# Experimental cross sections for electron capture from lithium by slow, highly charged, rare-gas projectiles

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Experimental cross sections have been measured for single-electron capture from Li by Ne, Ar, Kr, and Xe ions in charge states ($q$) between 2 and 10 and at projectile energies between $100q$ and $1000q$ eV. The cross sections are found to depend on the incident charge state but are nearly independent of both the projectile species and energy. Good agreement between the experimental results and those of absorbing sphere and classical barrier models is found.

## I. INTRODUCTION

The capture of electrons by multiply charged projectiles whose velocities are slow compared to the target electron orbital velocities has recently been the center of much experimental and theoretical interest. Numerous review articles and conference proceedings[1-8] have discussed the reasons for this, which include both the importance of this process in plasmas and the challenge of finding good theoretical descriptions of the long-range transfer. Because the transfer usually occurs at large internuclear distances, where crossings between covalent incident and ionic exit potential curves occur, the cross sections are quite large and often populate highly excited states on the projectile. These characteristics are particularly pronounced if the target electron is weakly bound, as is the case for the Li target used here. Some particular advantages which this target presents for plasma diagnostics have been discussed by Winter[9] and by Post.[10]

Theoretical approaches to such collisions have been of two major kinds. Detailed calculations for specific systems, using both eigenfunction-expansion and configuration-space techniques, have been given for a number of cases, usually involving an atomic hydrogen target.[1-4,6] Such an approach offers accurate treatments at the expense of generality. A more general approach has been the development of models which treat the process, within a one-electron picture, under general assumptions which allow the use of the results for analysis of a wide range of collision systems. Such models include the absorbing sphere model (OSAS) of Olson and Salop,[11] the tunneling model (GJ) of Grozdanov and Janev,[12] and the classical barrier model (CBM).[13,14]

The OSAS model, which is based on a multichannel Landau-Zener treatment of the coupling-matrix element, gives somewhat too high cross sections for $R^{q+}$ on $R$, where $R$ is used in this article to denote any of the rare gases Ne, Ar, Kr, or Xe.[11,15,16] For highly charged $Fe^{q+}$, $Xe^{q+}$, and $Ar^{q+}$ on atomic and molecular hydrogen,[17,18] the agreement is better. For the cases of $C^{q+}$ and $O^{q+}$ on H for $2<q<6$, the OSAS results are generally much higher than experiment,[19] as is the case for $R^{q+}$ on He.[20] Recent extensions of the multicrossing Landau-Zener model have been made to allow calculation of the population of individual final states.[21] The GJ model, which attributes the capture to under-barrier tunneling of the electron through the maximum in the double well formed by the two charge centers, predicts cross sections which tend to lie only slightly higher than the OSAS ones and tend also to somewhat overestimate the cross sections,[1,11,15-20,23] the worst cases being those for which the OSAS also fails.[19,20] The CBM, which describes the capture as a classical over-barrier transition, has proved quite successful in giving the principal quantum number ($n$) of the electron in its final state,[14,23-26] but is less reliable for cross-section calculations.[15,17,18,20,23,27,28] Each of these general treatments is expected to be most successful when the projectile core charge is high enough that the capture goes into states on the projectile characterized by very high principal quantum number ($n$), for which core structure is negligible. In many experimental cases studied above, this condition has not been well met, and comparison between theory and experiment has not always been particularly appropriate.

This paper describes an experimental study of electron capture from lithium by low-energy, highly charged (LEHQ) ions of Ne, Ar, Kr, and Xe. The Li target case is an especially appropriate one for the testing of the general models. The binding energy of the lone $2s$ electron in the $1s^2 2s$ ground-state configuration of Li is only 5.39 eV. The inner-shell electrons are relatively tightly bound, with the result that single capture completely dominates the total capture process at low energy. A one-electron treatment of the problem should be valid. The low binding energy of the $2s$ electron means that the capture proceeds at very large internuclear distances into very high $n$ states. Thus the captured electron sees only the core monopole charge, whose treatment as a point charge is better justified than is the case for capture of more tightly bound target electrons. It is interesting to note that, while the atomic hydrogen target is the only true one-electron target, the lower binding of alkali-metal targets make them in some sense better test targets for the general models than atomic hydrogen.

