Double-to-Single Target Ionization Ratio for Electron Capture in Fast *p*-He Collisions

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We have used the ion storage ring CRYRING and its internal gas-jet target and recoil-ion-momentum spectrometer to measure absolute cross sections for transfer ionization (TI: $p + \mathrm{He} \rightarrow \mathrm{H}^0 + \mathrm{He}^{2+} + e^-$) in 2.5–4.5 MeV p-He collisions with separate Thomas (TTI) and kinematic (KTI) TI contributions. The probability for electron emission in kinematical capture decreases with increasing velocity and appears to approach the photoionization shakeoff value (1.63%) [T. Åberg, Phys. Rev. A **2**, 1726 (1970)]. The velocity dependence of the TTI cross section is consistent with the theoretically predicted v^{-11} scaling [J. S. Briggs and K. Taulbjerg, J. Phys. B **12**, 2565 (1979)].

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The interaction between the two electrons in photoionization of helium leads to double ionization with a probability converging to 1.63% in the high photon energy limit [1–4]. An intuitive picture of this process, which is often used, is that the abrupt change in the screening of the nuclear charge as one electron is removed may lead to emission of the other electron into the He^{2+} continuum. However, it is necessary to include electron correlation beyond the central field approximation in the initial state to obtain quantitative agreement with experiments as shown already in 1967 by Byron and Joachain [5]. The high-photon-energy limit was confirmed experimentally by Spielberger *et al.* [4] who measured the double-to-single photoionization ratio using recoil-ion-momentum spectroscopy (RIMS) to isolate the photoelectric effect from Compton scattering, for which a lower asymptotic ratio of about 1% is found [6–8].

Ionization by charged-particle impact gives much lower probabilities for two-electron emission of ~0.25% in the high-velocity limit [9,10]. This is partly explained by the dominance of large impact parameters giving "first" emitted electrons with wide velocity distributions including important contributions from slower electrons [11]. DeHaven *et al.* [12] considered the special case of ionization with high momentum transfer to the first electron in a projectile angular differential measurement and found an enhanced probability of two-electron emission of about 1.2% compared to the angular integral value of 0.25%. This shows that the velocity of the first emitted electron influences the ratio strongly. As in the case of Compton scattering, the higher value (1.2%) is smaller than the photoabsorption shakeoff value of 1.63% as may be explained by the smaller momentum carried by the photon. Higher momentum components in the initial-state wave function are thus more important in photoabsorption than in ionization by charged-particle impact.

The probability for a proton to *capture* an electron released from the target is extremely small at projectile velocities $v \gg v_0$ (where v_0 is the Bohr velocity) [13]. These rare capture events are, however, similar to photoionization since very little momentum is transferred to the initially removed electron, which in both cases leaves the target with a velocity determined by the projectile energy. Thus, double-ionization of He following kinematical capture and photoionization alike rely on the (very small) momentum overlap between the final and initial-state wave functions.

Shah and Gilbody [14] measured the cross sections for transfer ionization (TI), $p + \mathrm{He} \rightarrow \mathrm{H}^0 + \mathrm{He}^{2+} + e^-$, and single-electron capture (SC), $p + \mathrm{He} \rightarrow \mathrm{H}^0 + \mathrm{He}^+$ in proton-He collisions in the $1.8v_0 < v < 4.5v_0$ velocity range and found an almost constant double-ionization probability of 2.8%. Mergel *et al.* [15] used the RIMS technique to separate the kinematical transfer ionization (KTI) process from the Thomas p-e-e transfer ionization (TTI), in which a fast electron from a proton-electron binary encounter interacts with the other electron in such a way that one is transferred to the projectile and the other one is emitted [16–19]. The two TI processes were separated by their different longitudinal recoil-ion momenta of (in atomic units) $p_{R\parallel}(\mathrm{KTI}) \simeq -v/2$ and $p_{R\parallel}(\mathrm{TTI}) \simeq 0$: The TTI process was then found to contribute with

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10–25% to $\sigma_{\mathrm{TI}} = \sigma_{\mathrm{KTI}} + \sigma_{\mathrm{TTI}}$ for $4.5v_0 < v < 7.5v_0$ [15]. From their data, we infer that the $\sigma_{\mathrm{KTI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{KTI}})$ ratio, which is the probability for double target ionization in a kinematical capture process, is increasing strongly with v for $v = 4.5$–$7.5v_0$.

