# $D^-$ production by multiple charge-transfer collisions in alkaline-earth-metal vapors

T. J. Morgan, J. Stone, M. Mayo, and J. Kurose
*Department of Physics, Wesleyan University, Middletown, Connecticut 06457*
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$D^-$ negative-ion equilibrium fractions of deuterium in collisions with Mg, Ca, Sr, and Ba alkaline-earth-metal vapors have been measured over the energy range 1.25–100 keV. For Ca, Sr, and Ba targets, the yield of $D^-$ ions is almost completely independent of the identity of the target over the energy range 4–35 keV. At low energies, an abrupt increase in $D^-$ production is observed for targets of Ca, Sr, and Ba but not Mg, and occurs at approximately the same energy, 4 keV, in all three cases. Possible mechanisms responsible for the structure in $D^-$ production are discussed.

## I. INTRODUCTION

Deuterium negative-ion formation and loss processes are of fundamental interest in atomic-collision physics[1] and of considerable importance in several areas of application. Intense $D^-$ beams are needed for neutral beam injection schemes in controlled thermonuclear fusion research; they can be accelerated to high energies, neutralized with efficiencies in excess of 60%, and the resulting $D^0$ beam used to both heat and fuel the plasma.[2] Also, methods of producing intense $D^-$ beams are of considerable interest in nuclear physics experiments using polarized $D^-$ ions.[3] Experimental data on $D^-$ production therefore have special significance and in view of the current interest are especially timely.[4]

$D^-$ production by charge-transfer collisions can be discussed in terms of the $D^-$ fraction of the beam $F_-^\infty$ after charge-state equilibration has been attained via multiple collisions in a thick target. In general, electron attachment and detachment processes, which govern the $F_-^\infty$ yield, follow trends associated with target electronic configuration. However, many anomalies exist and it is usually not possible to accurately predict the equilibrium yield of $D^-$ ions. In any case, Hiskes[5] has exploited these trends in an attempt to predict an effective $D^-$ charge exchange target for producing intense neutral beams at high energies for plasma heating and fueling, with the conclusion that the heavier alkaline-earth atoms might be attractive. Recent measurements by Berkner *et al.*[6] over the energy range 2.7–31 keV show that the behavior with energy of $F_-^\infty$ using a Sr metal vapor target is quite different from that of a gaseous target or an alkali-metal vapor target. Their data indicate a plateau in $F_-^\infty$ between 5 and 10 keV with an increase with decreasing energy below 5 keV. The purpose of the present experiment was to repeat the measurements of Berkner *et al.* in order to verify the observed low energy structure and to perform a systematic study of $F_-^\infty$ in several alkaline-earth–metal vapor targets over an extended energy range.

In this paper we report results for $F_-^\infty$ in Mg, Ca, Sr, and Ba over the energy range 1.25–100 keV. We find excellent agreement with the results of Berkner *et al.*[6] Furthermore, we find almost identical structure for Ca, Sr, and Ba targets. Also, we find that the production of $D^-$ ions is insensitive to the choice of target for Ca, Sr, and Ba over the energy range 4–35 keV. Possible mechanisms responsible for the structure in $F_-^\infty$ are discussed.

## II. EXPERIMENTAL APPROACH

A discussion of the apparatus has been published previously.[7,8] The present measurements involved only minor differences in the apparatus so we shall only describe the essential features here. The experimental arrangement consisted of an ion source, acceleration region, bending magnet, metal-vapor target, charge-analyzing electric field and an array of beam-particle detectors. The apparatus, which had been previously used in our charge-exchange cross-section measurements, was modified by placing a 6-mm-diam aperture between the metal-vapor cell and the electrostatic-analyzing plates. This arrangement ensured that for the thick targets required to reach apparent equilibrium (~$4\times10^{15}$ atoms/$cm^2$) the beam components fell within the detectors. The experimental procedure was basically the same as that given in Ref. 6 with one important difference. Berkner *et al.*[6] used a pyroelectric detector to measure the $D^0$ flux whereas we have chosen to use a secondary emission detector. The excellent agreement, throughout the energy range where there is an overlap, between the present results for Sr and those of Berkner *et al.* using different $D^0$ measuring techniques demonstrates the reliability of the results. The $D^+$

