STUDIES OF PHOTOABSORPTION AND COMPTON SCATTERING USING COLD TARGET RECOIL ION MOMENTUM SPECTROSCOPY

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We present experimental values of $R = \sigma^{++}/\sigma^{+}$ for photoabsorption at photon energies ranging from 90 eV to 400 eV and for Compton scattering at an energy of 58 keV. The photoabsorption values are significantly lower than previously published ones. The technique of Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS), that was used in these experiments, permits a detailed control of the experimental conditions either in measuring the recoil ion momentum distribution (*momentum mode*) or in imaging the target volume (*monitor mode*).

INTRODUCTION

The investigation of dynamical electron–electron correlation effects in multi-electron systems is one of the central goals of nowadays atomic physics. Most of these studies concentrate on the Helium atom, the simplest multi-electron system. Kinematical complete experiments on the double ionization, as (γ,2e) after photoabsorption [1, 2, 3] or (e,3e) after electron impact ionization [4, 5] yield the most detailed information on the dynamics of the ionization process and the initial correlations in the bound target atom. In the case of classical electron spectroscopy, these experiments suffer from a small coincidence solid angle in the range of $10^{-4} - 10^{-6}$ of 4π. Therefore, this type of experiments is extremely time consuming. For the Helium atom, they could only be realized for some selected arrangements of outgoing electron energies and angles in the case of photoabsorption at low photon energies [1, 6, 7]. In the case of electron impact ionization they could still not be performed [8]. The situation is getting even worse with decreasing cross sections at high photon energies or large momentum transfers for charged particle impact, respectively, where the results become to be mostly sensitive on correlation effects in the bound target atom.

Therefore, the investigation of the ratio of total cross sections for double to single ionization, $R = \sigma^{++}/\sigma^{+}$, which was historically the first quantity to be investigated in the field of correlation effects, is still essential. Here, much more efficient ion collection techniques can be used. In comparison with theoretical calculations, R probes the understanding of the dynamics in many body systems. Of special importance is R at high photon energies. It is reported to reach a constant asymptotic high energy value, R^{∞}, which is mostly probing the internal electron–electron correlations of the Helium atom [9].

At low photon energies from double ionization threshold up to several hundred eV the absorption of the photon is the only process leading to ionization. Several experimental investigations in this energy range culminated in recommended experimental values of R_{ph} [10, 11]. The recent advent of modern synchrotron radiation machines made higher photon energies above some keV accessible. In this regime the second ionization process, Compton scattering of the photon, becomes important. The cross sections for single ionization from photoabsorption and Compton scattering of Helium are equal at about 6 keV [12], Compton scattering dominates at higher energies. In this regime the double ionization cross section is in the range of 10^{-26} cm^2 and the available photon flux is decreasing. Therefore, only few experimental data points up to 20 keV were existing [11]. A constant asymptotic value, R^∞, is expected to exist for both processes.

In this paper we present experimental values of the ratio of double to single ionization of Helium obtained with the method of Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) [13] in the low energy regime from 90 eV to 400 eV for photoabsorption, R_{ph} [14], and at the highest energy reported so far, at 58 keV for Compton scattering, R_{C} [15]. Due to the intrinsic differences in momentum space between both processes COLTRIMS allowed the first experimental separation of photoabsorption and Compton scattering at about 8 keV [16]. Here the predicted asymptotic value for photoabsorption, $R^\infty_{\text{ph}} = 1.67\%$ could be confirmed. In this paper, we focus on the strongly increased experimental sensitivity of COLTRIMS with respect to classical time-of-flight techniques either in measuring the recoil ion momentum distribution for photoabsorption (*momentum mode*) or in a 3-dimensional monitoring of the target volume for Compton scattering (*monitor mode*). These techniques can also be applied to the investigation of charged paticle impact ionization.

RESULTS

Our data are shown in figure 1. The low energy pho-

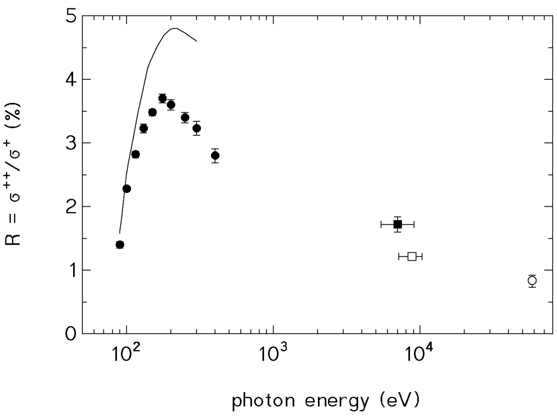

FIGURE 1. Ratios of double to single ionization for photoabsorption (full symbols) and Compton scattering (open symbols). The previously published data for photoabsorption are represented by the recommended values of [10] (full curve). Our results: full circles: [14], open circle: [15], squares: [16].

toabsorption values are significantly below the previously published results, which are represented by the recommended values from [10]. Our photoabsorption data show a close agreement with the calculations by Tang and Shimamura [17] and by Pont and Shakeshaft [18]. Our high energy Compton scattering result is also lower than the value published by Wehlitz *et al.* of $R_{\text{C}} = (1.25 \pm 0.3)\%$, however their significantly larger error bar almost overlaps with or data point.

