Equilibrium charge-state fractions of Li^-, Li^0, and Li^+ in Mg, Sr, and Cs vapors

J. Richard Mowat,* Emily E. Fisch,† Alfred S. Schlachter, and J. Warren Stearns
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

Young K. Bae
Molecular Physics Department, SRI International, Menlo Park, California 94025
(Received 26 December 1984)

Li^+ ions, accelerated to energies between 1 and 20 keV, pass through thick vapor targets of Mg, Sr, and Cs. The ions that emerge into a narrow, forward-directed cone are electrostatically separated into three charge-state components: Li^-, Li^0, and Li^+. The principal results over this energy range are the following: (1) The negative fraction peaks at 5% near 5 keV for a Cs target, and is less than 1% in Mg and Sr targets; (2) for Sr and Cs targets, the beam is greater than 90% neutral; and (3) for a Mg target, the beam does not neutralize at the lowest energies studied, remaining mostly positive between 1 and 10 keV.

I. INTRODUCTION

Describing the mechanisms for negative-ion formation in ion-atom collisions at adiabatic energies is an interesting challenge to our present understanding of the dynamics of quasimolecular systems. Furthermore, the production of negative-ion beams is of current practical interest to the Magnetic Fusion Energy Program. An important motivation for this work on Li^- formation is a recent proposal to measure the spatial distribution of fusion-product alpha particles through the double-electron-transfer reaction[1,2]

$$Li^0 + He^{++} \rightarrow Li^{++} + He^0 .$$

The most efficient way to produce the high-energy neutral lithium atoms needed for this diagnostic probe is by electron detachment of fast Li^-. Other uses of fast Li atoms have been proposed for space applications. The choice of target vapors used in the present experiment was suggested by previous work at the Lawrence Berkeley Laboratory on D^- formation,[3,4] in which negative-ion equilibrium fractions larger than 30% were found in Cs and Sr targets.

The detection of Li^- was first reported by Sloane and Love[5] who bombarded a Ni surface with Li^+ ions from a lithium thermionic source and analyzed the scattered ions with a mass spectrometer. The measured electron affinity 0.620(7),[6] agrees with accurate values calculated[7] for the $1s^2 2s^2$ configuration. Li^- is one of only two negative ions known to possess a second bound state; radiation from the core-excited configuration $1s\,2s\,2p^2$ has been observed[8] with a 2.3 ns lifetime. Since that state can autodetach and does not live long enough to be detected in the present experiment, the results presented below refer to production of ground-state Li^-.

The formation of negative ions of species heavier than hydrogen by charge transfer has recently been reviewed.[9] This review summarizes data for species as heavy as Cl.

II. EXPERIMENTAL ARRANGEMENT

A schematic diagram of the apparatus is shown in Fig. 1. $^7Li^+$ ions are formed by surface ionization in an indirectly heated, isotopically purified, β-eucryptite impregnated, porous tungsten plug.[10] A very stable ion current of about 1 μA is extracted from a Pierce electrode. The 1—20 keV acceleration potential is obtained from a well-regulated dc power supply. After electrostatic focusing and steering the beam is 100% square-wave modulated at a 50% duty cycle by a transverse electric field. Magnetic analysis then directs the beam into one of two beam lines where it passes through either a stainless-steel recirculating Cs-vapor heat-pipe target[3] or a differentially pumped iron oven[4] containing Mg or Sr vapor . A chromel-alumel thermocouple is used to measure the temperature at the reservoir. That temperature is used to estimate the target number density from a least-squares fit to temperature-vapor pressure data.[11] Neutral atoms and ions emerging from the target within approximately $\pm 3.5°$ of the forward direction pass through an electrostatic field which directs the positive and negative components into magnetically suppressed Faraday cups and which allows the neutral component to pass undeviated onto the face of a pyroelectric detector.[12] Both electrostatic and magnetic suppression are used to prevent secondary electrons from the front surface of the pyroelectric detector from reaching the Faraday cups; the small secondary-electron flux could mask the small Li^- signal. The two targets are coupled to identical analysis chambers[3,4] which terminate each beam line. Beam components travel approximately

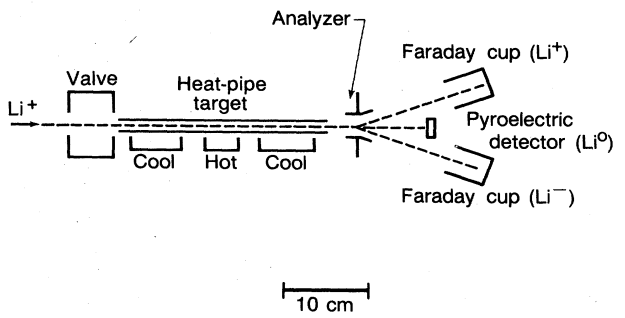

FIG. 1. Schematic diagram of the apparatus.

