

## Single-electron-capture cross sections for 1–10-keV $\text{Li}^+$ ions in alkaline-earth vapors

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Total charge-neutralization cross sections have been measured with use of an attenuation method for  $\text{Li}^+$  ions in the energy range from 1 to 10 keV incident on thin targets of Mg, Ca, Sr, and Ba vapors. These cross sections exhibit an energy dependence characteristic of asymmetric charge exchange, increasing with collision energy up to a broad maximum, then decreasing at higher energies. The  $\text{Li}^+ + \text{Ba}$  cross section reaches a maximum value ( $\sigma_m$ ) of  $4.0 \times 10^{-14} \text{ cm}^2$  at  $\sim 3.5$  keV impact energy. The corresponding values of  $\sigma_m$  for Sr and Ca are  $2.85 \times 10^{-14} \text{ cm}^2$  at 6.0 keV, and  $3.3 \times 10^{-15} \text{ cm}^2$  at 10 keV. The  $\text{Li}^+ + \text{Mg}$  cross section only attains a value of  $1.1 \times 10^{-15} \text{ cm}^2$  at the highest energy measured (10 keV), thus,  $\sigma_m$  clearly occurs at even higher energies for this system. The energies at which these cross-section maxima occur increase monotonically through the series from Mg to Ba, while the magnitude of  $\sigma_m$  decreases in that order. This ordering is consistent with the inverse ordering of the asymptotic energy defects for the process:  $\text{Li}^+(1s^2) + M(ns^2) \rightarrow \text{Li}(2s) + M^+(ns)$ , where  $M$  is an alkaline-earth element.

### I. INTRODUCTION

Near-resonant charge-transfer collisions involving asymmetric ion-atom pairs represent an interesting class of one-electron processes. In general, an electron-capture reaction is considered to be a near-resonant process if the "energy defect," i.e., the asymptotic energy difference between the initial and final channels,  $|\Delta E_\infty|$  is  $\ll I$ , the ionization potential of the target atom. For simple ion-atom collisions, the incoming channel will correspond to both species being in their electronic ground states, whereas the outgoing channel may involve one or more excited states of the neutralized atom. Extensive studies of ion-atom charge exchange have shown that the favored outgoing channel will typically have the smallest energy defect. This propensity has been exploited to generate beams of a number of electronically excited atoms, including all of the metastable rare gases,  $\text{H}(2s)$ , and  $\text{Li}(2p)$ .

A common characteristic of asymmetric charge-exchange collisions involving small energy defects is the energy dependence of the total neutralization cross section,  $\sigma_{+0}(E)$ . Typically,  $\sigma_{+0}(E)$  is small at low-collision energies, then rises with impact velocity until a maximum is reached, beyond which the cross section again decreases. A number of theoretical treatments<sup>1–5</sup> of this problem have sought to estimate the velocity ( $v_m$ ) at which the maximum in the cross section will occur. Experimentally, this energy dependence of  $\sigma_{+0}$  has been measured for many of the rare-gas-ion–alkali-metal-atom combinations,<sup>6–8</sup> alkali-metal-ion–alkali-metal-atom pairs,<sup>9</sup> proton–alkali-metal-atom pairs,<sup>10,11</sup> and proton–alkaline-earth systems.<sup>12,13</sup> The simple curve crossing formalism often used to describe near-resonant collisions also predicts that the electron transfer will occur at large internuclear separations, thus leading to large absolute cross sections. This behavior has been well documented for the

above cited collision systems, with many of the measured  $\sigma_m$  values exceeding  $10^{-14} \text{ cm}^2$ .

More rigorous theoretical treatments of ion-atom collisions have been developed using coupled-channel approaches, and used with success to model the proton–alkali-metal-atom systems. Until recently, the perturbed-stationary-state method had only been applied to one-electron, or pseudo-one-electron problems, however, Kimura, Sato, and Olson<sup>14</sup> have now reported  $\sigma_{+0}$  cross sections for the system,



They used an eight-channel impact-parameter perturbed-stationary-state method that included electron translation factors. Their resulting  $\sigma_{+0}$  displays the expected energy dependence, with a broad maximum occurring between 1.5 and 8 keV/amu. The predicted cross-section maximum in that energy range is  $\sim 4 \times 10^{-15} \text{ cm}^2$ . They also find that the cross section for the production of  $\text{Li}(2p)$  is typically one order of magnitude smaller than the  $\text{Li}(2s)$  production.

