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PROCESSES OF ENERGY DEPOSITION BY HEAVY PARTICLE IMPACT

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ELASTIC AND INELASTIC SCATTERING IN THE $\text{Li}^+ + \text{He}$ SYSTEM*

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ABSTRACT

Experimental studies of the $\text{Li}^+ + \text{He}$ interaction have been extended to smaller internuclear separations to clarify a discrepancy between experiment and theory in the elastic cross sections and to observe and measure cross sections for inelastic excitation. For $R \leq 0.5 a_0$ the elastic scattering differential cross section is observed to droop far below that expected from a single-state interaction based on ab initio ground-state potentials. In this same region of R both one- and two-electron excitations of He are observed to be strongly excited, and cross sections were measured for each of them. The lowest inelastic channel is an electron-capture state, which is observed indirectly to be strongly populated also. A diabatic correlation diagram is constructed that exhibits the basic interactions responsible for the excitation.

INTRODUCTION

Interest in the problem of understanding the $\text{Li}^+ + \text{He}$ interaction at small internuclear separations was kindled by the measurements of Zehr and Berry¹ and the calculation of Fischer.² Comparison of the potential derived from the data with the ab initio curve exhibited a large discrepancy in energy for $R \leq 0.5 a_0$. Questions were raised about the validity of both the experiment and the calculation, and for a time a situation similar to the well known discrepancy in the He-He interaction potential seemed to be developing.³ A detailed differential scattering measurement was carried out at about the same time by Aberth and Lorents⁴ in which the scattered ions were energy analyzed to separate and identify elastic and inelastic components. No inelastic component was observed, but elastic cross sections were measured and found to be smooth monotonic decreasing functions of θ . Olson, Smith, and Mueller⁵ inverted these data to obtain a two-term interaction potential that is in essential agreement with the Zehr and Berry potential and shows the same discrepancy with theory at small R . Meanwhile Junker and Browne⁶ performed a more accurate multiconfiguration interaction computation of the ground state and two excited states. Their ground-state potential is in good agreement with that calculated by Fischer. However, they observed that the ground $^1\Sigma$ state made a close approach to an excited $^1\Sigma$ state at $R = 0.5 a_0$ and suggested that inelastic scattering channels would open up in this range and thus reduce the flux in the elastic channel. A third single-state calculation by Catlow, et al.⁷ again confirmed Fischer's potential, but in

that work emphasis was placed mainly on the long range aspects of the potential. The possibility of errors in the experimental data must also be considered; in particular the Aberth and Lorents data exhibit a disturbing droop at large angles for each energy that indicates some inconsistency.

We have reinvestigated the $\text{Li}^+ + \text{He}$ interaction to answer the questions raised and to observe and study the excitation that occurs at small internuclear separations. To this end we extended our energy range to attain smaller internuclear separations than in our earlier measurements. The $\text{Li}^+ + \text{He}$ system is the simplest positive ion-atom collision having two filled K shells that one can study. For that reason it is an excellent reaction in which to learn about the electron promotion mechanisms that are effective when two filled K shells coalesce. Interest in the K-shell interactions has recently been stimulated by several observations, in higher Z systems, of surprisingly strong K-shell excitation detected by x-ray⁸ and Auger electron⁹ emission. Our expectations were rewarded in several ways. First, the suggestion of Junker and Browne that inelastic interactions were draining away the elastic flux at small R is confirmed. Second, we observed that at the smallest R values attained, corresponding to greatest shell overlap, the predominant excitation is two-electron promotion occurring exclusively in the He atom. The He electrons are apparently forced into the L shell as the united atom is approached and remain there when the atoms separate again. Third, by comparing the measured inelastic flux with that expected from the loss of elastic flux, we were able to deduce information about the electron transfer reaction producing neutral Li.

