Double Ionization of Helium and Neon for Fast Heavy-Ion Impact: Correlated Motion of Electrons from Bound to Continuum States

R. Moshammer,[1] J. Ullrich,[2,*] H. Kollmus,[1] W. Schmitt,[2] M. Unverzagt,[1] O. Jagutzki,[1] V. Mergel,[1] H. Schmidt-Böcking,[1] R. Mann,[2] C. J. Woods,[3] and R. E. Olson[3]
[1]*Institut für Kernphysik, Universität Frankfurt, D-60486, Frankfurt am Main, Germany*
[2]*Gesellschaft für Schwerionenforschung, D-64220, Darmstadt, Germany*
[3]*Department of Physics, University of Missouri-Rolla, Rolla, Missouri 65401*
(Received 8 April 1996)

Double ionization of He and Ne by charged particle impact was explored in kinematically complete experiments measuring the final momenta of the two emerging electrons ($\mathbf{P}_{e1}^f, \mathbf{P}_{e2}^f$) and of the recoiling target ion $\mathbf{P}_R^f$. The momentum transfer from the 3.6 MeV/u Se^{28+} projectile to the electrons is found to be negligibly small. Thus distinct patterns observed in the $\mathbf{P}_{e1}^f$-versus-$\mathbf{P}_{e2}^f$ spectra sensitively reflect the details of the electrons' correlated motion. Classical calculations reasonably describe the data only when the (e-e) interaction during the ionization reaction is included. [S0031-9007(96)00899-X]

PACS numbers: 34.50.Fa, 34.10.+x

The correlated dynamics of a quantum mechanical many-electron system, either perturbed during a collision or nonperturbed in an atomic state, is the subject of fundamental questions in atomic physics. In contrast to the enormous precision which is obtained in the investigation of the static structure of even many-electron atoms, basic and unsolved problems are still present in the understanding of the most simple dynamic reactions like double ionization of helium by photon or charged particle impact [1].

Double ionization by photon impact is the simplest and most fundamental multiple ionization process. The ejection of two electrons after absorption of one photon is prohibited in an independent electron approximation and therefore sensitively depends on the electron-electron (e-e) correlation. Numerous recent theoretical attempts to calculate R_γ, the total cross section ratio for helium double to single ionization, still differ by up to 74% at intermediate photon energies [2]. Predictions of R_γ at large photon energies have finally converged and are in agreement with the experimental results for photo absorption [3] and Compton scattering [4].

For charged particle impact the situation is more complicated for two reasons. First, in addition to the contribution due to the e-e correlation the projectile might interact independently with both electrons promoting them into the continuum [1]. So far one *ab initio* quantum mechanical theoretical model, the "forced impulse approximation" (FIM), includes both mechanisms [5] and correctly predicts total cross section ratios R_q for a large variety of projectile charges and velocities [1,5,6]. However, no results differential in the momenta of the emitted electrons are available due to the second major complication, namely, the inclusion of the four-particle Coulomb continuum. Our data are compared to results obtained in the classical trajectory Monte Carlo (CTMC) approach. Comparison with experimental data indicates that classical calculations reliably describe the momentum balance between the electrons and the heavy particles for both single and multiple ionization reactions [7,8].

Experimentally, kinematically complete experiments on multiple ionization are extremely difficult to perform if one applies conventional many-electron coincidence detection techniques. In fact, only a few results have been published in the literature on double ionization after photoabsorption where the two emitted electrons are measured in coincidence [9,10]. To our best knowledge, one investigation has been reported on the double ionization of Ar after electron impact [11], measuring three electrons under very special kinematic conditions. No such measurement on helium, the fundamental two-electron target, has been feasible up to now. Decisive progress has been achieved within the last 2 years with the development of extremely efficient, high-resolution, recoil-ion–electron coincidence techniques [12,13]. Here the full momentum vectors of the recoiling target ion and of one emitted electron are determined simultaneously with coincidence efficiencies of approximately 30%. Applying this method, several kinematically complete experiments on photon double ionization [2,3], single ionization by heavy projectiles [7], and transfer ionization have become feasible.