It is obvious from the deviation of the COLTRIMS data from the earlier work as well as from the large scatter between different experiments combined in the recommended values, that it is most cruical for such experiments to exclude systematical errors. COLTRIMS offers a detailed control of the experimental parameters and allows to check for all the different sources of systematical errors in a measurement of the ratio of total cross sections R that have been discussed in the literature so far. We first list those possible problems before we discuss how COLTRIMS allows to control them.

1. Contributions of lower energetic stray light resulting in double ionization with a different value of R. Especially at photon energies in the keV regime with the ionization cross section for photoabsorption in the $E_\gamma^{-3.5}$ scaling being by orders of magnitude smaller than at the threshold [12], even small impurities can cause large systematical errors on R. Photons with an energy below the double ionization threshold would lead to a reduction of the apparent R.

2. Ionization by low energetic secondary electrons produced by photons impinging on solids in the experimental setup. The effect on R is similar to the one due to point 1).

3. A different detection efficiency of doubly and singly charged Helium ions on the ion detector, most probable increasing the apparent R.

4. Secondary collisions of the recoiling ions with residual gas resulting in charge exchange, decreasing the apparent R.

5. A different detection solid angle for different charge states, which would result in a higher apparent R.

Due to the inherent difference between the outgoing recoil ion momentum distributions for photoabsorption and Compton scattering [16], the respective techniques yielding this experimental control within COLTRIMS are different. In the following, they shall be discussed separately.

Photoabsorption: The spectrometer setup, which is shown in figure 2, consists of the following basic elements: The well localized reaction volume that is created by the overlap region of the photon beam and a supersonic Helium gas jet, and a precisely controlled electrostatic field that extracts all ions onto a position sensitive detector. If the extraction field is "weak" (in the order of several V/cm) the detected position and time-of-flight are broadened due to the recoil ion momentum. The respective deviations from the mean values give the information on the ion momentum vector (*momentum mode*).

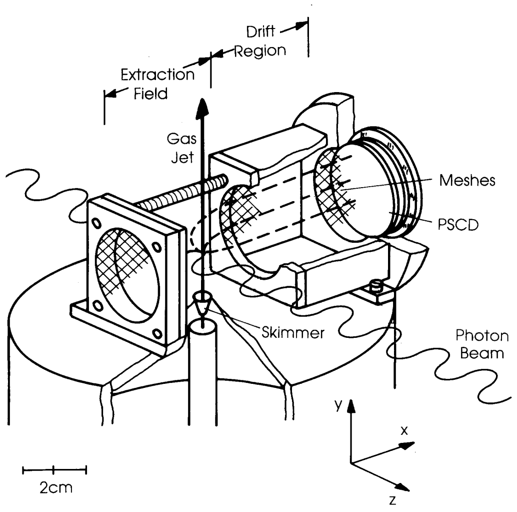

FIGURE 2. Sketch of the recoil ion momentum spectrometer.

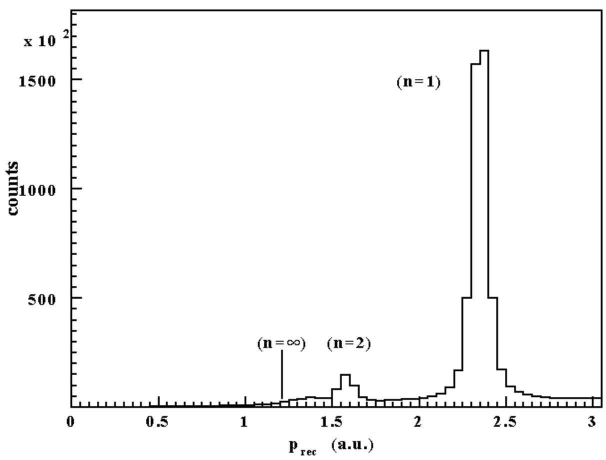

FIGURE 3. Momentum distribution of He^+ ions produced by 100 eV photons, integrated over all emission angles. No background was subtracted. (from [14])

For single ionization induced by the absorption of low energy photons, the outgoing recoil ion momentum vector mirrors the one of the photoelectron:

$$\vec{p}_{\mathrm{He}^+} = -\vec{p}_{\mathrm{e}^-} \, , \tag{1}$$

as the photon momentum k can be neglected and the incoming momentum of the Helium atom is small due to low internal temperature of the supersonic gas jet. With the ionization potential of Helium from the ground state without excitation of the remaining electron of 24.6 eV one obtains $|\vec{p}_{\mathrm{He}^+}| = |\vec{p}_{\mathrm{e}^-}| = 2.35$ a.u. for $E_\gamma = 100$ eV and 24.2 a.u. for $E_\gamma = 8$ keV. As $|\vec{p}_{\mathrm{He}^+}|$ is measured, the energy of the photon, that was absorbed in the ionization process is measured for each ionization event. The He^+ momentum distribution from absorption of 100 eV photons is shown in figure 3. Besides the groundstate ionization, ionization with excitation of the remainig electron can clearly be seen. No absorption of contaminating lower energetic photons *within the photon beam* (see point 1) listed above) is observed.