ENERGY LOSS SPECTRA

The differential scattering apparatus was used as described in the earlier publication of $\text{Li}^+ + \text{He}$ studies.⁴ The addition of a recording data acquisition system and extension of the energy range upward to 2 keV were the only improvements. Angular definition of the incident and scattered beams was determined by the slit system used in earlier measurements. No effort was made to improve the small angle resolution by reducing the slit height as was previously done. Energy analysis of the scattered ion beam was performed by reducing the energy in a retarding field and analyzing the remaining energy by electrostatic deflection. The energy resolution of the system was held constant by varying the retardation voltage in order to hold fixed the energy of the ions passing through the analyzer.

Two extensive sets of measurements were conducted at each energy:

- (1) fixed angle scans of the scattered ion kinetic energy spectra, and
- (2) angular distribution scans for each of the lines in the energy loss spectra.

Measurements of the energy loss spectra revealed three distinct energy loss peaks over most of the attainable $E\theta$ range of this experiment. A typical energy loss spectra taken at 2.0 keV and 5° is shown in Fig. 1. Spectra such as these were taken for each energy at one-degree intervals over the angular range from 2° to where the signal disappeared into the noise. The highest energy peak is due to elastic scattering, and the two lower energy peaks correspond to electronic excitation of one or the other of the collision partners. Identification

of the energy loss peaks with specific excited levels of Li^+ or He is based on the mean (lab) energy $\langle E_i \rangle$ of the peak relative to the mean (lab) energy $\langle E_e \rangle$ of the elastic peak at each angle θ , but the broad energy spreads of the peaks hampered the assignment of the energy losses to individual levels. The symmetrically shaped energy loss peaks allowed the mean energy $\langle E_i \rangle$ to be accurately determined, and the measurement of $\langle E_e \rangle - \langle E_i \rangle$ to ± 0.3 eV was easily attained at all angles. The exact relationship between $E_e - E_i$ and Q_i , the excitation energy, can readily be determined from the energy and momentum conservation laws which yield:

$$Q_i = (1 - \gamma)E_i + 2\gamma E_0 E_i \cos \theta \quad (1)$$

where $\gamma = M_1/M_2$, M_1 is the incident particle mass, M_2 is the target particle mass, E_i is the kinetic energy of M_1 scattered at angle θ , and E_0 is the incident energy ($E_e = E_i$ when $Q_i = 0$). For small angle scattering it is convenient to recognize that $\Delta E_{ei} \equiv E_e - E_i \simeq Q$, a useful relationship for obtaining an estimate of the levels involved.

In Fig. 2 are plotted observed values of $E_e - E_i$ as a function of θ along with computed values of ΔE_{ei} for specific excitation energies Q_i that might be responsible for the energy loss in the neighborhood of 20 eV. The only energetically possible excited states in this region are the single electron promotions in He. Since the initial state of the $\text{Li}^+ + \text{He}$ system is singlet, we assume (and demonstrate later) that the outgoing states are also limited to singlets. The ΔE_{ei} curves for 2^1S , 2^1P , and 3^1S states plotted in Fig. 2 together with the data show that the prominent excitation occurs in the $1s2\ell$ states and remains so

independent of angle. The 2^1P state seems to be favored, but unfortunately the inherent energy distributions of the scattered beam rapidly become very wide with increasing angle so that a strong contribution from 2^1S or weak contributions from higher lying states cannot be ruled out. That these energy distributions are inherent in the experiment and not necessarily generated by the excitation of a wide band of states is suggested by the fact that they have the same shape and width as the elastic energy distributions. This is shown in Fig. 3 where the observed $\delta E = \text{FWHM}$ (full width at half maximum) for the three energy losses is plotted against θ . Gaussian shaped contributions to the energy spread arise from three sources: the energy analyzer, the finite angular resolution, and the thermal velocity distribution of the target atoms. The measured energy distributions were obtained using constant analyzer resolution, and one can readily observe the shift from analyzer limited distributions at small angles to distributions limited by angular resolution and thermal effects at large angles. The angular resolution contribution to the energy distribution is due to the angular spread in the beam of scattered ions defined by the slits. This angular distribution converts to an energy distribution by means of Eq. (1). An estimate of the magnitude of the contribution is obtained (assuming an angular independent cross section over the angular range of the slits) from $(dE/d\theta)_{\theta_0} \cdot \Delta\theta$ where $\Delta\theta = \text{FWHM} \approx 0.34^\circ$ of the angular distribution. The component due to the thermal velocities of the target atoms was estimated by computing, using a modified Eq. (1), the energy distribution of scattered ions for a given θ resulting from collisions with each of the three thermal velocity components of the target atom.

