Photoionization and Compton Double Ionization of Helium from Threshold to 20 keV

Jon C. Levin, G. Bradley Armen, and Ivan A. Sellin
Department of Physics, University of Tennessee, Knoxville, Tennessee 37996-1200
(Received 22 September 1995)

We report new measurements of He^{++}/He^{+} ratios in three energy ranges. Near threshold, where only photoionization contributes, we find a ratio systematically lower than previously published measurements. From 2 to 6 keV, where Compton ionization gradually supplants photoionization as the dominant ionization mechanism, theory and experiment are in substantial agreement. Far above threshold where Compton ionization dominates, theories diverge markedly. Comparison with present results indicates asymptotic values have been established for photoionization but not Compton ionization.

PACS numbers: 32.80.Dz, 32.80.Fb

Theoretical prediction of the energy dependence of the photon double ionization 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 interacts directly with only one electron, double photoionization proceeds only by electron-electron correlation. As a result, double photoionization of helium has long been used as a definitive testing ground for understanding correlation phenomena.

Despite the importance of the problem, and the abundance of widely varying theoretical predictions at all energies, available data are definitive only for a few eV above the double-ionization threshold ($\approx$79 eV), where the ratio of double-to-single ionization of helium has been shown to be in good agreement with Wannier theory [1]. At higher energies, theory and experiment agree only that the He^{++}/He^{+} ratio rises rapidly to a maximum (3–5)% reached at a photon energy between 150 and 300 eV, subsequently falling to divergent values of $\approx$ (0.8–1.7)% reached at photon energies somewhere above 10 keV. The plethora of recent theory Letters leading to such divergent values indicates the urgency of new high-energy measurements.

The importance of including the contribution of Compton scattering in calculating double and single ionization of helium at photon energies above the few keV was first noted in a Comment by Samson, Greene, and Bartlett [2] in 1993. Several Letters have presented theoretical predictions of the behavior of the ratio of double-to-single ionization and Compton ionization interchange. These calculations are in substantial disagreement.

The first calculation to incorporate the Compton contribution, by Andersson and Burgdörfer [3], predicted an He^{++}/He^{+} ratio gradually falling to an asymptotic value of $\approx$0.8%, approached but still not reached by 20 keV. Subsequent investigations by these authors incorporating varying amounts of final-state correlation altered only details of the predicted rate at which the asymptote is reached, but not its value [4]. A substantially different result was found by Hino, Bergstrom, and Maceu [5] whose many-body perturbation-theory calculation found an asymptote of 1.6%, noted by the authors to coincide with predicted asymptotic ratios for photoionization. Further, Hino, Bergstrom, and Maceu [5] predicted a local minimum in the ratio at $\approx$6 keV, the energy at which the photoionization and Compton ionization cross sections are equal. Extension to 20 keV by Bergstrom, Hino, and Maceu [6] revealed that the result was, in fact, not yet asymptotic; at high energy, the discrepancy with the results of Andersson and Burgdörfer is attributed to a greater amount of final-state correlation (FSC) and to the approximate manner in which both calculations incorporate FSC. Both authors expect initial-state wave functions of Andersson and Burgdörfer and final-state wave functions of Bergstrom, Hino, and Maceu to be more accurate. Suric *et al.* [7] expect the same asymptotic value as Andersson and Burgdörfer but find it is approached from below and is reached much more rapidly. The energy dependence of these theories is illustrated in Fig. 1.

Some theorists believe that despite the difference between photoionization (in which the photon is annihilated, transferring all of its energy) and Compton ionization (where only relatively little energy is transferred inelastically), the He^{++}/He^{+} ratio should be the same for both processes. In particular, Amusia and Mikhailov [8] expect an asymptotic ratio of 1.68%, in close agreement with the first asymptotic calculations for photoionization, those of Åberg [9] and of Byron and Joachain [10].

In the first several hundred eV above threshold, calculations and measurements too numerous to discuss here (Refs. [11–26]) have been presented. In general, both experimental and theoretical results fall within the broad limits indicated in Fig. 2; thus, the location and magnitude of the peak of the He^{++}/He^{+} ratio is poorly known. Recent calculations [21] relating the He^{++}/He^{+} ratio for fast charged particle impact (for which the ratio is well established to be 0.26%) to that for photon impact, utilizing Bethe-Born theory, suggest that the photoionization ratio at low energies is overestimated, perhaps by as