We have measured the total attenuation cross sections of  $\text{Li}^+$  ions in the alkaline-earth targets Mg, Ca, Sr, and Ba under conditions such that charge transfer is the primary attenuation mechanism. The  $\text{Li}^+$  ion beam energy covered the range between 1 and 10 keV, corresponding to 0.14 to 1.4 keV/amu. The asymptotic energy defects for these collision pairs are listed in Table I, along with  $\Delta E_\infty$  for other  $\text{H}^+$  and  $\text{Li}^+$  systems for comparison. It is apparent from Table I that the charge exchange of  $\text{Li}^+$  in the alkaline earths should favor production of ground-state Li neutrals over the excited  $\text{Li}(2p)$ , provided that the asymptotic energy defect governs the outcome. This predominance of ground state over excited products is in agreement with the findings of Kimura *et al.*<sup>14</sup> This is in contrast to the  $\text{Li}^+ + \text{alkali-metal atom}$  pairs, where the

TABLE I. Asymptotic energy defects,  $\Delta E_\infty$ , in eV for  $H^+$  and  $Li^+$  charge transfer in alkali-metal and alkaline-earth targets.

Target	H(1s)	H(2s)	Neutral Product	
			Li(2s)	Li(2p)
Li	8.21	-2.00	0.0	-1.85
Na	8.46	-1.75	0.25	-1.60
K	9.26	-0.95	1.05	-0.80
Rb	9.42	-0.79	1.21	-0.63
Cs	9.70	-0.50	1.50	-0.35
Mg	5.95	-4.25	-2.25	-4.10
Ca	7.48	-2.72	-0.72	-2.57
Sr	7.91	-2.30	-0.30	-2.15
Ba	8.39	-1.81	-0.18	-1.67

particular target determines which final state is more likely, and to all of the  $H^+$  + alkali-metal and alkaline-earth systems, where excited H ( $n=2$ ) is expected to dominate. Table I also shows that the  $Li^+$  + alkaline-earth systems all have smaller energy defects than their corresponding  $H^+$  counterparts.

## II. EXPERIMENTAL METHOD

The data presented here were obtained by measuring the total attenuation of a  $Li^+$  ion beam in the alkaline-earth targets. The method has been described in detail recently<sup>15</sup> and used to determine the total charge-transfer cross sections for  $H^+$ ,  $H_2^+$ ,  $H_3^+$ ,  $N^+$ , and  $N_2^+$  in a Cs vapor target.<sup>16</sup> Basically, the method consists of measuring the  $Li^+$  beam current before ( $I_0$ ) and after ( $I$ ) interaction with a target of known thickness ( $\Pi=nl$ ). The total attenuation cross section,  $\sigma_A$ , is then given by

$$\sigma_A = -\Pi^{-1} \ln(I/I_0). \quad (2)$$

The major components of the apparatus are the ion source, a magnetic momentum analyzer, a movable target vapor cell with a set of stationary entrance and exit collimators, and a large aperture Faraday cup. The latter components are shown schematically in Fig. 1. The ion

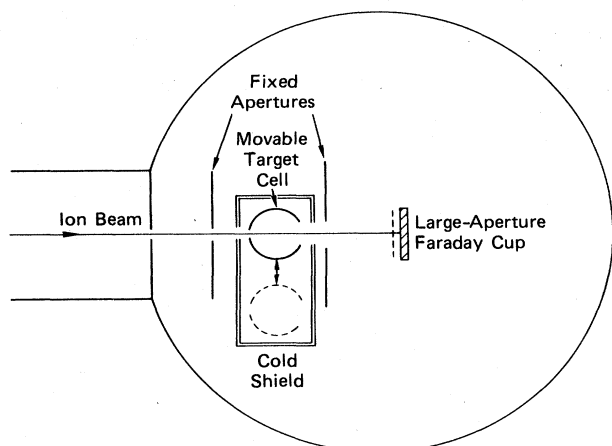


FIG. 1. Schematic diagram of the apparatus used to measure the total attenuation cross sections.

