PHYSICAL REVIEW A VOLUME 59, NUMBER 5 MAY 1999

Single-photon ionization of helium from 4.5 to 12 keV by Compton scattering and the photoelectric effect

D. V. Morgan and R. J. Bartlett
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 22 September 1998; revised manuscript received 11 January 1999)

We have measured the ratio of the cross sections for double-to-single ionization in helium for several monoenergetic photon energies between 4.5 and 12 keV using time-of-flight ion charge state spectroscopy. In this energy range, both the photoelectric effect and inelastic (Compton) scattering contribute significantly to the total cross section. The ionization states caused by Compton scattering were distinguished from those caused by the photoelectric effect by the different recoil energies of the helium ion associated with the two processes. The ratios of the double-to-single ionization cross sections of helium for the photoelectric effect (R_p) and for Compton scattering (R_c) are given, and compared with previous measurements and theoretical calculations. The measured value for R_c at 12 keV is $(1.21 \pm 0.27)\%$, which agrees well with the theoretical calculations of Andersson and Burgdörfer [Phys. Rev. A **50**, R2810 (1994)]. [S1050-2947(99)05905-3]

PACS number(s): 32.80.Cy, 32.80.Fb

I. INTRODUCTION

Helium is the simplest neutral atomic system in which electron-electron correlation effects can be studied. The existence of doubly ionized helium from the interaction with a single photon results directly from these correlations for both photoabsorption and inelastic (Compton) scattering. However, the final states associated with double ionization in these two processes differ considerably. For photoabsorption, there is a high probability that the photon energy is transferred almost entirely to the primary photoelectron [1], while for Compton scattering most of the initial x-ray energy is given to the scattered x ray [2], hence the ejected electrons are comparatively slow. At x-ray energies between 4.5 and 12 keV, the strongly energy-dependent photoionization cross section, σ_p, falls rapidly, whereas the Compton scattering cross section, σ_c, is a slowly increasing function of energy [3,4]. At approximately 6.3 keV, σ_p and σ_c are equal, therefore we call this energy range the ‘‘crossover’’ region. Theoretical calculations for the double-to-single ionization ratio in the crossover region, because of the intrinsic difference between Compton scattering and the photoelectric effect, are performed independently. Samson *et al.* [5] have shown that it is necessary to distinguish between these two effects to make a meaningful comparison between theory and experiment at energies where both effects contribute significantly. Here, we report measurements for the double-to-single ionization ratio using monoenergetic photons at energies below the crossover which distinguish between Compton scattering and the photoelectric effect.

II. EXPERIMENT

The experiments performed at photon energies of 4.5 and 5.5 keV were conducted at the Los Alamos National Laboratory beamline X8A at the National Synchrotron Light Source, which has been described previously [6,7]. This beamline provides monoenergetic x rays using a Si(111) monochromator with a bandpass of approximately 2 eV. The time-of-flight (TOF) spectrometer used in this experiment has been described in detail previously [6–8]. Helium atoms are injected into the ‘‘extraction region’’ by an effusive gas nozzle. The ambient helium pressure was maintained at 2.7×10^{-6} torr, following evacuation to a base pressure of $\sim 10^{-9}$ torr. The monochromatic synchrotron beam passes through the extraction region of the TOF spectrometer, in which He^+ and He^{2+} ions accumulate. A pulsed field extracts the ions and accelerates them into a field-free drift tube. The ions are then detected by a dual microchannel plate (MCP) biased at -5.46 kV. The field pulse and the signal from the MCP serve as the start and stop pulses, respectively, for the time-of-flight measurements. The TOF spectrometer axis was aligned parallel to the polarization of the x-ray beam. Because the photoionization process preferentially produces electrons along the polarization axis, the recoil velocity of the photoions either increases or decreases the time of flight relative to an ion produced with little or no recoil (Compton ionization process). Thus, the time-of-flight spectrum has three peaks for both He^+ and He^{2+}: the early and late time peaks are produced by photoions, and the central peak is produced by Compton ions.

Our pulsed-field type of TOF apparatus has collection efficiencies that depend on the speed and trajectory of the recoil ion. This is largely because the rapidly moving ions escape from the interaction region prior to application of the pulsed extraction electric field. In order to determine the collection efficiencies for both the photoelectric and Compton ionization processes, we have employed a Monte Carlo simulation of our TOF spectrometer. The recoil velocities of the different helium ion states are one input to the Monte Carlo simulation. Other inputs include the beam spot size, the pulse widths, pulse period, and amplitudes of the ion extraction electric field, and the TOF spectrometer dimensions. The collection efficiencies for both single (η_c^+) and double (η_c^{2+}) ionization from Compton scattering are much higher than the corresponding collection efficiencies for photoionization (η_p^+ and η_p^{2+}), because of the large recoil