The resulting gaussian distributions of scattered ions were folded to determine the resultant FWHM due to the thermal motion of the target atoms. The results of these estimates of the thermal and angular contributions of the energy spread are shown in Fig. 3. Convolution of these functions and the analyzer transmission function (also assumed gaussian for this computation) gives the estimated total $\delta E(\theta)$. In those cases where δE is dominated by the thermal and angular components, the magnitude and angular dependence of the widths are in reasonable agreement with the observed widths. The difference in magnitude of δE for the elastic scattering and for the 60 eV inelastic scattering is also explained. We can conclude on the basis of these calculations and from the observations on the elastic scattering that the observed energy spreads can reasonably be expected without requiring excitation of more than a single state. Of course, the excitation of more than one state is not ruled out provided the energy separation of the band of states is not wider than the δE at a given θ . Actually the most sensitive indicator of the participation of several states is the position of the center of the distribution. We conclude that the symmetrical energy distribution, which remains centered on the $1s2\ell$ states for all values of $E\theta$ attained in this experiment, strongly suggests that $1sn\ell$ states with $n > 2$ are not significantly excited.

A similar behavior is observed for the 60 volt energy loss as indicated in Fig. 4. Here the observed values of $\langle E_e - E_i \rangle$ fall between the curves expected for the $\text{He}(2s^2)^1S$ state at $Q = 57.8$ and the $\text{He}(2s2p)^1P$ and $\text{He}(2p^2)^1D$ states at 60.0 eV.¹⁰ The arguments given above for the one-electron excitation apply equally in this case, and we

conclude that the excitation remains predominantly on the three lowest doubly excited states for all E_0 . Consideration must be given to the possible excitation of $\text{Li}^+(1s2s)^1S$ since it lies at 60.7 eV and could contribute to the observed energy loss. However, arguments presented later show that excitation of Li^+ is highly improbable for the incident energies used in this experiment.

Some insight into the levels that might be expected to be excited is gained by constructing a correlation diagram such as shown in Fig. 5. The binding energies of each state are plotted in the separated and united atom limits for the several states that are believed to participate strongly in the collision. Because of the wide energy separation between the Li 1s and the He 1s electrons, it is clear that both Li^+ electrons correlate to the 1s state of B^+ . In fact, since the separation between the 1s and 2s levels for B^+ is considerably larger than for the separated atoms, it appears that the isolation of these shells increases as the internuclear separation decreases and that the 1s energy remains adiabatic. It follows that the Li^+ electrons cannot be excited in low energy collisions. Furthermore, since the Li^+ electrons are unavailable for exchange with the He electrons, no spin exchange can occur, from which it follows that only singlet states can be excited. The correlation of the He electrons is discussed later, but since these levels lie close to each other in the united atom limit, it is obvious that mixing of states due to the electron correlation interaction is important in the collision problem. We have, therefore, shown the He $1s^2$ electrons ingoing to the $n = 2$ level of B^+ and outgoing to the $(1s^2)$, $(1s2s)$, and $(2s^2)$ of He and also to $\text{Li}(1s^22s)$ and $\text{Li}^-(1s^22s^2)$.

INTENSITY MEASUREMENTS AND CROSS SECTIONS

Having identified the energy loss peaks, the intensity of the scattered beam for each of the peaks in the spectra was ultimately determined by the continuous angle scans during which the retardation voltage was continuously varied to follow a given energy loss. Since the angle scans were taken with a constant analyzer transmission function whose width δE_a was at times less than the line width, the observed signal was proportional to an angular dependent fraction of the total integrated intensity of a particular energy loss. Using the approximations described in the last section, we assume the analyzer transmission function and the natural line width of the scattered beam to be gaussian functions with FWHMs of δE_a and δE_n , respectively. The observed width is $\delta E_o = (\delta E_a^2 + \delta E_n^2)^{\frac{1}{2}}$. It follows that the ratio of the observed to the total scattered beam intensity is equal to $\delta E_a / \delta E_o$. This ratio is obtained from the experimentally determined curves of Fig. 3 and used to correct the intensity measurements. This correction was not made in the earlier elastic measurements, and it probably accounts for the large angle droop observed even in the low energy data previously published.

