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LETTER TO THE EDITOR

Revealing the effect of angular correlation in the ground-state He wavefunction: a coincidence study of the transfer ionization process

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Abstract

A joint theoretical–experimental study of the transfer ionization process $p + He \rightarrow H^0 + He^{2+} + e^-$ is presented. For the first time all particles in the final state have been detected in triple coincidence. This fully differential measurement is in good agreement with a theoretical model where the target is described by a wavefunction containing both radial and angular correlation terms.

1. Introduction

In the past few years, the problem of correlation in multi-electron systems has aroused renewed interest. Ionization experiments, especially those involving two active target electrons, are among the most promising method of directly studying such processes. The coincidence study of the single ionization of atoms and molecules is now well developed and has yielded major results [1, 2]. The extension of ionization coincidence spectroscopy to the study of double excitation processes (i.e. two active target electron processes) holds the possibility of even more spectacular breakthroughs. None of the spectroscopies currently used, for example studies of the satellites in photo ionization and the line shape in Auger spectroscopy, are able to supply direct information on the part of the wavefunction determined by correlations.

The double excitation cross section will be sensitive to correlation in the target, that is to a description of the target beyond the lowest level in Hartree–Fock and the collective interactions of the particles will provide the most stringent test on our scattering models. Examples of such processes are (γ, 2e), (e, 3e) excitation–ionization [3–6, 14]. Unfortunately

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these processes though sensitive to target effects are hugely dependent on post-collisional interactions between the charged particles in the final state [4–6]. Transfer ionization is another such double ionization process, for proton impact the projectile captures one electron to become atomic hydrogen, and one could therefore hope that post-collisional Coulombic interactions would be neutralized. At large impact parameters (i.e. very small H^0 deflection angles) in particular the full sensitivity to target correlation effects would become apparent.

Differential cross sections for the transfer ionization process,

$$p + He \to H^0 + He^{2+} + e^-,$$

have been measured by Mergel *et al* using the COLTRIMS technique [7, 8, 15, 16]. These experiments were not fully differential, only the H^0 projectile and the He^{2+} were measured in a double coincidence. In this letter, for the first time, we present experimental results where all three final-state particles are detected in coincidence (H^0, He^{2+}, e^-). In [8, 16, 15], the probability of the transfer ionization process was evaluated as a double overlap integral between the correlated helium ground state and the two electron continua, but no data for multiple differential cross sections were presented. Godunov *et al* [10] have produced a first-order theoretical model giving quantitative predictions for the triple differential cross section. This model demonstrates the sensitivity of the triple differential cross section to terms beyond ns^2 in a multiconfiguration Hartree–Fock description of the target, thereby confirming a suggestion first made by Schmidt–Böcking [8, 16, 15]. The Godunov *et al* [10] model contains two terms, one corresponding to ‘transfer first’, where the proton captures one of the He electrons and the second electron is shaken off into the ionization continuum, the second corresponding to ‘ionization first’ where the proton directly ionizes one of the He electrons and the second electron is shaken off into a bound state around the proton. Indeed, the same formalism was earlier investigated by Popov *et al* [17] in a study of single differential cross sections for transfer ionization but, in the calculations, only the ‘transfer first’ term was retained. Despite the fact that they used both correlated and uncorrelated wavefunctions for the He ground state, agreement with experiment was disappointing. Here we find good agreement with experiment. This agreement depends upon the inclusion of both the ‘transfer first’ and ‘ionization first’ terms and demonstrates the sensitivity of the transfer ionization cross section to high-level target radial and angular correlation effects.