source is a  $\beta$ -eucryptite thermionic emitter (Spectra-Mat, Inc.) mounted onto the base of a Colutron ion source. The focused, monoenergetic ion beam is passed through a  $60^\circ$  magnetic sector, thus giving a mass (momentum) selected primary beam. Following additional focusing, the beam is collimated by the 0.15 cm diameter entrance aperture, just prior to the collision cell which has a larger (0.25 cm diameter) entrance aperture. The ion current exiting a  $0.3 \text{ cm} \times 0.6 \text{ cm}$  aperture is then collected in a Faraday cup detector and measured with an electrometer. The detector has an acceptance angle  $\theta_{\text{det}} = \pm 10^\circ$ , and electrostatic secondary electron suppression. The target cell can be quickly translated into and out of the path of the ion beam, giving measured currents of  $I$  and  $I_0$ , respectively. Since the entrance and exit apertures on the movable cell are larger than the fixed apertures, these measured currents accurately represent the desired quantities. With the cell empty, or when the target material has a negligible vapor pressure,  $I$  and  $I_0$  are the same to within 1%.

Use of the attenuation method requires a knowledge of the effective target thickness  $\Pi$ . In the past, we have used this system only for studies involving alkali-metal vapors, principally cesium.<sup>15,16</sup> For that work, we determined the thickness directly by measuring the total attenuation of  $He^+$  in Cs. Then, using the previously measured values of  $\sigma_{+0}$  for that combination<sup>6</sup> we find  $\Pi$  directly from Eq. (2) without requiring separate determinations of  $n$  and  $l$ . However, for the present experiments, where the targets are alkaline-earth vapors, this direct method could not be used. Instead, separate estimates of  $n$  and  $l$  were made as follows: the target density,  $n$ , was determined from standard vapor pressure curves<sup>17</sup> using the measured oven temperature. The effective scattering length was taken to be the geometric path length between the oven apertures, plus the sum of their radii to account for the finite extent of the target vapor beyond the cell. Our previous experience with this target cell<sup>15</sup> shows that this estimate of  $\Pi$  should be accurate to  $\pm 10\%$ .

To test the efficiency of this method of obtaining the target thickness, we measured the total attenuation of  $H^+$  in Mg at several energies, and compared the results to the  $\sigma_{+0}$  cross sections of Morgan and Eriksen.<sup>12</sup> The data are in agreement to within the anticipated  $\pm 10\%$ .

To obtain the data presented here, the total attenuation of the primary  $Li^+$  beam was measured for each alkaline-earth target as a function of the kinetic energy of the projectile beam in the range between 1 and 10 keV (limited by the magnetic field attainable in the momentum analyzer). Each datum represents the average of several measurements made at different target densities that were chosen low enough to maintain the  $Li^+$  attenuation below 30%.

Because the attenuation method is only sensitive to the loss of positive current, several secondary processes can also contribute to the measured cross section. These have been enumerated and discussed in relation to the hydrogen and nitrogen ion collisions with Cs,<sup>16</sup> where it was shown that  $\sigma_{+0}$  is very well represented by  $\sigma_A$  for collisions of the present type in the intermediate energy range. However, except for the  $Li^+$  + Mg data, which are affected by

angular scattering at energies below 4 keV (see below), these attenuation cross sections can be considered to represent single-electron capture only.

### III. RESULTS AND DISCUSSION

The measured total attenuation cross sections for  $\text{Li}^+$  in the alkaline-earth vapors Ba, Sr, Ca, and Mg as a function of the collision energy are shown by the open symbols in Fig. 2. The experimental cross sections for barium and strontium are similar in magnitude and shape, with the

