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We report the first measurement of the ratio of double-to-single photoionization of helium well above the double-ionization threshold. Using a time-of-flight technique, we find $He^{++}/He^{+} = 1.6\% \pm 0.3\%$ at $h\nu = 2.8$ keV. This value lies between calculations by Amusia (2.3%) and by Samson, who predicts 1.2% by analogy with electron-impact ionization cross sections of singly charged ions. Good agreement is obtained with older shake calculations of Byron and Joachain, and of Åberg, who predict 1.7%.

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Helium is the simplest atom which exhibits electron-electron correlation. As a result, photoionization of He has been used as a testing ground for understanding correlation phenomena, such as autoionization, correlation-satellite production (shakeup), and double photoionization. Theoretical prediction of the energy dependence of the double photoionization of helium is a fundamental problem in atomic physics which requires solution of the Coulomb three-body problem. In the independent-particle framework, in which the photon can interact directly with only one electron, double photoionization can proceed only by electron-electron correlation [1]. Therefore, theoretical treatment of electron-electron correlation is of fundamental importance in calculations of double photoionization. Results also depend strongly on the choice of bound and continuum wave functions. In many-body perturbation theory (MBPT), there is no single lowest-order term which describes the double-excitation process; instead, there exists a delicate interference among the various amplitudes representing two-electron excitation [2]. Far above threshold, correlation between the continuum electrons is expected by some authors to be negligible [3]. In other formulations, scattering of the photoelectron by the remaining bound electron, called two-step-one (TS1), is a form of final-state correlation important at all energies [4]. Further, determination of the ratio of double-to-single photoionization of helium at high energy is of critical importance in clarifying the relationship of electron-electron correlation in double photoionization to that in double ionization by charged particles [4].

In a recent Letter, Samson has shown proportionality between the ratio of double-to-single photoionization for Ne, O, and N, and the cross section for single ionization of Ne^{+}, O^{+}, and N^{+} by electron impact, near threshold [5]. He argues that if direct 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. Predictions have not converged, and calculated values for the ratio, far above threshold, vary by a factor of 3 (Table I) [1,3,5–8]. Samson's semiempirical prediction for the ratio at high energy is lower than theoretical calculations and, unlike theory, predicts no asymptote.

Double photoionization of helium has been studied extensively near its threshold at 79 eV, and good agreement has been obtained between several theoretical predictions of the He^{++}/He^{+} ratio and experiment up to about 200 eV [1,2,6,7,9,10]. First, measurements obtained by Carlson [11] showed qualitatively the onset of double photoionization. Improved results employing synchrotron radiation by Schmidt *et al.* [12] and Holland *et al.* [13] revealed that He^{++} production is nearly 5% that of single photoionization at 160 eV, about twice the threshold energy. In a recent Letter, Kossmann, Schmidt, and Andersen [9] studied double photoionization with very high photon-energy resolution in the vicinity of threshold and reported quantitative information about the Wannier exponent, confirming the theoretical prediction in the energy region within about 2 eV above threshold.

Despite the divergence of theoretical results, only one measurement of He^{++}/He^{+} has been reported at photon energies above 300 eV. Carlson [11] used x-ray tubes and filters to produce a quasimonochromatic photon

TABLE I. Comparison of theoretical predictions of the He^{++}/He^{+} ratio with present measurement.

He^{++}/He^{+} (%)	Reference	Photon energy (keV)	Type of calculation
1.6 ± 0.3	Present	2.8	Experiment
0.3	24	10.0	Semiempirical
1.2	24	2.8	Semiempirical
1.7	1	Asymptotic	Sudden
1.7	3	Asymptotic	Sudden
2.3	7	Asymptotic	MBPT
3.4	8	Asymptotic	Acceleration form
3.8	6	2.8	Preliminary MBPT

beam at 625 eV and measured a ratio of $(3.5 \pm 1.2)\%$. As the photon energy is increased, however, the rapidly diminishing helium photoionization cross section (at 2.8 keV, $\sigma^+ \approx 20$ b [14], $\sigma^{++} \approx 0.3$ b) makes synchrotron radiation an ideal source. Therefore, it was the purpose of this investigation to perform a more precise measurement at high energy using a monochromatic beam of synchrotron radiation.

The present experiment was conducted on National Institute of Standards and Technology (NIST) beam line X-24A at the National Synchrotron Light Source (NSLS). Details of beam-line construction [15] and performance [16] have been provided elsewhere. Synchrotron radiation was tuned to 2.8 keV with a double-crystal monochromator employing Si(111) crystals. Horizontal and vertical collimating slits each of ≈ 1.5 mm width were mounted on linear-motion feedthroughs just upstream of a thin beryllium window located at the entrance to the experimental chamber. The 1.5-mm$\times$1.5-mm photon beam was positioned 1–2 mm away from the tip of the helium gas needle.