 0031-9007/96/76(8)/1220(4)$06.00

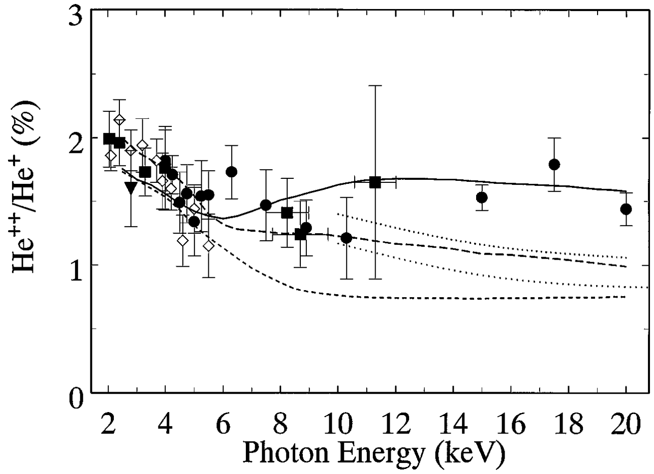

FIG. 1. Ratio of double-to-single ionization of helium in the photon energy range transitional between photoionization and Compton ionization compared with available theory. Individual symbols represent data: present results (circles), previous results (invested triangles [39], squares [40], diamonds [28]). Lines represent theory: the first calculation to incorporate Compton scattering, by Andersson and Burgdörfer [3] (long dashes) and subsequent Compton effect calculations [4] (dots) incorporating no final-state correlation (bottom) and Byron–Joachain-type configuration interaction final state (top), a recent MBPT calculation by Bergstrom, Hino, and Maceu [6] (solid line), and the results of Suric *et al.* [7] (short dashes).

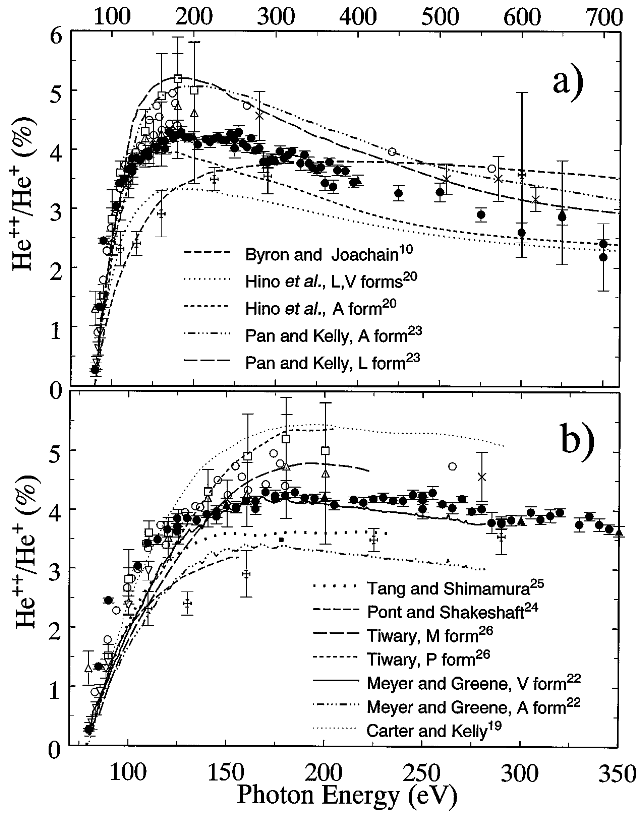

FIG. 2. Near-threshold ratio of double-to-single photoionization of helium using monochromatic undulator radiation (solid circles and solid triangles, present results) compared with available theory, as indicated. And published experimental results (open circles [11], Xs [12], crosses [13], triangles [14], diamonds [15], inverted triangles [17], squares [16]).

much as 25%, by existing measurements. Other work, in which dipole and nondipole contributions to the ratio for charged-particle ionization are related to photoionization and Compton ionization, respectively, places no such constraint on the magnitude of the ratio at low energies [18]. Further, new *R*-matrix calculations [22] and hyperspherical close-coupling results [25] have recently become available, as well as new results which circumvent the explicit inclusion of the asymptotic boundary conditions [24].

As a result of the large theoretical and experimental uncertainties, high statistics measurements are urgently required from threshold to many keV, a very broad energy range across which experimental requirements vary greatly. In this work, we report new data from threshold to 20 keV.

