Double and Single Ionization of Helium by 58-keV X Rays

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We have measured the ratio of cross sections for double to single ionization of helium by Compton scattering, $R_C = \sigma_C^{++}/\sigma_C^{+}$, at a photon energy of 58 keV using cold target recoil ion momentum spectroscopy. We find a value $R_C = (0.84^{+0.08}_{-0.11})\%$ that is in agreement with the asymptotic limits predicted by Andersson and Burgdörfer [Phys. Rev. A **50**, R2810 (1994)] and Surić *et al.* [Phys. Rev. Lett. **73**, 790 (1994)]. [S0031-9007(96)00468-1]

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During the last three decades the study of the process of double ionization in helium has been the focus of many experimental and theoretical investigations, and it continues to be a subject of fundamental interest (see, for example, the recent review [1] and references cited therein). Understanding the role of electron-electron interactions in helium, which is the simplest neutral atomic system exhibiting such correlation effects, is basic to an understanding of the more general cases of many-electron atoms and molecules. Among the different investigations, those concerned with double ionization induced by photons play a special role, because photons interact only with single electrons and do this in a way which can be formulated exactly. Thus photon-induced double ionization is an ideal probe for electron-correlation effects.

As a measure of the relative importance of electron correlation in the ionization process, determination of the ratio of cross sections for double and single ionization, $R = \sigma^{++}/\sigma^{+}$, is of particular interest. Calculations of this quantity are highly sensitive to approximations and assumptions about electron correlations both in the bound initial state and in the final state of the two electrons escaping from the ion. A special situation arises if the energy deposited into the atom by the photon is very large: then, the initially struck electron is removed practically instantaneously from the atom without being able to interact much with the other electron. A theoretical treatment of such a situation can be reduced to the so-called "sudden approximation" (cf. [2]). In this approach the probability for double ionization depends mainly on the overlap matrix element of the initial neutral $1s$ state with a continuum state ϵs of the ionized atom and is otherwise proportional to the probability of single ionization. In the limit of high photon energies the ratio R should therefore approach an asymptotic value, and calculations of this value depend very sensitively on an accurate representation of the highly correlated initial state of helium [2–6].

Photon-induced ionization can proceed in two ways depending on whether the photon is either absorbed (photoabsorption) or scattered (Compton scattering). The concept of a sudden process at high photon energies is in principle applicable in both of these situations, however, in a process-specific way. Also, the photon energies necessary to justify the treatment as a sudden process differ greatly in the two cases. In the absorption process the atom receives the photon's energy in full, whereas in the Compton process it is the energy loss of the scattered photon that is imparted to the atom. This latter energy transfer is not a constant, but it depends on the scattering angle of the photon and is maximum for backward scattering. As a consequence, in the process of Compton scattering substantially higher photon energies are required to assure the conditions for a sudden process than is the case for photoabsorption.

Whereas for the ratio $R_{\rm ph}$ of ionization by photoabsorption the theoretical prediction of an asymptotic value $R_{\rm ph} = 1.67\%$ has been solidified by several independent theoretical [2,3,5–8] (for a discussion of a breakdown of the asymptotic behavior, see Refs. [9,10]) and experimental studies [11–13], there are two general predictions for the asymptotic ratio R_C of ionization by Compton scattering which differ by more than a factor of 2. Amusia and Mikhailov [14] found the asymptotic value R_C to coincide closely with the photoabsorption value $R_{\rm ph}$, $R_C = 1.68\%$; Andersson and Burgdörfer [15], Surić *et al.* [16], and Kornberg *et al.* [17] calculated the asymptotic value to be about $R_C = 0.8\%$. The work of Bergstrom, Hino, and Macek [18] gives for the highest photon energy considered (20 keV) a value similar to the former prediction, $R_C = 1.6\%$. However, Bergstrom *et al.* pointed out that an asymptotic behavior had not been reached in their calculation—approaching 20 keV their calculated values R_C were still decreasing. In the experimental investigations of R_C the highest x-ray energy reported so far is also 20 keV: Levin, Armen, and Sellin [13] determined

$R_C = (1.45 \pm 0.13)\%$ in agreement with Samson *et al.* [19] who obtained $R_C = (1.4 \pm 0.42)\%$. It has to be emphasized that experiments become progressively more difficult at higher photon energies as the Compton double ionization cross section is at about 10^{-26} cm^2 and the available synchrotron-light-source flux diminishes. Because of the above stated concerns that an asymptotic value of R_C may not yet be reached at 20-keV x-ray energy, the present experiment was motivated by the desirability of establishing a firm value of R_C at a significantly higher photon energy.