10 cm between the target and the analyzing field. The vacuum, maintained by liquid-nitrogen-trapped oil diffusion pumps, is typically less than 2×10^{-6} Torr in all beam lines and chambers, except inside and immediately adjacent to the targets.

The signals from the two Faraday cups are amplified by electrometers whose outputs are digitized, integrated, and recorded by a microcomputer. The pyroelectric detector is insensitive to the charge of the particles impinging on its front surface, so it is calibrated by comparing its output to the output of a Faraday cup when the Li^+ beam is switched back and forth between the two. The output of the pyroelectric detector is rectified and amplified with a lock-in amplifier that is synchronized to the $\sim$1-Hz beam modulator. The output of the lock-in amplifier is also digitized, integrated, and recorded by the computer. Since charge-state fractions are measured, but not the beam intensity incident on the target, transmission through the target is estimated by assuming constant incident Li^+ intensity.

Typical beam currents reaching the Faraday cups are 200 nA for the Cs heat-pipe target, and 20 nA for the alkaline-earth oven, which requires a more highly collimated beam. At equilibrium thickness the beams are attenuated from these values by 10% to 90% depending upon the beam energy, target geometry, and target material.

III. EXPERIMENTAL PROCEDURE

Growth curves were taken for each target at the lowest and highest beam energies in order to determine the target thickness required for equilibrium. An example is shown in Fig. 2, for 4-keV Li^+ in cesium vapor. The target temperature was freely rising (i.e., driven at a constant power level), and the temperature change between points is typically 5 °C. Charge-state fractions are calculated from the integrated signals and from the pyroelectric-detector calibration constant, subject to the condition that the three charge-state fractions sum to unity. The densities that label the abscissa of Fig. 2 are the densities estimated at the center of the heat-pipe target, whose length[3] is about 5.3 ± 0.7 cm. Once the temperature corresponding to equilibrium thickness is determined for a given target and for each of the three charge-state fractions, the target is maintained at the fixed temperature and the fractions are measured as a function of beam energy.

Various systematic checks were performed to verify correct behavior of the apparatus. The measured equilibrium charge-state fractions are found to be insensitive to the polarity of the analyzing field and to small variations in the magnitude of the field. Likewise, the pyroelectric-detector calibration is found to be insensitive to the polarity of the analyzing field, the position of the beam on its 2.2-cm-diam face, and the position at which the charged beams strike the Faraday cups. The calibration constant is found to vary linearly with beam energy, as expected.

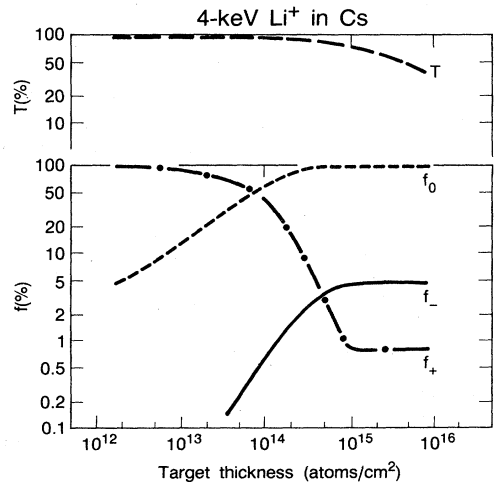

FIG. 2. Charge-state fractions as a function of target thickness for 4-keV Li^+ in cesium vapor.

