^2\Sigma$ charge-exchange channel (Fig. 1) or via direct dynamic transitions from the $X^2\Sigma$ state. The existence of the latter, nuclear motion-induced transitions is predicted in Demkov's zero-range potential model of detachment[6] and its existence has been clearly demonstrated in such a calculation of detachment in, e.g., $H^- + Ne$ collisions.[7]

Yet another aspect of this system has to be taken into account when considering electron detachment. This is related to the existence of the $Na^-(^3P)$ shape resonance which plays an important role in low-energy electron sodium-atom scattering.[8] It has been recently demonstrated[9] that electron detachment in negative-ion—molecule collisions may occur via charge exchange to shape resonances (CESR), e.g.,

$$H^- + N_2 \rightarrow H + N_2^-(^2\Pi_g) \rightarrow H + N_2(v \geq 0) + e^- .$$

This possibility does not exist for rare-gas— and H-atom—H^- collisions which we studied previously and was thus not observed. This could, however, be important in the case of a variety of atomic targets such as alkali-metal atoms, alkaline-earth atoms, etc., for which low-lying

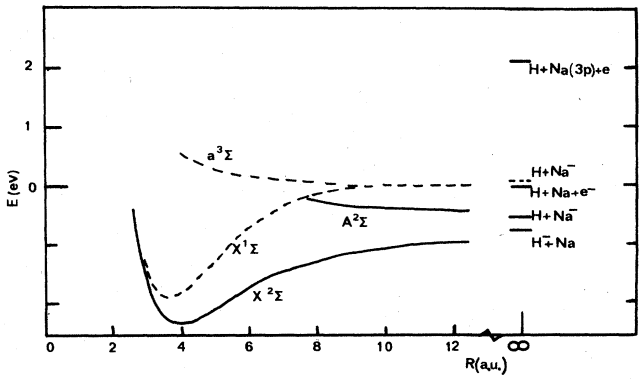

FIG. 1. Schematic diagram of the states of the $(HNa)^-$ system based on the calculations of Olson and Liu (Ref. 3).

shape resonances have been observed.[8] These peculiarities of the $H^- + Na$ system make a detailed investigation particularly interesting.

II. EXPERIMENT

Our measurements of the relative role of charge exchange and electron detachment were rendered possible by a differential time-of-flight (TOF) energy-loss study of scattered hydrogen atoms: a method which allows the separate study of the different reaction channels. The TOF spectra also give some indication about the energy distribution of detached electrons, since by energy conservation the neutral particle energy-loss peak reproduces it. Because a differential (in angle) study is performed, information about the impact-parameter dependence of this distribution may be obtained in some cases.[10]

The TOF spectrometer used has been described previously.[11] This setup allows use of a flight length that may be varied from 1.25 to 7.5 m. A good energy resolution ($\Delta E \cong 0.4$ eV) can thus be achieved in the energy range from about 100 eV to about 1 keV. The present measurements were therefore limited to this range. Extension to lower energies is rendered difficult by the decreasing efficiency of the microchannel plate detectors used. The upper energy limit was given by the energy resolution and also the angular resolution ($\Delta\theta \cong 0.15$ deg), which limits meaningful differential masurements at high energies in cases, as in the present one, of mainly forward scattering.

The chopped H^- beam was passed through a cell containing Na vapor. The temperature of the cell was adjusted for single-collision conditions, permanently monitored by a thermocouple, and kept constant by a feedback circuit. Time-of-flight spectra were recorded for angles on both sides of the beam direction in order to determine accurately the zero scattering angle. Measurements were also made for this angle. Relative differential cross sections were obtained from the measurement of the areas of the peaks corresponding to the different stripping channels. Data are presented as plots of the reduced differential cross section $\rho(\sigma(\theta)\theta \sin\theta)$ versus the reduced scattering angle $\tau(E\theta)$, where E is the laboratory collision energy, θ the laboratory scattering angle, and $\sigma(\theta)$ the differential cross section. The relative calibration of the ionic and neutral cross sections assumes a similar detector efficiency for both species. This assumption does not affect any results in this paper, however.

III. RESULTS AND DISCUSSION

A typical TOF energy-loss spectrum of scattered H atoms is presented in Fig. 2. The zero of the energy loss is taken to be the H + Na ground state, i.e., at 0.75 eV from the position of the elastic H^- peak.