The present experiment was conducted on beam lines U13UA, X24A, and X25 at the National Synchrotron Light Source (NSLS). Photoions produced in the source region of a time-of-flight (TOF) analyzer were extracted, accelerated, allowed to drift, and finally accelerated to 4.0–4.7 keV per charge with voltages selected to provide space focusing [27] and to minimize distortions in the extraction field due to the needle. Spectrometer apertures were covered with high transmission ($\approx$90%) mesh. Ions were detected by dual Galileo MCP25 chevroned microchannel-plate (MCP) detectors operated with 850–1000 V across each detector. The TOF spectrometer was designed to permit extraction and detection of helium ions well within the spacing between photon bursts characteristic of NSLS single-bunch operations, i.e., 568 ns on the x-ray ring, and 170 ns on the vacuum ultraviolet (VUV) ring. Availability of a ring timing signal coincident with each electron bunch employed as a stop input to a time-to-amplitude converter resulted in better resolution and shorter flight times than are obtained when the spectrometer extraction field is pulsed [1,14]. Pulsed extraction fields also result in varying ion collection efficiencies [28]. This variation is a result of large differences in the kinetic energies of ions produced by photoionization, in which the entire energy of the photon is imparted to the atom, and ions created by Compton ionization, which recoil slowly from the low-energy Compton electron. 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-collection and detection efficiencies [29].

A TOF spectrum obtained at 20 keV illustrates the timing resolution achievable using a ring signal to mark the arrival of each several-hundred-picosecond-wide photon burst (Fig. 3). In our technique, excellent timing resolution is critical to help eliminate single ionization of he-

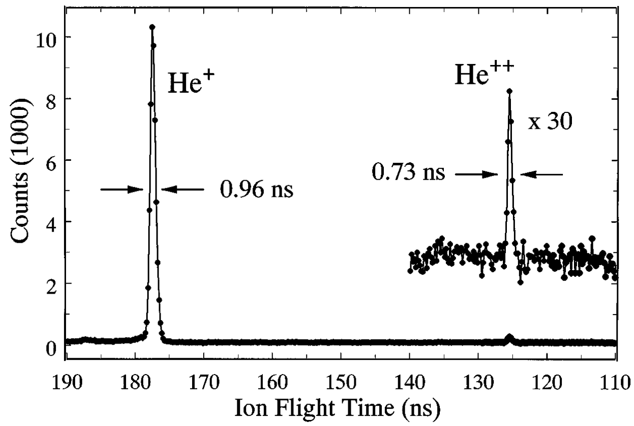

FIG. 3. Helium time-of-flight spectrum obtained with monochromatic x rays of energy 20 keV. At this energy, the double-ionization cross section is ≈15 mb.

lium by slow secondary electrons produced when photons interact with surfaces, thereby producing an artificially low measured ratio. Ion TOF peaks produced by slow electrons created at metal surfaces are displaced slightly in time, but sufficiently so to be observable with the subnanosecond resolution illustrated in Fig. 3. Since electron-impact ionization cross sections are many orders of magnitude higher than Compton ionization cross sections at high photon energies, care was taken to prevent the intense photon beam from interacting with surfaces near the spectrometer source region. The narrow, symmetric peaks and flat background of Fig. 3 demonstrate that contamination from slow electrons is negligible.

Beam contamination from low-energy photons, however, results in no such characteristic TOF peak shifts or distortions, although measured He^{++}/He^{+} rations will again by systematically lower. Eliminating such contamination is important because the photoionization cross section just above the double-ionization threshold is 5 orders of magnitude higher than at 3 keV [30]. As a result, substantial filtration, as discussed below, was employed at all energies to help assure spectral purity.

Several other experimental effects can result in a distortion of the measured ratio of double-to-single photoionization of helium. The most important were discussed by Schmidt, Sandner, and Kuntzemüller in connection with measurements following ionization by 2 keV electrons [31] and photons near threshold [15]. We find from Monte Carlo spectrometer simulation that biases resulting from collection inefficiencies are negligible [32]. Ratios of He^{++} to He^{+} studied as a function of ion kinetic energy and channel-plate voltage are found to be constant for the range of voltages employed here, consistent with earlier studies by Gao *et al.* [33]. Further, ratios reported here were obtained using very low electron discriminator thresholds (5–20 mV) and found to be independent of threshold, without statistical errors reported here. Inappropriately large thresholds result in discrimination against He^{+} and yield an artificially high He^{++}/He^{+} ratio. Finally, measured ratios are independent of target gas pressure for the range of pressures employed here, as found previously by researchers using a similar TOF apparatus [14].