IV. RESULTS AND DISCUSSION

The equilibrium charge-state fractions f_-^∞, f_0^∞, and f_+^∞ were measured for incident Li^+ ions over the energy range 1—20 keV in targets of magnesium ($Z=12$), strontium ($Z=38$), and cesium ($Z=55$) vapor. No previous experiments have measured Li charge-state fractions for this energy range in either Mg- or Sr-vapor targets. One previous report[13] has results for equilibrium fractions in Cs vapor for energies between 5 and 40 keV, while another[14] reports conversion efficiencies between 2 and 20 keV. Conversion efficiencies cannot be directly compared with equilibrium fractions, since the former are apparatus dependent. Measurements in Na vapor have also been reported.[15] Results for each target are described below and are shown in Figs. 3—5 and in Table I.

A. Magnesium-vapor target

The equilibrium fractions for 1—20-keV Li^+ in magnesium vapor are shown in Fig. 3. The f_0^∞ and f_+^∞ curves join smoothly with the results of an older, higher-energy experiment[16] which did not report measuring a negative component.

Previous studies[17] in a Mg-vapor target suggest that beams of elements from columns 6A and 7A of the

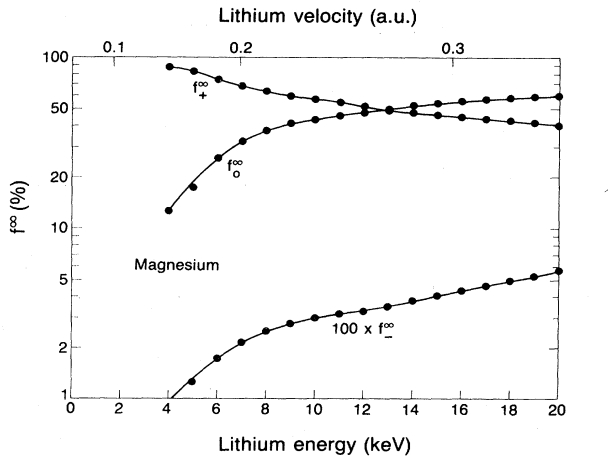

FIG. 3. Equilibrium charge-state fractions: Li in Mg vapor.

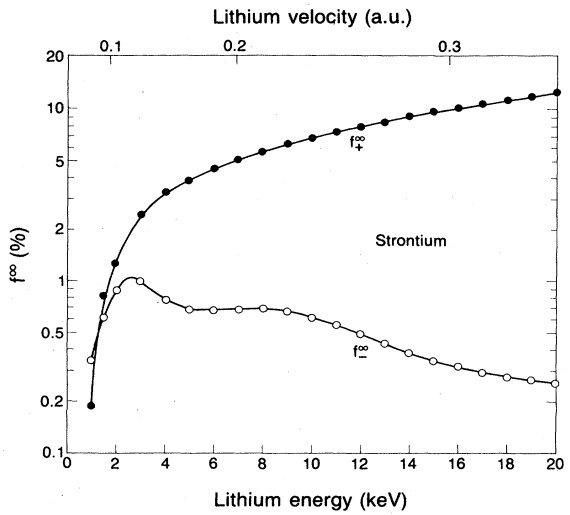

FIG. 4. Equilibrium charge-state fractions: Li in Sr vapor.

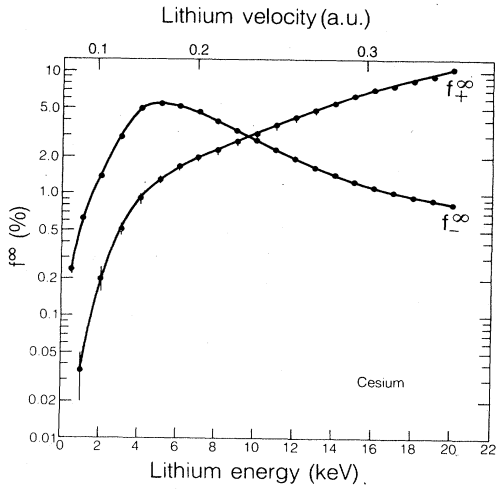

FIG. 5. Equilibrium charge-state fractions: Li in Cs vapor.

Periodic Table tend to have relatively small neutral equilibrium fractions ($<65\%$) for energies between 10 and 100 keV, while elements to the left in the periodic table tend to have relatively large neutral fractions ($>70\%$) for the same energies. The results reported here for Li ($f_0^\infty < 60\%$) do not fit into that trend.

