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Selective formation of He^{+}(n = 3) in He^{2+}-Li collisions

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Excited-state formation in inelastic He^{2+}-Li collisions at kinetic energies up to 7 kev (center of mass) has been diagnosed by dispersal and detection of both visible and soft x-ray radiation emanating from the intersection of He^{2+} and Li beams. The theoretical predictions of Shipsey *et al.*, that He^{+}(n = 3) is preferentially formed, are found to be valid. The cross section for this selective process is sufficiently high to support the suggested application for production of super-radiance at the He II Lyman α and Lyman β wavelengths, 304 Å and 256 Å, respectively.

I. INTRODUCTION

One of the most popular proposals for achieving the inverted state distribution needed for laser action in the vuv or soft x-ray region of the spectrum is through the use of electron-transfer processes in collision systems involving multiply charged positive ions.[1-3] In a recent theoretical paper, Shipsey *et al.*[4] predicted that He^{2+}-Li collisions in the approximate kinetic-energy range 1–6 keV would preferentially lead to He^{+}(n = 3) product ions. This product radiates the HeII Lyman-α (Ly-α) and Lyman-β (Ly-β) lines at 304 Å and 256 Å, respectively, as well as the Balmer-α (H-α) line at 1640 Å. The presumed selectivity of this process together with the relatively large calculated cross section (~ 70 Å^2) led Shipsey *et al.* to suggest this system as a candidate for attaining superradiance at the soft x-ray wavelengths. In order to test the theory, and because interactions of this type are important in the energy balance of controlled fusion plasmas,[5,6] our crossed beam apparatus was modified to permit analysis of collision-produced radiation in the wavelength range 200–1250 Å. The capability of analyzing radiation in the range 1800–8500 Å has been retained.[7] Preliminary data[8] generally supported the theoretical prediction that He^{+}(n = 3) was preferentially formed at kinetic energies up to 3 keV. In addition, from the ratio of the observed Ly-α and Ly-β signals qualitative information on the population of angular momentum states of He^{+}(n = 3) was also obtained. Figure 1 shows the allowed transitions between angular momentum states of the first four energy levels of He^{+}. Since the transition probabilities are known[9] the Ly-β/Ly-α ratio may be calculated for different sets of relative 3s, 3p, and 3d populations. The observed Ly-β/Ly-α ratio indicated that He^{+}(3p) was favored in varying degrees, depending upon the kinetic energy, over that of a statistical distribution, that is, 1 : 3 : 5, respectively. A simple kinematic[8] model for exchange between collisional and electronic orbital angular momenta was found to be in qualitative agreement with the data in predicting relative heights of Ly-α and Ly-β.

Since the preliminary report, substantial improvements and modification of the apparatus have been made. In this paper we present additional results of He^{2+}-Li experiments at kinetic energies up to 7 keV (c.m.). These data show that the Ly-α and Ly-β emission cross sections are of the same order of magnitude as predicted by Shipsey *et al.* In addition, it is found that the preference for formation of He^{+}(3p) persists up to 7 keV. Although excited states of He^{+} with principal quantum numbers greater than three are observed, especially at elevated kinetic energies, the cross sections for formation of these states are considerably smaller than the cross section for formation of He^{+}(n = 3).

II. APPARATUS

Figure 2 is a schematic diagram of the apparatus which, except for the vuv spectrometer, has been described elsewhere.[7] Ions are produced in an electron-impact emission-regulated source, focused into a beam by a set of cylindrical electrostatic lenses, and magnetically mass selected. Another set of lenses then focuses the beam, at the desired kinetic energy, into a collision cell.

