Ratio of Cross Sections for Double to Single Ionization of He by 85–400 eV Photons

R. Dörner,[1,2,*] T. Vogt,[2] V. Mergel,[2] H. Khemliche,[3] S. Kravis,[1] C. L. Cocke,[1] J. Ullrich,[4] M. Unverzagt,[2]
L. Spielberger,[2] M. Damrau,[2] O. Jagutzki,[2] I. Ali,[2] B. Weaver,[2] K. Ullmann,[2] C. C. Hsu,[6] M. Jung,[5] E. P. Kanter,[5]
B. Sonntag,[7] M. H. Prior,[3] E. Rotenberg,[8] J. Denlinger,[9] T. Warwick,[3] S. T. Manson,[10] and H. Schmidt-Böcking[2]
[1]*Department of Physics, Kansas State University, Manhattan, Kansas 66506*
[2]*Institut für Kernphysik, Universität Frankfurt, August Euler Str. 6, D60486 Frankfurt, Germany*
[3]*Lawrence Berkeley National Labortory, Berkeley, California 94720*
[4]*Gesellschaft für Schwerionen Forschung, D64291 Darmstadt, Germany*
[5]*Argonne National Laboratory, Argonne, Illinois 60439*
[6]*National Tsing Hua University, Taiwan*
[7]*Hasylab am DESY, D22603 Hamburg, Germany*
[8]*Department of Physics, University of Oregon, Eugene, Oregon 97403*
[9]*Department of Physics, University of Wisconsin, Milwaukee, Wisconsin 53211*
[10]*Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303*
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The ratio of double to single ionization of He by 85–400 eV photons has been measured using cold target recoil ion momentum spectroscopy. This technique allows the elimination of all possible systematic errors discussed so far in the literature on this subject. We find the ratio in this energy range to be about 25% lower than previously assumed.

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Double ionization of He by a single photon is the simplest and most fundamental many-electron process. The ejection of two electrons from an atom following absorption of one photon is strictly prohibited in an independent electron approximation. Thus, for photoabsorption by He, determining the probability of double ionization alone is already a challenging test case for our understanding of electron-electron correlation. The ratio of double to single ionization for photoabsorption ($R = \sigma^{++}/\sigma^{+}$) reaches its maximum $R_{\max}$ between 150 and 250 eV photon energy and drops to a value of 1.67% for asymptotic high photon energies. This value has only very recently been experimentally established [1,2]. Although all theoretical calculations today agree on this asymptotic value [3,4], they greatly disagree in the energy regime around $R_{\max}$. Values of $R_{\max}$ between 3.1% and 5.4% have been found by various theoretical approaches [4–12].

From the experimental side, values of $R_{\max}$ between 3.4% and 5.2% have been reported over the last 30 years [13–18]. The discrepancies between the various experimental results are larger than most of the statistic errors, indicating unknown systematic problems in some of these experiments. The goal of this work is to provide reliable benchmark data on R in the region of the maximum.

We have used cold target recoil ion momentum spectroscopy (COLTRIMS) to determine R. This technique allows the measurement of the momentum vector of the ion as well as its charge state [1,19]. The experiment was performed at the Advanced Light Source (ALS) at LBNL during double bunch operation. The light from the U7 undulator passed through a spherical grating monochromator and was focused to a beam spot of $0.1 \times 0.1\ \mathrm{mm}^2$. The photon beam intersected a precooled supersonic He gas jet. The latter had a diameter of 1 mm at the intersection region and an internal momentum spread of below 0.07 a.u. (atomic units). The local He pressure in the jet was about 10^{-5} mbar dropping by approximately 2 orders of magnitude at the edge of the jet. The residual gas pressure was about 2×10^{-8} mbar in the scattering chamber. The He ions created in the $0.1 \times 0.1 \times 1\ \mathrm{mm}^3$ overlap region of beam and gas jet were accelerated out of the target region by a 20–40 V/cm electric field. After passing the electrostatic extraction field and a drift region they were postaccelerated by 2600 V onto a position sensitive channel-plate detector. For each event the time of flight (measured by a coincidence of the ion signal with the machine trigger), the pulse height, and position on the detector were recorded in list mode. From this information the charge state and the three-dimensional momentum vector of each ion were obtained in the off-line analysis. The experimental setup is discussed in more detail in Refs. [1,19].