 

and $D^-$ components of the beam were measured with Faraday cups. As a further check on our experimental technique we measured $F_-^\infty$ using argon gas with both hot and cold targets. The results are in excellent agreement with previous measurements.[9] The data also agree with previous measurements above 3 keV in Mg.[10] The present results were obtained using $D^+$, $D^0$, and $H^+$ incident beams and no difference in the equilibrium fractions was observed. This observation confirms the fact that equilibrium had been reached. Target thicknesses required for the $D^-$ fraction to equilibrate were typically $\lesssim 4\times10^{15}$ atoms/$cm^2$ and measurements were usually made for target thicknesses up to $\sim 1\times10^{16}$ atoms/$cm^2$. Over the target thickness range from apparent equilibrium to $1\times10^{16}$ atoms/$cm^2$ the charge state fractions did not change appreciably even though at low energies the total beam transmitted was attenuated by as much as a factor of 10. The low vapor pressure of Ba imposed added experimental difficulties since the temperature of the oven had to be raised to 1050 K in order to reach apparent equilibrium. With the present experimental setup we were unable to reach $1\times10^{16}$ atoms/$cm^2$ in the case of Ba and consequently we estimate that our results for Ba may be $\lesssim 10\%$ too low.

In order to analyze the data to obtain the equilibrium fractions the secondary emission coefficient $\gamma$ for $D^0$, $D^+$, and $D^-$ impact on our neutral detector surface must be known. A study of $\gamma^-$ and $\gamma^+$, using incident $D^-$ and $D^+$ beams, was carried out several times during the course of the experiment. Although the absolute value of $\gamma^-$ and $\gamma^+$ changed between measurements, the ratio $\gamma^-/\gamma^+$ remained constant and equal to $1.5\pm0.3$ over the present energy range. This ratio is in excellent agreement with recent measurements of Meyer and Barnett at 1.5 keV.[11] During the accumulation of data the value of $\gamma^+$ was measured at every data point and the relation $\gamma^-/\gamma^+=1.5$ was used in the data analysis. $\gamma^0$ was obtained from the relation $\gamma^0/\gamma^+=1.1$, recently verified experimentally down to energies well below the present work.[11]

## III. RESULTS AND DISCUSSION

In Fig. 1 we present the measured $D^-$ equilibrium fractions versus incident $D^+$ energy for Mg, Ca, Sr, and Ba targets. The data were taken over a period of several months and were reproducible to within ±12%. The results are in excellent agreement with previous measurements by Baragiola *et al.* for Mg and by Berkner *et al.* for Mg and Sr throughout the energy region where there is an overlap.[6,10] The significant feature of the data

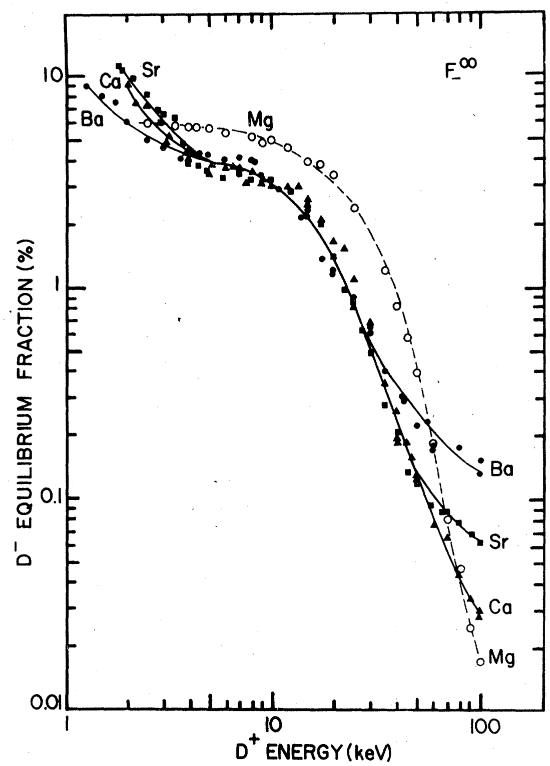


FIG. 1. Measured $D^-$ equilibrium fraction $F_-^\infty$ vs incident $D^+$ energy for several alkaline-earth—metal vapor targets. $D^+$, $D^0$, and $H^+$ incident beams were used, and the $H^+$ data are plotted at twice the incident energy. An isotope effect was not observed. The lines through the data points are drawn in to guide the eye, and have no other significance.

is the plateau in the $D^-$ production in the energy range 10–4 keV followed by an abrupt rise with decreasing energy. Typically, $D^-$ equilibrium fractions pass through a single rather broad maximum at low energies. Measurements of $F_-^\infty$ in several gases and alkali metal vapors do not show any examples of structure.[12,13] The present results for the alkaline-earth–metal vapors Ca, Sr, and Ba are very different from those in gases or alkali-metal vapors and indicate that the qualitative ideas used for estimating $D^-$ production are invalid for the alkaline earths.

