the r process is days, this would still not be a very serious limitation. In any case the r process is likely to increase the amount of matter in the abnormal state very substantially. From terrestrial studies on the abundance of various isotopes above Fe, one may estimate that $\sim 10^{-3}$ of all matter has been processed through the s process and perhaps one quarter as much through the r process, as discussed in D. D. Clayton, W. A. Fowler, T. E. Hull, and B. A. Zimmerman, Ann. Phys. (N. Y.) 12, 331 (1961).

Evidence for Two-Electron Excitation Producing Li K Vacancies in Slow Li^+ + He Collisions

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Significant Li K excitation is observed in Li^+ + He collisions at incident energies as low as 6 keV. To interpret the results a multistep process is proposed including two-electron excitation in the quasi molecule formed during the collision. The importance of this process is discussed for heavier collision systems.

In the past, K-shell excitation in slow ion-atom collisions has often been described in terms of the molecular orbital (MO) model.[1,2] According to this model available L-shell vacancies may be transferred to the K shell by rotational coupling of the $2p\pi$ and $2p\sigma$ MO's.[2,3] In the case of asymmetric collisions,[4] K vacancies are produced primarily in the particle with the lower atomic number Z. However, there is a finite probability for K excitation of the higher-Z particle as the $2p\sigma$ and $1s\sigma$ MO's couple. Meyerhof,[5] using the charge-transfer model by Demkov,[6] has derived a simple expression to calculate the $2p\sigma$-$1s\sigma$ transition probability in good agreement with a large number of experimental data.[5,7] Only recently, measurements[8] from our laboratory have shown discrepancies between experiment and Meyerhof's theory when light collision systems such as C + B are used.

In the present work we report on the even lighter system Li + He. Similar studies[9-14] have previously been made to investigate primarily excitation of He. Measurements of Li K excitation have been carried out at relatively high energies,[15,16] and corresponding cross sections have not been reported. In this work cross sections for Li K excitation were measured in Li^+ + He collisions at energies as low as 6 keV for which the promotion model is expected to be applicable. It is found that Li K vacancies are produced with much higher probability than predicted from Meyerhof's theory. We suggest that the application of Meyerhof's formalism needs revision with respect to the possibility of multielectron excitation in the lighter collision partner. The significant double excitation of He in Li^+ + He collisions has been studied extensively in the past.[9-14] Here, it is shown that double excitation of the lighter particle produces enhanced K-vacancy production in the heavier collision partner.

The experiments were carried out using the crossed-beam apparatus described in detail previously.[17] The chamber was operated at a 50-kV ion accelerator equipped with a universal ion source. Beams of 6- to 30-keV Li^+ were magnetically analyzed and, after the passage through 2-mm collimator slits, currents of typically 2 μA were obtained. In the center of the chamber the ion beam was crossed by an atomic He beam producing a gas target of $\sim 2\times 10^{-3}$ Torr pressure and ~ 3 mm length. Electrons ejected from the target region were detected by an electrostatic parallel-plate spectrometer. In Fig. 1 a typical electron spectrum is shown for 10-keV Li^+ + He. The plot indicates two groups of peaks at about 35 and 65 eV originating from autoionizing He and Li, respectively. The two groups were separately integrated with respect to electron emission angle and energy to obtain total cross sections for electron production. Electron emission was found to be significantly anisotropic. Therefore, the angular integration was performed under the assumption[19] that the cross-section differential in electron-emission angle θ is given by $d\sigma/d\theta = a + b\cos^2\theta$, where a and b are constants determined from separate measurements at 30° and 90°.