For 100 eV photon impact the measured momentum distribution of He$^+$ in the plane given by the polarization axis and the gas jet is shown in Fig. 1(a). The momentum of the recoil ion (p_{rec}^{+}) for single ionization at a photon energy E_γ is given by energy and momentum conservation to be

$$p_{\mathrm{rec}}^{+} \approx \sqrt{2(E_\gamma - E_{\mathrm{bind}} - E_{\mathrm{exc}})}\,. \tag{1}$$

E_{bind} is the binding energy of the He atom and E_{exc} is the internal excitation energy of the He$^+$ product ion. The momentum of the photon and the kinetic energy of the ion are negligible for our purpose. The outer ring in Fig. 1(a) results from ions in the ground state. Rings with smaller

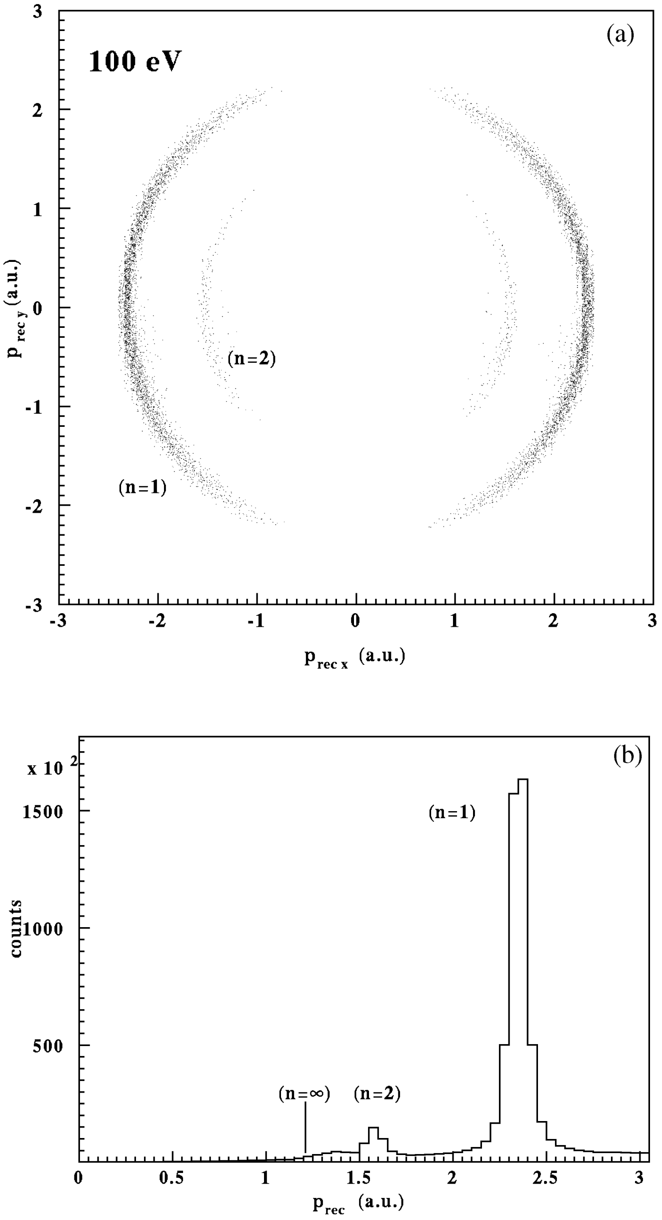

FIG. 1. (a) Momentum distribution of He^+ ions produced by 100 eV photons. The X axis is the direction of the electric field vector of the linearly polarized light. The Y axis is the direction of the gas jet. The data are integrated over a momentum range of ±0.3 a.u. in the third direction, which is the direction of the photon beam. (b) Momentum distribution of He^+ ions produced by 100 eV photons, integrated over all emission angles (raw spectrum, no background subtracted).

diameter are from ions in excited states. Light from the undulator was linearly polarized with the polarization axis parallel to the electric field in the spectrometer, which is the X axis in Fig. 1(a).

This momentum measurement for each ion, combined with the measurement of the height of the channel-plate pulse, allows us to eliminate all possible sources of systematic errors discussed in the literature on this subject [2,16,20].

Low energy stray light and higher order harmonic light from the monochromator would yield different p^+_{rec} and thus show up in Fig. 1. Levin *et al.* [20] have reported peaks and shoulders on the time of flight peaks resulting from ionization by secondary electrons from the residual gas, gas needle, or spectrometer walls. Our experiment has an inherent double check against this process, which would primarily produce He^+ ions. These electrons would not be restricted to the $0.1 \times 0.1\ \mathrm{mm}^2$ area of the photon beam but produce ions all along the gas jet and thus appear as a line on our position sensitive detector. In addition, electron impact ionization produces recoil ions with a continuous momentum distribution peaked close to zero momentum [21].

