Collective Behavior of Electrons Emitted in Multiply Ionizing Collisions of 5.9 MeV/u U⁶⁵⁺ with Ne

M. Unverzagt,¹ R. Moshammer,¹ W. Schmitt,⁴ R. E. Olson,² P. Jardin,³ V. Mergel,¹ J. Ullrich,⁴

and H. Schmidt-Böcking¹

¹Institut für Kernphysik, August-Euler-Strasse 6, D-60486 Frankfurt, Germany

²Department of Physics, University of Missouri-Rolla, Rolla, Missouri 65401

³Centre Interdisciplinaire de Recherches avec les Ions Lourds, Rue Claude Bloch BP 5133, 14040 Caen, Cedex, France

⁴Gesellschaft für Schwerionenforschung, m.b.H., Planckstrasse 1, D-64291 Darmstadt, Germany

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The collective behavior of up to six target electrons emitted in a single collision of 5.9 MeV/u U^{65+} with Ne was investigated using high-resolution recoil-ion momentum spectroscopy. With an increasing number of ejected electrons the recoil ions are increasingly scattered into the backward direction, providing evidence that the electrons are emitted into the forward hemisphere. Their mean longitudinal sum energy varies from about 5 eV for single ionization up to 1.1 keV for Ne⁶⁺. Experimental recoil-ion momentum distributions are in excellent agreement with results of classical many-particle calculations.

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In a single collision of a fast, heavy, and highly charged ion with a complex atom the simultaneous transfer of many target electrons from bound to continuum states (multiple target ionization) occurs with large cross sections [1,2]. In many cases, for large perturbations q/v_P by the incoming projectile (q is the projectile charge and v_P is the projectile velocity), up to 50% of all ionizing collisions are multiple ionization events. These reactions, therefore, decisively contribute to the energy loss and straggling of heavy ions in gases [3], plasma [4], tissue [5,6], or solid matter [7,8].

Theoretically not much is known on the behavior of a many-electron system under the action of a timedependent perturbation. For fast heavy-ion impact where large projectile charges of up to q = 92 occur, q/v_P typically exceeds unity up to comparably large velocities of $v_P \leq 100$ a.u. and perturbation theories cannot be used over a broad v_P regime. Quantum mechanical ab initio coupled-channel calculations are usually not practicable in the regime of strong perturbations due to the tremendous increase in the number of coupled channels. The *ab initio* calculation for triple and multiple ionization is definitely beyond present computing capabilities. Thus, in addition to its relevance to applied physics, the timedependent quantum mechanical many-particle problem remains among the most important and very fundamental questions to be investigated in the future.

In the past classical *n*-body classical trajectory Monte Carlo (nCTMC) [9] and semiclassical quantum statistical [10] many-particle methods have been developed with tremendous success in the prediction of total multiple ionization cross sections. Such theories were even able to provide a reliable description of differential multiple ionization cross sections in dependence of the projectile scattering angle [11,12] or the recoil-ion transverse momentum [12,13]. A strongly collective behavior of the emitted electrons was predicted for multiple ionization

after fast heavy-ion impact [14,15]. (Following these authors "collective" is used in the sense of "behaving in a similar or same way." This does not necessarily imply a strong interaction between the individual electrons or a "correlated" motion.) According to these calculations electrons are scattered to the side of the incoming projectile and strongly into the forward direction opposite the recoiling target ion which compensates for most of the electron longitudinal and transverse sum momentum. The projectile was found to be deflected to negative angles onto the side of the recoil ion for a major part of all ionizing collisions [15,16].

Experimentally, as well, only few investigations have been reported on many-electron transitions going beyond the determination of total cross sections. In particular, a conclusive proof of the above predictions, most important to test the abilities of the theoretical approximations, is still missing. This is due to the enormous difficulties one faces in many-electron coincidence experiments. Two alternative ways to achieve information on the collective properties of emitted electrons are the measurement of the projectile energy loss or of the recoil-ion momentum. Energy-loss experiments at large v_P are extremely difficult to perform with sufficient resolution of $\Delta p/p < 10^{-5}$ [17] and no information on the direction of the emitted electrons can be deduced from these experiments. The other way, the determination of the recoiling target-ion momentum (recoil-ion momentum spectroscopy) also suffered from limited transverse [12,18] or longitudinal [19] resolution for experiments reported until now.

