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We report the ratio of double-to-single photoionization of He at several photon energies from 2 to 12 keV. By time-of-flight methods, we find a ratio consistent with an asymptote at 1.5%±0.2%, essentially reached by $h\nu \approx 4$ keV. Fair agreement is obtained with older shake calculations of Byron and Joachain [Phys. Rev. **164**, 1 (1967)], of Åberg [Phys. Rev. A **2**, 1726 (1970)], and with recent many-body perturbation theory (MBPT) of Ishihara, Hino, and McGuire [Phys. Rev. A **44**, 6980 (1991)]. The result lies below earlier MPBT calculations by Amusia *et al.* [J. Phys. B **8**, 1248 (1975)] (2.3%), and well above semiempirical predictions of Samson [Phys. Rev. Lett. **65**, 2861 (1990)], who expects no asymptote and predicts $\sigma(He^{2+})/\sigma(He^{+})=0.3\%$ at 12 keV.

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The Coulomb three-body problem is fundamental in many branches of physics. For this reason, helium has long been studied, both experimentally and theoretically, with the goal of quantitatively understanding the mechanisms by which two electrons can be ejected into the continuum following single-photon ionization. Photons are ideal probes because they impart all their energy to the ionized electron, while charged particles impart a continuous distribution of (mostly) relatively small energies [1]. Twenty five years ago, Carlson showed qualitatively the onset of double photoionization in helium using x-ray tubes and filters [2]. Since then, the threshold region has been studied extensively by many authors, e.g., Schmidt *et al.* [3], Holland *et al.* [4] and, more recently Kossmann, Schmidt, and Andersen [5], who find excellent agreement with Wannier theory. Theoretical work spans a similar time period, including the calculation of shake effects in double ionization using dipole operators by Dalgarno [6], Byron and Joachain [7], Brown [8], and Åberg [9]; many-body perturbation theory (MBPT) calculations of Amusia *et al.* [10]; and more recent MBPT by Carter and Kelly [11], and by Ishihara, Hino, and McGuire [12].

Each of these theoretical treatments predicts a high-photon-energy limit for the ratio of double-to-single photoionization of He, generally at about 1.6–1.7% [6,7,9,12] (Brown and Gould find 2.0% [8]; Amusia *et al.*, 2.3% [10]; Carter and Kelly do not report a numerical asymptotic value [11]). The question arises whether this agreement is fortuitous. In many-body perturbation theory there is no single lowest-order term that describes the double-excitation process; instead, there exists a delicate interference among the various amplitudes representing two-electron excitation [11]. In addition to ground-state correlation (GSC) and shakeoff (SO), Carter and Kelly [11], Amusia *et al.* [10], and Ishihara, Hino, and McGuire [12] considered a final-state "two-step 1" (TS1) amplitude, in which the photoelectron ejects the remaining electron as it leaves the atom. Ishihara, Hino, and McGuire claim TS1 to be very important, and assert

that each of these effects plays an important role across a broad energy range. Dalgarno and Sadeghpour instead find the importance accorded TS1 to be gauge dependent and state that, in the acceleration form of the electric-dipole operator, initial-state correlation suffices to obtain an accurate result in the asymptotic limit [13].

In two recent papers, Samson and Samson, Bartlett, and He have shown a remarkable proportionality between the ratio of double-to-single photoionization of He and the cross section for single ionization of He^+ by electron impact, from near threshold [14] to almost 500 eV above threshold [15]. They argue that if double photoionization of He is considered as a two-step process in which a single energetic photoelectron ionizes the remaining electron, then this proportionality may continue far above threshold. This process is analogous to TS1 included in the calculation of Ishihara, Hino, and McGuire [12]. Samson's semiempirical prediction for the ratio at high energy is lower than theoretical calculations and, unlike theory, predicts no asymptote.

We reported previously the ratio of double-to-single photoionization of He at $h\nu=2.8$ keV [16]. Our result, $\sigma(He^{2+})/\sigma(He^+)=1.6\%\pm0.3\%$, while in excellent agreement with the most recently quoted *ab initio* theory [12], tells nothing about the photon-energy dependence of the ratio in this energy range. Therefore, it was the purpose of this investigation to perform a series of measurements at a number of widely spaced energies far above threshold.