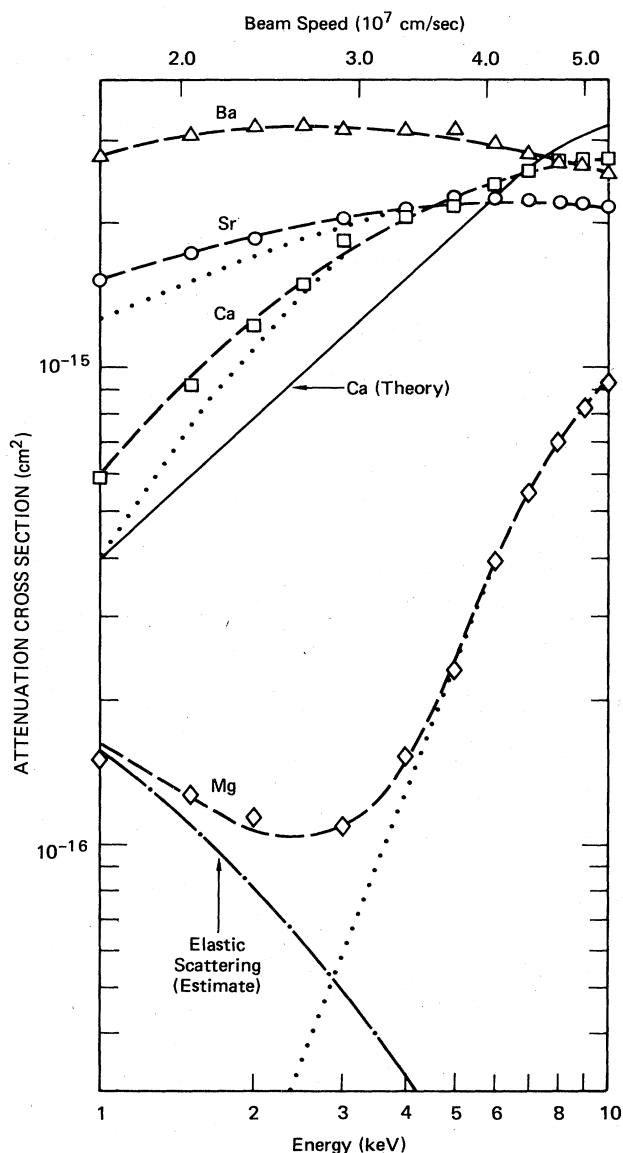


FIG. 2. Total attenuation cross sections for  $\text{Li}^+$  ions on Ba, Sr, Ca, and Mg vapor as a function of impact energy. The open symbols are the present experimentally measured points, the dotted lines are corrected for elastic scattering effects using the calculated cross sections of Olson (Ref. 18). The dashed lines through the data are a visual guide only, while the solid line is the calculated  $\sigma_{+0}$  cross section of Kimura *et al.* (Ref. 14) for  $\text{Li}^+ + \text{Ca}$ .

barium results being larger at all energies studied. Both of these cross sections show only a relatively weak energy dependence over the range from 1 to 10 keV.

The cross section data for  $\text{Li}^+ + \text{Ca}$  are markedly smaller in magnitude at the lower energies than either the Ba or Sr data, rising to comparable values at the higher energies.

The solid line included in Fig. 2 is the calculated cross section of Kimura *et al.*<sup>14</sup> for the production of  $\text{Li}(2s)$  from  $\text{Li}^+ + \text{Ca}$ . The general agreement between it and the present data is very good, both in regard to the overall shape and the absolute magnitude. Because the measured total attenuation cross section,  $\sigma_A$ , agrees well with the calculated  $\sigma_{+0}$ , we can conclude that single electron capture is the dominant process in this energy range. Kimura *et al.*<sup>14</sup> also calculate the cross section for  $\text{Li}(2p)$  production, and find it to have an energy dependence similar to the  $\text{Li}(2s)$  result, but an order of magnitude smaller. Since the present experimental method cannot distinguish among the final products state, any  $\text{Li}(2p)$  production will result in a larger  $\sigma_A$ . By summing the  $\text{Li } 2s$  and  $2p$  contributions of Kimura *et al.*, the low-energy portion of their cross section would be more nearly equal to the measured data, although the high-energy region would then be overestimated.

For the three heaviest alkaline-earth targets, it is possible to estimate the energy (velocity) at which  $\sigma_{+0}$  reaches a maximum. The approximate values of the corresponding  $v_m$  for Ba, Sr, and Ca are  $3.2 \pm 0.5 \times 10^7$  cm/sec,  $4.1 \pm 0.4 \times 10^7$  cm/sec, and  $5.4 \pm 0.5 \times 10^7$  cm/sec, respectively. The large uncertainties associated with these values of  $v_m$  result from the very broad nature of the plateau regions in the energy dependence of  $\sigma_A$ .