With neglect of competing radiative transitions, the deduced electron-production cross sections were set to be equal to cross sections σ_2(He) and

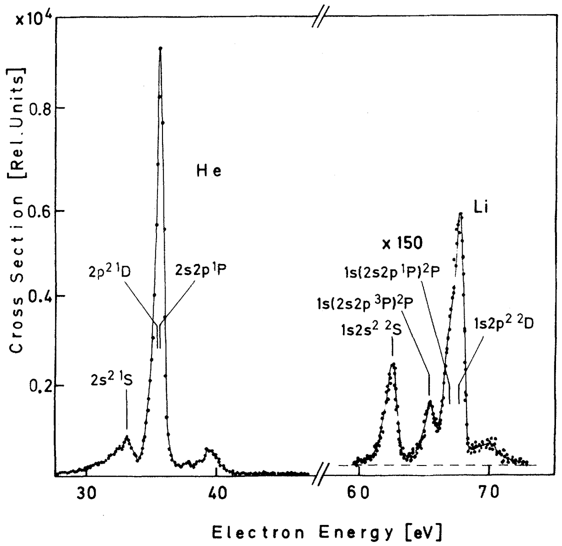

FIG. 1. Autoionization spectrum of He I and Li I excited in 10-keV $Li^+ + He$ collisions. Electron observation angle is 30°. Energy resolution is 0.4 eV (full width at half-maximum). For line identification see, e.g., Refs. 16 and 18.

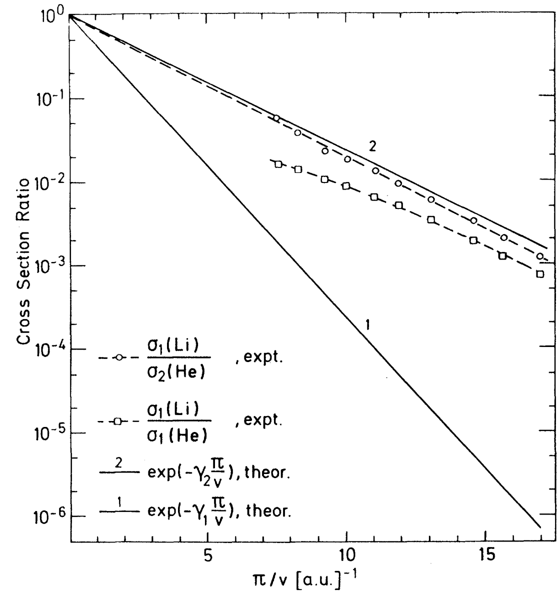

FIG. 2. Ratios of cross sections for $Li^+ + He$ collisions as a function of the inverse projectile velocity v. σ_1(Li) and σ_1(He) are cross sections for production of, respectively, Li and He singly excited in the 1s level. σ_2(He) is the corresponding cross section for doubly excited He. The theoretical data are calculated with $\gamma_1 = 0.83$ a.u. and $\gamma_2 = 0.376$ a.u. (see Ref. 5 and text).

σ_1(Li) for production of doubly excited He and singly excited Li in the 1s level, respectively. The results are plotted in Fig. 2 showing the ratio $\sigma_1(\text{Li})/\sigma_2(\text{He})$ versus π/v where v is the projectile velocity. In the following, we discuss also the cross section σ_1(He) for production of singly excited He. These data were derived including previous results from energy-loss measurements. François, Dhuicq, and Barat[10] and Park *et al.*[11] have measured He-excitation cross sections differential with respect to the projectile scattering angle β. To obtain total cross sections, we carried out the β integration yielding the ratio $\sigma_2(\text{He})/\sigma_1(\text{He})$. Combining these values with the present results, we derived the ratio $\sigma_1(\text{Li})/\sigma_1(\text{He})$ as shown in Fig. 2.

To discuss the experimental results we use the molecular-state diagram shown in Fig. 3. Compared to previous schematic plots,[9,10] we have simplified the diagram to show only prominent states which are expected to be most important (e.g., see Fig. 1) and each of which is considered to be representative of a group of closely lying levels. However, we added reaction channels with Li excited in the K shell. As the 2p levels of He and Li are very close in energy, the correlation of the 2$p\pi$ MO is uncertain[9,10] at large R. Therefore, we split the molecular states leading to both the direct dissociation channel and its corresponding charge-exchange channel. This should indicate that corresponding dissociation channels are roughly equally populated.[9,10] In

