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Excitation of the He(2^1S and 2^1P) States by 15–100-keV Li^+ Bombardment*

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Excitation of the He(2^1S) state by 15–100-keV Li^+ bombardment has been studied as a function of the angular scattering of Li^+. The ratio of He(2^1S) to He(2^1P) excitations is about 0.6 and appears to be independent of both Li^+ energy and scattering angle. Excitation of the He(2^1S) state indicates an inability of correlation diagrams based on the electron promotion model to predict all of the excitation products for Li^+ + He collisions in this energy range.

The Li^+ + He interaction provides a study of the collision of two completed K shells. Sets of high-resolution energy-loss spectra for 15–100-keV Li^+ ions scattered at various angles provide information on the possible states of excitation of the target-projectile system. The excitation of the He(2^1S) state is of particular interest since correlation diagrams based on a straightforward application of the electron-promotion model do not predict the excitation of this state. The sets of spectra shown in Fig. 1 clearly display a contribution from the He(2^1S) state with energy loss at 20.6 eV.

The apparatus and general method employed in heavy-ion energy-loss spectrometry have been discussed in detail elsewhere.[1-5] In the current experiment the system has been modified to permit the angle of the beam entering the collision chamber to be varied.

A set of spectra taken for 30-keV Li^+ ions at various scattering angles is shown in Fig. 1. The angular divergence of the incident beam and the angular acceptance of the deceleration-analyzer system are functions of ion energy. At this energy the measured convolution of the angular divergence of the incident beam and the angular acceptance of the decelerator-analyzer system is approximately 8.5×10^{-4} rad full width

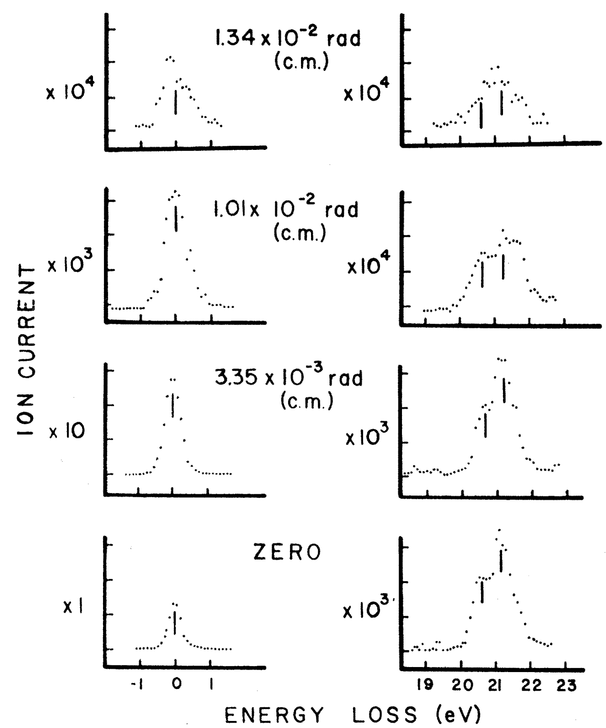

FIG. 1. Data for 30-keV (lab energy) lithium ions incident on helium gas ($nl = 6.37\times10^{15}$ atoms/cm^2). Transmitted ion current in arbitrary units plotted against energy loss at various center-of-mass angles for scattering of the incident lithium ion.

at half-maximum. The measured energy spread is a convolution of the incident-beam energy distribution with the analyzer resolution function, and is 0.5 eV full width at half-maximum.

The 21-eV energy-loss peak contains contributions from both the He(2^1S) peak and the He(2^1P) peak. The He(2^3S) peak which dominates the He^+ + He spectrum is completely absent, as would be expected because the spin must be conserved and triplet excitation with spin conservation requires promotion of an electron in both the He and Li^+ K shells with a correspondingly larger energy loss. Both the He(2^1S) peak at 20.611 eV energy loss and the He(2^1P) peak at 21.213 eV energy loss are clearly present with 0.5 eV energy resolution and are partially resolved at all scattering angles where the ion intensity permits measurements.

The procedure used to determine the relative magnitude of the two peaks[1] was to assume the presence of peaks, each having the same energy distribution as the primary ion beam, at the spectroscopic locations of the transitions involved. A least-squares fit to the data then yields the appropriate magnitude of each peak which when summed over both transitions will reproduce the data. The fraction of the 21-eV peak due to He(2^1S) excitation appears to be independent of scattering angle. The study involved 182 spectra taken over the course of 5 months. The resolution was varied and several apparatus changes and realignments were completed during this period. The measurements have a low signal-to-noise ratio, particularly at scattering angles greater than 1×10^{-2} rad (c.m.), but in no case is the peak fitting satisfactory if the He(2^1S) fraction is set to zero. This excitation fraction appears to be approximately the same at all the ion energies and scattering angles. The overall average for the He(2^1S) contribution to the He(2^1S+2^1P) peak was 38% with a standard deviation of 11%.

Using sets of spectra like those shown in Fig. 1, an apparent differential cross section was calculated. The data, shown in Fig. 2 in plots of ρ versus τ, have a broad angular dependence. The plots of ρ versus τ display a striking similarity in general features over a large ion energy range. Our data exhibit essentially the same features as those of Lorents and Conklin[6] and Francois, Dhuicq, and Barat[7] taken at energies much lower. While the data at various ion energies do not quite fit a common curve, they seem to indicate that the same excitation mechanism is dominant over the entire energy range. The observed variation in the locations of the maxima in the plots of ρ versus τ as a function of ion energy was observed by Lorents and Conklin,[6] and the current data, covering a much larger and higher energy range, substantiate this effect.