2. Theory

Transfer ionization at intermediate collision velocities may proceed via a number of different channels [11]; however most of the mechanisms are sensitive to collision energy. Since the experimentally observed features were insensitive to collision energy Godunov *et al* [10] argued that the process was likely to be dominated by target correlation and that the ionizing mechanism would correspond to a single interaction between projectile and target; they drew a parallel between transfer ionization and double ionization, the design of the transfer ionization experiment corresponding to a very particular double ionization kinematics, i.e. one electron being ionized and the second being ejected due to correlation, since the electrons are indistinguishable there is no way of knowing whither the electron which is finally captured is the one first ionized and the ejected one comes out because of correlation or vice versa. Consequently in the model of [10] the transition amplitude consisted of the sum of two terms. A ‘transfer first’ term is

$$t_{\mathrm{tr}} = -\frac{\sqrt{\mu_{\mathrm{i}}\mu_{\mathrm{f}}}}{(2\pi)^4} \int \frac{-4\pi Z_{\mathrm{p}}}{|\vec{s}_0 - \vec{s}|^2}\, \varphi^{\mathrm{F}}_{1\mathrm{s}}(\vec{s})\, \mathrm{d}\vec{s} \int \psi^*_{\vec{\mathrm{k}}_2}(\vec{r}_2) \\ \times \exp[\mathrm{i}\vec{r}_1\vec{Q} - \mathrm{i}\vec{r}_2\vec{Q}/(M_{\mathrm{t}} + 1)]\Phi_{\mathrm{i}}(\vec{r}_1, \vec{r}_2)\, \mathrm{d}\vec{r}_1\, \mathrm{d}\vec{r}_2, \qquad (1)$$

where $\mu_{\mathrm{i}} = M_{\mathrm{p}}(M_{\mathrm{t}} + 2)/(M_{\mathrm{p}} + M_{\mathrm{t}} + 2)$ is the reduced mass of the projectile and the helium atom, $\mu_{\mathrm{f}} = (M_{\mathrm{p}} + 1)(M_{\mathrm{t}} + 1)/(M_{\mathrm{p}} + M_{\mathrm{t}} + 2)$ is the reduced mass of the hydrogen atom and the helium ion He^{+}, M_{p} is the mass of the proton and M_{t} is the mass of the helium nucleus, $\psi_{\vec{\mathrm{k}}_2}(\vec{r}_2)$ is the Coulomb wavefunction for the ionized electron in the field of the He^{2+} ion, $\varphi_{1\mathrm{s}}^{\mathrm{F}}(\vec{s})$ is the Fourier transform of the hydrogen ground state, the momentum transfer is $\vec{Q} = \frac{M_{\mathrm{t}}+1}{M_{\mathrm{t}}+2}\vec{K}_{\mathrm{i}} - \vec{K}_{\mathrm{f}}$ and $\vec{s}_0 = \vec{K}_{\mathrm{i}} - \vec{K}_{\mathrm{f}}\frac{M_{\mathrm{p}}}{M_{\mathrm{p}}+1}$; $\vec{K}_{\mathrm{i}}$ and $\vec{K}_{\mathrm{f}}$ are the momenta of the incoming projectile and the scattered particle respectively, $\Phi_{\mathrm{i}}(\vec{r}_1, \vec{r}_2)$ is the ground state of the helium atom. The 'ionization first' amplitude is given by

$$t_{\mathrm{ion}} = -\frac{\sqrt{\mu_{\mathrm{i}}\mu_{\mathrm{f}}}}{(2\pi)^4} \int \frac{-4\pi Z_{\mathrm{p}}}{|\vec{s}_0 - \vec{s}|^2} \varphi_{1\mathrm{s}}^{\mathrm{F}}(\vec{s})\,\mathrm{d}\vec{s} \int \psi_{\vec{\mathrm{k}}_2}^{*}(\vec{r}_2) \exp[\mathrm{i}\vec{r}_1(\vec{s} - \vec{s}_0 + \vec{Q}) \\ - \mathrm{i}\vec{r}_2(\vec{s}_0 - \vec{s} - \vec{Q}/(M_{\mathrm{t}} + m))]\Phi_{\mathrm{i}}(\vec{r}_1, \vec{r}_2)\,\mathrm{d}\vec{r}_1\,\mathrm{d}\vec{r}_2. \tag{2}$$

We note that in the transfer first amplitude t_{tr}, the transfer and ionization process are separable; this is not the case for the ionization first amplitude t_{ion}. Consequently the dependence on the initial state $\Phi_{\mathrm{i}}(\vec{r}_1, \vec{r}_2)$ is much more transparent in the 'transfer first' case. The triple differential cross section as a function of the scattered angle, Ω_{f}, and the energy E_{e} and the angle Ω_{e} of the ionized electron is the coherent sum of both amplitudes i.e.