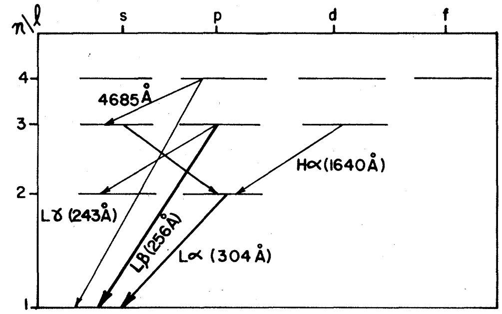

FIG. 1. Partial level diagram for He^{+} showing the electric dipole allowed transitions: Lyα, Lyβ, Lyγ, Hγ, and Paschen α.

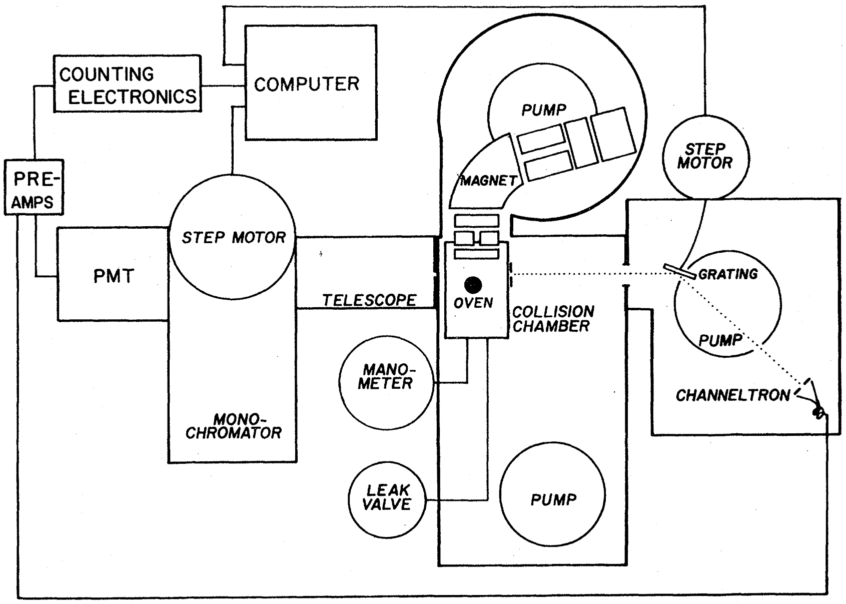

FIG. 2. Schematic diagram of the apparatus.

The ion current is collected in a Faraday cup behind the collision cell; He^{2+} beam currents were typically 20 nA.

An oven beneath the collision cell produces an effusive beam of lithium atoms which crosses the ion beam at right angles in the center of the collision cell. A fraction of the radiation emanating from the intersection of the two beams passes through a slot in the collision cell oriented parallel to the ion beam. This radiation then enters an optical system (1800–8500 Å) consisting of a light-collecting telescope, a 0.25-m scanning monochromator and a cooled photomultiplier tube used in the counting mode. The absolute spectral efficiency of the optical system has been calibrated with standard lamps, and all data displayed have been corrected for this efficiency.

During the course of an experiment the Li atom density was determined by observation of the signal from the 6708-Å resonance line of Li I in conjunction with our measured cross section for collision-induced excitation. Figure 3 is a plot of this cross section as a function of kinetic energy. The Li atom density required for calculation of the cross section from the data was determined with a surface ionization detector as discussed previously.[10,11]

The vuv system is located on the side of the collision cell opposite the optical system, and consists of a grazing incidence spectrometer and a channeltron particle multiplier used in the counting mode. The vuv spectrometer employs a platinum-coated grating on a Seya-Namioka mounting which permits fixed-slit operation. The grating, which is blazed at 300 Å, is toroidal for low astigmatism and wide band, 150–1500 Å, with