In the region of internuclear separations where no inelastic scattering is observed and the interaction is purely elastic potential scattering, accurate scattering cross sections can be calculated from ab initio computations of the ground state of LiHe^+ . Our relative elastic cross sections obtained from the intensity versus angle measurements were normalized at low τ values to the cross sections obtained

using the potential curve of Catlow, et al.⁷ Unfortunately in the low τ region corresponding to laboratory scattering angles $\leq 5^\circ$, the observed cross sections drooped below the true values because of the use of slits (a slit geometry weights angles larger than the mean angle so that a cross section decreasing in the lab system gives a low scattering signal). In the region of purely elastic scattering $\rho(\tau)$ plot ($\rho = \theta_{cm} \sin \theta_{cm} \sigma(\theta_{cm})$ and $\tau = E_{cm} \theta_{cm}$) of the data taken at various energies should overlay to produce a common curve.¹¹ Such a plot of the experimental data shows the points following a common curve for a region of τ but falling rapidly away at the low τ end of each energy. A correction function for the small angle droop was generated by taking the ratio $\rho_{600}(\tau)/\rho_{1500}(\tau)$ and applying it to the data above 600 eV in the region of τ corresponding to $\theta_L \leq 5^\circ$. The 600 eV data was corrected by comparison with data obtained in an earlier measurement⁴ where improved angular resolution at small angles was used. These corrections were necessary for the higher energy data in order to obtain reliable $\rho(\tau)$ curves in a region of τ , where they could be normalized to the calculated cross section. The final corrected elastic data plotted in ρ - τ coordinates is shown in Fig. 6, together with the cross sections computed from the potential of Catlow, et al.⁷ It is readily seen that the observed elastic scattering rapidly falls far below that expected from a single-state interaction. Such behavior is consistent with the onset of inelastic scattering wherein part of the scattered flux exits through excited state channels.

The inelastic data were normalized to the measured elastic cross sections. The ratio of the scattered intensity in an elastic peak to

that in the elastic peak was obtained from the energy loss measurements at several angles and used to determine the cross sections. ρ - τ plots of the data corresponding to the 21 eV loss are given in Fig. 7, and for the 60 eV loss in Fig. 8. In Figs. 9, 10, and 11 are shown at each energy the reduced cross sections for each of the energy losses, together with the elastic cross sections. These plots clearly show the buildup of the inelastic scattering in the region where the elastic droops below that expected from a single-channel case. Examination of these plots reveals that the difference between the theoretical and experimental elastic curves cannot be fully accounted for by the observed inelastic cross sections, particularly in the region of τ where the elastic first begins to droop. This behavior is most probably due to one or more inelastic transitions that are not observable in this experiment. The obvious unobservable reactions are electron-capture events where the Li^+ becomes neutral or negative. No attempt was made to observe these products, but we can think of no other possibilities. A reasonable estimate of the cross section of the unobserved transitions is easily obtained. The observed total inelastic cross section as a function of τ is obtained by summing the two measured inelastic curves. The theoretical total inelastic cross section is determined from the difference between the observed and calculated elastic cross section. We assume here that the single-state cross section determined from the ab initio potential is equal to the total differential cross section including all elastic and inelastic processes. The total unobserved inelastic cross section is then equal to the difference between the theoretical and observed total inelastic