In this Letter we report a measurement of the ratio $R_C = \sigma_C^{++}/\sigma_C^{+}$ of cross sections for double and single ionization by Compton scattering with 58-keV x rays. Our measurements employed the cold target recoil ion momentum spectroscopy (COLTRIMS) method [20] which is optimally suited for the present purpose in that it is able to separate Compton scattering events from photoabsorption events [11]. Furthermore, COLTRIMS offers a powerful tool to detect and avoid any contaminating ionization events which are caused by stray lower-energy photons or electrons, for both of which the cross section can be higher by several orders of magnitude than the low cross section for ionization by Compton scattering.

We used the 58-keV x-ray beam at the ID 15, beam line 25 of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France [21]. It arises from a wiggler insertion device and a Bragg-type monochromator employing a single bent Si(311) crystal. It was focused and collimated by two upstream sets of tungsten 4-jaw slits to a spot 1 mm wide by 4 mm high at the target. The beam entered the target chamber through a 1 mm thick Be window. Between the window and the target region a third set of 4-jaw slits (tantalum) was included to shield against possible scattering products generated in the transition of the beam through the window. The flux was typically around 10^{12} photons/sec. The incident photon-energy spectrum peaked at 57.85 keV and had a bandwidth of 0.1%, i.e., about 60 eV. The experiment was performed during the 16-bunch operation mode of the storage ring.

The target in our experiment was a cold (<1 K) supersonic jet produced by allowing He gas at a pressure of 30 bars to expand vertically through a 30-μm nozzle into a first vacuum chamber. The jet that resulted then passed through a 0.6-mm skimmer into the target chamber, which was maintained at a pressure of 10^{-6} mbar. At the position where the jet was intercepted by the photon beam, its diameter was about 3 mm and the density was about 10^{12} He atoms/cm^2. Ions created at the intersection of the photon beam with the jet were accelerated out by means of a carefully constructed uniform electric field of 225 V/cm. After traveling 4.5 cm these ions passed into a 9 cm long field-free drift tube and were then accelerated further by a 1500-V postacceleration potential onto a two-dimensional position-sensitive chevron-channel-plate detector (PSCD), 40 mm in diameter, and with a wedge-and-strip readout. The ions were detected in coincidence with a timing signal derived from the ESRF storage ring. Identification of the helium charge states was then obtained from the analysis of flight times. Additional information on the detection properties was obtained from the pulse height of the detector signal. A more detailed description of the experimental method can be found elsewhere [11,20,22].

Because of the smallness of the relevant cross sections, the low rate of ionization events was the main difficulty in the experiment. According to the compilation of Veigele [23], the total attenuation cross section of helium is about 1.1×10^{-24} cm^2 at an x-ray energy of 58 keV. Virtually the entire attenuation cross section is to be attributed to Compton scattering, photoabsorption at this energy is suppressed by roughly a factor of 1/6000 [6] (due to the $E^{-7/2}$ scaling of the cross section in dipole