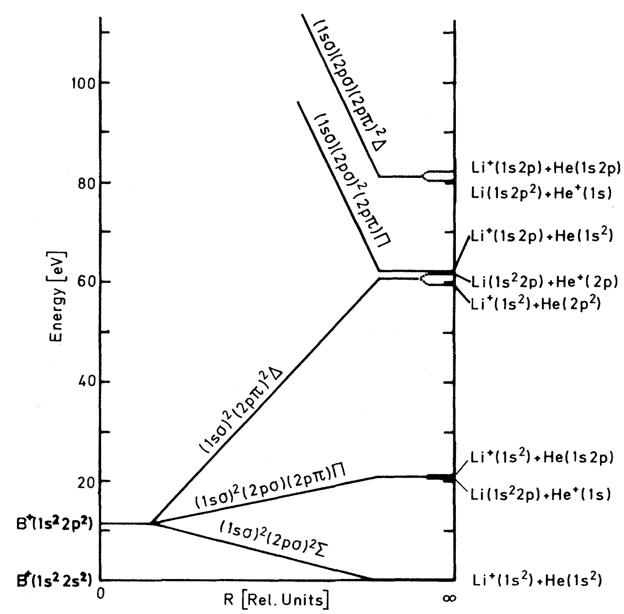

FIG. 3. Schematic molecular-state diagram for the $(LiHe)^+$ system. R is the internuclear distance.

the following we shall consider only one branch of each pair of channels and we suggest that the cross section ratios being discussed are not significantly altered as the corresponding channels are added. For simplicity the states $(1s\sigma)^2(2p\sigma)^2$-Σ, $(1s\sigma)^2(2p\sigma)(2p\pi)\Pi$, $(1s\sigma)^2(2p\pi)^2\Delta$, $(1s\sigma)(2p\sigma)^2\times(2p\pi)\Pi$, and $(1s\sigma)(2p\sigma)(2p\pi)^2\Delta$ are abbreviated by Σ, Π, Δ, $K\Pi$, and $K\Delta$, respectively. The K-vacancy-sharing process discussed by Meyerhof corresponds to the transition Σ-Π-$K\Pi$. To account for the observed Li K excitation probability we propose an alternative transition mechanism, namely Σ-Π-Δ-$K\Delta$. We note that the Δ state implies two-electron excitation which is not explicitly accounted for in the MO diagram. Hence, the application of the molecular-state diagram appears to be most useful in the present case.

The multistep process Σ-Π-Δ-$K\Delta$, though rather complex, appears to be dominant in slow Li^+ + He collisions for the following reasons: (i) The excitation of the Δ state is strong. This is seen from the ratio $\sigma_2(\mathrm{He})/\sigma_1(\mathrm{He})$ which was calculated as mentioned above; it increases from about 0.3 to 0.7 as the incident energy decreases from 30 to 6 keV. (ii) The probability for Δ-$K\Delta$ transitions is comparatively high at large internuclear distances R as the corresponding energy difference is relatively small. At the separated-atom limit the energy spacing is ~20 eV for Δ and $K\Delta$, whereas it is ~40 eV for Π and $K\Pi$ (see Fig. 3).

To support the proposed mechanism we show first by means of Meyerhof's theory for K-vacancy sharing[5] that the Π-$K\Pi$ transition probability is rather small. In atomic units the sharing ratio follows as $\sigma_K(H)/\sigma_K(L) = \exp(-\gamma\pi/v)$, where $\gamma = (2I_H)^{1/2} - (2I_L)^{1/2}$ with I_H and I_L being the K binding energies of the heavier and the lighter collision partner, respectively. $\sigma_K(H)$ and $\sigma_K(L)$ are the corresponding K-excitation cross sections. In Fig. 2 are shown theoretical results (curve 1) obtained with $I_H = 64.4$ eV and $I_L = 24.6$ eV, the binding energies of the 1s electrons of Li and He, respectively. Since Meyerhof's theory describes the Π-$K\Pi$ transition, curve 1 should be compared with $\sigma_1(\mathrm{Li}^+)/\sigma_1(\mathrm{He})$, where $\sigma_1(\mathrm{Li}^+)$ is the cross section for producing Li^+ excited in the K shell. Unfortunately, we were not able to observe Li^+ in the present experiments; however, comparison is possible with Li. It is seen that the calculated results are orders of magnitude lower than the experimental $\sigma_1(\mathrm{Li})/\sigma_1(\mathrm{He})$. This indicates that the Π-$K\Pi$ transition can hardly contribute to Li K excitation.[20]