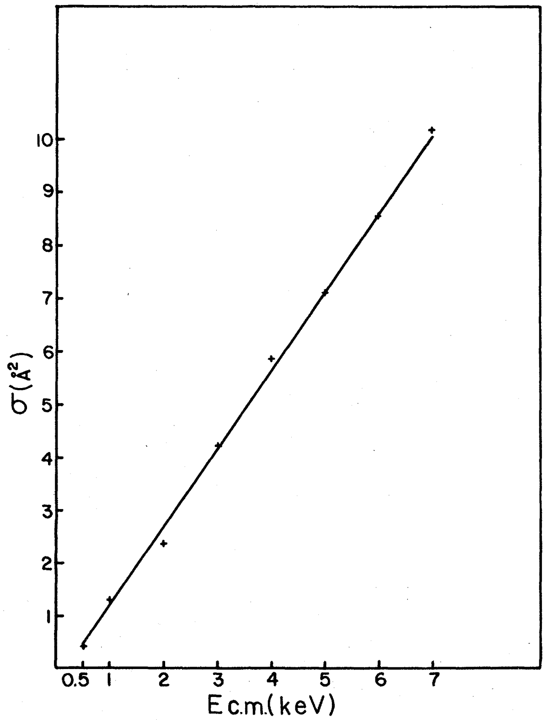

FIG. 3. Emission cross section σ for the 6708-Å Li I resonance line as a function of kinetic energy (c.m.) for He^{2+}-Li collisions.

an ultimate resolution of 4 Å. Wavelength calibration was performed with a differentially pumped rf discharge source using hydrogen, helium, and neon. Scans of the region 500–800 Å were taken with a tin filter in place to suppress second-order radiation from the He II Lyman series. The relative spectral response of the vuv system was obtained by folding together the grating and channeltron responses as given by the respective manufacturers. The absolute spectral efficiency of the vuv system was obtained by calibration against the optical system, the calibration of which is discussed elsewhere,[12] using the branching ratio technique with $He(3\,^1P)$.[13,8] Transitions from this state occur at 537 Å ($3\,^1P \rightarrow 1\,^1S$) and 5016 Å ($3\,^1P \rightarrow 2\,^1S$). The statistical uncertainty in the ratio of the counts at 537 Å to those at 5016 Å was less than 10%.

The entire experiment is controlled by two microprocessor-based computers, one of which controls the optical system and the other the vuv. The two systems are interfaced with the vuv controller acting as the master, supplying synchronization and system control. Two modes of operation are employed. In the first, emission spectra are assembled at a fixed kinetic energy by stepping through the wavelength range of interest and counting photons at each setting. In the second mode of operation a spectrometer is set at the wavelength of a given transition and the collision energy varied to obtain the energy dependence of the emission cross section. For the 6708 Li I resonance line, the ion beam is electronically chopped and photons detected synchronously to correct for interfering blackbody background.

Relative cross sections reported here are considered reliable to within 15%. As discussed previously,[10-12] uncertainties in the absolute calibration of the optical system together with the difficulties associated with precise determination of the Li atom density make it possible to assign absolute cross sections in the optical only to within a factor of 3.

TABLE I. Observed ratios of the Ly-α to Ly-β signals as a function of center-of-mass kinetic energy.

E (keV)	Ly α/Ly β	σ [a]
0.5	0.78	0.15
1.0	1.15	0.31
1.5	0.89	0.21
2	0.76	0.11
3	0.58	0.15
4	0.57	0.11
5	0.51	0.13
6	0.47	0.10
7	0.50	0.11

[a] Standard deviation of the Ly-α/Ly-β ratios.

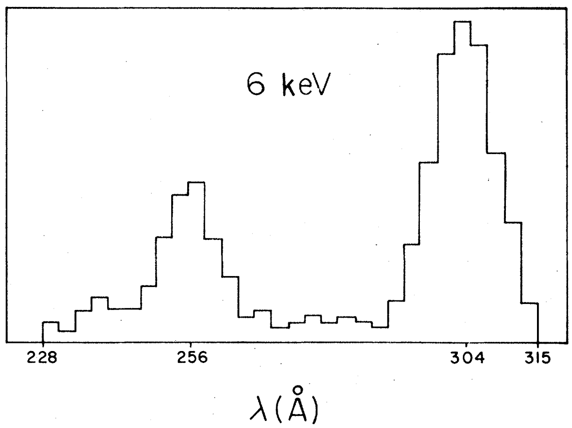

FIG. 4. Emission spectrum (228–315 Å) from He^{2+}-Li collisions at 6 keV (c.m.); the resolution is 18 Å FWHM.