The first peak A in the spectrum corresponds to nonresonant charge exchange to Na^-.

The second peak B corresponds to direct detachment leading to the production of both H and Na atoms in their ground states. This peak appears to be composed of two contributions: a fairly well-defined, narrow low-energy-loss peak and a broader distribution which results in a

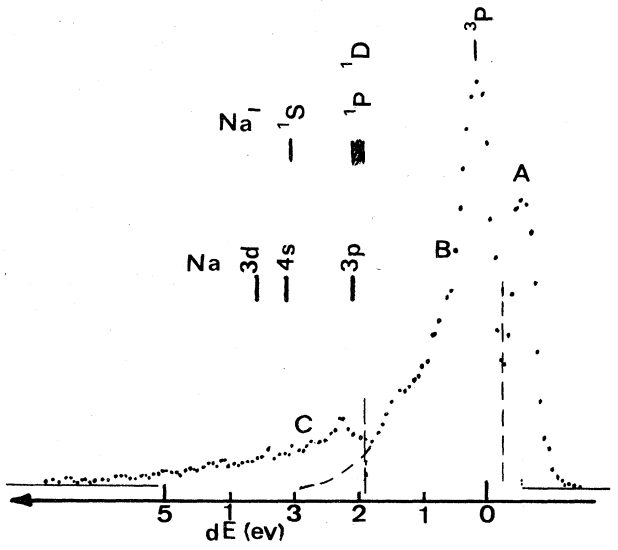

FIG. 2. Hydrogen-atom energy-loss spectrum for a 500-eV collision energy and a 0.3-deg scattering angle.

hump at energies between 1 and 2 eV. This characteristic may be attributed to the superposition of different direct-detachment channels and CESR. Contrary to what we have previously observed insofar as the position of direct-detachment peaks is concerned (see, e.g., Tuan *et al.*[10(a)] and Esaulov[10(b)]), the position of the maximum of this peak was not found to change, as a function of energy (150—500-eV range) or in the studied angular range. It was found to lie at an energy loss of (0.11 ± 0.05) eV, close to the position of the low-lying Na^- resonance reported at (80 ± 20) meV by Johnston and Burrow.[8]

In our previous work on H^- molecule collisions[9] the position of peaks in the TOF spectra due to CESR was not found to change as a function of an angle. Changes with energy were observed but these correspond to the "non-Franck-Condon" population of negative molecular ion "vibrational energy levels," not present in the case of $H^- + Na$ collisions. It thus appears that charge exchange to the Na^- shape resonance plays a role in electron detachment.

The third peak C in the spectrum corresponds mainly to excitation of Na and perhaps the production of autodetaching Na^- states. Excitation of the Na($3p$) level is clearly observed. Because of broadening effects related to electron ejection (see above), our measurements do not give a clear indication as to the production of higher-lying states. No H-atom excitation was observed.

Figure 3 shows the reduced differential cross sections for a 300-eV energy. The elastic differential cross section appears much larger than that for hydrogen-atom production. This is due to the large value of the elastic cross section. Indeed the calculations of Olson and Liu[3] show that at 100 eV the total elastic cross section is 399 Å^2, whereas the total stripping cross section reaches only about 22 Å^2. These calculations predict a rainbow maximum in the elastic cross section for a 103 eV deg angle, which is not observed in Fig. 3. The absence of any clear oscillatory structure in the elastic cross section is attributed to the de-

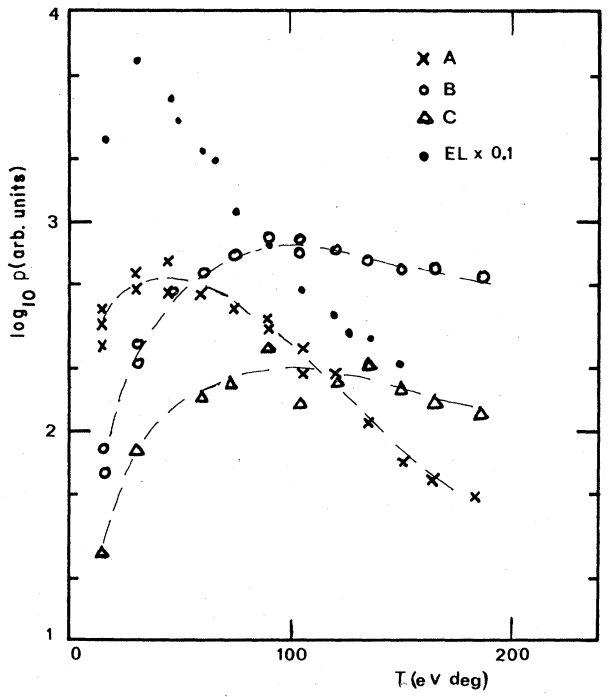

FIG. 3. Reduced differential cross setions vs the reduced scattering angle. Note that for the purpose of presentation the elastic cross section has been multiplied by 0.1.