The total attenuation cross section of  $\text{Li}^+$  in Mg vapor shown in Fig. 2 is significantly different from the results for the three heavier alkaline-earth targets. For this target, the cross section exhibits a much stronger energy dependence in the range from 4 to 10 keV, with the maximum,  $\sigma_m$ , occurring at energies beyond 10 keV. While at the highest energies measured here, the cross section is reasonably large,  $\sim 9 \text{ \AA}^2$ , it is considerably smaller than any of the other cross sections. More interesting perhaps is the low-energy portion of the data where the cross section shows an apparent minimum near 3 keV, rising again at the lowest energies. Initially, the origin of this behavior was unclear. Our measurements of  $\sigma_{+0}$  for protons on Mg that were used to test the target thickness calibration had given good agreement with the results of Morgan and Eriksen,<sup>12</sup> even down to energies of 1 keV. However, since  $\text{Li}^+$  ions at 1 keV have a much smaller relative collision velocity than 1 keV  $\text{H}^+$ , the most likely cause of the increasing cross section at low energy is elastic scattering beyond  $\theta_{\text{det}}$ . To test for the effects of elastic scattering of low-energy  $\text{Li}^+$  ions, we measured the total attenuation of  $\text{Li}^+$  on a potassium target between 1 and 10 keV. The  $\sigma_{+0}$  cross section for this system had been previously measured by Perel and Daley.<sup>9</sup> The two sets of data are compared in Fig. 3, where it is apparent that the present results are, in fact, smaller at the low energies, and in very good agreement at the high energies. The excellent agreement in absolute value at the high energies implies that

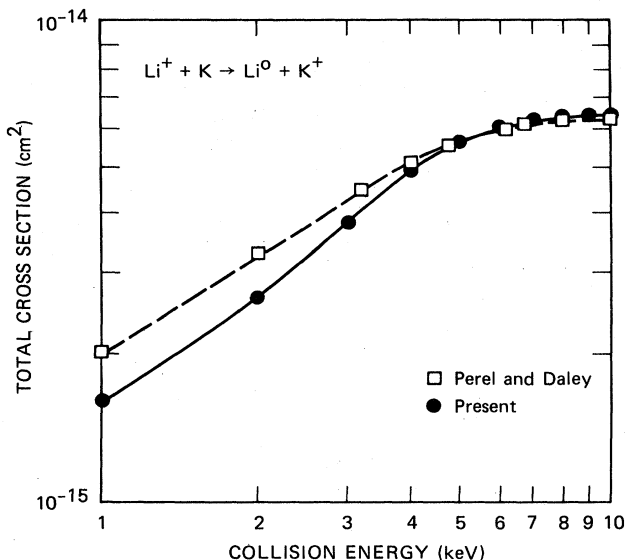


FIG. 3. Total charge neutralization cross section for  $\text{Li}^+$  ions in potassium vapor as a function of ion energy. The solid points are the present results, and the open symbols are the results of Perel and Daley (Ref. 9).

the larger cross section found by Perel and Daley at low energies includes an elastic scattering contribution. However, because of the much smaller magnitude of the  $\text{Li}^+ + \text{Mg}$  cross section at low energy relative to potassium, this test is not definitive in determining the effects of elastic scattering in our measured  $\sigma_{+0}$  for  $\text{Li}^+ + \text{Mg}$ .

The role of elastic scattering in the  $\text{Li}^+ + \text{alkaline-earth}$  charge transfer was confirmed, however, by Olson,<sup>18</sup> who calculated the cross section for  $\sigma_{++}$  beyond  $\theta_{\text{det}}$  for a Mg target at low  $\text{Li}^+$  collision energy. When these calculated values of  $\sigma_{++}$  are subtracted from the measured cross sections, the dotted line shown in Fig. 2 is obtained. The resulting cross section now shows a much smoother, monotonic energy dependence.

Olson<sup>18</sup> has also calculated the elastic scattering cross sections for the calcium and strontium targets at 1 and 2 keV  $\text{Li}^+$  energy. Those results were similarly subtracted from their respective measured cross sections to yield the dotted curves shown in Fig. 2. Note that although  $\sigma_{++}$  for  $\text{Li}^+$  on Ca and Sr are larger than for Mg, their effect on the total attenuation cross sections is very much less due to the magnitudes of the Ca and Sr data relative to the Mg. In the calcium case, the  $\sigma_{+0}$  cross section is lowered from 5.8 to 4.0 Å at 1 keV, while for Sr,  $\sigma_{+0}$  is reduced from 19 to 16.5 Å at the same energy.

