Experimental separation of the Thomas charge-transfer process in high-velocity *p*-He collisions

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We present differential cross sections of electron capture in 7.5 MeV and 12.5 MeV proton-helium collisions. Complete experimental separations of the Thomas and the kinematic single electron capture processes in the two-dimensional He⁺ momentum distribution in the plane perpendicular to the fast ion beam have been achieved. We compare the resulting projectile angular differential cross section with the two most recent theoretical calculations and expose significant deviations.

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Charge transfer in fast ion-atom collisions has been the subject of a tremendous number of theoretical studies since the early days of quantum mechanics [1-3] until today (e.g. [4-7]). In contrast to direct target ionization, electron transfer cannot be described by a single interaction between the projectile and the target electron in the high velocity limit, as the electron has to undergo a large momentum and energy charge transfer is a benchmark reaction to test higher order theories and few-particle collision dynamics. Moreover, for a helium target, the study of fast charge transfer processes gives insight into the screening of the target potential and even into the correlated dynamics of few electron systems [8,9].

A theoretical first order description of charge transfer in fast ion-atom collisions was formulated by Oppenheimer, Brinkman, and Kramers [2,3]. In their approach the capture process-often referred to as "kinematic" capture-can be understood as a matching of the electron velocity before and after the collision, caused by the overlap of the Compton profiles of the target and projectile bound states (see [10]). A classical double scattering model was developed by Thomas [1] in 1927, and was extended to a quantum mechanical description (see [11]). In this scenario, the electron (e) is first scattered by the projectile (*p*) and then by the target nucleus (N) in two consecutive binary encounters in such a way, that after the collision the electron propagates parallel to the projectile and with almost the same velocity. Thus, the electron and the projectile can become bound. For this (p-e-N) Thomas reaction the projectile is scattered with an angle of $\theta_{\text{Thomas}} = \sqrt{3}/(2M_p)$ (in atomic units, M_p : projectile mass), which leads to a peak at θ_{Thomas} in the projectile angular distribution. For the Thomas mechanism the cross section decreases less steeply with the projectile velocity (v_p^{-11}) than for the kinematic capture process (v_p^{-12}) . It is thus to be expected that the Thomas mechanism will become dominant at sufficiently high velocities.

A considerable number of more sophisticated quantum mechanical calculations have been developed until today (for

overviews, see [12,13]). In the case of proton-helium collisions the second Born [14], the continuum distorted wave (CDW) [15], the CDW with eikonal initial state (CDW-EIS) [6], and the Born-Faddeev approximation [5,16] have been applied. In these calculations the region around the Thomas angle θ_{Thomas} is of particular interest, since there the shape and magnitude of the angular distribution is very sensitive to the theoretical approach. First, it is crucial how the calculations account for higher order terms in the Born series and their interference [17]. Second, for collisions with helium, the electron which remains bound at the target has to be considered in an adequate way [6,18].

In spite of the large theoretical interest, there are only very few earlier experimental investigations of electron capture in the velocity regime, where the contribution of the Thomas process is significant. The (*p-e-N*) mechanism has been observed as a peak in the differential cross section at scattering angles around θ_{Thomas} (0.47 mrad for proton projectiles) for 5 MeV proton-hydrogen [19] and for protonhelium collisions with projectile energies between 2.82 and 7.40 MeV [20]. Also in collisions of 3 MeV protons with molecular hydrogen a signature of the Thomas peak has been found [21]. In the helium experiment [20] the resolution was, however, not sufficient to show details around the Thomas scattering angle.

For the helium target, the two most recent calculations [5,6] have both, after convolution with the appropriate resolution, been claimed to be in excellent agreement with the only earlier available experimental results of Horsdal-Pedersen *et al.* [20]. However, with access to the unconvoluted theoretical data of [5,6] and the experimental resolution of [20], we could only reproduce such agreement with the data of Abufager *et al.* [6]. We will argue below that the results of Ghanbari Adivi and Bolorizadeh [5], in spite of the claims in their paper, are in disagreement with [20] and that their erroneous statement was caused by a mistake in the convolution procedure. In this work, we present experimental data with high resolution which allows a more detailed comparison to the theoretical results from [5,6] than the older dataset from [20].