In this study, we have increased the experimental sensitivity level by 3 orders of magnitude over the previously best experiment [15] using the intense beam of the ion storage and cooler ring CRYRING with its precooled internal He gas-jet target and a novel switching RIMS technique. We were thus able to measure the $\sigma_{\mathrm{KTI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{KTI}})$ ratio and the absolute σ_{KTI} and σ_{TTI} cross sections in the earlier inaccessibly high-velocity range 10.0–$13.4v_0$. In contrast to the result of Mergel *et al.* below $7.5v_0$ [15], we report a *decrease* with increasing v for the $\sigma_{\mathrm{KTI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{KTI}})$ ratio for $v =$ 10.0–$13.4v_0$, demonstrating for the first time the approach of the asymptotic region for p-He transfer ionization processes. We further expose the close connection to photoionization processes and show that the present results agree with experimental data [4,20–23] and theory [2,24] on the double ionization in *photoabsorption* as functions of the velocity of the first emitted electron. We find the velocity scaling for σ_{TTI} above $10v_0$ to be consistent with v^{-11}, which differs significantly from the $v^{-7.4}$ behavior below $7.5v_0$ [15].

The experiment is performed in the ion storage ring CRYRING [25] at the Manne Siegbahn Laboratory in Stockholm. Protons are injected into the storage ring, accumulated, accelerated, and electron cooled [26] at 2.5, 3.5, and 4.5 MeV yielding currents of 20–60 μA. This intense ion beam intersects the gas-jet target [27] ($\phi = 1.0$ mm and density $\sim 10^{11}$ cm^{-3}) yielding luminosities of (2–5) $\times 10^{24}$ cm^{-2} s^{-1} [27] at a background pressure in the 10^{-12} mbar range. Neutral atoms formed in electron transfer processes leave the ring and are detected by a position-sensitive microchannel-plate detector, which starts a multihit time-to-digital converter (TDC). A homogeneous dc extraction field of 11.5 V/cm accelerates the recoil ions towards a second position-sensitive microchannel-plate detector. A fast signal from this detector serves as a stop for the TDC, which stores the time-of-flight information yielding the recoil-ion charge-state distribution. The longitudinal recoil-ion momenta are deduced from the positions along the beam on the recoil detector (cf. Fig. 1).

The ratio between the cross sections for TI and single ionization (SI) is extremely low. At our highest velocity ($13.4v_0$) the SI cross section is about 6×10^{-18} cm^2 [28], whereas we find a TI cross section of $(3.7 \pm 0.7) \times 10^{-26}$ cm^2. This suggests a TI rate of ~ 1 min^{-1} and a SI rate of $\sim 10^7$ s^{-1}. It is, thus, essential to introduce a charge selection prior to detection, which prevents random SI ions from reaching the detector at the same time as He^{2+} from TI. This is achieved by switching off the voltage on a deflector in the spectrometer only after a neutral hydrogen atom formed in a SC or TI collision has

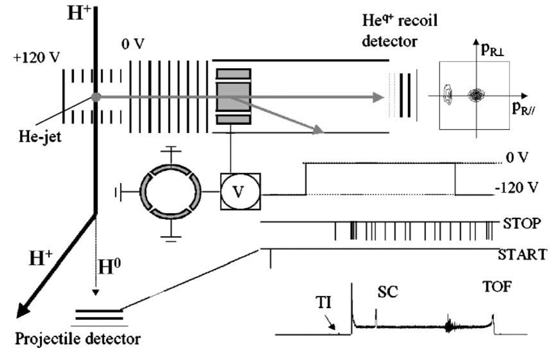

FIG. 1. The experimental setup as described in the text.