Photoions produced in the source region were extracted, accelerated, allowed to drift, and finally accelerated to 3.3 keV per charge with voltages applied to the spectrometer as indicated in Fig. 1. The voltages were selected to provide space focusing [17] and to minimize distortions in the extraction field due to the needle. Apertures were covered with high-transmission ($\approx 90\%$) mesh. Ions were detected by dual Galileo MCP25 chevroned microchannel-plate (MCP) detectors operated with 1 kV across each detector. The time-of-flight (TOF) spectrometer was designed to permit extraction and detection of ions as slow as Ar^+ within the 568-nsec spacing between photon bursts characteristic of NSLS single-bunch operations. 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 result when the spectrometer extraction field is pulsed [9,13]. Use of the ring timing signal also permitted TOF measurements without the need for counting electrons ionized from helium, a common technique in many TOF measurements, with attendant uncertainties concerning electron-detection efficiency [18].

Ion flight times are known [17] to scale as $(m/q)^{0.5}$. Consequently, introduction of argon into the source region permitted precise determination of the expected flight times for He^+ and He^{++}. When ionized above the K edge at ≈ 3206 eV, argon TOF spectra contain measurable amounts of Ar^+ through Ar^{7+}, thus permitting accurate prediction of flight times for other m/q. Figure 2 shows both charge states of helium against a flat background. Several experimental effects can result in a distortion of the measured ratio of double-to-single photoionization of helium. The most important effects were discussed by Schmidt, Sandner, and Kuntzemüller in connection with measurements following ionization by 2-keV electrons [19] and photons near threshold [12]. Relevant here are the following.

(i) In addition to direct photoionization in the source region, spurious helium ions can be created by ionization due to stray light or electrons. A small amount of stray low-energy light scattered through the monochromator can have a profound effect since the helium photoionization cross section just above threshold [14] is nearly 5 orders of magnitude higher than at 2.8 keV [20]. A 5-μm carbon foil located before the monochromator and a beryllium window located just upstream of the source region eliminated this contamination. The window also provided vacuum isolation from the NSLS storage ring. Low-energy secondary electrons produced at collimating slit edges or in the experimental chamber by photons interacting with, e.g., various parts of the TOF apparatus itself pose a more serious problem. These low-energy electrons produce primarily singly ionized helium whose

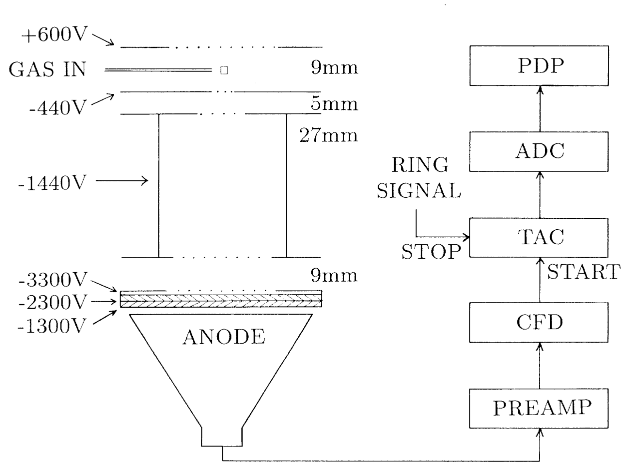

FIG. 1. Schematic of the time-of-flight apparatus and associated electronics showing approximate dimensions of the spectrometer and size of the collimated x-ray beam.

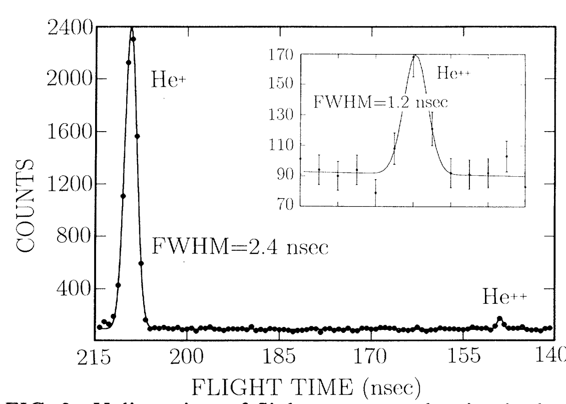

FIG. 2. Helium time-of-flight spectrum showing both charge states following photoionization by 2.8-keV x rays. Spectrum illustrated represents about one-third of the total sample reported here and required about 8400 sec.

flight times are different from He^+ formed in the interaction region by photons. With poor x-ray collimation, as many as five additional He^+ peaks appeared clustered near, and partially overlapped with, the desired photoionization peak. Without the high resolution of the TOF spectrometer, only one broader He^+ peak would appear and the ratio He^{++}/He^+ would be underestimated. This problem was eliminated by careful beam collimation and by locating the beryllium window downstream of both collimating slits. The window removed both stray low-energy scattered light and low-energy electrons from the slits. The collimated x-ray beam traversed the TOF spectrometer without interaction with any surfaces.