cross sections. We are assuming here that $\tau(R_0)$ is the same for the elastic and inelastic scattering, which is only approximately true because of the difference in trajectories. For values of τ in the region where inelastic collisions are occurring, the potential energy is an order of magnitude larger than the energy separation of the states at large R . This means that the elastic and inelastic scattering angles for a given R_0 and E will differ only slightly. The errors incurred by this assumption are such that the observed inelastic cross sections appear at smaller values relative to the elastic τ scale than they should; thus the deduced inelastic cross section is underestimated at low τ and overestimated at large τ . The differential cross sections obtained by this method are plotted in Fig. 12. They appear to have a very reasonable shape and to exhibit a velocity-dependent threshold similar to the other excitation channels. These electron capture curves are also included in Figs. 9, 10, and 11, together with the cross section curves for each of the outgoing channels. Here one observes that the electron-capture process is indeed the first channel to be excited and also the predominant inelastic channel over a considerable range of τ . For each of the four energies the electron-capture cross sections have been integrated to yield total cross sections. These cross sections are compared with recently reported measurements of the single electron-capture cross sections for $\text{Li}^+ + \text{He}$ at energies from 0.6 to 3.0KeV^{12} in Table I. The points at 1.5 and 2.0 keV are in excellent agreement, whereas the lower energy points lie somewhat high. The 50% errors possible in each of these measurements, however, are sufficient to account for the discrepancy so that the two measurements are in

agreement. It is thus established with reasonable certainty that the unobserved reactions are single electron-capture reactions.

DISCUSSION

The nature of the interactions that produce excitation in the $\text{Li}^+ + \text{He}$ system appear to differ markedly from the curve-crossing interactions that are commonly found in many other systems. Notable is the observation that the onset of excitation moves toward larger R as the energy increases. This behavior is in contrast to the case of an isolated curve-crossing interaction where the threshold in R remains essentially independent of energy. Another characteristic feature of curve-crossing interactions is the appearance of an interference pattern in the differential cross section. Oscillatory structure is not observed in the $\text{Li}^+ + \text{He}$ case except at large τ values where the doubly excited levels predominate. These observations suggest that the transitions are occurring between close lying molecular states that are approaching each other with very small difference in slope. LiHe^+ excited and ground-state curves computed by Junker and Browne⁶ are seen to behave in exactly this manner. Transitions are then possible over an extended range of R , and the localized curve-crossing characteristics will not appear. In general the range of R over which transitions take place with appreciable probability becomes more extended as the velocity increases. This effect will tend to be more pronounced when the states approach each other slowly and may account for the observed threshold shift with increasing v . The concept of well-defined classical trajectories and their associated phases that are identified with individual potential energy curves loses its validity under these circumstances, and we do not expect to see interference phenomena.

The simplified correlation diagram presented in Fig. 5 which is based on single electron binding energies shows that it is the interactions between the two He electrons approaching the 2ℓ levels of B^+ that lead to excitation in the collision. The Li^+ electrons play only a passive role of filling the K shell. This is a particularly simple illustration of the effect observed in K-shell x-ray excitation by heavy particles wherein the probability of excitation in the particle of lower Z can be several orders of magnitude larger than that of its partner.⁸ Taking this result and applying the correlation rules for asymmetric one-electron systems worked out by Gershtein and Krivchenkov,¹³ we have constructed a diabatic correlation diagram based on total energies. This is a correlation diagram constructed on the basis of Lichten's¹⁴ independent particle definition of diabatic states, which assumes perfect shielding and no electron-electron interaction. This diagram, shown in Fig. 13, gives a much clearer picture than does the adiabatic correlation of the interactions among states near the united atom limit and where these states are expected to lead when the system separates. Obviously this correlation should not be taken to indicate that the molecular states involved can actually be described as single configurations but rather to indicate which configurations will participate most strongly in the description of the various collision interactions. The correlation of the ground state $Li^+ + He$ system with $B^+(2p^2)^1D$ is the key that illustrates that the first important interactions are the three-state Σ - Σ couplings leading to single and double charge transfer as is inferred in the experiment. The interactions leading to the excited states observed in this experiment appear to come from two Σ - Π .

interactions and a Π - Δ interaction, all of which make asymptotic approaches in the united atom limit.