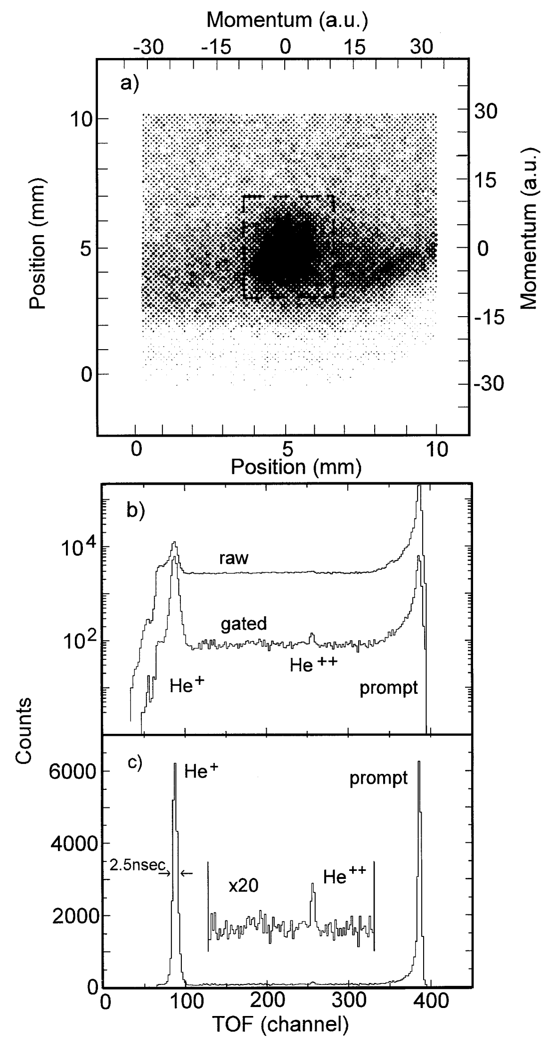

FIG. 1. (a) Raw image of the position sensitive ion detector. The software window on the jet spot is indicated. (b) Ion time-of-flight spectrum. On top the experimental raw spectrum, below the spectrum after off-line data analysis. (c) Gated TOF spectrum as in (b) on a linear scale.

approximation). Therefore, the vast majority of events observed can be expected to arise from ionization by Compton scattering. The raw data accumulated during 40 h of continuous data taking are shown in Fig. 1(a) as the PSCD image in an enlarged scale and Fig. 1(b) as the time-of-flight (TOF) spectrum. In the 16-bunch mode of the storage ring the timing signal synchronous with the x-ray pulses arrives at intervals of 176 ns. The ion flight times, however, are several hundred nsec and so the TOF spectrum shown in Fig. 1(b) is "wrapped around." The signal delays were adjusted to bring the peaks due to He^+ and He^{++} ions into regions of the time spectrum where background subtraction could be performed effectively. The peak labeled "prompt" on the right end of the TOF spectrum is attributable to photons and high-energy electrons scattered towards the PSCD. The strong background is mainly due to "dark" pulses in the PSCD.

The COLTRIMS technique allows one to overcome some of the problems inherent in traditional ion-counting techniques. The noise level can be significantly reduced and, moreover, it is possible to identify ionization products from contaminating events of different origins. In the Compton-ionization process very little momentum is transferred to the residual ion. Therefore, the (x, y) position and arrival time of each ion at the detector served to create a three-dimensional image of the source of the ions. On the PSCD, the ions originating in the region of intersection of x-ray beam and cold helium jet generate a sharply bounded image as can be seen in Fig. 1(a). The size of this "jet spot" is caused by the extended target volume. The jet spot overlays a weaker diffuse distribution from thermalized ambient helium gas ionized along the x-ray beam path. Because of the well-defined vertical momentum of the jet atoms of 6 a.u., the image of the jet spot is somewhat displaced vertically against this other contribution. By setting a suitable software window in the off-line analysis on the jet-spot image one can sort out those ions with low momentum, i.e., events from ionization of helium atoms in the cold jet by Compton scattering [24]. At the same time the noise level from "dark" pulses in the spectrum is significantly reduced. Carrying out this procedure produced the "gated" TOF spectrum also shown in Fig. 1(b) as well as in Fig. 1(c) on a linear scale. Now the weak peak of He^{++} ions is clearly visible and the ratio R_C can be calculated from the respective peak areas. Furthermore, setting another software window displaced along the beam direction permits determination of possible disturbances of R_C arising from the underlying background of residual gas and thermalized helium, for which the conditions are less well defined than for the jet spot. Within the statistical uncertainty the He^{++}/He^+ ratios from the jet-spot window and the "warm" helium gave the same result. Similarly, if helium ions in the jet spot were created by photons of lower energy or electrons produced in beam-solid interactions, such events should be also found outside the beam path and the well-defined jet spot. No such events were detected. We therefore conclude that our result reflects the uncontaminated ratio R_C.

The possibility of different detection efficiencies for the two He charge states due to their different impact velocities when hitting the PSCD was investigated in measurements with postaccelerations of 3000, 2100, and 1500 V/charge. In comparing the normalized count rates for the He^+ charge state, we found the detection efficiency to be constant within 10%, the statistical accuracy of the procedure. Therefore, no such correction had to be applied in the determination of R_C. A velocity dependency of the detection efficiency would tend to increase the observed ratio He^{++}/He^+, and this uncertainty has been incorporated in the lower error limit of our result. The value then derived for the ratio was $R_C = (0.84^{+0.08}_{-0.11})\%$. The low rate of production of He^{++} ions was the limiting factor influencing the accuracy with which a value for R_C could be extracted.