1050-2947/99/59(5)/4075(4)/$15.00

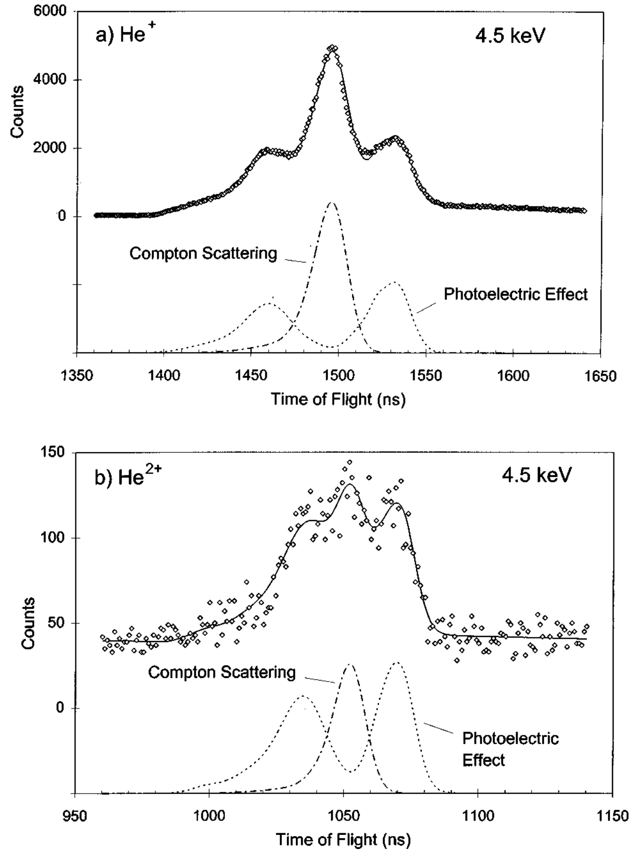

FIG. 1. TOF spectrum (◇) and least-squares fit to the data (solid line) at 4.5 keV for (a) He$^+$ and (b) He^{2+}. The Monte Carlo calculated efficiency curves for Compton scattering (dot-dashed line) and the photoelectric effect (dotted line) are also shown.

velocities associated with the photoionization process. Because of the narrower divergence of the He^{2+} ions with respect to the axis of the TOF spectrometer, η_c^{2+} is slightly higher than η_c^{+}. For our calculation of η_p^{+} and η_p^{2+}, we have assumed that the directional distribution probability of ejected photoions is proportional to $\cos^2\theta$, where θ is the angle of the photoion trajectory with respect to the TOF spectrometer axis [9].

The experiment to determine R_c at 12 keV was performed on the Los Alamos National Laboratory beamline X8C at the National Synchrotron Light Source [10], which provides focused monochromatic x rays for energies up to 20 keV, with a bandwidth of approximately 4 eV at 12 keV. The count rate for He$^+$ was obtained by subtracting the background counts from the time-of-flight spectrum, then integrating the spectrum over the single ionization flight time. In this energy range, the Compton cross section is larger than the photoionization cross section, and the TOF spectrometer collection efficiency, as determined by the Monte Carlo simulation, is much larger for the Compton scattering process than for the photoionization process. Although both processes contribute to the He$^+$ signal, we estimate that the photoabsorption component of the experimental spectrum is only about 1%. The corresponding total collection efficiency for the TOF spectrometer for He^{2+} Compton ions was virtually identical to the collection efficiency for He$^+$ ions.

TABLE I. Comparison of photoelectric and Compton scattering cross sections in the crossover region.

	σ_c^+/σ_p^+		σ_I/σ_p
Photon energy (keV)	This work	Hino *et al.*[a]	Viegele[b]
4.0		0.14	0.15
4.5	0.21		0.24[c]
5.0			0.38
5.5	0.43		0.56[c]
6.0		0.81	0.83
12.0		10.94	10.83

[a] Reference [11].
[b] Reference [3]; σ_I is the incoherent scattering cross section.
[c] Reference [3], determined by cubic interpolation.

TABLE II. Values for R_c and R_p obtained in this work.

Photon energy (keV)	R_c (%)	R_p (%)
4.5	1.07 ± 0.49	2.00 ± 0.24
5.5	0.72 ± 0.29	2.04 ± 0.37
12.0	1.21 ± 0.27	

III. RESULTS

Time-of-flight spectra for He^{2+} and He$^+$ ions are shown in Fig. 1 for 4.5 keV photons. We observe good agreement between the overall shape of the efficiency curves calculated by the Monte Carlo program and the singly ionized helium TOF spectrum. The experimental values for σ_c^+/σ_p^+ were obtained by a least-squares fit of the amplitude and time-shift parameters of the calculated efficiency curves to the singly ionized peaks in the TOF data. To our knowledge, these are the only experimental values for σ_c^+/σ_p^+ in the crossover region obtained with monoenergetic x rays. The experimen-