triggered the projectile detector. The delay is chosen to let the He^{2+} recoils from TI events through without deflection (normally the deflector is set to -120 V and no recoils reach the detector). At the time of switching, there are typically 10–20 He^{+} ions (randoms) on their way towards the deflector. They are, however, slower than the He^{2+} ion (from TI) and will thus be detected at a later time. This can be seen in Fig. 2 where the very small TI peak sits on a small background due to random events from double ionization. After about 0.5 μs, there is a sharp rise in the random level due to the arrival of the first He^{+} from single ionization. The increase in the random level is by a factor ~ 400 as given by the ratio of single to double ionization in fast charged-particle impact [9]. On top of the SI random level, there is a peak due to single-electron capture (SC). The relative detection efficiency for He^{+} and He^{2+} ions was obtained through normalization at 0.3 MeV to the $\sigma_{\mathrm{TI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{TI}})$ ratio by Mergel *et al.* [15] yielding $(83.3 \pm 6.5)\%$. The ratios $\sigma_{\mathrm{TI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{TI}})$ were then extracted from the time-of-flight spectra for measurements at 2.5, 3.5, and

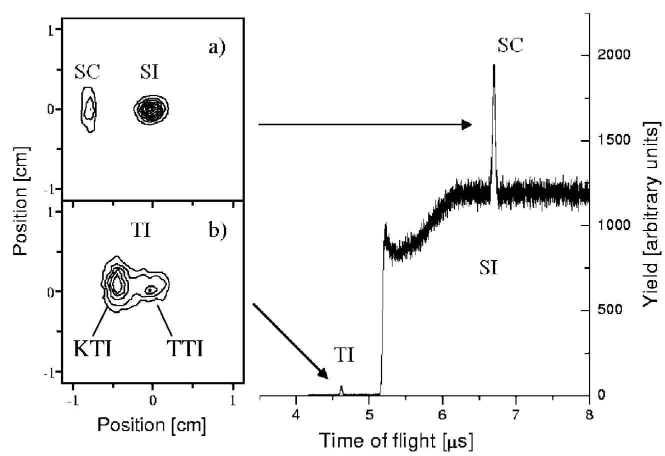

FIG. 2. The time-of-flight spectrum recorded in the time-switched mode with 2.5 MeV protons. At the time for the TI peak there is only a weak random signal from DI, whereas the SC peak rides on the large SI random level. Inset (a) shows a contour plot of the recoil positions recorded within a narrow time interval around the SC peak. Inset (b) shows the recoil positions for the TI peak.

4.5 MeV. The absolute cross section scale was determined through the total electron capture cross sections by Schwab *et al.* [29]. We separate the KTI and the Thomas p–e–e TI mechanism through their different recoil-ion-momenta along the projectile axis as deduced from the position distribution of the TI recoil ions. Figure 2(a) shows the density of hits on the recoil-ion detector for a narrow time window around the SC peak. The maximum to the left is due to true SC events, whereas the wider peak is due to SI randoms. Figure 2(b) shows the recoil ions from TI. The structure to the left stems from the KTI process, whereas the distribution closer to the detector's center is mainly the Thomas p–e–e scattering process with a small contribution from DI randoms. The latter part was isolated in a separate measurement in which a random pulse generator started the TDC yielding a time-of-flight spectrum without real coincidences.

In Fig. 3, we show the ratios $\sigma_{TI}/(\sigma_{SC} + \sigma_{TI})$ and $\sigma_{KTI}/(\sigma_{SC} + \sigma_{KTI})$ as functions of the proton velocity together with earlier results of Shah and Gilbody [14] and Mergel *et al.* [15]. Our $v > 10v_0$ data show for the first time a *decrease* with increasing velocities for both ratios in sharp contrast to the trends for $v < 10v_0$ reported by the earlier highest energy investigation [15]. The probability for emission of the second electron following electron transfer appears to approach the shakeoff limit 1.63% [1,2] as v increases.