In this Letter we provide first experimental evidence for the collective forward emission of electrons from multiply ionizing 5.9 MeV/u U⁶⁵⁺ on Ne collisions exploiting high-resolution recoil-ion momentum spectroscopy at large projectile velocities ($v_P = 15$ a.u.). The complete recoil-ion momentum vector was determined with a resolution of about $\Delta p_R = \pm 0.35$ a.u., a factor of 15 better than the best resolution ever reported [19] for a target heavier than He. At large projectile velocities the longitudinal recoil-ion momentum distribution mirrors the longitudinal sum momentum of the emitted electrons within an accuracy of $\sum E_e^n/v_P$ (E_e^n is the continuum energy of the *n*th electron) for a multiple ionization event [20,21]. Thus experimental information on $\sum p_{e\parallel}^n$ and on the collective behavior of emitted electrons can be obtained by a measurement of $p_{R\parallel}$ alone.

The experiments were performed using a 5.9 MeV/u stripped, well-collimated $1 \times 1 \text{ mm}^2$ and charge state analyzed U⁶⁵⁺ beam from the UNILAC of GSI. After the collision the outgoing projectiles were deflected in a magnet and only U65+ ions (no charge exchange) was recorded by a fast scintillation detector at a rate of up to 1 MHz. Emerging U^{65+} ions, recoiling low-energy target ions of various charge states as well as one of the electrons emitted in each event were measured in a triple coincidence as illustrated in Fig. 1. This was the final charge states of the projectile and of the target ion are controlled. In addition, the recoil momentum vector was determined as described below (due to the complexity of the triple-coincidence measurement only a brief and incomplete outline of the experiment can be provided in this Letter; details can be found in two recent experimental papers [22,23]).

A single stage supersonic jet provided a well-localized Ne target of 2.8 mm diameter and a density of about 3×10^{11} atoms/cm² at the intersection point with the ion beam at a rest gas pressure of 10^{-7} Torr [21,24]. Recoil ions are created at the ion-beam jet intersection with small energies of typically well below 1 eV for the present multiple ionization reactions. They were extracted by a weak uniform electric field of 0.75 V/cm transverse to the ion beam. The field was generated between two resistive-layer covered ceramic plates of 20 cm length (see Fig. 1). After 10 cm of acceleration, the recoil ions drift over 20 cm to be focused in time. They are postaccelerated by 2000 V over 2 mm and detected by a two-dimensional position sensitive (2D PS) channel plate detector of 40 mm diameter. The



FIG. 1. Schematic drawing of the combined recoil-ion electron spectrometer.

Ne^{*i*+} time of flight (TOF) as large as 48 μ sec for *i* = 1 was measured by the recoil-ion-projectile coincidence. The TOF provides the target-ion charge state and the recoil momentum along the extraction direction. From the position on the detector and the measured TOF the other two momentum components can be calculated and the full recoil-ion momentum vector is determined.

In principle the additional fast coincidence with one of the emitted target electrons is not necessary but turned out to be essential for the experiment: In order to sufficiently reduce the background contributions in the 50 μ sec time window only projectiles coincident with one of the electrons were accepted as true start signals for the recoil-ion TOF measurement. To provide this trigger signal all electrons with a total energy of less than 70 eV were detected with a solid angle of $\Delta \Omega > 2\pi$ by a 2D PS channel plate placed opposite to the recoil-ion detector (a postacceleration of 200 V guarantees optimum detection efficiency). The large solid angle is obtained by extracting the electrons with the electric field and forcing them onto spiral trajectories in an additional solenoidal magnetic field of 30 G along the extraction direction [21,22].

The absolute calibration of the recoil-ion longitudinal and transverse momentum components has been checked by applying different extraction voltages and is accurate within about 0.5 a.u. For Ne¹⁺ and 1.2 a.u. for Ne⁶⁺. Since in all previous experiments for single ionization the theoretical maximum in the longitudinal momentum distribution was in excellent agreement with experiment [21,25], the present position of the maximum for i =1 was set equal to the theoretical value. The relative accuracy in the $p_{R\parallel}$ scale for different *i* is much better and experimental uncertainties are less than ±0.1 a.u.

The sum of the events over the recoil-ion momenta and charge states was normalized to the previously measured absolute ionization cross section $\sigma = \sum_{i=1}^{6} \sigma^{i}$ of $\sigma = 1.1 \times 10^{-14}$ cm² [15]. The cross sections for recoil ions with different charge states *i* are in excellent agreement with *n*CTMC calculations where the total energy deposition to the target is considered properly.

In Fig. 2 the experimental longitudinal momentum distributions for recoiling target ions with charge states up to i = 6 are shown in comparison with theoretical results calculated in the *n*CTMC approach. Recoil ions of all charge states are mainly ejected with negative longitudinal momenta, i.e., into the backwards direction. Already for Ne¹⁺ this feature is obvious and comparable to that observed before for single ionization of He [21] indicating the influence of the strong and long ranging potential of the emerging projectile. The backwards emission of the recoil ions becomes more and more pronounced with increasing electron multiplicity and a maximum in the momentum distribution at about -4 a.u. is observed for i = 6. For charge states up to i = 4 the theoretical prediction is in excellent agreement with the experimental data in shape, in absolute magnitude, as well as in the prediction of the position of the maximum in $p_{R\parallel}$.