Exploiting these techniques we have performed a first kinematically complete measurement on double ionization of helium and neon by charged particle impact. This was accomplished applying a recoil-ion–electron-electron–projectile coincidence to simultaneously determine the full final momentum vectors (nine Cartesian momentum components) of the recoiling target ion and of the two electrons. In this Letter the various longitudinal momentum balances (along the beam direction) are presented. Transverse momenta will be discussed in a following paper. All relative emission angles between the three emerging particles are accepted with a 4π solid angle for electron energies up to 50 eV and superior momentum resolution in the longitudinal direction is obtained. The recoil ion longitudinal momentum resolution

 0031-9007/96/77(7)/1242(4)$10.00

$\Delta P_{R\parallel}$ compares to an equivalent energy resolution as small as $\pm 20\ \mu$eV; for low-energy electrons an equivalent energy resolution of ± 5 meV was achieved. The energy change of the 284 MeV Se^{28+} projectile during the double ionization reaction is controlled with a resolution of $\Delta E_P = \pm 33$ eV $(\Delta E_P/E_P = \pm 1.2 \times 10^{-7})$ corresponding to a momentum resolution of $\Delta P_P = \pm 0.1$ a.u. (atomic units) or $\Delta P_P/P_P = \pm 6 \times 10^{-8}$.

The experiments were performed at the UNILAC of GSI using a 3.6 MeV/u stripped, charge state analyzed and well collimated (1 mm $\times$ 1 mm) Se^{28+} beam. The final charge state was analyzed after the collision, and Se^{28+} ions (no charge exchange) were recorded by a fast scintillation detector at a rate of up to 1 MHz. A two-stage supersonic jet provided a well defined (2.8 mm diameter), cold (50 mK), and dense (10^{12} atoms per cm^2) helium or neon target (Fig. 1). Low-energy ions (typical energies $E_R \ll 1$ eV) and electrons (typical energies $E_e < 50$ eV) are accelerated into opposite directions by applying a weak electric field (1–5 V/cm) along the ion beam generated between two ceramic plates which are covered with burned-in resistive layers. The field is sufficiently strong to project recoiling target ions with a large solid angle $(\Delta\Omega_R/4\pi > 98\%$ for He^{2+} or $Ne^{(1-6)+})$ onto a two-dimensional position sensitive (2DPS) microchannel plate detector (MCP). An additional solenoidal magnetic field is generated by two Helmholtz coils (1.3 m diameter) with its field vector along the ion beam. It efficiently guides the electrons $(\Delta\Omega_e/4\pi > 50\%$ for the same reactions) onto three independent 2DPS MCP's, each of them capable to accept "multihits" for time intervals between individual hits larger than 6 ns. From the measured absolute positions and flight times the ion and electron trajectories are reconstructed and their initial momenta are calculated [13].

In Fig. 2 the longitudinal momentum balances, along the beam direction, for helium double ionization are shown. The experimental data are normalized on 50% of the total double ionization cross section of $\sigma^{2+} = 5 \times 10^{-16}$ cm^2 to account for the estimated loss of solid angle due to the limited electron energy acceptance of $E_e < 50$ eV. Two important and surprising features are visible. First, the momentum transfer by the projectile in each individual ionization reaction is negligibly small compared to the measured final momenta of the recoil ion $(P_{R\parallel})$ and electrons $(\sum P_{ei\parallel})$. These, then, are not a result of direct momentum transfers during the collision but must have been "stored" in the bound state of the helium before the encounter. Therefore, they both closely reflect the correlated longitudinal sum-momentum distribution of the electrons in the bound state, i.e., the two-electron Compton profile. Since the total momentum of the helium atom is zero in the initial state it follows that $\sum P^i_{e\parallel} \approx -P^i_{R\parallel}$, a feature which is clearly visible in the data. Like a photon field the fast projectile mainly delivers energy but only little momentum. Second, electrons are found with positive momenta emitted into the forward direction whereas recoil ions are emerging backwards, which is due to the final state interaction between the target fragments and the projectile. This postcollision interaction (PCI) has been observed before for single ionization of helium [7,14] as well as for multiple ionization of Ne [8] and is well understood.

Both features are correctly predicted by the classical calculations (lines in Fig. 2). Here, the two independent, distinguishable, and "classical" electrons are set on different Kepler orbits bound with the sequential experimental binding energies (nCTMC for n-body CTMC; [15]). In this model the total binding energy of the initial state is correct and the initial state sum-momentum distribution of the electrons in the bound state is well reproduced. In addition, the collisional asymmetry is correctly predicted indicating the ability of this model to account for the PCI. Surprisingly, explicit quantum mechanical features like the spatial (momentum) correlation of the electrons due to the symmetry of the wave function (Fermi statistics) or the direct (e-e) interaction due to the $1/r_{12}$ potential between the two electrons do not have to be included to describe the longitudinal sum momenta of the emitted electrons.