In Fig. 4, we show the probabilities for emission of the second electron as functions of the velocities with which the first electron leaves the target for photoionization and electron capture. In the latter case, this velocity is simply the projectile velocity. The double-ionization threshold energy is subtracted from the photon energy to get the photoelectron kinetic energy in the photoionization data, which range from 400 eV to 4.0 keV (the upper limit chosen to limit the influence of Compton scattering [2,23]), except for the single data point of Spielberger *et al.* [4], where the Compton scattering contribution was isolated by means of RIMS. The dashed curve is a very recent result of Shi and Lin [24], who have calculated the shakeoff probability as a function of the velocity of the first outgoing electron without any reference to the nature of the projectile. Their results agree with our experimental data.

An important difference between photo-double ionization (PDI) and TI is the final-state correlation between the two free electrons in PDI. Dalgarno and Sadeghpour showed, however, that the effect of final-state correlation vanishes in the high photon energy limit [3]. This is consistent with the observation that the target double-ionization probabilities for electron transfer and photoionization are different for low first electron velocities but merge for higher v (cf. Figure 4).

The present absolute total TI and TTI cross sections are shown in Fig. 5 together with the results of Mergel *et al.* [15] and Shah and Gilbody [14] at lower velocities. The line has a slope corresponding to the theoretically predicted asymptotic v^{-11} dependence of the Thomas cross section [16,17]. We conclude that our data are consistent with this velocity dependence. A power-law fit to the measured TTI cross sections over a range covering the present data and the highest energy point of Ref. [15] yields an exponent of -11.1 ± 0.4.

In this work, we have measured *separate* absolute cross sections for the kinematic (KTI) and Thomas (TTI) transfer ionization processes for p-He collisions at high

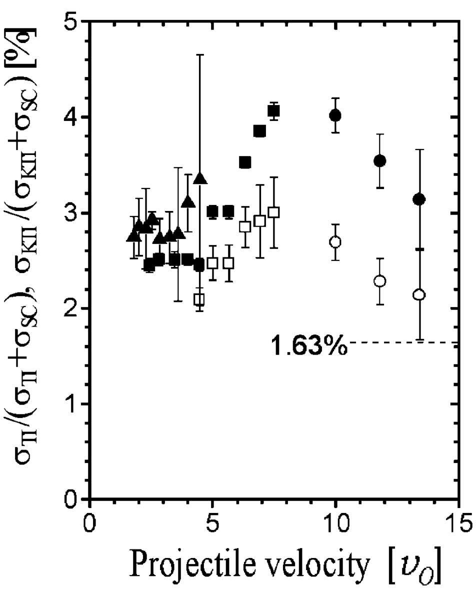

FIG. 3. The ratios of TI to total electron capture (• present work, ▲ Shah and Gilbody [14], ■ Mergel *et al.* [15]) and $\sigma_{KTI}/(\sigma_{KTI} + \sigma_{SC})$ (∘ present work, □ Mergel *et al.* [15]) as functions of the projectile velocity. The latter ratio is expected to approach the 1.63% shakeoff limit for $v \gg v_0$. The error bars show (± 1) standard deviations.