At the highest energies used here, the neutral fraction dominates. In most beam-target systems, the neutral fraction becomes larger as the energy is lowered. An unexpected finding is that the neutral fraction *decreases* in favor of the positive fraction as the energy is lowered from 20 keV. For energies below about 12 keV and extending to the lowest energy measured to this experiment, the beam does not neutralize to a high degree, and the positive fraction dominates.

The probable reason for this observation is that the electron-capture cross section for $Li^+ + Mg \rightarrow Li + Mg^+$ is very small below 10 keV. Such behavior is expected if recent calculations[18] on the similar $Li^+ + Ca \rightarrow Li + Ca^+$ system are scaled to the endoergicities ΔE of the respec-

TABLE I. Equilibrium charge-state fractions f_i^∞ for Li in Mg, Sr, and Cs vapors (in %). Uncertainty is 5% or less unless otherwise indicated.

	Mg			Sr			Cs		
E (keV)	f_+^∞	f_0^∞	f_-^∞	f_+^∞	f_0^∞	f_-^∞	f_+^∞	f_0^∞	f_-^∞
1				0.19[a]	99.5	0.34[a]		99.3	0.62[b]
1.5				0.83[b]	98.6	0.61[b]			
2				1.3[b]	97.8	0.88[b]	0.19[b]	98.5	1.3[c]
3				2.5[b]	96.5	0.99[b]	0.48[b]	96.8	2.8[c]
4	87[c]	12.8[b]	0.0096[c]	3.3[c]	96.0	0.76[c]	0.85[b]	94.5	4.6
5	82.4	17.6[c]	0.0126[c]	3.8	95.5	0.68	1.3[c]	93.5	5.2
6	74.3	25.7	0.017	4.5	94.8	0.68	1.6	93.5	4.9
7	67.3	32.6	0.021	5.1[c]	94.2	0.68[c]	1.9	93.7	4.4
8	62.8	37.2	0.025	5.6	93.7	0.68	2.2	94.1	3.7
9	59.1	40.8	0.027	6.2	93.1	0.66	2.5	94.4	3.1
10	57.3	42.6	0.030	6.7	92.7	0.61	3.0	94.4	2.6
11	54.8	45.2	0.032	7.2	92.2	0.56	3.4	94.4	2.2
12	51.8	48.1	0.034	7.8	91.7	0.49	4.0	94.2	1.8
13	49.0[c]	50.0	0.036	8.3	91.2	0.44	4.6	93.8	1.56
14	47.8	52.2	0.038	9.2	90.4	0.39	5.2	93.4	1.36
15	46.1	53.9	0.041	9.8	89.9	0.34	6.0	92.8	1.22
16	44.7	55.3	0.044	10.3	89.4	0.32	6.7	92.2	1.09[c]
17	43.5	56.5	0.047	10.8	88.9	0.29	7.4	91.6	1.05[c]
18	42.2	57.7	0.050	11.3	88.4	0.28[c]	8.2	91.8	0.97[b]
19	41.4	58.5	0.054	11.8	87.9	0.27[c]	9.0	90.1	0.91[b]
20	40.7	59.2	0.058	12.4	87.4	0.26[c]	9.7	89.4	0.84[b]

[a] ±30%.
[b] ±20%.
[c] ±10%.

tive reactions. The ΔE for $Li^+ + Ca$ electron capture is 0.72 eV, while that for $Li^+ + Mg$ is 2.26 eV. For $Li^+ + Ca$, the maximum cross section is realized[18] at $E \approx 10$ keV. Using the adiabatic criterion[19] which predicts that the velocity at the maximum in the cross section will scale inversely with ΔE, one is led to predict the $Li^+ + Mg$ electron-capture cross section will not maximize until the energy is above those studied here. Thus, our observations of a small neutral fraction are in accord with theoretical predictions. It should be noted that the heavier alkaline earths, such as Sr, where $\Delta E = 0.30$ eV, will necessarily have large electron-capture cross sections at low energies and will not exhibit behavior similar to that of Mg.