These characteristics change dramatically if the correlated two-electron emission is explored in detail. In Fig. 3 the experimental $P_{e1\parallel}$-versus-$P_{e2\parallel}$ spectrum for helium double ionization (upper right figure) is compared to the results of various classical calculations. This coordinate system can also be considered as plotting the electron

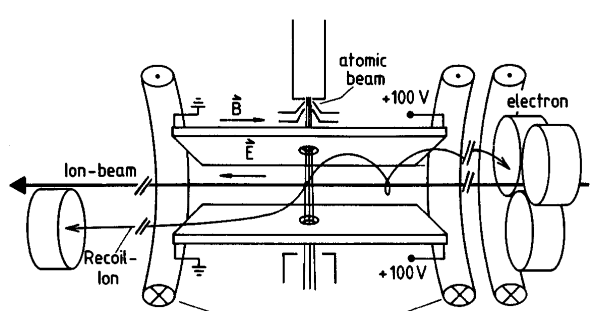

FIG. 1. Schematic drawing of the spectrometer [13].

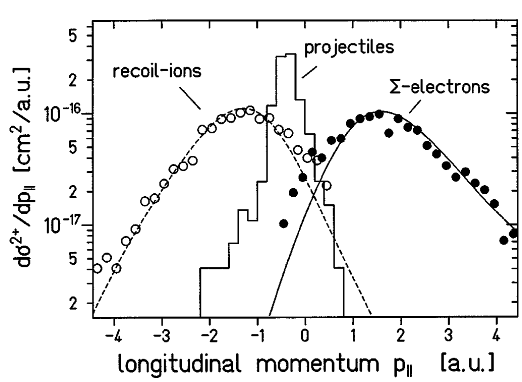

FIG. 2. Longitudinal momentum distributions $d\sigma^{2+}/dP_{\parallel}$ for the sum momentum of the electrons, the He^{2+} ions and the momentum change of the 3.6 MeV/u Se^{28+} projectile for He double ionization. Lines: nCTMC (see text).

longitudinal sum momentum ($P_{e1\|} + P_{e2\|} = P^{+}_{e\|}$) versus their difference ($P_{e1\|} - P_{e2\|} = P^{-}_{e\|}$) if the figure is rotated by 45°. Two major features are visible: First, both electrons are mainly emitted into the forward hemisphere which has been identified before to be a result of the PCI [7,8]. Second, a distinct pattern is found which is even more pronounced for neon double and triple ionization (Fig. 4). If one electron is slow the other is most likely fast or, in other words, with increasing $P^{+}_{e\|}$ larger $P^{-}_{e\|}$ are observed. Such a feature cannot be explained in *any* independent particle model or, more precisely, in the absence of the explicit $1/r_{12}$ interaction between the two electrons. Independent of any theory this demonstrates that our results are extremely sensitive on the correlated motion of the electrons.

We have performed several calculations to illuminate the facets of the correlated dynamics. In the first one the electrons are independent and distinguishable (nCTMC). The second one (lower left part of Fig. 3) includes the monopole part of the (e-e) interaction in the ground (initial) state as well as during the collision (dCTMC for dynamical screening [16]) thus taking care of the fact that the electrons are indistinguishable. A reasonable, but still not perfect description of the experimental result is "only" obtained when the $1/r_{12}$ interaction is included after both electrons have a positive energy relative to the target nucleus during the collision (lower right part in Fig. 3).

The basic dilemma of any classical calculation is that it is impossible to correctly describe the bound initial two-electron ground state. The explicit inclusion of the $1/r_{12}$ potential causes autoionization of the unperturbed classical atom due to the missing quantization of the electronic orbits. In the third model the monopole part of the $1/r_{12}$ interaction in the ground state is simulated by a model potential obtained from Hartree-Fock calculations. Moreover, the electron screening is dynamically included during the collision [16] and the full $1/r_{12}$ potential is finally turned on as the second electron of the helium proceeds in a continuum state. Thus the complete Hamiltonian is considered during that part of the collision where both electrons are in continuum states and the final state interaction between all particles is correctly described classically [17]. Since the initial state correlation is not completely included, however, one can neither decide what its influence really is nor estimate to what extent it is included by turning it on after the second electron proceeds in the continuum.