Contaminations of $H_2{}^+$ have the same time of flight in a strong electric field as He^{++} ions. These ions would, however, be produced in our spectrometer all along the photon beam. In addition, they have momenta very different from those of the He^{++} ions.

In all experiments using warm He gas as a target the momentum distribution of the ions is given by the sum of their momentum from the photoabsorption and their initial thermal momentum (the second being about 4.4 a.u. for He at room temperature). Depending on the extraction field this might result in a higher effective collection efficiency for He^{++} in the spectrometer, in particular if only a small diameter channeltron is used for detection. The use of a position sensitive detector with an active area of 48 mm diameter and a localized target allows us to rule out this problem.

Finally, the ion detector could have a different detection efficiency for different ion charge states. In this and all previous experiments the ions are accelerated by an electric field onto the detector. Therefore He^{++} ions hit the detector with a velocity higher by a factor of $\sqrt{2}$ than He^+ ions, which could result in a different pulse height. Since the signals from the detector have to be processed by a discriminator with a threshold to suppress electronic noise, this may lead to an enhanced probability to detect the He^{++} ions. To control all problems arising from this, we recorded the pulse height of the signal from the channel-plate detector for each event (together with its position and its time of flight). We used a Z stack of three channel plates. In addition, the threshold of the constant fraction discriminator was set so low that about 5% of all counts were due to electronic noise well below the lowest pulses. Figure 2 shows the pulse height distribution of the channel plate for He^+ and He^{++} ions. For the amplification which was used during the experiment we find the distribution to be independent of the ion charge state.

Figure 3(a) shows our results and those of previous experiments. Our data show a similar energy dependence but are about 25% lower than all older data except for those from the very first experiment by Carlson [14], which are lower as well but have a very different shape.

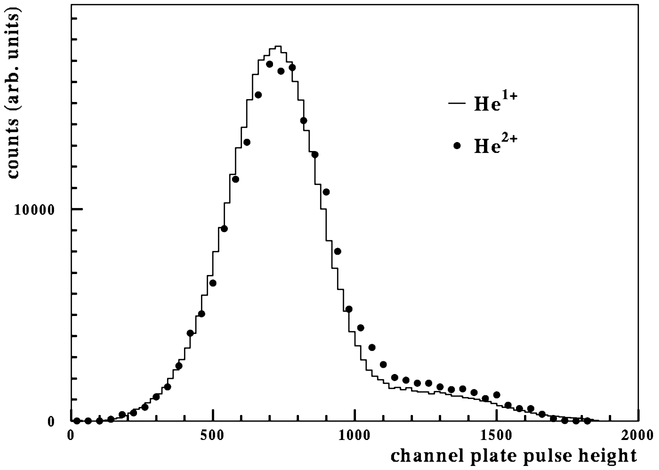

FIG. 2. Pulse height distribution from the channel-plate detector for He^{+} and He^{++} ions (see text).

The error analysis described in detail above gives us confidence that the remaining error in our data is below 6% of the measured value. These 6% are the maximum error arising from an uncertainty on the shape of the background. Most probably the error is smaller. One year before the current experiment we had performed a pilot experiment with the same technique at a bending magnet beam line at Hasylab (Hamburg). For this experiment a slightly different spectrometer and a different channel-plate detector with only two channel plates were used [22]. The results of this experiment at 130 and 150 eV are shown by the full squares in Fig. 3(a). According to [13], the data given by the open circles have been scaled up by a factor of 1.3 in order to match them with all the other available data at that time. If one takes their unscaled data (although the authors did not claim their absolute values to be correct, suspecting some unknown systematic error), one obtains the open circles shown in Fig. 3(b) which are in general agreement with our data. There are other sets of recent data obtained by Levin and co-workers [23] and by Stolte and Samson [24]. Both experiments were performed by detecting the ion time of flight only. They agree with our finding that the ratio is significantly below the previous experiments. The data of Stolte and co-workers are in very close agreement with our data.

Our new data have significant impact on evaluating the available theoretical results [Figs. 3(b) and 3(c)]. The previous experiments favored the results by Tiwari [5] and those obtained in many body perturbation theory (MBPT) calculations by Carter and Kelly [8] and Pan and Kelly referenced in [25]. These calculations clearly overestimate our data. Hino and co-workers [10,11] also performed a calculation using MBPT. The present data lie between their results in the length and acceleration gauge, respectively. Hino and co-workers used only the lowest order diagrams but a larger basis set, while in Refs. [8,25] higher order terms were included but a smaller basis set was used [25]. The calculations by Meyer and Greene [6] and by Tang and Shimamura [9] are both coupled channel calculations. The first uses the R-matrix method while the second uses wave functions in hyperspherical coordinates to represent the bound states and the continuum [26]. The latter yields identical results in the length and acceleration form and the results show close agreement with our data. The results of Pont and Shakeshaft [12] also compare favorably with our data.