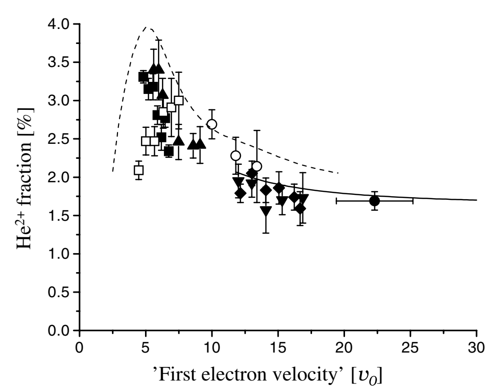

FIG. 4. The probability for emission of the second electron as a function of the velocity of the first electron as it leaves the target. Electron transfer data: ∘ present work, □ Mergel *et al.* [15]. Photoionization data: (Levin *et al.* ■ [20], ▲ [21], ▼ [22]), (Sagurton *et al.* ◆ [23]), (Spielberger *et al.* • [4]). The full curve is the $(h\nu)^{-1}$ expansion for the photoionization ratio of Andersson and Burgdörfer keeping only the constant and first order terms [2]. The dashed curve is the result of the generalized shakeoff theory of Shi and Lin [24].

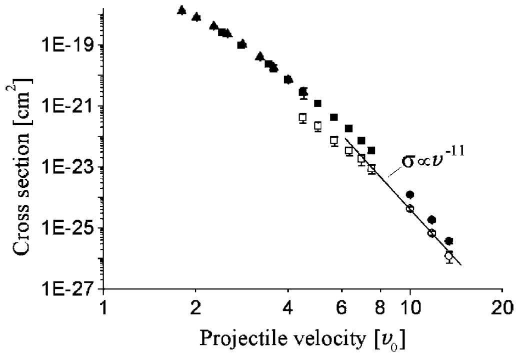

FIG. 5. The total TI (• present work, ▲ Shah and Gilbody [14], ■ Mergel *et al.* [15]), and Thomas *p-e-e* TI (TTI) (∘ present work, □ Mergel *et al.* [15]) cross sections as functions of the projectile velocity. The line through the present TTI data points (∘) has a slope corresponding to v^{-11}. A power-law fit to the four highest velocity TTI points yields a $v^{-11.1\pm0.4}$ dependence (cf. text).

velocities ($v = 10$–$13.4v_0$). At the highest velocity, we measure $\sigma_{\mathrm{TTI}} = (1.22 \pm 0.46) \times 10^{-26}\ \mathrm{cm}^2$, which required truly novel experimental techniques including an intense electron-cooled stored proton beam (CRYRING), a supersonic gas-jet target, and a novel time-switched RIMS spectrometer. This unique combination improved the sensitivity by 3 orders of magnitude over the previously best experiment [15]. The probability for target double ionization, i.e., the $\sigma_{\mathrm{KTI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{KTI}})$ ratio, was found to decrease with increasing v in the range between 10 and $13.4v_0$, exposing a clear break in the trend from the strongly increasing ratio for $v < 7.5v_0$ reported by Mergel *et al.* [15]. Our high-velocity Thomas cross sections also show a much steeper velocity dependence (consistent with v^{-11}) than the data by Mergel *et al.* [15], which indicated a $v^{-7.4}$ dependence. The present $\sigma_{\mathrm{KTI}}/(\sigma_{\mathrm{SC}} + \sigma_{\mathrm{KTI}})$ ratios agree with very recent generalized shakeoff calculations of the same quantities by Shi and Lin in which photo-double ionization and the KTI processes are treated on the same footing [24]. The present data are, in addition, in agreement with measured double-to-single photoionization ratios (considering only photoabsorption) when the velocity of the first removed electron (photoionized or captured) is taken to be the important parameter. Finally, the present data and those of Mergel *et al.* [15] give a broad maximum in the double-ionization probability, which peaks around 7–$8v_0$. Shi and Lin [24] also calculated a maximum in the same ratio, but with a different location. Very recently, it has been suggested that the increase below $7.5v_0$ is related to electron transfer from non-s^2 components in the He ground state wave function yielding high probabilities for emission of the second electron in the backward direction [30]. In the near future, we will measure the momentum distributions (calculated in [24]) of the emitted electrons in the high-velocity domain, which may now be accessed using the new technique presented in this work.

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