We report on the measurement of the (p-e-N) Thomas process using COLd Target Recoil Ion Momentum Spectros-

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

copy COLTRIMS [22,23] and an ion storage ring. The COLTRIMS technique has proven to be a powerful tool to reveal the details of the dynamics of atomic collisions (see, e.g., [24]). Unlike in earlier experiments, where the projectile scattering angle was measured directly, the angular resolution in our experiment is much less sensitive to the emittance and collimation of the projectile beam, since here the scattering angle is calculated with the measured momentum transfer to the recoiling target ion. The present excellent luminosity and the high detection efficiency allowed us to measure at the so far highest projectile energies (7.5 and 12.5 MeV). Much better resolution and statistics than in previous experiments could be achieved. Due to our improved resolution, the Thomas peak can be clearly separated and the angle of the minimum between the kinematic and Thomas single capture processes can be determined with higher precision than earlier.

The experiment was carried out at the internal gas-jet target |25| and its momentum spectrometer |9,26,27| in the ion storage ring CRYRING [28] at Stockholm University. The electron capture processes take place in the overlap region of the proton beam and the cold atomic helium beam. The ionized helium is extracted by a homogeneous electric field of 1.2 V/mm oriented in the x direction, which is perpendicular to the projectile and target beams (see Fig. 1). After a 78 mm long field-free drift region the recoil ions are detected by a two-dimensional position sensitive microchannel-plate detector. The momentum of the recoil ions along the extraction direction is calculated by means of their time of flight, while the perpendicular momentum is given by the position of the hit on the detector. In order to compensate for the finite size of the reaction volume and to improve the resolution, an electrostatic lens is placed between the acceleration and drift regions. To optimize the time resolution, a time focusing geometry is chosen, i.e., the drift region is twice as long as the acceleration region. The recoil ions are measured in coincidence with the charge changed, neutral projectiles detected 3.2 m downstream of the reaction volume. The projectile scattering angle θ is calculated from the measured recoil ion momentum perpendicular to the projectile beam axis p_{\perp} as $\theta = p_{\perp} / (M_{\rm p} v_{\rm p})$ (for $\theta \leq 1$).

In Fig. 2(a) a typical time-of-flight spectrum of ionized target atoms is shown. Clearly, two peaks can be identified, one for doubly charged helium ions created in transferionization processes and a larger peak due to single electron capture events. Note, that the higher transverse momentum for the Thomas process in comparison with the kinematic capture gives rise to the shoulders on the single capture peak. We make a momentum calibration in the x direction (cf. Fig. 1) from the measured positions of the He^+ and He^{2+} peaks and the assumption of a homogeneous electric extraction field in the spectrometer. However it should be kept in mind that field inhomogeneities as well as the electrostatic lens at the entrance of the drift region may cause small deviations from this calibration in the x direction. Ion trajectories and flight times have been simulated with the SIMION software and we estimate an accuracy of 5% of the resulting momentum calibration depending on the exact geometry and voltages applied. The estimated time resolution of 3 ns, which is limited by electronics and fringe field effects, corresponds to a momentum resolution of about 0.3 a.u. (2σ) .

In Fig. 2(b) the position spectrum of all recoil ions is shown. Due to energy and momentum conservation, the ions which are created by nonradiative single electron capture have momenta in the z direction of $p_z = -v_p/2 + Q/v_p \approx$ $-v_p/2$ (Q is the inelasticity of the collision, e.g., [22,23]) and form the off center, vertically extended intensity on the recoil ion detector. The momentum resolution in the y and z directions can be estimated by the width of the p_z distribution and amounts to about 2 a.u. Knowing the x momenta, a calibration for the y direction can be performed by taking advantage of the cylindrical symmetry of the reaction.

The cross section for direct ionization is about seven orders of magnitude higher than that for electron capture. Thus, the detection of the helium ions originating from direct ionization events has to be suppressed, in order to limit the number of random coincidences. These ions have predominantly a smaller momentum in the z direction, and are blocked by a round plate with a diameter of 1 cm at the center of the detector. This is the reason for the circular area



FIG. 2. (Color online) Time-of-flight (a) and position spectrum (b) of the recoiling target ions for 7.5 MeV proton-helium collisions.