The data of figure 3 are integrated over all recoil ion emission angles. They show a dipolar angular distribution [16, 3, 14]. Ions from ionization induced by stray light or secondary particles would not be restricted to the well localized target volume and would be visible as background in figure 3 or in the dipole emission pattern. No such contamination, according to the above listed points 1) and 2) can be seen in both distributions.

For each ionization event, the detector pulse height was additionally recorded which therefore can be checked during the experiment and in the off-line data analysis. The effect of any threshold to discriminate against noise can thus be controlled in the off-line data analysis. The pulse height distribution was found to be identical for both Helium charge states (see figure 2 in [14]). Therefore, a difference in the detection efficiency according to point 3) can be excluded within the given error bars.

Charge exchange in secondary collisions of the recoiling ions with the residual gas (point 4.) is strongly unlikely due to the well localized gas jet target with a background pressure of a few 10^{-7} mbar. In addition such collisions would strongly change the measured momenta of the ions and thus be visible in figure 3.

In the *momentum mode* the ion extraction field is strong enough to project all ions onto the detector and both helium charge states have the same detection solid angle. This excludes errors according point 5).

Compton scattering can be regarded as a scattering of the photon from an atomic electron with the nucleus remaining as a spectator. The outgoing recoil ion momentum is therefore close to the one of the atomic nucleus in the atom, which is the compensated bound electron momentum distribution [16, 15] with the FWHM of about 1 a.u.. If the details of this momentum distribution are not of interest, the ion extraction voltage can be increased until the momentum-broadening does not affect the recoil ion TOF and position and both quantities reduce to an image of the localized target volume in three dimensions (*monitor mode*). With the help of software windows in the off-line analysis the correct target size and TOF window can be selected and one has the ability to check for sources of background ions. This is demonstrated in figure 4. The background that was strongly suppressed in the "gated" spectrum shown in figure 4 was detector dark counts, that are randomly distributed in time and position. Due to the limited photon flux at the photon energy of 58 keV their rate was roughly a factor 30 higher than the ion rate. No ionic background arising from other other sources as the target volume, according to the above point

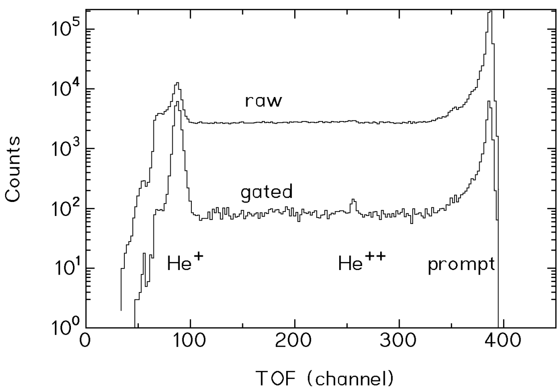

FIGURE 4. Ion TOF spectrum for Compton scattering at 58 keV. The spectrum on top is the experimental raw spectrum, the one labeled with "gated" was obtained in the off-line analysis. Here, only events with a position in the target area are selected.

1) and 2) could be found.

The remaining sources of possible error are avoided or can be checked like in the photoabsorption case.

DISCUSSION

With the above described capability of controlling the experimental conditions COLTRIMS provides a much more sensitive control of possible sources of systematic errors than all previous experiments in this field, that measured only the time-of-flight of ions produced in an extended effusive gas target. A recent consistency check of the data on $R_{\rm ph}$ for photoabsorption with the well established ratio for charged particle impact, $R_{\rm cp}$, strongly supports this claim [3]. Furthermore, our values of $R_{\rm ph}$ in the low energy regime are in excellent agreement with the most recent calculations by Tang and Shimamura [17] and by Pont and Shakeshaft [18]. Our low energy data are also in good agreement with new results by Stohlte and Samson [19] unpublished so far. A further set of data which is in between the recommended data and the new COLTRIMS values has been published by Levin *et al.* [11]. These data have been taken by detecting the ion charge state only. The reason for the disagreement is not obvious to us.

Our value of $R_{\rm C} = (0.84^{+.08}_{-.11})\%$ at 58 keV for Compton scattering is in agreement with the asymptotic high energy ratio of 0.8% [20, 21, 22] but is in contrast with another prediction of $R_{\rm C}^{\infty} = 1.67\%$ being the same value as the asymptotic value for photoabsorption [23].

Beyond its powerful abilities in determing R, COLTRIMS in the *momentum mode* is well suited for measuring highly differential cross sections. Completed by a position sensitive electron detector the above described spectrometer based on the ion collecting technique represents an extremely efficient tool for coincidence measurements [24, 25, 3]. Recently we have obtained fully differential cross sections for all angles and energies of the photoelectrons close to threshold [3].

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