Several previous experiments on electron capture at low energy from Li by multiply charged ions above He have been reported, although much of the data is for lower incident projectile charge states and higher incident energies

 

than used here. Rille and Winter[29] used photon spectroscopy to investigate the final-state populations in $Ne^{2+}$ on Li, and have measured total capture cross sections between 20 and 60 keV. Recently Dijkamp *et al.*[30] have measured final-state distributions and total cross sections for capture from Li by $C^{q+}$ and $O^{q+}$ projectiles with $2<q<6$ at accelerating voltages between 15 and 150 kV. A single cross section for capture by $Xe^{10+}$ at $3.8\times10^{7}$ cm/sec from Li was reported by Müller *et al.*[31]

## II. EXPERIMENT: PRODUCTION AND DETECTION OF LEHQ IONS

The technique used to produce LEHQ ions for this experiment has been reported previously in the literature.[15,20,32] Fast, pulsed 19-MeV $F^{4+}$ and $F^{8+}$ beams from the Kansas State University EN tandem Van de Graaff accelerator were passed through a gas cell containing rare gases to produce a source of ions. The fluorine beam was typically pulsed to a width of 5 ns or less at a repetition rate of 8 $\mu$sec. Figure 1 shows the experimental system which was kept at base pressures on the order of $2\times10^{-6}$ Torr. Ions created in the gas cell, called projectile ions, were extracted by applying voltages to the cell and the three-grid system attached to the cell. These ions then passed through a resistively heated Li vapor oven, and into a double-focusing spherical sector electrostatic analyzer. For a fixed acceleration voltage $V_1$ the analyzer voltage $V_a$ at which an ion of original charge state $q$ will pass through to detection is given by

$$V_a = K\frac{qV_1}{q'},$$

where $K$ is a constant depending on the geometry of the analyzer and $q'$ is the charge of the ion in the analyzer. The voltage applied to the analyzer was swept from $KV_1$ to slightly above $2KV_1$ through the use of a triangle wave generator. The total flight time of each ion from production to detection, proportional to $\sqrt{m/q}$, was recorded in coincidence with $V_a$ for the same event to form a two-dimensional spectrum, such as that shown in Fig. 2. Events for which no charge exchange has occurred lie along a line at 500 eV/$q$ in this spectrum, while single-capture events lie above them along a curved line. No double-capture events are visible above the discriminator level for that figure.

A side view of the Li vapor oven is shown schematically in the lower half of Fig. 1. The stainless-steel cylindrical oven was 1.90 cm in diameter and 1.85 cm in length with entrance and exit apertures of 1.0 and 2.0 mm diameter, respectively. The oven was attached to the mounting system of the analyzer by two ceramic standoffs on one end, and a tapped loading port was located on the other end. The oven was heated by passing a dc current through heater wire wrapped around the oven. Two Chrommel-Alumel thermocouples were silver soldered to opposite faces of the oven. The reference junction of each thermocouple was immersed in an ice bath at 0 °C and tabulated Chrommel-Alumel voltage temperature characteristics were used to determine the temperature of the oven.[33] The Li vapor pressure inside the cell was determined using this temperature and known temperature-pressure relationships.[34] From this pressure the absolute gas target density was then calculated. The oven was typically in the range from 625 to 670 K corresponding to target densities of 0.34 to $2.4\times10^{12}$ atoms per cm$^2$.