FIG. 3. (Color online) (a) Present differential cross sections $d\sigma/d\Omega$ of electron capture in 7.5 MeV proton-helium collisions (full triangles) compared to data from Horsdal-Pedersen *et al.* [20] (open circles). The solid curve represents our data convoluted with the experimental resolution from [20]. As inset, the recoil ion momentum distribution in the plane perpendicular to the projectile beam is shown. (b) Differential cross section of 12.5 MeV proton-helium collisions.

of low intensity in the center of the position spectrum in Fig. 2(b).

In Fig. 3(a) we present the results for 7.5 MeV protonhelium collisions in the form of the projectile angular differential cross section extracted from the recoil ion momentum distribution. We compare our data to those of Horsdal-Pedersen et al. [20], who used an incident energy of 7.4 MeV. A difference in the total cross section of less than 10% is expected for these two energies (see [29]), whereas the energy dependence of the angular distribution can be neglected. Since in our measurements no absolute cross sections were obtained and in order to facilitate the comparison with the previous experiment, we normalized our data to the total cross section from Horsdal-Pedersen et al. [20]. Further, we convoluted our experimental data with the resolution of Horsdal-Pedersen et al. [20] [solid curve in Fig. 3(a)]. Except for small deviations close to the minimum, the curve (our convoluted data) and the experimental results of [20] are consistent.

The structure featured by the previous data from [20] and by our results is similar with maxima at 0 mrad caused by kinematic capture and the smaller Thomas peak around $\theta_{\text{Thomas}}=0.47$ mrad. Even though a clear signature of the Thomas peak is visible in both cases, the minimum between the two peaks is completely filled out in the earlier direct projectile scattering measurement [20]. In the present data, however, there is a factor of five between the Thomas peak height and the minimum and, thus, there is a clear separation of the kinematic and the Thomas capture processes. Assuming that all events with a scattering angle larger than the angle of the minimum at about 0.32 mrad originate from a Thomas process, this mechanism contributes with $(9\pm0.5)\%$ to the total single electron capture cross section at 7.5 MeV projectile energy. As an inset in Fig. 3(a), the recoil ion momentum distribution in the plane perpendicular to the projectile beam is shown. Here, the Thomas process can be identified as a separate ring with a radius of about 15 a.u. around the central peak due to kinematic capture processes.

Compared to the experimental results for 5 MeV protonhydrogen collisions [19], the position of the minimum is shifted by about 0.05 mrad towards larger angles in our data for 7.5 MeV proton-helium collisions. Moreover, the Thomas peak is narrower for the hydrogen target and gives a significantly larger relative contribution (30%) to the total capture cross section than in the case of helium (9%). Hence, for the helium target a better resolution is required than for hydrogen, in order to clearly separate the Thomas peak. The larger relative contribution of the Thomas mechanism in the hydrogen case is somewhat surprising in view of the calculations of Shakeshaft and Spruch [10]. In their "hydrogenlike target" treatment they found that the scaling with the projectile and target nuclear charges was rather similar for the kinematic and the Thomas capture processes.

In Fig. 3(b) the present experimental results for 12.5 MeV proton energy are shown. For this reaction the total cross section is extremely small ($\sim 10^{-27}$ cm²). Hence, the statistical quality of this data set is somewhat limited, but at the expected position of the Thomas peak a pronounced shoulder can be identified that yields (14±3.5)% of the total cross section.

In Fig. 4, we compare our data for 7.5 MeV incident energy with the two most recent calculations from Ghanbari Adivi and Bolorizadeh [5] and Abufager et al. [6]. Ghanbari Adivi and Bolorizadeh calculated the differential cross section with a Faddeev-Watson-Lovelace (FWL) treatment within a second-order approximation. Here the two-body potentials of the second Born amplitudes are replaced by twobody T matrices, thereby automatically including infinite perturbative orders of each of the individual interactions [16]. Abufager et al. used the continuum distorted waveeikonal initial state (CDW-EIS) approach in order to calculate the cross section. In this approximation the electronic wave function in the final state is modified by a Coulomb distortion. In the initial state, the long-range interaction between the projectile and the bound target electron is considered through an eikonal phase. Hence, also here multiple scattering terms are partially included up to infinite order. Both calculations consider the internuclear potential and use an effective one-electron picture. The effective target potential, which is used to describe the electronic states, is obtained by the Hartree-Fock theory and in [5] approximated by a combination of long-ranging Coulomb and shortranging Yukawa potentials.