Measurements from the double-ionization threshold at 79 eV up to 700 eV were obtained using focused undulator/wiggler radiation monochromatized using 300 grooves/mm or 1200 grooves/mm spherical gratings on NSLS VUV beam line U13UA. Flux was estimated to be $\approx 10^{14}$ photons/sec. A 1500 Å Al window located downstream of the monochromator and upstream of a set of 1 mm horizontal and vertical collimating slits served to provide experimental chamber isolation from both ring vacuum and filtration of low-energy scattered light. A second, movable 1567 Å Ti filter positioned downstream of the collimating slits was employed at six energies from 150 to 650 eV to assess the effect of low-energy photon contamination from slit scattering and any remaining scattered light from elsewhere in the beam line. These latter measurements (Fig. 2, triangles) resulted in higher He^{++}/He^{+} ratios at all energies (≈10% at 200 eV) and were used to derive an energy-dependent upward correction which has been applied to the remaining data (Fig. 2, circles).

Higher energy TOF spectra were obtained using monochromatized bending-magnet radiation from National Institute of Standards and Technology beam line X24A [34,35] (3–6 keV) and NSLS wiggler beam line X25 (10–20 keV) [36]. Apertures and Be windows were used to guard against contamination of the measured ratio due to low-energy photons or electrons. The results are shown in Fig. 1 and compared with several recent calculations.

Our results are in substantial agreement with those of Sagurton *et al.* [28] although consistently somewhat higher (Fig. 1). This is likely due to greater collection efficiency in their apparatus for low-energy ions produced by Compton ionization than for higher-energy photoions; predicted ratios in this energy range are lower for the Compton process than for photoionization [4].

Recently, Spielberger *et al.* [37] employed recoil-ion momentum spectroscopy to the determination of He^{++}/He^{+} for Compton ionization and separately for photoionization using broadband synchrotron radiation ($h\nu = 5.4$–9.1 keV) and found a ratio of 1.72(0.12)% for photoionization. Since our data below 4 keV are due almost entirely to photoionization, a comparison with the results of Spielberger *et al.* show that for photoionization, an asymptotic ratio has been reached, essentially by 3 keV, with a value of ≈1.7% in excellent agreement with early predictions of Åberg [9] and of Byron and Joachain [10].

At intermediate energies, 7–10 keV, our results and the Compton ionization results of Spielberger *et al.* [37], who found a ratio of 1.22(0.06)% with broadband radiation ($h\nu = 5.4-9.1$ keV), are in better agreement with the results of Andersson and Burgdörfer [3] than with Bergstrom, Hino, and Maceu [6]. At higher energies, our data suggest that the ratio is still decreasing, with an asymptotic value still not reached by 20 keV. In absolute magnitude, the many-body perturbation result of Bergstrom, Hino, and Maceu [6] is in better agreement with our measurements, although no conclusive evidence for the predicted dip is observed. The bifurcation of theoretical ratios at high $h\nu$ thus remains a partially unresolved problem, with an asymptotic value still not reached.

At lower energies (Fig. 2), we find a rather broad maximum at about 180 eV, in agreement with earlier measurements. Our measured ratio, however, is consistently lower, reaching a maximum value of $\approx$4.2%, in excellent agreement with the recent velocity form *R*-matrix calculation of Meyer and Greene [22]. Their acceleration form results, however, differ substantially, and so the good agreement with the velocity form may be fortuitous. Our results are consistently about 10% higher than recent measurements by Spielberger *et al.* [38].

This work was supported in part by the NSF. NSLS is supported by U.S. DOE under Contract No. DE-AC020-76CH00016. We appreciate the hospitality and assistance of Dr. Lonny Berman and Dr. Steven Hulbert of NSLS and the able technical support of Barry Karlin.