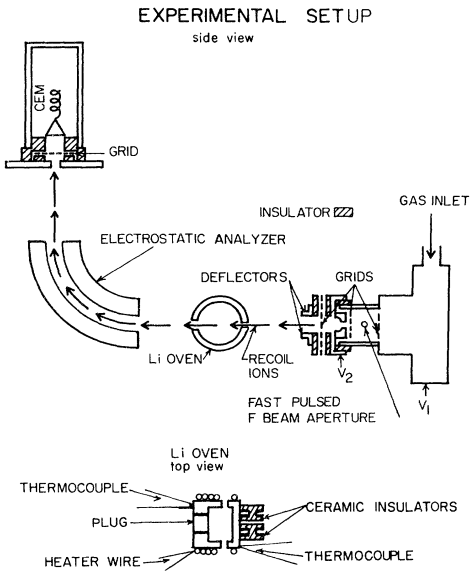


FIG. 1. Schematic of apparatus.

Although the silver-soldered thermocouples gave consistent and reproducible results, we sought an independent check on the absolute target density by measuring, with the same oven, cross sections for single capture by protons and $He^{2+}$ on Li. In the former case, our results were in good agreement with previously published data of Grüeber *et al.*[35] In the latter case, our absolute scale was found to be consistent with published results of Murray *et al.*[36] and McCullough *et al.*[37] although we were not

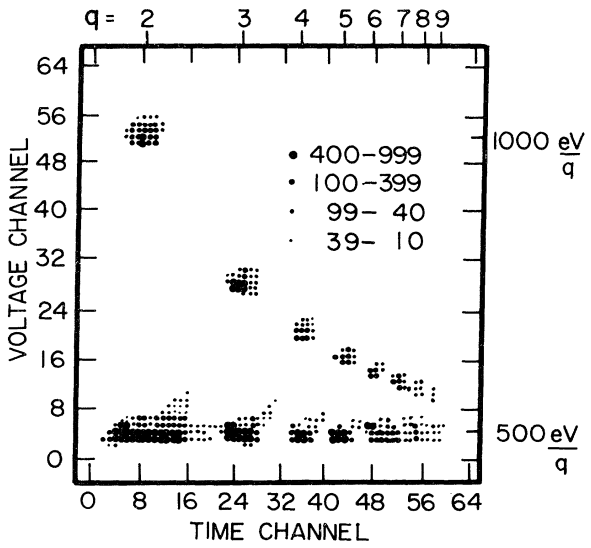


FIG. 2. Two-dimensional spectrum of events from $Ar^{q+}$ on Li at $E=500q$ eV. Horizontal axis is the flight time, dispersing $q$, and the vertical axis is the electrostatic analyzer voltage, dispersing the post-collision charge states $q'$.

able to go quite high enough in energy to overlap their data and some extrapolation is necessary for comparison of the two sets of data. Some small differences in the energy dependence of the cross sections for single capture by $He^{2+}$ on Li were found between our results and those of Murray *et al.*, and will be reported in a separate publication.

Cross sections were obtained by projecting slices of two-dimensional spectra onto the $V_a$ axis for each $q$ (see Fig. 2), and taking the ratio between charge-exchange and direct peaks. Suitable corrections for the $V_a$-dependent spectrometer acceptance window and computer dead time were made. Linearity of the yield with target thickness was verified experimentally for several representative cases. Details of the data-reduction procedure are given elsewhere.[15,20]

The uncertainties in our cross-section measurements are of two types, relative and absolute. In most cases, enough independent runs for each cross section were made that meaningful relative standard deviations could be calculated. For cases where an insufficient number of runs were made, relative error bars either reflect the counting statistics or were estimated on the basis of known reproducibility for similar cases. Assignment of the absolute error bar is more problematic. The data presented here were found to be reproducible over a series of modifications to the oven, including the use of two entirely separate ovens, the removal and reattachment of the thermocouples, and the disassembly and assembly of the entire apparatus several times, to within approximately 10%. Our agreement with other data (see above) is at approximately this same level. Combining this quadratically with an assumed 9% uncertainty in effective target cell length we assign an overall absolute error bar of 13% to the data. The error bars in the figures represent only the relative errors; an additional 13% uncertainty in absolute scales attend all data shown in this paper.