The present measurements were obtained at the National Synchrotron Light Source (NSLS) on two beam lines. Monochromatic light from National Institute of Standards and Technology and Argonne National Laboratory (NIST-ANL) beam line X-24A was used in the 2–4-keV range. Focused broadband radiation from Atomic Physics beam line X-26C was employed at higher energies, where the rapidly diminishing He photoionization cross section makes use of monochromatic light less practical (at 10 keV, $\sigma^{2+}\leq 3$ mb) [17].

Details of beam line X-24A construction [18] and performance [19] have been provided elsewhere. Synchrotron radiation was tuned to 2.05, 2.4, 3.3, or 4.0 keV with a double-crystal monochromator employing Ge(111) crystals. Horizontal and vertical collimating slits each of 1.5-mm width were mounted on linear-motion feedthroughs just upstream of a 0.125-mm Be window located at the entrance to the experimental chamber. The 1.5×1.5 mm^2 photon beam was positioned 1–2 mm away from the tip of a He gas needle.

Helium ions produced in the source region were analyzed by a time-of-flight (TOF) mass spectrometer previously described [16]. Photoions were extracted, accelerated, allowed to drift through apertures covered with high transmission ($\approx 90\%$) mesh, and detected by dual chevroned Galileo MCP25 microchannelplate (MCP) detectors [28] operated with 1 kV across each plate. Spectrometer voltages were chosen to provide first-order focusing in the flight time of ions created across the ≈ 1-mm source region [20], to minimize distortions in the extraction field due to the needle, and so that flight times of typical vacuum chamber contaminants were small compared to the 567-ns spacing between stored electron bunches, thus reducing background below the He^{q+} peaks due to "wraparound." Availability of a ring timing signal coincident with each electron bunch as a stop input to a time-to-amplitude converter resulted in better resolution and shorter flight times than those that result when the spectrometer extraction field is pulsed [4,5,21]. Use of the ring timing signal also permitted TOF measurements without the need for counting electrons ionized from He, a common technique in many TOF measurements, with the penalty of increased uncertainties concerning electron-detection efficiency [22]. Dependence on a ring timing signal, however, precludes the use of this technique during normal multibunch operations (when photon bursts are separated by only 19 ns), limiting useful beam time in this mode to the few days per year reserved at NSLS for timing.

Ion flight times are known [20] to scale as $(m/q)^{0.5}$. Consequently, introduction of Ar into the source region permitted precise determination of the expected flight times for He^+ and He^{2+}. When ionized above the K edge at ≈ 3206 eV, argon TOF spectra contain measureable amounts of Ar^+ through Ar^{7+}, thus permitting accurate predictions of flight times for other m/q. Figure 1 shows both charge states of He produced by 3.3-keV photons. Both peaks attributed to He appear in exactly the location predicted and only when He is introduced into the chamber.

Data from 8–12 keV were obtained on beam line X26C using broadband light focused by a 1:1 Pt-coated mirror that accepts four horizontal mrad of the bending magnet synchrotron radiation fan and all of the vertical divergence. Because the photoionization cross section at $h\nu=200$ eV, where the $\sigma(He^{2+})/\sigma(He^+)$ ratio peaks (at about 4.8%), is more than five orders of magnitude higher than at 10 keV [17], steps were needed to assure that no low-energy photons reached the source region to avoid biasing the measurement. The presence of 1.125 mm of Be in several windows along the beam line reduced

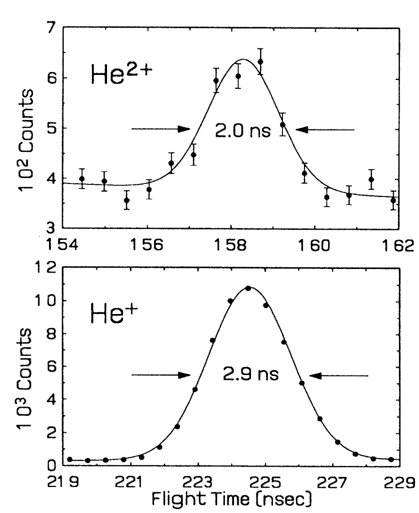

FIG. 1. Helium time-of-flight spectra obtained with monochromatic light at $h\nu=3.3$ keV.

transmission to below 10^{-5} at 2.2 keV.