The electron promotion model developed by Lichten and others[8-10] was used by McCarroll and Piacentini[11] for the Li^+ + He case. They used the building-up principle and the form of the molecular orbitals in their separated-atom limit to obtain their energy correlations. McCarroll and Piacentini found several possibilities for rotationally induced transitions at or near the the B^+ $(1s^22p^2)\,^1D$ core, including

$$Li^+(1s^2)\,^1S+He(1s^2)\,^1S \to Li^+(1s^2)\,^1S+He(1s2p)\,^1P,$$

$$\Delta\epsilon=21.21 \text{ eV}.$$

They also considered radially induced transitions from the molecular states at intermediate impact parameters. McCarroll and Piacentini

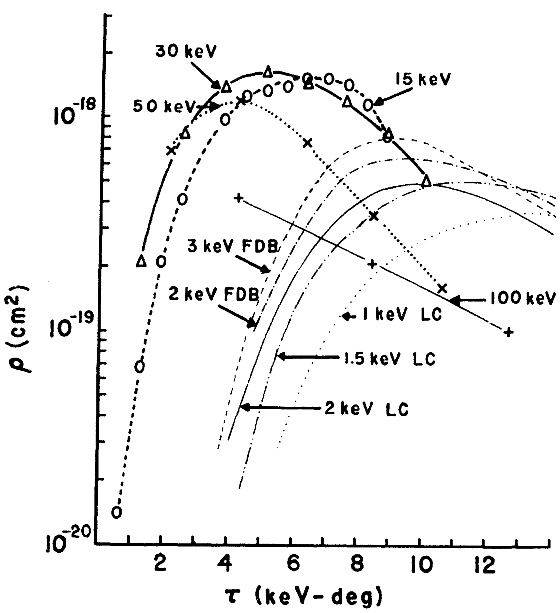

FIG. 2. Apparent reduced-cross-section measurements for excitation of the 21-eV peak, ρ versus τ. $\rho=\theta\sin\theta\, d\sigma/d\Omega\approx\theta^2 I(\theta)/I_0 nl(\Delta\Omega)$, $\tau=E\theta$, where $I(\theta)$ is the current inelastically scattered into the solid angle $\Delta\Omega$ in c.m. coordinates, I_0 is the total current density in the elastic peak integrated over 4π sr, n is the target particle density, θ is the mean scattering angle in c.m. coordinates, and l is the ion path length in the target gas. Curve are labeled by ion lab energies. Curve LC is the data of Lorents and Conklin (Ref. 6). Curve FDB is the data of Francois, Dhuicq, and Barat (Ref. 7).

did not expect the Σ-Σ (radial) interactions to affect the direct excitation interaction, while it might of course affect the charge-transfer process. Since our experimental system ignores neutral projectiles, the only interaction providing for an energy loss of about 21 eV is the excitation of the $He(1s2p)\,^1P$ state, with a loss of about 21.21 eV. The energy correlations given by McCarroll and Piacentini,[11] Lorents and Conklin,[6] or Francois, Dhuicq, and Barat[7] do not provide for a direct coupling to the $He(2^1S)$ state. These correlation diagrams only permit the excitation of the $He(2^1S)$ state by multiple crossing processes. Multiple crossing processes would be expected to result in angular distributions quite different from those involved in the single crossing process exciting the $He(2^1P)$ state.

The Li^+ ions are in an energy range (2.1 to 14 keV/amu), where the electron-promotion model is routinely applied; however, the electron-promotion model is primarily intended to explain inner-shell excitation.[9] It may prove to be inapplicable to problems involving the determination of the specific state being excited except for very low-velocity collisions. It may be necessary to include long-range interactions to explain the excitation of specific states. In a recent Letter Lesech, McCarroll, and Baudon[12] state that the branching ratio between the $Li(2^2P)$ charge-exchange channel and the $He(2^1P)$ excitation channel depends on the interaction between the two channels at large internuclear separations. Such a process would be dependent on velocity but not on scattering angle. This explains the similarity in the shape of the charge-exchange cross section curve and the 21-eV peak excitation cross-section curve reported by Francois, Dhuicq, and Barat.[7] The assumption of a strong interaction at large internuclear distances which mixes the $He(2^1S)$ and $He(2^1P)$ states following excitation would explain our results; however, the potential energy curves connected to the $He(2^1S)$ and $He(2^1P)$ states are believed to lie further apart than the curves connected to the $Li(2^2P)$ and the $He(2^1P)$ states.

The similarity in the plots of ρ versus τ of the 21-eV peak imply that the primary excitation mechanism for the 21-eV peak has not changed over the entire energy range from 1 to 100 keV. The shift in the peaking of the plots of ρ versus τ at lower values of τ (larger impact parameters) with increasing energy is observed over this entire energy range and would seem to be a property of the excitation mechanism rather than being due to a change in excitation mechanism. In our experiment covering 15 to 100 keV the ratio of $He(2^1S)$ to $He(2^1P)$ excitations appears to be independent of both ion energy and scattering angle. This does not prove that the ratio of the $He(2^1S)$ to the $He(2^1P)$ cross sections does not drop dramatically at some ion energy below 15 keV, perhaps because of a change in an interaction at large internuclear distances. The data imply that if such a change occurs, its onset is at energies lower than 15 keV. The available data also do not show any features which indicate a change in the primary excitation mechanism. Such a change in excitation mechanism could of course pass undetected in the energy range under 15 keV if it did not produce dramatic changes in the appearance of the angular distribution.

Neither Lorents and Conklin[6] nor Francois, Dhuicq, and Barat[7] were able to resolve the 2^1S transition and only reported the cross section for the 21-eV peak as a whole. It would be very interesting to restudy this transition with higher resolution at the lower Li^+ energies to determine if the $He(2^1S)$ state is excited by collisions at these low energies.

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