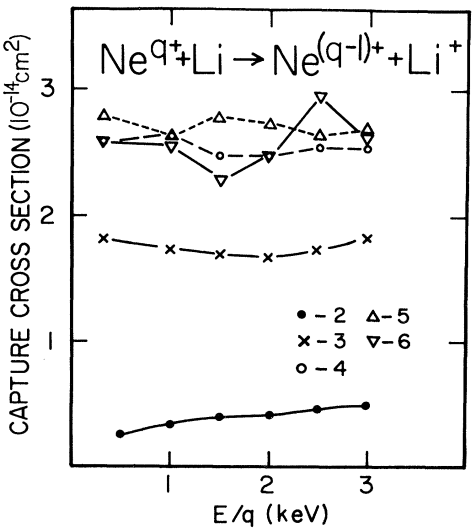


FIG. 3. Energy dependence of single-capture cross sections for $Ne^{q+}$ on Li.

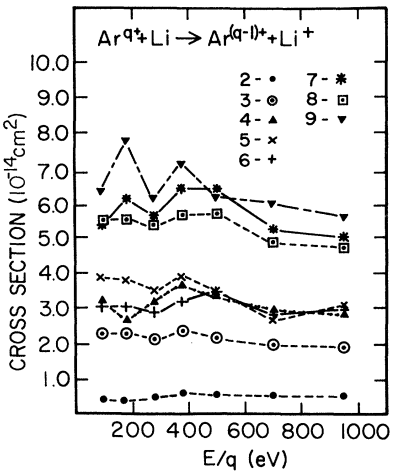


FIG. 4. Energy dependence of single-capture cross sections for $Ar^{q+}$ on Li.

## III. RESULTS

The energy dependence of total electron capture cross sections for collisions involving Li target atoms with Ar and Ne projectiles are presented in Figs. 3 and 4, respectively. Except for the case of Ne projectiles of charge state $q=2$, both Ne and Ar cross sections show almost no dependence on projectile energy, an effect which is now well documented for such collisions. The total electron capture cross sections for Ne, Ar, Kr, and Xe on Li are presented in Fig. 5 as a function of charge state $q$. The Ar, Kr, and Xe data, as well as the Ne $q=2$ data, are for a fixed projectile energy of $500q$ eV. For higher Ne charge states some data collected at higher energies were used in the Ne cross-section determinations.

The cross sections in Fig. 5 are seen to be nearly independent of projectile species for a given value of $q$ for $2<q<7$. A departure from this result occurs for $q=7$–9. These are the charge states for which the beams

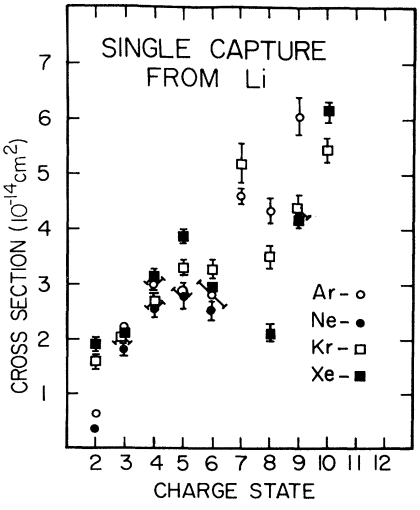


FIG. 5. Cross sections for single capture from Li by $Ne^{q+}$, $Ar^{q+}$, $Kr^{q+}$, and $Xe^{q+}$ at a projectile energy of $500q$ eV.

from the recoil ion source are known to possess appreciable electronically excited metastable components. If some fraction of the beam is in an electronically excited metastable state (for example, the $2p^5 3s\ ^3P_{0,2}$ states for $Ar^{8+}$), capture of a single electron may form a state on the projectile which can promptly autoionize, and thus the projectile charge state will revert to its original one. Such an event is lost in the present experiment from the charge-exchange channel, but such events were detected, for the same ion source and beams, by Justiniano *et al.*[15,20] who measured in coincidence the charge states of both projectile and target after the collision for various rare-gas targets. The metastable fraction in the beam is interpreted to be the ratio of the measured cross section for producing singly charged target ions in coincidence with projectiles experiencing no charge change to that for producing all singly charged target ions. Using the metastable beam fractions deduced in this way from the results of Refs. 15 and 20 we have corrected the present results for event losses due to this effect by dividing each cross section in Fig. 5 by 1 minus the appropriate metastable fraction for $q=7$–9. The resulting corrected cross sections are shown in Fig. 6, where the scatter for $q=7$–9 is seen to be considerably reduced. The $Xe^{8+}$ point remains anomalously low.