$$\frac{\mathrm{d}^3\sigma}{\mathrm{d}E_{\mathrm{e}}\,\mathrm{d}\Omega_{\mathrm{e}}\,\mathrm{d}\Omega_{\mathrm{f}}} = \frac{2}{(2\pi)^3}\frac{K_{\mathrm{f}}k_{\mathrm{e}}}{K_{\mathrm{i}}}|t_{\mathrm{tr}} + t_{\mathrm{ion}}|^2, \tag{3}$$

it thus depends on both mechanisms and their interference.

3. Experiment

The present experiment was performed at the van de Graaff accelerator of the Institut für Kernphysik at the University of Frankfurt. The H^{+} beam of 630 keV was collimated by two sets of adjustable slits to a beam size of about $0.5 \times 0.5\ \mathrm{mm}^2$ at the target. The beam was cleared from charge state impurities by a set of electrostatic deflector plates 15 cm upstream the target. Fifteen centimetres downstream a second set of electrostatic deflector plates separated the primary (charged) beam from the—now—neutral H^0 ejectiles. This H^0 beam intersected a supersonic He gas jet with a density of 5×10^{11} atoms cm^{-2} and 1 mm diameter at the intersection. The H^0 particles were detected with a position- and time-sensitive 40 mm multi-channel plate (MCP) detector. The recoil ions were accelerated by an electrostatic field of 4.8 V cm^{-1} at the target. A three-dimensional time and space focusing field geometry [9, 12] was used to minimize the degrading influence of the extended reaction volume on the momentum resolution. A resolution $\leqslant 0.1$ au was achieved. The electrons were guided by a magnetic field of 13.5 Gauss and accelerated 20 cm by the same electrical field onto a 120 mm MCP-detector with delay line anode; a time focusing geometry was used here, too. Events were recorded in a three-particle coincidence (e^{-}–H^0–He^{2+}). By measuring the time of flight and the position of impact on the detectors, we obtained the initial momentum vectors of the recoil ion and the electron. Six of a total of nine momentum components were thus measured directly. The projectile momentum vector i.e. the projectile scattering was calculated from the measured He^{2+} and the electron distribution by using momentum conservation. Energy conservation was used for offline background suppression. The hydrogen atom is detected at angle θ_f with respect to the beam direction, the triple differential cross section is presented as a function of the detected electron angle θ_e, again defined with respect to the beam direction; both angles are measured in the clockwise direction.

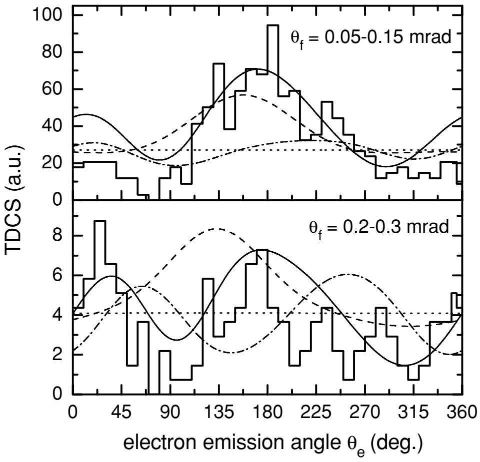

Figure 1. The triple differential cross section of transfer-ionization of helium by 630 keV proton impact as a function of the electron emission angle θ_e for several scattering angles. The cross sections were averaged over the electron emission energies ($E_e = 2.5$–7.5 eV). Theory: solid line—calculations include both 'transfer first' and 'ionization first' mechanisms with angular and radial correlation in the initial state; chain line—the initial state includes radial correlation only; dashed line—calculations with 'transfer first' only that includes both angular and radial correlation; dotted line—'transfer first' calculations with radial correlation only. Experiment: COLTRIMS measurements normalized to theoretical cross sections around the peak value.