pletion of this channel by electron detachment. A similar absence of oscillations in the elastic cross section due to detachment was noted in our earlier study of $H^- + H$ collisions. In the case of $H^- + Na$ collisions a maximum in the elastic cross section has been observed, however, at lower energies, close to 20 eV.[12]

The division of the neutral differential cross section (DCS) into cross sections for DD and DE posed some problems because of the width of peak *B*. The cross sections in Fig. 3 were obtained by integrating the peak *B* between the limits shown in Fig. 2 and peak *C* for energies above 1.9 eV. The peak *C* cross section thus gives an upper limit to the summed excitation cross section. An estimate of the effect of the error introduced by this cutoff of peak *B* was obtained by an extrapolation of it as shown in Fig. 2. This suggests that the excitation cross section may be overestimated by about 15%.

Integration of the neutral DCS allows an estimate of the evolution of the relative importance of the different electron stripping channels as a function of collision energy. In order to integrate the DCS, in cases of peak *B* and *C* these had to be extrapolated. Based on the general behavior of $\sigma(\theta)$ an exponential extrapolation was adopted. This was *not* necessary in the case of the charge-exchange cross section, which dropped off rapidly. Measurements of the ratios of the total charge-exhange (peak *A*) cross section to the total detachment cross section (peaks *B* and *C*) after summing over the *measured* angular range thus give the *upper limit* of this figure. The exponential extrapolation adopted gives an estimate of the *lower limit*. The ratio (R) thus obtained is shown in Fig. 4 as a function of collision energy. As can be seen the im-

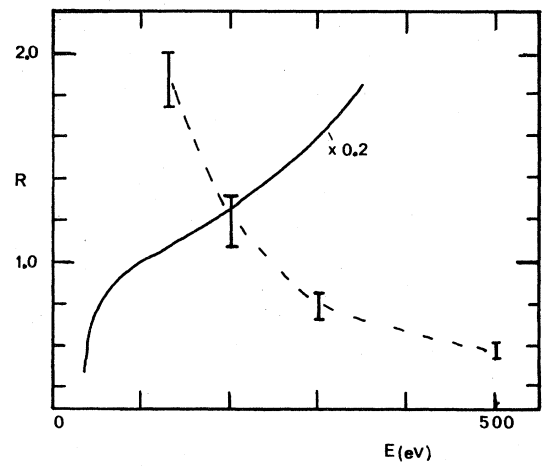

FIG. 4. Ratio (R) of the total charge-exchange cross section to the total detachment cross section. Vertical bars: this experiment; solid line: theoretical estimate of Olson and Liu (Ref. 3). The theoretical results have been multiplied by 0.2 in the figure.

portance of charge exchange versus detachment decreases as a function of collision energy. A detailed study of the behavior of these cross sections at lower energies (< 100 eV) would be interesting. In particular it would be interesting to see if the observed trend would not be reversed, since as the energy decreases the charge-exchange probability should decrease, whereas detachment via the $X^2\Sigma$ state may persist to very low energies.

Also shown in Fig. 4 are the results of a prediction of the above ratio by Olson and Liu.[3] In this calculation only detachment via the $A^2\Sigma$ "charge-exchange channel" was taken into account in order to obtain this estimate. The neglect of other detachment channels, and in particular that via the $X^2\Sigma$ state must be at the origin of the discrepancy between theory and experiment.