FIG. 4. (Color online) Experimental data for 7.5 MeV protonhelium collisions compared to CDW-EIS results from [6] (solid curve) and the second order Faddeev calculation from [5] (dashed curve). Both theoretical cross sections were calculated for 7.4 MeV impact energy and were convoluted with the experimental resolution. The unconvoluted calculated data is shown as dotted curves. As inset, the Thomas peak is shown on a linear scale.

Both calculations are state of the art, and agreement with the data from Horsdal-Pedersen *et al.* [20] was first reported after convoluting the theoretical results with the experimental resolution. The cross section calculated with the CDW-EIS approach [6] showed only slight deviations from the experimental data in the region around the Thomas peak and the minimum at smaller angles. In the case of the FWL calculation [5] the agreement between the convoluted theoretical result and the experiment was first reported to be excellent. However, as mentioned above it was not possible to reproduce the latter agreement by using our convolution procedure, the experimental resolution from Horsdal-Pedersen et al., and the unconvoluted theoretical results of [5]. The mistake in [5] might be caused by an unfortunate misprint in Eq. (6) of [21] which was used in [5] to convolute the theoretical results. From this we conclude that there is a significant disagreement between the theoretical results [5] and the earlier experimental data [20]. It should be noted that the result of a FWL calculation by Alston [16] also agreed with the experimental cross section from Horsdal-Pedersen et al. [20]. Unfortunately we do not have access to the unconvoluted data of Alston and are therefore unable to compare it to the present experimental data.

In order to compare the calculations from [5,6] with our experimental data, the theoretical results were convoluted with a two-dimensional Gaussian function with a momentum width of 0.3 a.u. in the *x* and 2 a.u. in the *y* direction, which corresponds to angular uncertainties of 0.009 mrad and 0.063 mrad, respectively. The cross sections are shown in

Fig. 4 together with the present experimental results. Some features are similar in both calculations. The position of the Thomas peak is for both approaches at the classically predicted angle θ_{Thomas} and in agreement with the present experimental results.

However, the two calculations differ from each other and from the experimental results as can be seen most clearly in the inset of Fig. 4, where the Thomas peak is plotted in a linear-linear graph. The total cross section is more than 50% larger in the FWL calculation [5] than in the CDW-EIS approach [6]. The minimum in the angular distribution occurs at smaller and larger angles than in the experiment for the CDW-EIS calculation and for the FWL approach, respectively. Also the relative contributions of the Thomas peak to the total cross sections are very different in the two calculations. In the CDW-EIS calculation, a contribution of 15% is obtained, i.e., the Thomas process is overestimated by a factor of almost 1.8 in relation to the experimental result. In the FWL approach, the Thomas peak is found to contribute with only about 3% and is thus underestimated by a factor of about 3.

Even though in Fig. 4 the agreement with our data is better for the CDW-EIS calculation than the FWL method it cannot be concluded that the CDW-EIS approach is in general more suitable to describe the collision dynamics. As mentioned already above, in an earlier publication by Alston [16] excellent agreement with the previous experimental data from [20] was achieved using the FWL method. However, in the same work, the results from the FWL approach showed slight deviations from the experiment on the atomic hydrogen target [19]. This demonstrates, that it is in fact difficult to disentangle the role of the higher order contributions in the projectile-target interaction and the relevance of the target potential and the resulting electronic states.

In summary, we obtained experimental results for fast, charge-changing proton-helium collisions for projectile energies of 7.5 and 12.5 MeV by exploiting COLTRIMS in a cooler storage ring. We achieved an unprecedented angular resolution which provides detailed and unique information on the differential cross sections, in particular on the relative contribution of the Thomas peak and the position of the minimum at smaller angles. In comparison with the two most recent state-of-the-art calculations we could show that important discrepancies remain even with the most advanced modern theoretical descriptions. The data provides an extremely sensitive test not only for higher-order terms and their interference, but also for the description of the two-electron target, which is necessary to understand the dynamics in few-particle Coulomb systems.

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