III. RESULTS AND DISCUSSION

In our preliminary report of He^{2+}-Li collisions it was noted that the relative intensities of the Ly-α and Ly-β signals indicated nonstatistical population of $He^+(n=3)$ up to about 3 keV (c.m.) with preferential formation of $He^+(3p)$. If only $He^+(n=3)$ were formed, and if the orbital angular momentum states were statistically populated then the ratio Ly-β/Ly-α would be 0.44. Any higher value of this ratio indicates preferential population of $He^+(3p)$. This is, in fact, true even if states having $n > 3$ are also formed. Of course formation of $He^+(n=2)$ along with $He^+(n=3)$ can only lower this ratio. Table I is a listing of the measured Ly-β/Ly-α ratios up to 7 keV (c.m.). Also included in this table is the statistical un-

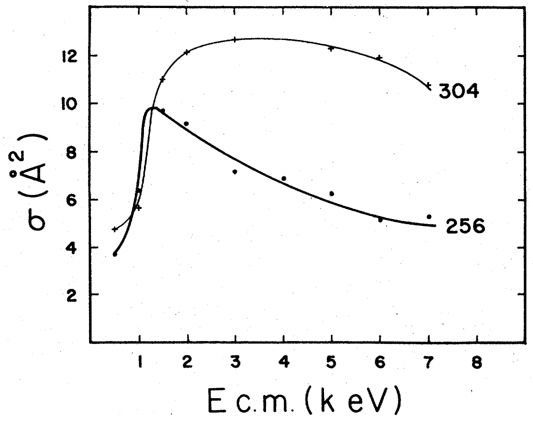

FIG. 5. Emission cross sections σ for He II Lyman lines as functions of kinetic energy (c.m.) for He^{2+}-Li collisions.

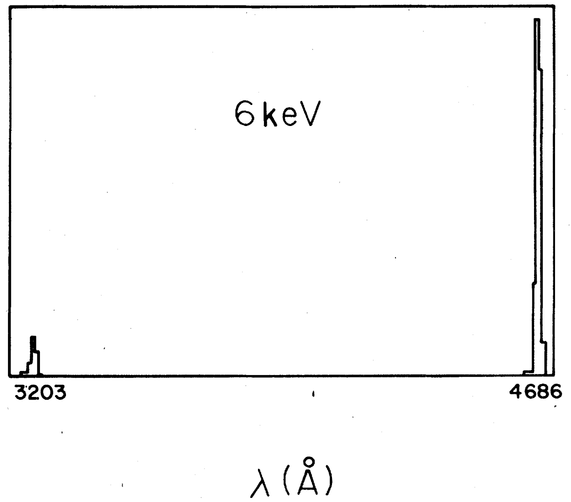

FIG. 6. Emission spectrum (3200–4690 Å) from He^{2+}-Li collisions at 6 keV (c.m.) the resolution is 5 Å FWHM. The lines shown are the He II Paschen-α and Paschen-β emissions.

certainty σ, in this measured ratio. Although this ratio approaches that of a statistical distribution as the kinetic energy is increased, the preferential formation of $He^{+}(3p)$ persists up to 7 keV. Figure 4 is a collision-produced spectrum taken at 6 keV (c.m.) over the wavelength range 228–315 Å which illustrates the nonstatistical nature of the process. Additionally, although Ly γ would be resolvable, its absence in this spectrum is indicative of a relatively low population of $He^{+}(4p)$ at 6 keV. Spectra obtained at the highest kinetic energy studied, 7 keV, are essentially identical to the 6-keV spectrum. The emission cross sections for Ly α and Ly β are plotted separately as functions of kinetic energy in Fig. 5. As noted in our preliminary report the magnitudes of these cross sections are sufficiently high to support the potential x-ray laser application suggested by Shipsey *et al.*