This simple diagram shows that, with one exception, all the observed excited states are directly connected with the incoming state by either Σ - Σ crossings or Σ - Π and Π - Δ asymptotic approaches to a common united atom state. The exception is the $\text{He}(2s^2)^1S$ state, which does not correlate to any of the first three united atom states that interact with the incoming state. It appears that a strong interaction must be assumed between the $(3p\sigma^2)^1\Sigma$ and $(2s\sigma^2)^1\Sigma$ states so that the crossing is avoided and the $(2s\sigma^2)^1\Sigma$ is shunted to the $\text{He}(2s^2)^1S$ state. Clearly an experiment designed to observe and measure the intensity in the $\text{He}^{++} + \text{Li}^-$ channel would provide valuable additional information about this state. It would also be desirable to do a high resolution study of the autoionization spectra arising from $\text{Li}^+ + \text{He}$ collisions to establish more precisely the states that are excited and their relative populations. It is noteworthy that the autoionization spectrum recently reported for $\text{H}^- + \text{He}^{15}$ shows two peaks corresponding to the $(2s^2)^1S$ and $(2p^2)^1D$ states of H^- . These are just the states that are expected to be excited on the basis of the model for asymmetric four-electron systems presented in this paper.

Junker and Browne have calculated the ground and first three excited adiabatic states of the LiHe^+ ion using a multiconfiguration interaction program that produces very accurate results.⁶ Their results also include interaction matrix elements between these states that permit the calculation of inelastic cross sections. The Σ - Π and Π - Δ interactions have been recently calculated in an approximate way by McCarroll

and Piacentini.¹⁶ Although they did not include the Σ - Σ interactions in their calculation, the Σ - Π and Π - Δ couplings appear to give a reasonable explanation of the observed excitations and confirm the importance of the suggested basic mechanisms.

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TABLE I. TOTAL ELECTRON CAPTURE CROSS SECTIONS

E keV	Q_{LC} 10^{-18} cm^2	Q_{LS} 10^{-18} cm^2
0.6	2.0	0.78
1.0	1.5	0.9
1.5	1.16	1.1
2.0	1.64	1.52

Q_{LC} total electron capture cross sections, calculated from the differential cross sections of Fig. 12, and Q_{LS} , obtained from the measurements of Latypov and Shaporenko, Ref. 12.

FIGURE CAPTIONS

Figure No.

- 1 Spectrum of energy losses at 2.0 keV and 5°.
2. Observed and calculated values of ΔE_{ei} as a function of laboratory scattering angle. The solid curves are calculated for the indicated Q_i values and the points are measured values of $E_e - E_i$.
- 3 Observed and estimated values of the energy spread as a function of laboratory scattering angle. Solid lines are estimated as described in the text, and the points are measured values.
- 4 Observed and calculated values of ΔE_{ei} in the region of 60 eV energy loss.
- 5 Correlation diagram based on electronic state binding energies.
- 6 $\rho(\tau)$ plot of measured elastic scattering cross sections compared with cross sections calculated from the potential of Catlow, et al., assuming a single-state interaction. The curves for each of the energies have been displaced for clarity.
- 7 Measured scattering cross sections for excitation of the 21.2 eV energy loss.
- 8 $\rho(\tau)$ plot of cross sections for excitation of the 58.7 eV energy loss.
- 9 Elastic and inelastic cross sections at 1.0 keV. The electron capture cross section was obtained indirectly as described in the text.

Figure
No.

- 10 Elastic and inelastic cross sections at 1.5 keV. The electron capture cross section was obtained indirectly as described in the text.
- 11 Elastic and inelastic cross sections at 2.0 keV. The electron capture cross section was obtained indirectly as described in the text.
- 12 $\rho(\tau)$ plot of electron capture cross sections obtained at four energies.
- 13 Correlation diagram based on total energies of united and separated atoms.

TA-8037-5R

FIGURE 1

FIGURE 2

TB-8037-21

TC-8037-23

FIGURE 3

TB-8037-20

FIGURE 4

FIGURE 5

TA-8037-9R

FIGURE 6

TC-8037-14

FIGURE 7

TB-8037-19

FIGURE 8

TC-8037-16

FIGURE 9

TC-8037-18

FIGURE 10

TB-8037-17

FIGURE 11

TC-8037-15

FIGURE 12.

TC-8037-22

FIGURE 13