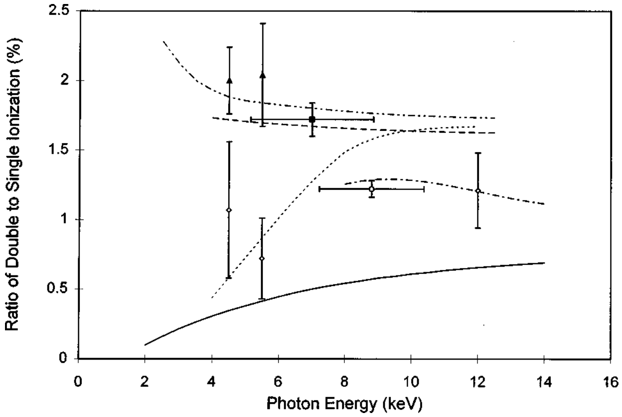

FIG. 2. Ratio of double-to-single helium ionization from Compton scattering (R_c) and from the photoelectric effect (R_p). Present experimental results for R_c (◇) and R_p (▲), $R_c=(1.22\pm0.06)\%$ at $8.8^{+1.5}_{-1.65}$ keV (○), and $R_p=(1.72\pm0.12)\%$ at $7.0^{+2.1}_{-1.6}$ keV (■), obtained by Spielberger *et al.* [9]. Also shown are the calculations of R_c by Suric *et al.* [14] (solid line), Andersson and Burgdörfer [13] (dot-dashed line), and Hino *et al.* [11] (dotted line), as well as the calculations of R_p by Andersson and Burgdörfer [20] (two dot-dashed line) and Hino *et al.* [1] (dashed line).

TABLE III. Values for R_c at 12.0 keV and at the high-energy limit.

	R_c (%)							
	Experiment			Theory				
$\hbar\omega$ (keV)	This work	Spielberger *et al.*[a]	Wehlitz *et al.*[b]	Andersson *et al.*[c]	Suric *et al.*[d]	Hino *et al.*[e]	Kornberg *et al.*[f]	Amusia and Mikhailov[g]
12.0	1.21 ± 0.27			1.2	0.66	1.7		
$\hbar\omega\rightarrow\infty$		$0.84^{+0.08}_{-0.11}$	1.25 ± 0.30	0.84	0.8	1.6	0.8	1.68

[a] Reference [17], measured at $\hbar\omega = 58$ keV.
[b] Reference [18], measured at $\hbar\omega = 57$ keV.
[c] Reference [13].
[d] Reference [14].
[e] Reference [11].
[f] Reference [15].
[g] Reference [16].

tal values for σ_c^+/σ_p^+ at 4.5 and 5.5 keV are compared with the ratio of the incoherent scattering cross section to the photoelectric cross section of Viegele [3] and with the values for σ_c^+/σ_p^+ given by Hino *et al.* [11] in Table I. Our results fall somewhat below the values ascribed by Viegele [3], however this may be caused in part by the inclusion of nonionizing inelastic (Raman) scattering events in the attenuation coefficient calculations which do not contribute to the Compton ionization process [12]. We do not believe significant systematic errors caused by our TOF spectrometer efficiency calculations exist in our measurements of R_c and R_p because both η_p^{2+}/η_p^+ and η_c^{2+}/η_c^+ are close to unity. The very low number of He^{2+} ions produced by Compton scattering observed at 4.5 keV and the reduced peak separation caused by the small photoion recoil velocity at this energy make the uncertainty in our measurement of R_c at 4.5 keV rather large. Our experimental values for R_c and R_p and their estimated uncertainties at 4.5, 5.5, and 12.0 keV are given in Table II. The values for R_c and R_p obtained in this experiment are compared with experimental and theoretical results for photon energies in the crossover region in Fig. 2.

IV. DISCUSSION

Several calculations of the double-to-single ionization ratio for Compton scattering, $R_c = \sigma_c^{2+}/\sigma_c^+$, have been performed from 4 keV to the high-energy (nonrelativistic) limit [2,11,13–16]. These results for 12 keV and the high-energy limit are summarized in Table III. Measurements of R_c by Spielberger *et al.* [17] at 58 keV give a value of $R_c = (0.84^{+0.08}_{-0.11})\%$, which favors the lower calculated values in Table III, and, in particular, those of Andersson and Burgdörfer [13]. Wehlitz *et al.* [18] have measured a higher value, $R_c = (1.25\pm0.30)\%$ at 57 keV, but with a relatively large error bar. This measurement does not favor any of the calculations in Table III. Our value of $(1.21\pm0.27)\%$ at 12 keV, even with the large error bar, agrees only with the calculations of Andersson and Burgdörfer [13]. At 4.5 and 5.5 keV our results for R_c are consistent with the calculations of Hino *et al.* [11], but somewhat higher than the values calculated by Suric *et al.* [14].