Further attenuation of the light was achieved by the use of Cu, Zn, or Al filters, which, in addition, served to help define the photon energy and reduce the energy bandwidth. The He photoionization cross section at 5 keV is less than one order of magnitude higher than at 9 keV, while transmission is more than three orders of magnitude lower [17]. Thus, one can assign a mean photon energy corresponding to the Cu and Zn foils in the vicinity of their respective K edges without concern for significant spectral contamination by low-energy photons. Increasing transmission with energy through Al is negated above 11.6 keV by absorption in the Pt mirror coating. Further reduction in effective energy bandwidth can, in principle, be achieved using balanced filters [23]. When inclined at an angle of $\approx 26°$, a 0.05-mm Zn foil has a transmission curve matching almost exactly that of a 0.05-mm Cu filter below the Cu K edge. The angle was adjusted so that transmission through the two foils was equal using Fe $K\alpha$ radiation at 6.4 keV. Calculated photon flux for each filter is indicated in Fig. 2. Differences in data taken alternately with the Cu or Zn inserted into the beam can be ascribed almost solely to the effects of light of energies between the K edges of Cu and Zn, i.e., from 8979 to 9659 eV. The large uncertainties associated with the necessary subtraction prevented use of this technique here. The data obtained with each foil individually, however, are statistically significant, although with a larger uncertainty in photon energy.

Several additional experimental effects also can result in a distortion of the measured ratio of double-to-single photoionization of He. The most important were discussed by Schmidt *et al.* in connection with measurements following ionization by 2-keV electrons [24] and photons near threshold [3]. These effects have been discussed in detail with regard to the present experimental apparatus [16], and are summarized here.

(i) Spurious ionization: In addition to biases introduced by spurious light, electrons, created primarily by photoemission from the stainless-steel gas needle, can ionize He in the source region. Although not initially obvious, additional peaks near, and structure in, the He^{2+} peak appeared as the time-of-flight spectrometer resolution was improved. Care taken to collimate the beam so that it came no closer than 1–2 mm from the tip of the gas needle eliminated the contamination.

(ii) Unequal collection efficiency: In a TOF spectrometer, ion extraction efficiency is an increasing function of charge state. Monte Carlo simulation of the spectrometer confirms that no charge-state discrimination exists in the present experiment [25].

(iii) Unequal detection efficiency: Helium ions were accelerated to 4.6 keV per charge with results reproducing our earlier measurements [16] at 3.3 keV per charge. Similarly, Gao *et al.* [26] found that Varian MCP [28] detection efficiencies reached a plateau near 60% (the channel plate open-area ratio) above 3 keV.

(iv) Pressure effects: We find no bias at pressures employed, although errors are large due to the low cross sections at high photon energies. Holland *et al.* [4], using a similar technique to measure the ratio $\sigma(He^{2+})/\sigma(He^{+})$ near threshold, found pressure effects to be less than 3% for background pressures in the range $(6\text{–}130)\times 10^{-4}$ Pa for a TOF apparatus with a much longer flight path (>0.5 m) and flight times (>10 μs) than employed in the present measurement. Data reported here were collected at a measured background pressure of 10^{-3} Pa.

(v) No contamination of either peak was found in background TOF spectra obtained without He.

A systematic bias introduced by any of these effects would result in an incorrect value for the measured $\sigma(He^{2+})/\sigma(He^{+})$ ratio. The first effect would push the measured ratio too low, while the last four would render it incorrectly large. We believe these possible biases have been kept much smaller than the statistical errors reported here.