As discussed above, the Ly-γ line, indicative of formation of $He^{+}(4p)$ is not present in Fig. 4. In addition, although there are known lines of Li II in this wavelength range, none were observed. Scans of the wavelength region above 500 Å show no other lines, indicating that double charge transfer, with excited-product helium atoms, is not important in this system. The absence of Ly γ in the vuv spectrum does not preclude production of the 4*s*, 4*d*, and 4*f* states of He^{+}. In order to further investigate this point the optical system was employed. The most prominent line in the He^{2+}-Li spectrum in the wavelength range 1800–8500 Å is the 6708-Å Li I resonance line which is indicative of collision-induced excitation with product Li(2*p*). This reaction channel, which was observed in previous work in this laboratory,[5,8] was not discussed by Shipsey *et al.* However, the maximum observed cross section for this process is about 10 $Å^2$ at 7 keV. Figure 3 shows the measured emission cross section for 6708 Å as a function of kinetic energy. It is this line, as discussed in the previous section of this paper, which was used to measure the Li atom density. Because no emissions having Li (2*p*) as a lower state were observed, the emission cross section for 6708 Å is essentially the same as the excitation cross section for Li(2*p*).

Also observed in the 1800–8500 Å wavelength range were lines from the He II Paschen series, that is the $n = 4 \rightarrow 3$ transition at 4685 Å, the $5 \rightarrow 3$ transition at 3202 Å and the $6 \rightarrow 3$ transition at 2733 Å. All of these lines were observed with lower emission cross sections than that for the 6708-Å Li I resonance line. Figures 6 and 7 are 6-keV collision-produced spectra taken with the optical system. These spectra show the relative strengths of the 3202, 4685, and 6708 Å lines. Two spectra are required for this purpose because a change of grating was necessary. Emission cross section for the He II Paschen lines as functions of kinetic energy are shown in Fig. 8. The magnitudes of these cross sections are less than 10% of those for Ly α and Ly β, which further support the predictions of Shipsey *et al.* that He^{2+}-Li collisions below 6 keV result primarily in formation of $He^{+}(n = 3)$.

According to Shipsey *et al.* the selective formation of $He^{+}(n = 3)$ is effected by transfer of the He^{2+}-Li system to a molecular Π state which has separated atom products $He^{+}(3p)$ and Li^{+}. Mixing of the angular momentum states is then presumed

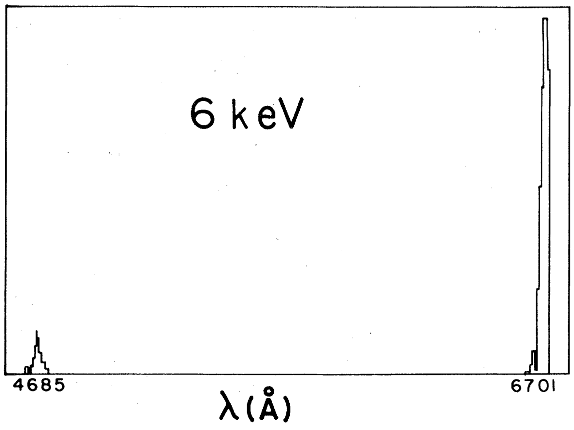

FIG. 7. Emission spectrum (4680–6710 Å) from He^{2+}-Li collisions at 6 keV (c.m.); the resolution is 10 Å FWHM.