For the photoelectric case, theoretical studies [1,19,20] generally agree, with the exception of Drukarev [21], that the double-to-single ratio for the photoelectric effect, $R_p = \sigma_p^{2+}/\sigma_p^+$, decreases asymptotically to a value of approximately 1.67 in the high-energy limit. Recently, Spielberger *et al.* [9] have obtained values for R_c and R_p in the crossover region using cold target ion momentum spectroscopy (COLTRIMS), which is quite similar to the method used here and described above. Using broadband synchrotron radiation, these authors have separated the components of the photoelectric effect from Compton scattering to obtain values for both R_c and R_p. They have measured $R_c = (1.22\pm0.06)\%$ at a mean energy of 8.8 keV and $R_p = (1.72\pm0.12)\%$ at a mean energy of 7.0 keV for photoabsorption. This is the highest energy for which an experimental value for R_p has been obtained. Our monoenergetic values for R_p at 4.5 and 5.5 keV, shown in Fig. 2, are consistent with both the theoretical calculations and the experimental work of Spielberger *et al.* [16].

V. CONCLUSION

This paper reports monoenergic x-ray measurements of R_c and R_p as well as experimental values for σ_c^+/σ_p^+ for helium in the crossover region. In addition, our measurement of R_c at 12.0 keV and that of Spielberger *et al.* [17] at 58 keV are in agreement with the calculations of Andersson and Burgdörfer [13], and together favor this calculation over all others reported.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy through Los Alamos National Laboratory. We would like to thank M. Sagurton for his contributions to this work.

[1] K. Hino, T. Ishihara, F. Shizumi, N. Toshima, and J. H. McGuire, Phys. Rev. A **48**, 1271 (1993).
[2] P. M. Bergstrom, K. Hino, and J. H. Macek, Phys. Rev. A **51**, 3044 (1995).
[3] W. T. Viegele, At. Data **5**, 51 (1973).
[4] J. A. R. Samson, Phys. Rev. Lett. **72**, 3329 (1994).
[5] J. A. R. Samson, C. H. Greene, and R. J. Bartlett, Phys. Rev. Lett. **71**, 201 (1993).
[6] M. Sagurton, R. J. Bartlett, J. A. R. Samson, Z. X. He, and D. Morgan, Phys. Rev. A **52**, 2829 (1995).
[7] D. V. Morgan, R. J. Barlett, and M. Sagurton, Phys. Rev. A **51**, 2939 (1995).
[8] D. V. Morgan, M. Sagurton, and R. J. Bartlett, Phys. Rev. A **55**, 1113 (1997).
[9] L. Spielberger, O. Jagutzki, B. Krässig, U. Meyer, Kh. Khayyat, V. Mergel, Th. Tschentscher, Th. Buslaps, H. Bräuning, R. Dörner, T. Vogt, M Achler, J. Ullrich, D. S. Gemmell, and H. Schmidt-Böcking, Phys. Rev. Lett. **74**, 4615 (1995).
[10] R. W. Alkire, M. Sagurton, F. D. Michaud, W. J. Trela, R. J. Bartlett, and R. Rothe, Nucl. Instrum. Methods Phys. Res. A **352**, 535 (1995).
[11] K. Hino, P. M. Bergstrom, and J. H. Macek, Phys. Rev. Lett. **72**, 1620 (1994).
[12] A. Sommerfeld, Phys. Rev. **50**, 38 (1936).
[13] L. Andersson and J. Burgdörfer, Phys. Rev. A **50**, R2810 (1994).
[14] T. Suric and K. Pisk, Phys. Rev. Lett. **73**, 790 (1994).
[15] M. A. Kornberg and J. E. Miraglia, Phys. Rev. A **53**, R3709 (1996).
[16] M. Ya. Amusia and A. I. Mikhailov, Phys. Rev. Lett. **71**, 50 (1993).
[17] L. Spielberger, O. Jagutzki, B. Krässig, U. Meyer, Kh. Khayyat, V. Mergel, Th. Tschentscher, Th. Buslaps, H. Bräuning, R Dörner, T. Vogt, M. Achler, J. Ullrich, D. S. Gemmell, and H. Schmidt-Böcking, Phys. Rev. Lett. **76**, 4685 (1996).
[18] R. Wehlitz, R. Hentges, G. Prümper, A. Farhat, T. Buslaps, N. Berrah, J. C. Levin, I. A. Sellin, and U. Becker, Phys. Rev. A **53**, R3720 (1996).
[19] A. Dalgarno and H. R. Sadeghpour, Phys. Rev. A **46**, R3591 (1992).
[20] L. R. Andersson and J. Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993).
[21] E. G. Drukarev, Phys. Rev. A **51**, R2684 (1995).