The results obtained are shown in Fig. 3 along with our earlier data point at 2.8 keV, measurements near threshold by Holland *et al.* [4], and recent measurements of Samson, Bartlett, and He up to 560 eV [15]. Other researchers [3,5] have reported results near threshold in excellent agreement with those of Holland *et al.* Asymp-

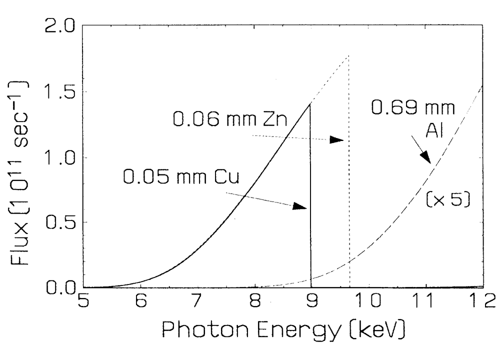

FIG. 2. Calculated flux through foil filters of the indicated thickness. Included in each curve is the absorption due to 1.125 mm (45 mils) of Be present in several windows along the beam line. In practice, the rapidly increasing transmission through Al with energy is negated above 11.6 keV by increased absorption due to the Pt mirror coating.

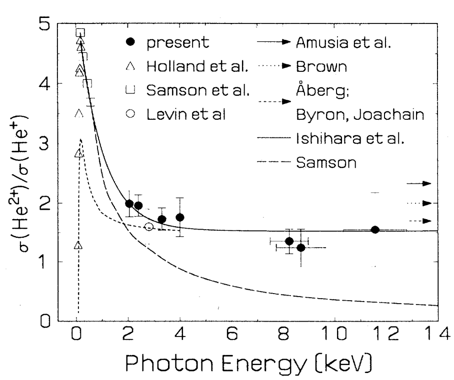

FIG. 3. Comparison of present data and measurements of $\sigma(He^{2+})/\sigma(He^{+})$ near threshold with predictions of Samson [14] and Samson, Bartlett, and He [15]. Solid curve is at least-squares fit to a form $\alpha+\beta e^{-\gamma\omega}$, where ω is photon energy, and is drawn primarily to guide the eye. Present data are consistent with an asymptote of $\approx 1.5\% \pm 0.2\%$.

totic ratios as predicted by Åberg [9], Byron and Joachain [7], and Amusia *et al.* [10] are indicated. The MBPT result of Ishihara, Hino, and McGuire, which underestimates the ratio near threshold, is in substantial agreement with our data near 4 keV, the upper energy limit of the MBPT calculation [12]. We find the double-to-single photoionization prediction of Samson [14] and Samson, Bartlett, and He [15], which agrees well with data up to $h\nu=560$ eV, falls increasingly below present measurements as photon energy is increased. Although in disagreement with our data, Samson's prediction that the double photoionization of helium has no asymptote is not definitively ruled out by present measurements.

For electron, proton, and antiproton probes of energies from 10 MeV/amu [27] to 80 GeV/amu (40-MeV electrons) [1], the $\sigma(\mathrm{He}^{2+})/\sigma(\mathrm{He}^{+})$ ratio has been determined to be $\approx 0.27\%$. In simple shake theory, the ratio of double-to-single ionization is independent of projectile charge and velocity, and should therefore be the same for charged particles and photons [1]. This ratio, lower by a factor of 6 than shake theory predicts, has been ascribed to the difference in the continuum-electron energy distribution produced by charged particles and photons [1,27]. Nonetheless, the ratios obtained with photons and charged particles are expected to be related [1], emphasizing the importance of establishing the comparative high-energy $\sigma(\mathrm{He}^{2+})/\sigma(\mathrm{He}^{+})$ photoionization ratio. The broad range over which Samson, Bartlett, and He find proportionality between double photoionization and electron ionization of He^{+} is remarkable. Our data suggest a breakdown of this direct relationship, beginning in the 1–2-ke V photon-energy range. Further work is needed to help elucidate the roles played by ground-state correlation, shakeoff, and "two-step 1" in determining the ratio of double-to-single photoionization as a function of energy.

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