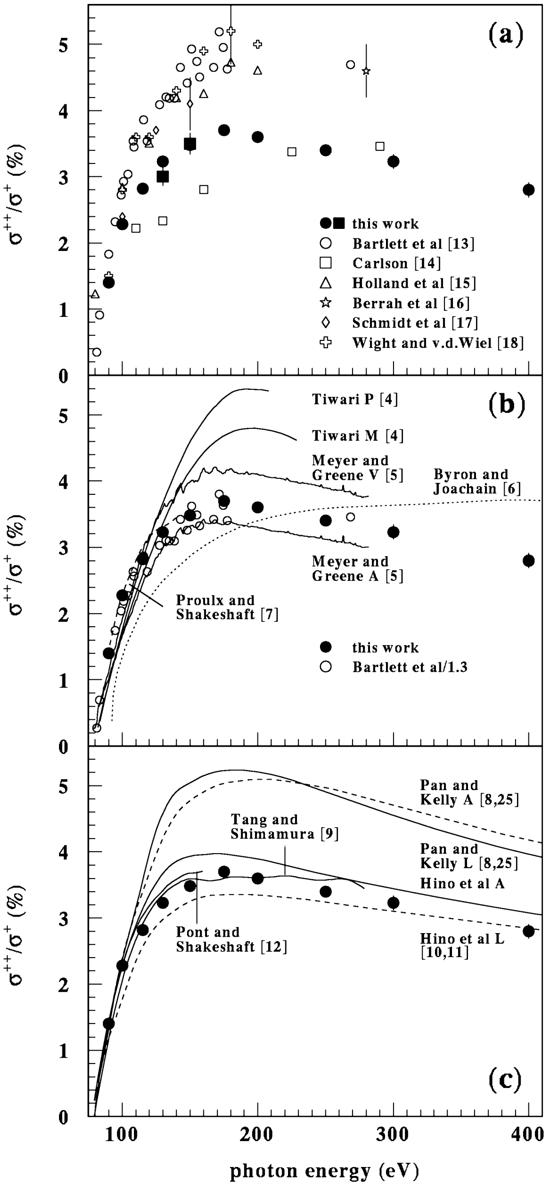

FIG. 3. R as a function of photon energy. Full circles, this work (ALS); full squares, this work (Hasylab) (see text). The open circles in (b) are the same data as in (a) but scaled down by 1.3 (see text). Tiwari P and M position and momentum matrix element [4]; V, A, and L stand for results obtained in the velocity, acceleration, or length form, respectively.

Samson and co-workers suggested [27,28] viewing double ionization as single ionization followed by an internal electron impact ionization (the so-called TS1 term of many body perturbation theory [10]). They succeeded in reproducing the previous data on R by using the experimental electron impact ionization cross section scaled by a factor which is associated with an effective radius of the atom. Since the shape of our data is not different from the previous work, this instructive model will also fit our data after adjustment of the scaling factor.

As pointed out by Manson and McGuire and others [25,29–32], R for photon impact is closely related to R for fast charged particle impact for fixed energy loss. However, using the previous photon impact data and the Bethe-Born theory, an asymptotic ratio for fast heavy particle impact, which is about 30% higher than the experimental value for these collisions [33], resulted, while another analysis using the full Born theory rather than the Bethe-Born limit [32] found no inconsistency. The inconsistency in the analysis of [30] is resolved by our new data. Using them as input to the calculation outlined in [30] we obtain ratios of $\mathrm{He}^{++}/\mathrm{He}^{+}$ for 1, 4, 25, and 200 MeV p impact on He of 0.45%, 0.38%, 0.33%, and 0.29% extrapolating to about 0.27% in the high energy limit. This is now in excellent agreement with the experimentally and theoretically well established asymptotic ratio for charge particle impact of 0.26% [33]. In addition, these new results do not substantially affect the analysis of [32].

In conclusion, we have determined the ratio of double to single ionization of He by photons from 85 to 400 eV using recoil ion momentum spectroscopy and list mode data acquisition. This technique provides a much more sensitive control of possible sources of systematic errors than all previous experiments in this field. We find the ratio to be about 25% lower than found by most of the previous work. COLTRIMS offers a unique combination of 4π solid angle and high resolution in momentum space. By adding the coincident detection of one or more electrons this technique allows a complete determination of the final state for multiple ionization by photon electron or ion impact.

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*Electronic address: doerner@ikf.uni-frankfurt.de

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