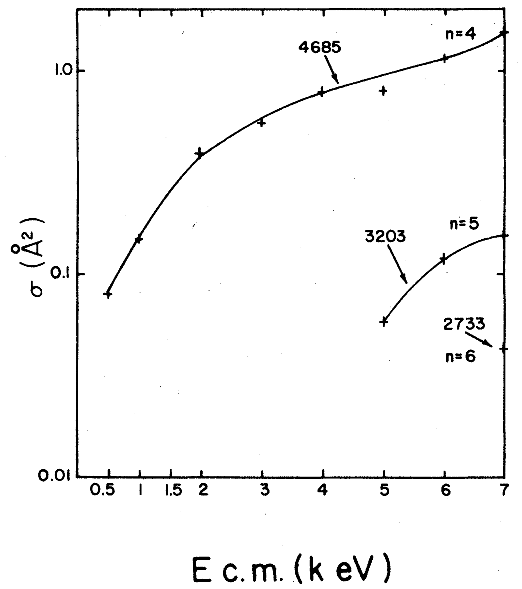

FIG. 8. Emission cross sections σ for the He II Paschen lines as functions of kinetic energy for He^{2+}-Li collisions.

to result in product $He^{+}(3s)$ and $He^{+}(3d)$ as well. While Shipsey *et al.* were unable to predict the specific product distribution, our data show that mixing is not complete and that $He^{+}(3p)$ predominates. Although only small amounts of $He^{+}(n=4)$ are formed, our data show that $He^{+}(4p)$ is populated at a lower than statistical level. This conclusion is based on the measured 4685-Å signal, the fact that Lyγ was not observed, and the relative vuv-to-optical efficiencies. This result is not inconsistent with the kinematic model for angular momentum exchange discussed earlier, but, in the absence of more specific information on the formation of the $4s$, $4d$, and $4f$ states, further conclusions are unwarranted.

If, as suggested by the data, it is assumed that virtually all He^{+} products are formed in the $n=3$ state then the Ly-β cross section, corrected for $3p \rightarrow 2s$ branching and summed with the Ly-α cross section, gives the cross section for formation of $He^{+}(n=3)$. Figure 9 contains a plot of the measured values of this cross section as a function of kinetic energy up to 7 keV (c.m.). The functional dependence of this cross section up to 3 keV is identical to that reported earlier. However, the

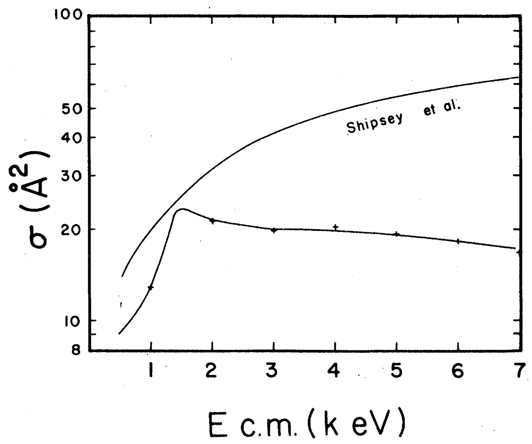

FIG. 9. Cross section for production of $He^{+}(n=3)$ calculated from the data as discussed in the text. The curve represents theoretical values and was taken from Ref. 4. The points were obtained in the present work.

absolute values are approximately twice as large as those reported in the preliminary version of this work. This difference is due to the revised calibration of the instrument. While our measured cross sections are generally smaller than those predicted by Shipsey *et al.*, they are, as noted above, supportive of the potential x-ray laser application of this system.

Finally it would be interesting to monitor the H-α 1640-Å radiation resulting from these collisions. Unfortunately this wavelength can be neither dispersed nor detected with the present apparatus. Comparison of the 1640-Å signal with those of Lyα and Lyβ would yield more definitive information on the direct formation of $n=2$. In addition, because the lower state of the H-α transitions is not the ground state, as it is for the Lyman series, lasing at 1640 Å may be more practical in this system than at 256 or 304 Å.

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