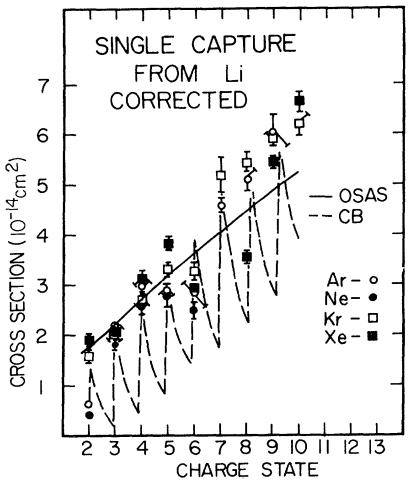


FIG. 6. Similar to Fig. 5, but corrected for metastable beam components. Theoretical curves are as follows: solid line, OSAS adsorbing sphere (Ref. 11); dashed line, CBM (Refs. 13 and 14).

## IV. DISCUSSION

The velocity independence for $q>2$ is well documented in the literature and occurs because of the large number of curve crossings which occur near the favored internuclear distance where the transfer takes place. The characteristic velocity dependence of any single crossing is lost in the sum over many channels. This result is predicted by all of the general models discussed above. One exception noted to this behavior occurs for $Ne^{2+}$ on Li (see Fig. 7). The previous results of Rille and Winter[29] show only a weakly velocity-dependent behavior of the cross section between 20- and 60-keV bombarding energy. They conclude from a comparison of their measured photon emission and total capture cross sections that the capture probably proceeds mainly to $3p$ and $3d$ orbitals about the $^3P$ and $^1D_2$ $Ne^{2+}$ projectile cores. The corresponding crossing radii are near 8.5 and 25 a.u., respectively. Our cross sections at projectile energies up to only 6 keV show a very strong energy dependence for this capture, indicating that the multichannel argument does not hold in this case. The maximum geometrical cross section which a crossing at 8.5 a.u. can give is only $0.6\times10^{-14}$ cm$^2$. Thus a cross section as large as $2.2\times10^{-14}$ cm$^2$ at 60 keV requires dominant $3d$ capture. It seems unlikely, however, that behavior at the $3d$ crossing could be other than diabatic at as high an energy as 20–60 keV since it would require a coupling-matrix element orders of magnitude larger than would be expected for such a distant crossing.[11,38] Our total cross sections are more consistent with the $3s$ and $3p$ capture cross sections deduced by Rille and Winter from their emission cross sections. An extension of the Landau-Zener treatment presented by Rille and Winter into our velocity regime gives a good representation of the energy dependence of our cross section, suggesting that the $3p$, and to a lesser extent, $3s$ capture is dominating. The $3d$ crossing should be characterized by an even smaller Landau-Zener velocity than are the $3s$ and $3p$ crossings, and would be expected to maximize very much below 20–60-keV region. There appears to be an unresolved discrepancy between the two sets of total cross-section measurements. (See note added in proof.)

For the higher-$q$ cases the conjecture that, for such a loosely bound target, effects due to projectile core structure should recede is confirmed by the dependence of the cross sections only on incident charge state. In the spirit of this result, we choose to treat the projectile as a point charge of size $q$ in comparing our results with model calculations. This choice is deemed preferable to the use of effective charges which, if treated correctly, would have to be both species and $q$ dependent. In particular, it would be inappropriate to choose for each projectile ion an effective charge based on that ion's ground-state binding energy, since capture is to much higher levels usually for which the point-charge approximation is much better than for the ground-state configuration.