Next, we estimate the probability for Δ-$K\Delta$ transitions. For this we use Meyerhof's formalism with modified energy parameters. The Δ-$K\Delta$ process involves a one-electron transition from the 1$s\sigma$ MO into the completely empty 2$p\sigma$ MO. At large R such transitions change the total energy of the molecular system by an amount equal to the difference in the 1s binding energies for Li($1s^2 2p$) and He($1s2p$). (Here, we assumed that the two $2p\pi$ electrons excited in the Δ and $K\Delta$ states are equally shared between Li and He.) Using the corresponding binding energies $I_H = 64.8$ eV and $I_L = 44.4$ eV, we calculated curve 2 as shown in Fig. 2. The theoretical results agree well with $\sigma_1(\mathrm{Li})/\sigma_2(\mathrm{He})$, suggesting that the $K\Delta$ state is preferentially populated by the Δ-$K\Delta$ transition.

The Σ-Π-Δ-$K\Delta$ excitation mechanism involves transitions of two $2p\sigma$ electrons into the $2p\sigma$ electrons into the $2p\pi$ MO. This requires (at least) two vacancies in the $2p\pi$ MO prior to the collision.[3] For Li^+ + He the $2p\pi$ MO is completely empty, which might partly explain the dominance of the proposed mechanism. For heavier collision systems, say C + B, the $2p\pi$ MO is partially filled and the Σ-Π-$K\Pi$ process gains relative importance. Assuming that the two processes contribute independently to K-vacancy production of the heavier particle, we extend Meyerhof's formula by setting approximately

$$\sigma_K(H)/\sigma_K(L) = P_1 \exp(-\gamma 1\pi/v) + P_2 \exp(-\gamma_2\pi/v),$$

with $P_1 + P_2 = 1$ where P_1 and P_2 are the probabilities for producing one and two vacancies in the $2p\sigma$ MO, respectively. As above, γ_1 and γ_2 are obtained using K binding energies for the lighter particle with zero or one K vacancy, respectively. As an example, the formula is applied to B + C as measured previously in disagreement with Meyerhof's original expression.[8] It was found that the data are well reproduced by the extended formula with $P_2 \approx 0.15$. This value is consistent with the probability for double $2p\sigma$-$2p\pi$ transitions which was estimated using the statistical methods of Briggs and Macek.[3]

In summary, we note that there is strong evidence for two-electron excitation followed by significant K-vacancy production in the heavier collision partner. The latter process was shown to correspond most likely to a one-electron transition which might be accounted for in a one-electron model. However, it is still an open question whether the preceding process of two-electron excitation can be treated statistically on the basis of the one-electron model.[3] We suggest that further work is needed to calculate multielectron excitation in the framework of the many-electron model.

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Raman-Induced Kerr Effect*

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A new effect is demonstrated in which a laser pulse can be made to induce a Kerr effect only at Raman-shifted frequencies. This permitted the observation of a Raman spectrum with a single dye-laser pulse. The pulse power required is a small fraction of that required to produce stimulated instabilities. The spectral information emerges in a coherent beam, phase matching is not necessary, and spectra can be obtained at any scattering angle.

Spontaneous Raman scattering has been a productive tool for studying the excitations of the scattering medium. However, low signal levels have precluded the use of Raman scattering in short-lived or high-background media such as explosions, flames, shocks, and sparks. Nonlinear optical effects, such as the stimulated and inverse Raman effects, and coherent anti-Stokes Raman scattering, have been employed in attempts to increase the effective signal-to-noise ratio in Raman spectroscopy.[1-4] Each of these effects has disadvantages that have limited its usefulness. In this paper we demonstrate a new nonlinear optical effect which can produce a complete Raman spectrum in a coherent beam with high scattering efficiency, without phase-matching requirements, and without a concomitant background from nonresonant nonlinearities. We first describe this "Raman-induced Kerr effect" by several experimental examples, and then outline the theory, which suggests useful variations on our experiments.

In the Raman-induced Kerr effect, a strong polarized monochromatic pump beam at frequency ν induces complex, anisotropic changes in the refractive indices experienced by a weak probe