An estimate of the electron-detachment cross section σ_{0+} can be made from f_0^∞ when the electron-capture cross section σ_{+0} is known, if the negative fraction is sufficiently small for a two-component model to be valid. In this model the neutral fraction is

$$f_0^\infty = \sigma_{+0}/(\sigma_{+0}+\sigma_{0+}) .$$

Cross sections σ_{+0} for electron capture have recently been measured by Coggiola, Bae, and Peterson.[20] These values combined with present measurements of f_0^∞ provide an estimate of the electron-detachment cross section σ_{0+}, which lies between 1.0×10^{-15} and 1.2×10^{-15} cm^2 over the energy range 4—10 keV.

B. Strontium-vapor target

Figure 4 summarizes the energy dependence of f_-^∞ and f_+^∞ in strontium vapor. Previous measurements[4] of the D^- charge-state fraction in Sr vapor showed a peak at about 50% for a D^- energy near 0.5 keV. Note in the figure that there is a small peak at about 2.5 keV, with a broad shoulder at higher energies. This is qualitatively similar to the D^- yield in the same target at the same velocities, although the maximum Li^- yield of 1% is very small compared to 50% for D^-.

C. Cesium-vapor target

The largest negative fraction (5.4%) observed for the three targets studied was for a cesium-vapor target. This result is shown in Fig. 5. The negative fraction also exceeds the positive fraction at energies less than 10 keV. Our observations confirm a trend noted earlier[13] for other alkali-metal projectiles in cesium vapor; the negative-ion fraction peaks at a projectile velocity of about 0.17 a.u. As with the Sr target, but unlike the Mg target, the positive-ion fraction is monotonically decreasing as the energy is reduced.

D. Scattering

The incident beam is not tightly collimated in this experiment, nor is it continuously monitored, hence little can be said about scattering. The collimation of the beam after the target is designed so that only ions and atoms that scatter into a cone of half-angle approximately 3.5° about the forward direction enter the analysis chamber. For the cesium target the following quantitative statements can be made. At equilibrium, by definition, almost all of the ions undergo more than one collision that is close enough for an electron to be captured or lost. We find that at least 80% of the beam reaches the analysis chamber; it thus follows that most of the ions are not scattered by more than 3.5°. For the Mg and Sr targets, because the target collimation is tighter, scattering loss is much greater within the target, and the downbeam collimation does not determine an upper bound for the scattering angle.

E. Uncertainties

The systematic errors associated with the data in Figs. 3—5 are estimated to be equal to or smaller than the plotting symbols except where noted by error bars. These uncertainties are a measure of the repeatability of the charge-state fractions over a period of time and arise mainly from variations in the neutral-atom detector. Additional uncertainties occur for very small fractions, where instrument drift and various backgrounds increase the uncertainty of the charge-state-fraction measurements. Typical uncertainty for an equilibrium yield is ±5%. Charge-state fractions are, of course, determined at equilibrium thickness, so there is no error associated with target temperature measurement or target density determination. The error associated with the ion energy and q/m is determined by the stability and calibration of the accelerator and magnet power supplies, which are well enough regulated so that those errors are negligible.

V. CONCLUSION

The negative-ion equilibrium charge-state fractions of lithium ions passing through vapors of Mg, Sr, and Cs show quite different trends, as seen in Fig. 6. The maximum negative-ion fraction occurs near 5 keV in Cs va-

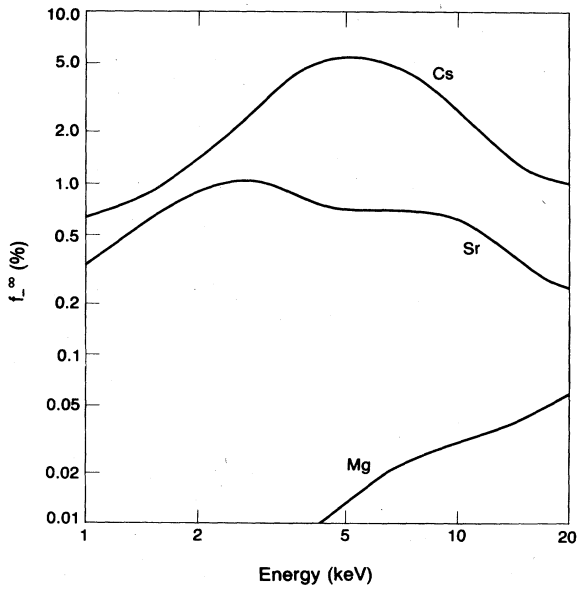

FIG. 6. Summary of f_-^∞ for Li ions in metal vapors.

por, and for energies below 10 keV, the emergent Li beam has a net negative charge. While negative-ion formation is extremely small in a Mg target, interesting behavior is observed, indicating an inhibition of neutralization at low projectile energies, so that the emergent beam has a net positive charge. In contrast to production of D^- at comparable velocities, very little Li^- is produced in Sr vapor.