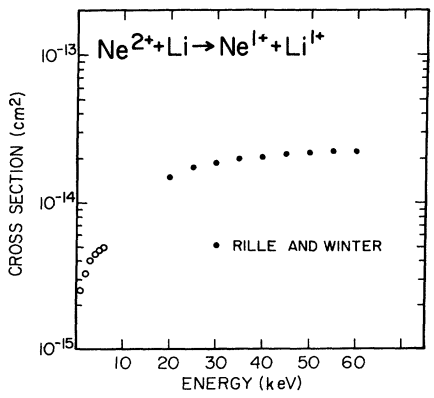


FIG. 7. Cross sections for single capture from Li by $Ne^{2+}$ vs projectile energy. Open circles, present data; solid circles, Rille and Winter (Ref. 29).

The solid line in Fig. 6 shows the result of the OSAS model, calculated for Ar projectiles at $E=500q$ eV. The results of this model are sufficiently weakly velocity dependent that the predicted cross sections for all projectile masses used are nearly the same as for this case (within 10%). The predicted "absorption" radii lie between 13 and 25 a.u. (for $q=2$ to 10), and correspond to exoergicities of approximately 2—10 eV, respectively. Examination of the relevant energy-level diagrams shows that, for all cases except $Ne^{2+}$ and $Ar^{2+}$, there are many possible level crossings near the absorption radii and thus the principal assumption of the model is well justified (see Fig. 8). The agreement between the model prediction and the present results is thus gratifying, and gives strong support to the formulation of the model. The GJ model results are not directly available to us for this case, but have been found previously to be slightly higher than the OSAS results for other cases.

The dashed line in Fig. 6 shows the CBM result using the strictly hydrogenic model for the projectile. The cross sections were calculated from $\sigma=\frac{1}{2}\pi R_x^2$, with

$$R_x=\frac{2(q-1)}{(q^2/n^2-2I_t)}$$

and

$$n=\frac{q}{\sqrt{2I_t}}\left[\frac{1+2\sqrt{q}}{q+2\sqrt{q}}\right]^{1/2},$$

where $I_t$ is the Li target ioniziation potential in atomic units.[12,14]

In order to avoid showing unphysical behavior from this model, the projectile charge state has been treated as a continuous variable in Fig. 6. The principal quantum number fed is indicated on the falling part of each corresponding segment of the curve. It is an interesting accident that, for this target, an increment of the projectile charge by one unit raises $n$ by one unit as well. In reality, one expects the oscillatory behavior to be substantially washed out by subshell splitting of the final states and by electron tunneling through the final barrier. While the maxima in the CBM result lie near the data, one should not take the strict interpretation of this result too seriously in this case. For $q<6$, examination of the real level structure of the final projectile ions shows that the states fed are not characterized by the $n$ values given by the hydrogenic model of the projectile. For example, population of $n<5$ on Xe is not even open for $q<6$. Populations of states with $n=6$ and 7 is likely for Xe even for $q$ as low as 2. The OSAS result, which does not depend on assignment of any particular quantum numbers to the populated states, remains valid. Above about $q=5$, the levels populated are sufficiently hydrogenic that the CBM values are approximately correct. Dijkamp *et al.* have found cross sections of $2\times10^{-14}$ and $2.6\times10^{-14}$ cm$^2$ for $C^{4+}$ and $O^{6+}$ on Li at higher energies. These cross sections are quite consistent with those reported here for charge states 4 and 6. They find that, for $O^{6+}$, population of $n=5$—7 dominates, consistent with the CBM result given in Fig. 6. The cross section for $Xe^{10+}$ on Li reported by Müller *et al.*[31] is about a factor of 2 above ours.