[1] H. Kossmann, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1266 (1988).
[2] J. A. R. Samson, C. H. Greene, and R. J. Bartlett, Phys. Rev. Lett. **71**, 201 (1993).
[3] L. R. Andersson and J. Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993).
[4] L. R. Andersson and J. Burgdörfer, Phys. Rev. A **50**, R2810 (1994).
[5] K. Hino, P. M. Bergstrom, Jr., and J. H. Macek, Phys. Rev. Lett. **72**, 1620 (1994).
[6] P. M. Bergstrom, Jr., K. Hino, and J. H. Macek, Phys. Rev. A **51**, 3044 (1995).
[7] T. Suric, K. Pisk, B. Logan, and R. Pratt, Phys. Rev. Lett. **73**, 790 (1994).
[8] M. Ya. Amusia and A. I. Mikhailov, Phys. Lett. A **199**, 209 (1995); J. Phys. B **28**, 1723 (1995).
[9] T. Åberg, Phys. Rev. A **2**, 1726 (1970).
[10] F. W. Byron and C. J. Joachain, Phys. Rev. **164**, 1 (1967).
[11] R. J. Bartlett, P. J. Walsh, Z. X. He, Y. Chung, E-M Lee, and J. A. R. Samson, Phys. Rev. A **46**, 5574 (1992).
[12] N. Berrah, F. Heiser, R. Wehlitz, J. Levin, S. B. Whitfield, J. Viefhaus, I. A. Sellin, and U. Becker, Phys. Rev. A **48**, R1733 (1993).
[13] T. A. Carlson, Phys. Rev. **156**, 5574 (1967).
[14] D. M. P. Holland, K. Codling, J. B. West, and G. V. Marr, J. Phys. B **12**, 1465 (1979).
[15] V. Schmidt, N. Sandner, H. Kuntzemüller, P. Dhez, F. Wuilleumier, and E. Källne, Phys. Rev. A **13**, 1748 (1976).
[16] G. R. Wight and M. J. Van der Wiel, J. Phys. B **9**, 1319 (1976).
[17] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[18] J. Burgdörfer, L. R. Andersson, J. H. McGuire, and T. Ishihara, Phys. Rev. A **50**, 349 (1994).
[19] S. L. Carter and H. P. Kelly, Phys. Rev. A **24**, 170 (1981).
[20] K. Hino, T. Ishihara, F. Shimizu, N. Toshima, and J. H. McGuire, Phys. Rev. A **48**, 1271 (1993).
[21] S. T. Manson and J. H. McGuire, Phys. Rev. A **51**, 400 (1995).
[22] K. W. Meyer and C. H. Greene, Phys. Rev. A **50**, R3573 (1994).
[23] C. Pan and H. P. Kelly, referenced in J. H. McGuire, N. Berrah, R. J. Bartlett, J. A. R. Samson, J. A. Tanis, C. L. Cocke, and A. S. Schlachter, J. Phys. B **28**, 913 (1995).
[24] M. Pont and R. Shakeshaft, Phys. Rev. A **51**, 494 (1995).
[25] J.-Z. Tang and Isao Shimamura, Phys. Rev. A **52**, R3413 (1995).
[26] S. N. Tiwary, J. Phys. B **15**, L323 (1982).
[27] W. C. Wiley and J. H. McClaren, Rev. Sci. Instrum. **26**, 1150 (1955).
[28] M. Sagurton, R. J. Bartlett, J. A. R. Samson, Z. X. He, and D. Morgan, Phys. Rev. A **52**, 2829 (1995).
[29] P. Lablanquie, J. H. D. Eland, I. Nenner, P. Morin, J. Delwiche, and M.-J. Hubin-Franskin, Phys. Rev. Lett. **58**, 992 (1987).
[30] W. H. McMaster, N. Kerr Del Grande, J. H. Mallett, and J. H. Hubbel, Lawrence Radiation Laboratory, University of California, Livermore, Report No. UCRL-50174 Sec. II, Rev. 1, 1969.
[31] V. Schmidt, N. Sandner, and H. Kuntzemüller, Phys. Rev. A **13**, 1743 (1976).
[32] J. C. Levin, C. S. O, and I. A. Sellin, Bull. Am Phys. Soc. **33**, 1002 (1988).
[33] P. S. Gao, P. S. Gibner, J. H. Newman, K. A. Smith, and R. F. Stebbings, Rev. Sci. Instrum. **55**, 1756 (1984).
[34] P. L. Cowan *et al.*, Nucl. Instrum. Methods Phys. Res. Sect. A **246**, 154 (1986).
[35] S. Brennan *et al.*, Rev. Sci. Instrum. **60**, 1603 (1989).
[36] L. E. Berman, J. B. Hastings, T. Oversluizen, and M. Woodle, Rev. Sci. Instrum. **63**, 428 (1992).
[37] L. Spielberger, O. Jagutzki, R. Dörner, J. Ullrich, U. Meyer, V. Mergel, M. Unverzagt, M. Damrau, T. Vogt, I. Ali, Kh. Khayyat, D. Bahr, H. G. Schmidt, R. Frahm, and H. Schmidt-Böcking, Phys. Rev. Lett. **74**, 4615 (1995).
[38] Preliminary ratios at several energies presented by Spielberger *et al.*, at ICPEAC, Whistler, Canada, 1995, are $\approx$10% below results reported here.
[39] J. C. Levin, D. W. Lindle, N. Keller, R. D. Miller, Y. Azuma, N. Berrah Mansour, H. G. Berry, and I. A. Sellin, Phys. Rev. Lett. **67**, 968 (1991).
[40] J. C. Levin, I. A. Sellin, B. M. Johnson, D. W. Lindle, R. D. Miller, N. Berrah, Y. A. Azuma, H. G. Berry, and D.-H. Lee, Phys. Rev. A **47**, R16 (1993).