Our value R_C is plotted in Fig. 2 (open circle) along with calculations and other experimental data in the photon-energy range 8–60 keV. Some of these experiments were not able to distinguish between photoabsorption and Compton scattering events, however, for photon energies above 15–20 keV the contribution of photoabsorption to the total cross section decreases to less than 1%, and one can expect that $R \approx R_C$. Among the available theoretical predictions some quote a value for the asymptotic high-energy limit [14–17]. Others calculate the energy dependence of R_C; however, these studies do not extend up to the x-ray energy of our experiment. The present result would appear not to sup-

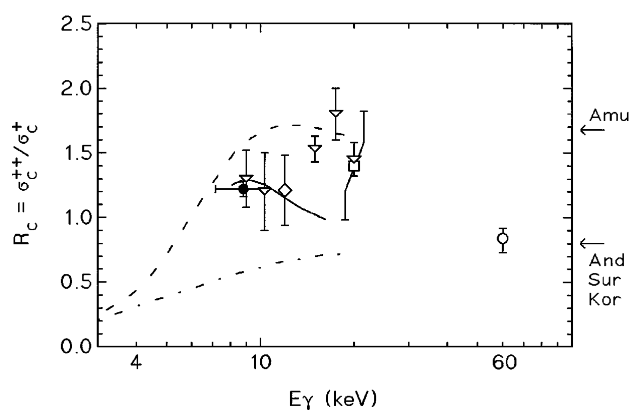

FIG. 2. Ratios of double to single ionization, R_C, after Compton scattering. Experimental data—open circle: this work; closed circle: Spielberger *et al.* [11]; open triangles: Levin *et al.* [13]; open diamond: Sagurton, Morgan, and Bartlett [25]; open square: Samson *et al.* [19]. Theoretical results—broken line: Bergstrom, Hino, and Macek [18]; full line: Andersson and Burgdörfer [15]; dash-dotted line: Surić *et al.* [16]. The arrows indicate the predicted asymptotic ratios $R_C = 1.68\%$ of Amusia and Mikhailov (Amu) [14] and $R_C = 0.8\%$ of Andersson and Burgdörfer (And) [15], Surić *et al.* (Sur) [16], and Kornberg *et al.* (Kor) [17].

port the prediction of Amusia and Mikhailov [14] for the asymptotic high-energy limit of $R_C = 1.68\%$. As indicated in Fig. 2, our value of R_C is more closely in agreement with the asymptotic values calculated by Andersson and Burgdörfer [15], Surić *et al.* [16], and Kornberg *et al.* [17]. It must be pointed out, though, that the calculation of Surić *et al.* [16] predicts an approach to the asymptote from below, which seems to be clearly at variance with all of the available experimental points. Thus, our result, especially when taken in conjunction with other determinations of R_C [11,25] made at lower photon energies, appears to be in the best agreement when compared with the theoretical values calculated by Andersson and Burgdörfer. Clearly the comparisons with theory would benefit significantly with additional data filling the wide gaps of the photon-energy range displayed in Fig. 2.

In the energy regime accessed in the present experiment—an x-ray energy of 58 keV amounts to 11% of the electron rest mass—the question of the necessity of a fully relativistic treatment has to be raised. The currently available predictions of the asymptotic value R_{ph} and R_C refer to an asymptotically high, but nonrelativistic limit. Relativistic calculations for R_C will certainly soon be required for comparison as experiments that push towards even higher photon energies in the range 50–150 keV are clearly within reach. The more intense photon beams soon to be available at the Advanced Photon Source at Argonne National Laboratory and at SPring8 in Japan and the very high x-ray energies accessible at the storage ring PETRA of HASYLAB in Germany will permit this situation to be addressed.

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Note added.—A similar measurement of $R = \sigma^{++}/\sigma^{+}$ at this energy was also performed recently on the same beam line by another group [26] using a traditional TOF technique. A discussion of the result for R_C of Amusia and Mikhailov [14] can be found in Surić, Pisk, and Pratt [27].

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