The basic dilemma of any quantum mechanical calculation, on the other hand, is that it is extremely complicated if not impossible to correctly describe the time-dependent correlated many-particle dynamics during and after the collision [17]. In contrast, the static initial state wave function can be calculated and the binding energy can be predicted with extreme precision. Such a correlated wave function [18] was used to calculate the ground-state probability distribution of the two electrons in the longitudinal momentum space, i.e., in the $P_{e1\|}$-versus-$P_{e2\|}$ plane [19]. This distribution is symmetric around $P_{e1\|} = P_{e2\|} = 0$ and would be the result expected due to the initial state correlation alone. For a qualitative comparison with the experimental data the electron-projectile PCI was estimated from the classical calculations and approximately accounted for

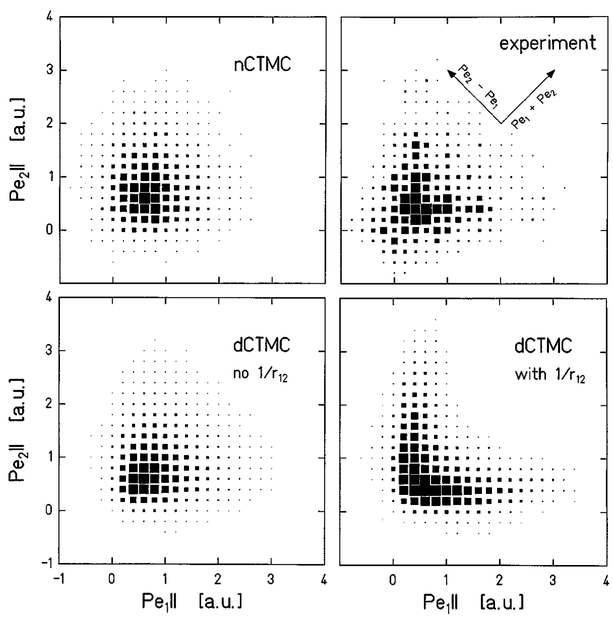

FIG. 3. $P_{e1\|}$ versus $P_{e2\|}$ of the two electrons for the same collision system. Experiment: different box sizes (15) represent the cross sections $d^2\sigma^{2+}/dP_{e1\|}dP_{e1\|}$ in 1.0×10^{-16} cm^2/a.u.^{2} (largest box) on a linear scale. Various theoretical results (see text, same representation) are scaled on the maximum differential cross sections (largest box) for each of the calculations.

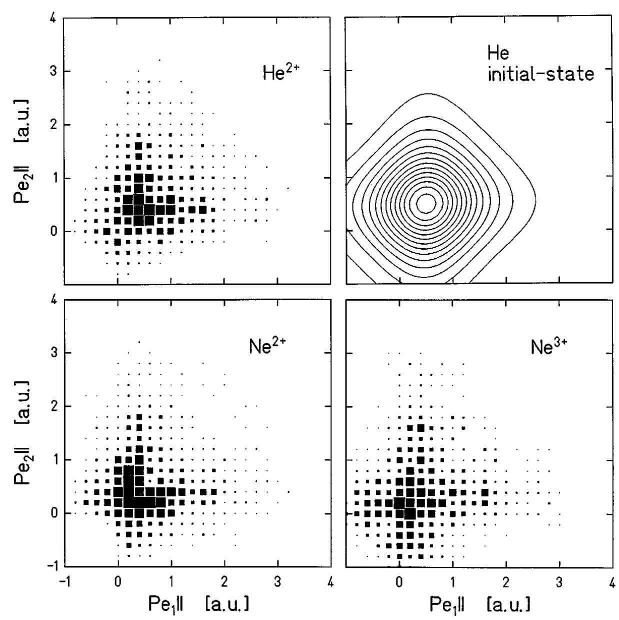

FIG. 4. $P_{e1\|}$ versus $P_{e2\|}$ of the two electrons emitted for He and Ne double and triple ionization. Various results (see text, same representation as in Fig. 3) are scaled on the maximum differential cross sections, respectively.

by shifting the whole distribution by $P_{e\|}^{+} = 0.6$ a.u. (upper right corner of Fig. 4). Surprisingly, the shape of the experimental distribution is well reproduced leading to the conclusion that the measured patterns may strongly reflect the initial state correlated two-electron wave function.

This interpretation can be understood viewing the attosecond ($\sim 10^{-18}$ sec), extremely intense ($\sim 10^{18}$ W/cm^2) electromagnetic pulse which is generated by the passing projectile as a field of virtual quanta (Weizäcker-Williams formulation for the ionization by relativistic projectiles). Each one of both He electrons is independently "photoionized" in the broadband field by absorbing one virtual photon with an energy corresponding to the individual electron momentum at the instant of ionization. A negligibly small momentum is transferred and no significant momentum exchange between the electrons themselves or between each electron and the helium nucleus may take place since the collision time is short compared to the electron revolution frequency in the bound state. The emission of two electrons as a result of independent interactions with the projectile is the dominant double ionization mechanism at strong perturbations [1,2] leading to a magnitude of $R_q \approx 15\%$ for the investigated collision system which is much larger than for ionization by a single photon of $R_\gamma \leq 3\%$. Whereas the $(e\text{-}e)$ interaction is demanded to achieve double ionization by a single photon at low and intermediate energies this interaction contributes negligibly to double ionization in the present case. Accordingly, the total double ionization cross section changes only by a few percent by the implementation of the $(e\text{-}e)$ interaction in the classical calculations but strongly influences the details of the emitted electron momentum distributions. Thus from these considerations there are good arguments that the final electron momenta may closely reflect the initial state correlated two-electron wave function. Because of the impossibility to correctly describe the two-electron initial state within a classical model only a full quantum mechanical calculation which includes the $(e\text{-}e)$ correlation in the initial state, during the collision, and in the final state can provide a definite answer to what extent the observed patterns are influenced by the initial or final state interaction.