If we assume that the equilibration of $D^-$ ions is determined primarily by collisions involving only $D^+$, D(1*s*), and $D^-$ we may perform the following analysis. We have measured[8] the double-electron-capture cross section in Mg and Ba over the present energy range and found that the cross section does not exceed $3\times10^{-17}$ $cm^2$. Consequently, we are confident that the primary $D^-$ collisional formation mechanism is single-electron attachment. Neglecting double-charge exchange processes and using the relation that at low energies single-electron capture by $D^+$ is large compared to electron loss from $D^0$, the negative-ion equilibrium fraction at low energies can be written in rather simple form[14]

$$F_-^\infty = (1 + \sigma_{-10}/\sigma_{0-1})^{-1}, \quad (1)$$

where $\sigma_{-10}$ and $\sigma_{0-1}$ are the single-electron-detachment and -attachment cross sections respectively.

This equation reveals that within the framework of a three-component model the plateau in $F_-^\infty$ indicates that the cross-section curves for $\sigma_{-10}$ and $\sigma_{0-1}$ are parallel. Also, the large rise in $F_-^\infty$ at low energies is due to an abrupt change in the energy dependence of either $\sigma_{-10}$ or $\sigma_{0-1}$ or both. Oscillations in the electron-attachment cross section $\sigma_{0-1}$ are predicted by the two-state Stucklebburg-Landau-Zener (SLZ) theory of curve crossings and have been suggested by Berkner *et al.*[6] as a possible explanation for the $D^-$ rise at low energies in Sr. If $\sigma_{-10}$ remains constant over the energy range 4–2 keV, the present results indicate that $\sigma_{0-1}$ must increase by almost a factor of 4. We would not expect the amplitude of the SLZ oscillations to be so large. However, it is also possible that the electron detachment cross section $\sigma_{-10}$ is a rapidly decreasing function of energy below 4 keV. At low energies the detachment process occurs via electron association with the target. For alkaline-earth–metal atoms, with an $ns^2$ configuration and a negative-electron affinity, detachment at low energies is unlikely and $\sigma_{-10}$ might decrease rather abruptly.[15] In summary, within the framework of a three component model there are two possible mechanisms which can contribute to the observed rise in $D^-$ production at low energies. However, we note that both processes suggest that the rise should also occur for a Mg target yet we do not observe any structure for energies down to 2 keV.

An alternative approach to the explanation of the rise in $D^-$ production is based on our experimental data for the cross section for D(2*s*) metastable atom formation $\sigma_{+m}$.[16] The cross section $\sigma_{+m}$ is large at low energies for Ba and reaches a maximum value of $5.6\times10^{-16}$ cm$^2$. However, for Mg the cross section reaches a maximum value of $3.2\times10^{-16}$ cm$^2$ at ~14 keV and decreases rapidly with decreasing energy. The behavior of $\sigma_{+m}$ suggests that the present results for $D^-$ formation might be related to the formation of metastable atoms. If so, electron attachment by D(2*s*) may be the dominant $D^-$ formation mechanism at low energies for the heavier alkaline earths.

Inspection of the appropriate alkaline-earth energy levels[17] reveals that low-energy $D^-$ formation by D(2*s*) attachment collisions occurs predominately via curve crossings of the incident D(2*s*) + *X* covalent potential with several attractive $D^- + X^{+*}$ Coulomb potentials. ($X^{+*}$ indicates an alkaline-earth ion in an excited state.) Assuming the interaction potential can be accurately approximated by a constant potential for the covalent reactant channel and by an attractive Coulomb potential for the ionic product channels, the crossings occur at internuclear separations $R = 14.4/\Delta E_\infty$, where $\Delta E_\infty$ is the energy defect in eV and $R$ is in angstroms. Using this relation we find many $D^- + X^{+*}$ Coulomb channels which cross the D(2*s*) + *X* channel at internuclear separations at which a transition can occur. Furthermore, there are many more of these crossings for Ca, Sr, and Ba than for Mg. These observations coupled with the measured D(2*s*) formation cross-section energy behavior in Mg and Ba suggest that the measured $D^-$ production at low energies might be due to electron attachment by D(2*s*). We note that it should be possible to use a multichannel Landau-Zener calculation to compute the relevant cross sections using estimates of the coupling matrix elements at the curve crossings based on semiempirical formulas.[18]

A possible objection to the above discussion is that at low energies, the D(2*s*) atoms will be collisionally deexcited via the 2*p* state. In the case of a Cs target it is known that D(2*s*) does not play a role in $D^-$ equilibrium production for this reason.[19] However, as discussed above, for alkaline-earth targets there are many $D^-$ formation channels via D($n = 2$) collisions which are not present in the case of Cs. At low energies, the availability of these collision channels should increase the cross section for $D^-$ formation from D($n = 2$) atoms and reduce the deexcitation cross section. We note that experimental and theoretical investigations of the relevant cross sections using alkaline-earth targets are needed. Furthermore, the rising $D^-$ yield in Ca, Sr, and Ba warrants further experimental study at lower energies.

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