(ii) In a TOF spectrometer, multiply charged ions are extracted faster than singly charged ions. As a result of the near-thermal photoion energy [21], the possibility of unequal collection efficiency exists. Since the source volume, the perpendicular intersection of a 1.5-mm × 1.5-mm x-ray beam with an effusive gas source, is small compared to the 5-mm-diam extraction aperture located 4 mm away, collection efficiency should be equal for both charge states of helium for the spectrometer fields employed for this measurement. Monte Carlo simulation of the spectrometer confirms this feature [22].

(iii) When a particle strikes a channel plate, it may initiate an electron avalanche. If the outgoing pulse is sufficiently large (10^8 gain is typical for a dual chevroned MCP) the particle may be detected. The probability that an avalanche will be initiated depends on the energy, charge state, and mass of the impinging particle, with energetic, highly charged, light projectiles detected more efficiently. Helium ions were accelerated to 3.3 keV in the work reported here in order to obtain equal detection efficiency for both charge states. Earlier studies by Gao *et al.* [23] found that detection efficiencies for He^+ and He^{++} with energies greater than about 3 keV reach a plateau near 60% (the channel-plate open-area ratio) using a Varian MCP. Electronic thresholds were set much lower than the smallest pulses associated with He^+, as determined from oscilloscope examination of the pulse-height distribution.

(iv) The ratio of double-to-single photoionization of helium was determined at background pressures of 19×10^{-6}, 10×10^{-6}, and 5×10^{-6} torr, as measured by a nude ion gauge calibrated for N_2. The gas pressure in the source region near the tip of the gas needle is, of course, higher but drops off rapidly with distance. The short ion flight path of only about 45 mm is traversed in less than 215 nsec for both charge states of helium, thus minimizing the possibility of interaction with background gas. A linear fit of He^{++}/He^+ versus pressure (torr) results in a slope of -0.03 ± 0.07, consistent with no pressure dependence. Statistical uncertainties associated with fitting peak areas are large, however, due to the low cross section with which He^{++} is produced at 2.8 keV. Holland *et al.* [13] used a similar technique to measure the ratio He^{++}/He^+ near threshold. These authors found pressure effects to be less than 3% for background pressures in the range $(5-100)\times10^{-6}$ torr for a TOF apparatus with a much longer flight path (> 50 cm) and flight times (> 10 μsec) than employed in the present measurement. Data reported here were collected at a measured background pressure of 10^{-5} torr.

(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 He^{++}/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 error reported here.

The present results are compared in Table I with several predictions of the ratio of double-to-single photoionization at high energy. We obtain best agreement with early shake calculations [1,3,10] which emphasized the importance of electron correlation in the initial state. Recent preliminary MBPT calculations by Hino and Ishihara [6], including effects due to ground- and final-state correlation, shakeoff, and two-step processes in which the photoelectron interacts with the remaining electron, yield a high-energy ratio of 3.8%; improved calculations have been performed and will soon be published. Carter and Kelly [2] found final-state correlation effects to be as important as ground-state correlation in MBPT calculations at low energies but did not derive an asymptotic limit. Both predictions agree well with other calculations and measurements up to energies of 300 eV. Our results are also within 1 standard deviation of Samson's prediction, considering the errors in measured electron-ion cross sections employed in his picture and other uncertainties, estimated to be about 20% [24].

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 [4]. Measurements with beams of electrons, protons, and antiprotons reveal He^{++}/He^+ has an asymptotic value of $\approx 0.27\%$ obtained for beam energies from 10 MeV/amu [25] to 80 GeV/amu (40-MeV electrons) [26]. 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 [4,25]. Nonetheless, the ratios obtained with photons and charged particles are expected to be related [4], emphasizing the importance of establishing the high-energy He^{++}/He^+ photoionization ratio.

We have reported the first measurement ever of the ratio of double-to-single photoionization of helium above 600 eV and the first above 300 eV since 1967. The result, $1.6\%\pm0.3\%$, is much lower than expected by most predictions, and is in best agreement with the oldest calculations and the newest semiempirical predictions. This agreement may be fortuitous and a measurement of the energy dependence of the ratio will provide a much more stringent test of these calculations. We will soon conduct measurements at several photon energies and with smaller statistical errors to determine the high-energy slope of the He^{++}/He^{+} ratio. These new measurements will help settle the issue of whether the ratio has an asymptotic value, and at what energy it is reached.

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