For neon double and triple ionization (Fig. 4) similar but different and even more pronounced patterns are observed. The pattern observed for Ne^{3+} is exciting since only two out of three electrons are detected, thus effectively integrating over all momenta of the unobserved electron. This pattern can occur only if all three electrons in the continuum are strongly correlated. Since the momentum transfer by the projectile is so extremely small, the initial state correlation evolves in a dynamically correlated way into the continuum and is finally observed in the strongly correlated three-electron continuum.

In conclusion, we have measured for the first time the complete momentum balance for double ionization of helium and neon by charged particle impact. The important finding of our work is that the fast highly charged projectiles are an extremely "soft" and "sensitive" tool to multiply ionize the target; i.e., it merely transfers any momentum to the target electrons, acting very much as an intense broadband photon field. Thus the measured two-electron momentum distributions are found to sensitively reflect the $(e\text{-}e)$ interaction as demonstrated by the comparison with various classical calculations. Moreover, there are strong indications that details of the correlated initial state wave function are reflected in the data in a very direct and unperturbed way.

Finally, we want to stress that quantum mechanical *ab initio* calculations are urgently required for the fundamental two-electron helium target as a starting point to explore more complex situations which will experimentally be investigated in the very near future. The calculations might not be too complicated if relativistic heavy ions are used as projectiles, reducing the PCI and making the Weizäcker-Williams method applicable. Then, one might envision that this technique will become a standard "attosecond microscope" for the investigation of bound-state electron wave functions in atoms, molecules, clusters, or even solids.

Theoretical support by N. Grün and discussions with S. Keller and M. Horbatsch are gratefully acknowledged. We would like to thank the UNILAC crew of GSI for providing an excellent beam. This work was supported by Gesellschaft für Schwerionenforschung, Bundesministerium für Bildung und Forschung, Deutsche Forschungsgemeinschaft, and the National Science Foundation No. Int-9112815.

*Electronic address: J.Ullrich@gsi.de

[1] J. H. McGuire, J. Phys. B **28**, 913 (1995).
[2] R. Dörner *et al.*, Phys. Rev. Lett. (to be published).
[3] L. Spielberger *et al.*, Phys. Rev. Lett. **74**, 4615 (1995).
[4] L. Spielberger *et al.*, Phys. Rev. Lett. **76**, 4685 (1996).
[5] J. F. Reading and A. L. Ford, Phys. Rev. Lett. **58**, 543 (1987); J. Phys. B **20**, 3747 (1987).
[6] J. Ullrich *et al.*, Nucl. Instrum. Methods **B87**, 70 (1994).
[7] R. Moshammer *et al.*, Phys. Rev. Lett. **73**, 3371 (1994).
[8] M. Unverzagt *et al.*, Phys. Rev. Lett. **76**, 1043 (1996).
[9] O. B. Schwarzkopf *et al.*, J. Phys. B **27**, L347 (1994).
[10] A. Huetz *et al.*, J. Phys. B **27**, L13 (1994).
[11] A. Lahmam-Bennani, J. Phys. B **24**, 2401 (1991).
[12] J. Ullrich *et al.*, Comments At. Mol. Phys. **30**, 285 (1994).
[13] R. Moshammer *et al.*, Nucl. Instrum. Methods **B108**, 425 (1996).
[14] T. Vajnai *et al.*, Phys. Rev. Lett. **74**, 3588 (1995).
[15] R. E. Olson, J. Ullrich, and H. Schmidt-Böcking, Phys. Rev. A **39**, 5572 (1989).
[16] V. J. Montemayor and G. Schiwietz, Phys. Rev. A **40**, 6223 (1989).
[17] J.-M. Rost, J. Phys. B **28**, 3003 (1995).
[18] P. E. Regier and A. J. Thakkar, Phys. Rev. A **30**, 30 (1984).
[19] N. Grün (private communication).