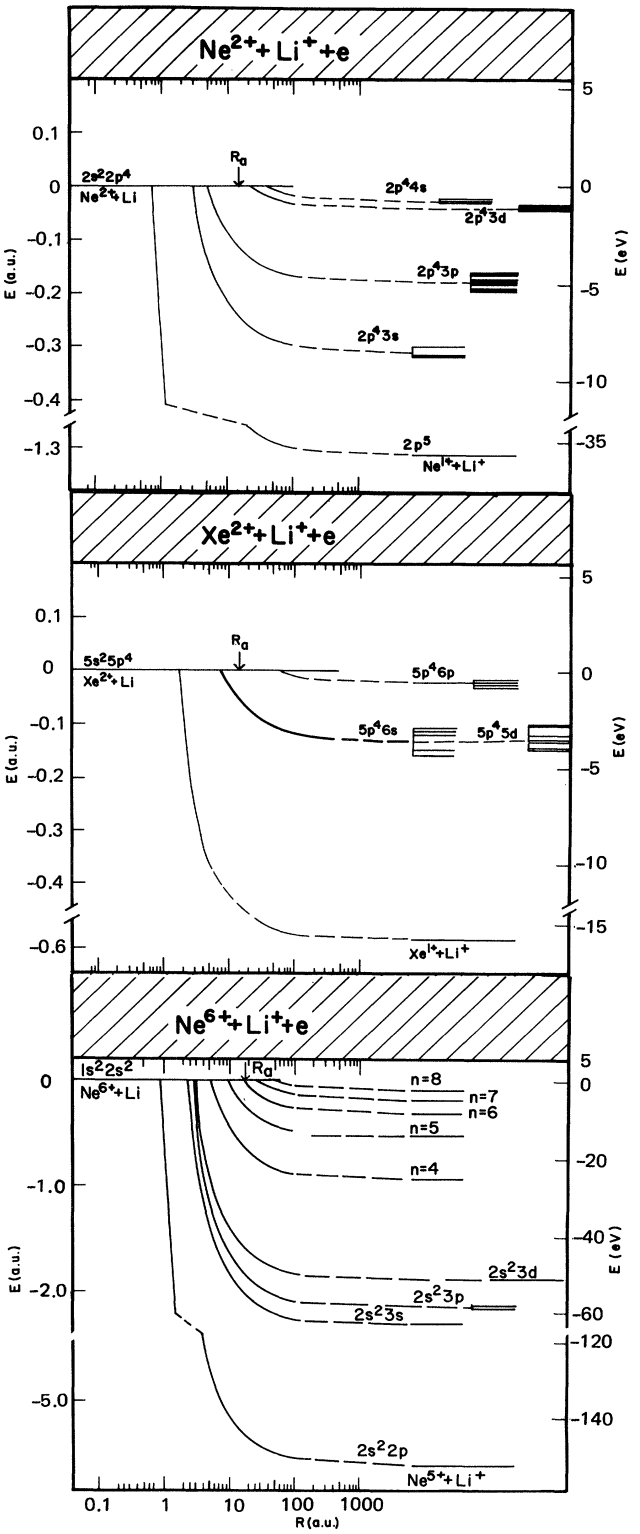


FIG. 8. Potential energy curves for the ground-state ions of $Ne^{2+}$, $Xe^{2+}$, and $Ne^{6+}$ on Li. Absorbing sphere radii from Ref. 11 are indicated by arrows (see text).

In summary, except for $Ne^{2+}$ and $Ar^{2+}$, we find that electron capture cross sections from Li by highly charged projectiles are dependent mainly on the projectile core charge, and not on the structure of the projectile or its velocity. The cross sections are huge, ranging up to

$7\times10^{-14}$ cm$^2$, corresponding to a transfer radius of at least 25 a.u. Plasma diagnostic schemes which rely on capture from beams of Li injected into the plasma are rendered more feasible by the enormous size of such cross sections. The absorbing sphere and classical barrier models appear to give a good description of the process. Although direct experimental evidence is not given here, the size of the cross sections coupled with model interpretations make it clear that the population of high-$n$ levels, up to $n=10$ for $Xe^{10+}$ and $Kr^{10+}$, will occur in such collisions, offering interesting opportunities for spectroscopy of selectively populated Rydberg states on highly charged ions.

*Note added in proof.* Recent measurements by Winter *et al.* indicate that their total cross sections shown in Fig. 7 should be lowered by a factor of about 2, bringing them into agreement with the present results. We thank H. Winter for conveying these unpublished results to us.

## ACKNOWLEDGMENTS

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