4. Results

In figures 1 and 2 we compare the experimental data for the triple differential transfer-ionization

$$p + He \rightarrow H^0 + He^{2+} + e^-$$

cross sections for 630 keV collisions with theoretical cross sections. The calculations have been carried out in several variations to study effects of the target correlation and collision mechanisms. We have considered the effect of having only one of the mechanisms, either transfer first or ionization first only, or including both and their interference. We have used a multi-configurational Hartree–Fock (MCHF) representation [13], for the helium target. Two sets of wavefunctions $\Phi_i(\vec{r}_1, \vec{r}_2)$ for the ground state of helium were calculated, namely, one set with allowing for both radial and angular correlations, including $(ns)^2$, $(ps)^2$ and $(nd)^2$ terms with $n \leqslant 5$, and the second set with $(ns)^2$ terms only. Having only $(ns)^2$ terms in the wavefunction means that only radial correlation is included, the addition of the other terms that account is taken of angular correlation as well. Because of the delicacy of the experiments it was necessary to average over an angular range of the scattering angle and a number of electron energies. For consistency the theoretical calculations are averaged over the same angular and energy ranges, exact details are given in the figure captions, it should be noted that theoretical calculations which use the middle energy point and middle angle are not significantly different from those presented here. Experimental data were normalized to theoretical cross sections around at the principal peak.

Experimental data on figures 1 and 2 show two broad peaks, one at around 150° and the second at 30°–60°. From our calculations follow that the first peak corresponds to 'transfer-first' mechanism when angular correlation (non-s^2 terms) is included. The second peak corresponds to 'ionization-first' mechanism. Allowing for angular correlation has rather quantitative effect on the peak position and intensity. It is clear from our results that we

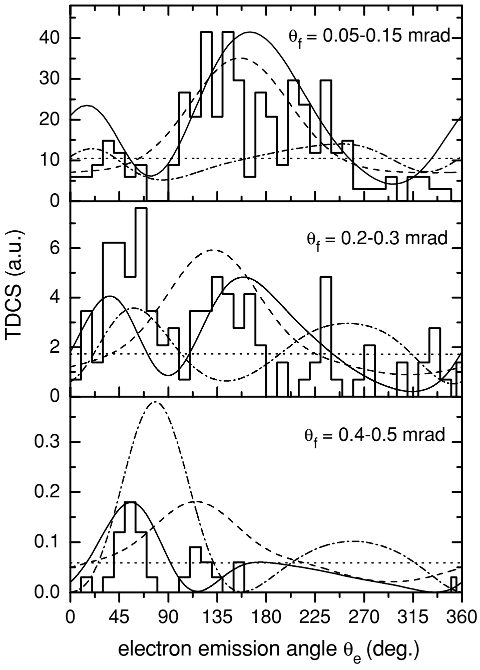

Figure 2. The triple differential cross section of transfer-ionization of helium by 630 keV proton impact as a function of the electron emission angle θ_e for several scattering angles. The cross sections were averaged over the electron emission energies (E_e = 15–25 eV). Notation as in figure 1.

need to include both transfer-ionization mechanisms and target correlation in the form of the non-s^2 terms to provide qualitative agreement with the experimental data. The principal contribution to angular correlation comes from the $(n\mathrm{p})^2$ terms, while effect of the $(n\mathrm{d})^2$ terms is small. In the model of Popov *et al* [17] only one term was included in the calculations of the single differential cross section for transfer-ionization. That term corresponds to the 'transfer-first' term in our model. From figures 1 and 2 above one may see that the 'ionization-first' term and its interference with the 'transfer-first' play a significant role depending on collision kinematics.

In conclusion experiments have been performed which for the first time give fully differential cross sections for the transfer ionization process. The theoretical calculations we have presented demonstrate a clear dependence on initial-state correlation. In terms of a multi-configurational Hartree–Fock description of the helium ground state we have shown that terms other than the $(n\mathrm{s}^2)$ give the dominant contributions to the features observed experimentally in the transfer ionization triple differential cross section. We have, we believe, demonstrated conclusively that transfer ionization in fast proton collisions at small scattering angles is very sensitive to high-level target correlation effects. While the present calculations and experiment are in good accord, it still remains to explore mechanisms of the transfer-ionization in a broader range of collisional kinematics.

Acknowledgments

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