Using the measured DCS we also attempted to estimate the relative contribution of excitation processes to the detachment cross section. The ratio of the measured excitation cross section (peak *C*) to the total stripping cross sec-

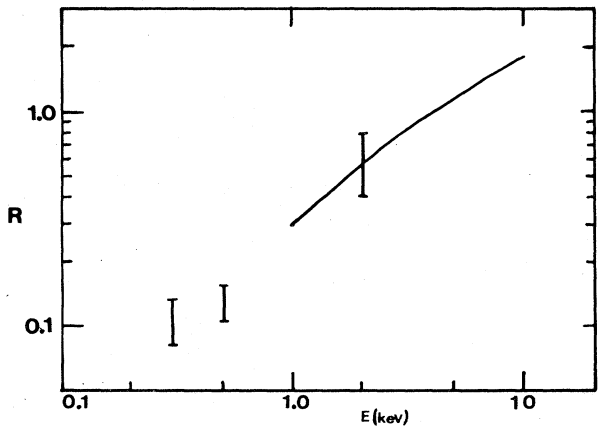

FIG. 5. Ratio (R) of the total excitation cross section to the total stripping cross section. Vertical bars ($E < 1$ keV): this work; line with error bar: data of Howald *et al.* (Ref. 2).

tion (peaks A, B, and C) is shown in Fig. 5. This ratio gives the *upper limit* of the contribution of all excitation processes. The *lower limit* shown is based on the estimation of the effect of (a) the angular cutoff obtained using the exponential extrapolation of the DCS discussed above and (b) of the contribution of the tail of peak B, estimated to be 15%. This lower limit should therefore be treated with some caution. The only other study of excitation processes in $H^- + Na$ collisions concerns the measurement of Na($3p$) excitation in the 1—25-keV energy range.[2] Using this data of Howald *et al.*[2] and their measurements of total stripping cross section[2] we obtain the ratio shown in Fig. 5. The rather large error bar is due to uncertainties in the absolute value of the total cross sections in two different experiments. Our data extrapolates reasonably well to these higher-energy measurements. It is interesting to note that the excitation cross section becomes larger than the stripping cross section, indicating that at high energies substantial excitation occurs without electron stripping. This channel was not observed in our low-energy measurements.

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[1]F. W. Meyer, J. Phys. B **13**, 3823 (1980); see also A. S. Schlachter, Brookhaven National Laboratory Report No. 50727 (unpublished), p. 11.

[2]A. M. Howald, L. W. Anderson, and C. C. Liu, Phys. Rev. A **24**, 44 (1981); Phys. Rev. Lett. **51**, 2029 (1983).

[3]R. E. Olson and B. Liu, J. Chem. Phys. **73**, 2817 (1980); see also A. M. Karo, M. A. Gardner, and J. R. Hiskes, J. Chem. Phys. **68**, 1942 (1978).

[4]V. A. Esaulov, J. Phys. B **13**, 4039 (1980).

[5]K. D. Jordan and J. J. Wendoloskii, Mol. Phys. **35**, 223 (1978); O. H. Crawford, Proc. Phys. Soc. London **91**, 279 (1967).

[6]Yu. N. Demkov, Zh. Eksp. Teor. Fiz. **46**, 1126 (1964) [Sov. Phys.—JETP **19**, 762 (1964)]; A. Z. Devdariani, Zh. Tekh. Fiz. **43**, 399 (1973) [Sov. Phys.—Tech. Phys. **18**, 255 (1973)].

[7]J. P. Gauyacq, J. Phys. B **13**, L501 (1980).

[8]A. R. Johnston and P. D. Burrow, J. Phys. B **15**, L745 (1982).

[9]Vu Ngoc Tuan and V. A. Esaulov, J. Phys. B **15**, L95 (1982); Vu Ngoc Tuan, V. A. Esaulov, J. P. Gauyacq, and A. Hertzenberg, J. Phys. B **18**, 721 (1985); R. I. Hall and V. A. Esaulov, in *Proceedings of the Twelfth International Symposium on the Physics of Ionized Gases, Invited Lectures and Progress Reports, Sibenic, 1984,* edited by M. Popovic (Institute of Physics, University of Belgrade, in press).

[10](a) Vu Ngoc Tuan, J. P. Gauyacq, and V. A. Esaulov, J. Phys. B **16**, L95 (1983); (b) V. A. Esaulov, *ibid.* **14**, 1303 (1981).

[11]C. Reynaud, J. Pommier, Vu Ngoc Tuan, and M. Barat, Phys. Rev. Lett. **43**, 579 (1979).

[12]R. L. Champion and L. D. Doverspike (private communication).