The adiabatic treatment of curve crossing interactions by Massey<sup>19</sup> and Demkov<sup>3</sup> predicted that  $v_m$  should be proportional to the absolute value of the energy defect,  $|\Delta E_\infty|$ . A subsequent treatment by Drukarev<sup>5</sup> found that the dependence of  $v_m$  on  $|\Delta E_\infty|$  could vary according to the magnitude of the energy defect. When  $|\Delta E_\infty| \ll I$ , the ionization potential of the target atom, then the Massey adiabatic criterion would hold. However, when the energy defect was on the same order as the binding energy of the transferred electron, then Drukarev showed that  $v_m \propto |\Delta E_\infty|^{1/2}$ . The predictions have been subjected to numerous experimental tests involving both

alkali-metal and alkaline-earth charge transfer with a variety of projectile ions. Perel and Daley<sup>9</sup> measured the charge transfer for a number of mixed alkali-alkali systems, and found that their results could be parametrically represented as

$$v_m = a |\Delta E| / I^{1/2}, \quad (3)$$

with the constant  $a$  determined to be  $22 \times 10^7$  cm/sec  $\text{eV}^{1/2}$ . The theoretical treatment of Olson<sup>4</sup> for charge transfer at large internuclear distances similarly gives (3) for the dependence of  $v_m$  on  $\Delta E$ , with  $a = 14.5 \times 10^7$ . For protons on alkali-metal vapors, Gruebler *et al.*<sup>11</sup> also find that  $v_m$  is best described as being proportional to  $|\Delta E_\infty|^{1/2}$  rather than  $|\Delta E_\infty|$ . The values in Table I for the energy defects for these systems [assuming excited H(2s) products for the proton collisions] substantiate these findings. For proton collisions with alkaline-earth targets, Morgan and Eriksen<sup>12</sup> similarly obtain a reasonable correlation between their measured values of  $v_m$  and  $|\Delta E_\infty|^{1/2}$ . In any case, this type of representation for  $v_m$  should be viewed only as a simple approximation.

For the present collision systems, Table I shows that the energy defect is generally in the range where the velocity maximum should also be proportional to  $|\Delta E_\infty|^{1/2}$ , with the possible exception of  $\text{Li}^+ + \text{Ba}$ . In Fig. 4, our experimentally estimated values of  $v_m$  are plotted as a function of  $|\Delta E_\infty|^{1/2}$ , along with some of the previously mentioned results. As expected, the  $\text{Li}^+ + \text{alkaline-earth}$  charge-transfer systems show a good correlation between  $v_m$  and  $|\Delta E_\infty|^{1/2}$ . The slope of the fitted straight line in Fig. 4 gives a proportionality constant of  $6.1 \times 10^7$  cm/sec  $\text{eV}^{1/2}$ , which is in close agreement with the value of  $6.4 \times 10^7$  cm/sec  $\text{eV}^{1/2}$  found by Morgan and Eriksen.<sup>12</sup> Using this result, we can estimate the value of  $v_m$  for the  $\text{Li}^+ + \text{Mg}$  case to be  $9.1 \times 10^7$  cm/sec, corresponding to a collision energy of 28.5 keV. Thus, it is not unreasonable that the measured  $\sigma_A$  does not reach a maximum in the energy range studied here.

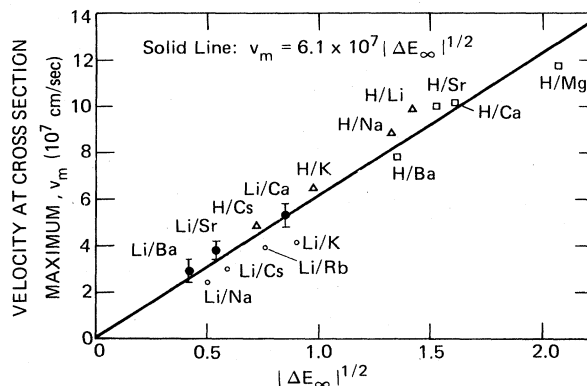


FIG. 4. The velocity,  $v_m$ , corresponding to the maximum in the electron-transfer cross section as a function of  $|\Delta E_\infty|^{1/2}$ . The points are labeled by ion/target. The values plotted include the present results (●), results from Gruebler *et al.* (Ref. 11) (Δ), Perel and Daley (Ref. 9) (○), and the data of Morgan and Eriksen (Ref. 12) (□). The line is the best fit to the data shown.

In summary, the total attenuation of  $\text{Li}^+$  ions in alkaline-earth vapor targets is very well described as a single electron capture process leading to ground-state neutral lithium atoms. Both the magnitude of the total cross section, and the velocity maximum,  $v_m$ , are found to correlate well with the asymptotic energy defect,  $|\Delta E_\infty|$ . The present results for  $\text{Li}^+ + \text{Ca}$  also confirm the recent theoretically determined  $\sigma_{+0}$  cross section of Kimura *et al.*<sup>14</sup>

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