ACKNOWLEDGMENTS

We would like to thank Dr. R. E. Olson for helpful conversations, Dr. J. R. Peterson for providing data on charge transfer of Li^+ in Mg vapor, prior to publication, and Dr. R. V. Pyle for his support of this project. One of us (E.F.) would like to acknowledge support from the Student Employment and Training Program at the Lawrence Berkeley Laboratory. This work was supported by the Director, Office of Energy Research, Office of Fusion Energy, Applied Plasma Physics Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, by the U.S. Department of Defense, and by the U.S. Air Force Office of Scientific Research under Contract No. F47620-82k-0030.

*Permanent address: Department of Physics, North Carolina State University, Raleigh, NC 27695-8202.

†Present address: Department of Physics, Bryn Mawr College, Bryn Mawr, PA 19010.

[1]R. W. McCullough, T. V. Goffe, M. B. Shah, M. Lennon, and H. B. Gilbody, J. Phys. B **15**, 111 (1982).

[2]G. A. Murray, J. Stone, M. Mayo, and T. J. Morgan, Phys. Rev. A **25**, 1805 (1982).

[3]A. S. Schlachter, K. R. Stalder, and J. W. Stearns, Phys. Rev. A **22**, 2494 (1980).

[4]R. H. McFarland, A. S. Schlachter, J. W. Stearns, B. Liu, and R. E. Olson, Phys. Rev. A **26**, 775 (1982).

[5]R. H. Sloane and H. M. Love, Nature (London) **159**, 302 (1947).

[6]T. A. Patterson, H. Hotop, A. Kasdan, D. W. Norcross, and W. C. Lineberger, Phys. Rev. Lett. **32**, 189 (1974).

[7]Recent calculations are reviewed by H. Hotop and W. C. Lineberger, J. Phys. Chem. Ref. Data **4**, 539 (1975).

[8]R. L. Brooks, J. E. Hardis, H. G. Berry, L. J. Curtis, K. T. Cheng, and W. Ray, Phys. Rev. Lett. **45**, 1318 (1980).

[9]A. S. Schlachter, in *Production and Neutralization of Negative Ions and Beams,* edited by K. Prelec (AIP, New York, 1984), Vol. III, p. 300.

[10]O. Heinze and R. T. Reaves, Rev. Sci. Instrum. **39**, 1229 (1968).

[11]R. R. Hultgren, P. A. Desai, T. Hawkins, M. Gleiser, K. K. Kelley, and D. Wagman, *Selected Values of the Thermodynamic Properties of the Elements* (American Society of Metals, Metal Park, Ohio, 1973).

[12]K. H. Berkner, B. R. Meyers, and R. V. Pyle, Rev. Sci. Instrum. **39**, 1204 (1968).

[13]P. Tykesson, in Proceedings of the 1978 Symposium of Northeastern Accelerator Personnel, Oak Ridge, Tennessee, 1978 (unpublished).

[14]E. Steffens, IEEE Trans. Nucl. Sci. **NS-23**, 1145 (1978).

[15]J. Heinemeier and P. Hvelplund, Nucl. Instrum. Methods **148**, 425 (1978).

[16]L. I. Pivovar, L. I. Nikolaichuk, and V. M. Gurov, Zh. Eksp. Teor. Fiz. **49**, 1072 (1965) [Sov. Phys.—JETP **22**, 746 (1966)].

[17]J. Heinemeier and P. Hvelplund, Nucl. Instrum. Methods **148**, 65 (1978).

[18]M. Kimura, H. Sato, and R. E. Olson, Phys. Rev. A **28**, 2085 (1983).

[19]J. B. Hasted and A. R. Lee, Proc. Phys. Soc. London **79**, 702 (1962).

[20]M. J. Coggiola, Y. K. Bae, and J. R. Peterson (unpublished).