FIG. 2. Recoil-ion longitudinal momentum distributions for i-fold target ionization. Circles: experimental data. Solid lines: theoretical results in the *n*CTMC approach. Dashed straight line: Recoil-ion momentum equals zero.

The recoil-ion longitudinal momentum distribution can be related to the double differential multiple electronemission cross sections as has been pointed out before [21,25] and described in detail recently for single ionization [20]. In brief, for collisions with small energy, momentum, and mass transfers (perfectly fulfilled in the present case on the level of 10^{-6}), it follows from momentum and energy conservation for the longitudinal momentum balance in multiply ionizing collisions (all in atomic units):

$$p_{R\parallel}^{(i)} = \sum_{n=1}^{i} (U_n + E_e^n) / v_P - \sum_{n=1}^{i} p_{e\parallel}^n.$$
(1)

 U_n are the well-known sequential ionization potentials for Ne, E_e^n is the continuum energy, and $p_{e\parallel}^n$ is the longitudinal momentum of the *n*th electron emitted in the collision.

At large projectile velocities and for small ionization potentials as well as continuum electron energies, the first two terms are negligibly small and Eq. (1) reduces in good approximation to $p_{R\parallel}^{(i)} = -\sum_{n=1}^{i} p_{e\parallel}^{n}$. Thus direct information on the longitudinal sum momentum of the emitted electrons can be obtained for Ne charge states up to i = 3, where these conditions are reasonably well fulfilled (for single ionization of He by fast Ni impact [21] it has been demonstrated in a kinematically complete experiment that the recoil-ion longitudinal momentum mirrors that one of the electron on the level of $\Delta p_{e\parallel} \approx 0.2$ a.u.). The backwards emission of the recoil ions for all charge states provides a first and unambiguous experimental proof that the electron sum momentum is directed into the forward hemisphere for multiple ionization by fast highly charged ion impact.

With an increasing number of emitted electrons the contribution to the longitudinal recoil-ion momentum due to the $\sum E_e^n/v_P$ term, throwing the recoil ions into the forward direction, can no longer be neglected. The sum energy of the continuum electrons may be roughly estimated to be equal to the sum of the ionization potentials of all emitted electrons (virial theorem). However, in previous experiments for 10 MeV C⁶⁺ impact, projectile energy losses were found to be about a factor of 2 larger than expected by the above consideration nearly independent of the electron multiplicity [17] (a detailed discussion can be found in [12]; the data are in good agreement with theoretical results). Assuming this to be similar for the present collision system, the maximum in the recoil-ion longitudinal momentum for i = 6 would be at $p_{R\parallel} \approx +4$ a.u. if the electrons were emitted isotropically. Instead, a strong net shift to the backward direction of about -9 a.u. is observed demonstrating that the electron sum momentum is increasingly pointing into the forward direction with increasing electron multiplicity. For i = 6 a considerable forward sum energy of the electrons of about 1.1 keV can be estimated.

The recoil-ion transverse momenta (Fig. 3) are not unambiguously connected to the emission characteristics of the electrons. The balance between the nuclear and electronic contributions has to be considered which may change with increasing electron multiplicity. Therefore the transverse momentum distributions for the different recoil-ion charge states sensitively monitors the full dynamics of the collision where the interplay of all active particles has to be taken into account. The excellent agreement of the experimental $p_{R\perp}$ distributions with theoretical results in Fig. 3 demonstrates the validity of the nCTMC approach in the regime of highly perturbative collisions. Moreover, it is an additional hint of the nonisotropic emission of electrons even in the transverse direction as predicted by theory [14]: The electrons are calculated to be preferentially ejected to the side of the incoming projectile.

The question arises whether the experimentally observed large forward directed electron sum momentum is a proof that most of the individual electrons are emitted into the forward hemisphere resulting in a collective forward motion as predicted by theory. One might assume that one fast "binary encounter" electron would yield a backward scattered recoil ion the other electrons being emitted more or less isotropically. However, as pointed out recently [20], electrons which suffer a hard binary encounter with the projectile emerge with an energy of $E_e = 2v_P^2 \cos^2 \vartheta_e$ and, thus, with a longitudinal momen-



FIG. 3. Differential cross sections for the transverse momenta of Ne^{*i*+} recoil ions. Circles: experimental data. Solid lines: results of the *n*CTMC calculation.

tum of $p_{e\parallel} = 2v_p \cos^2 \vartheta_e$. Putting this into Eq. (1) yields $p_{R\parallel} = U_n/v_P$ for this individual electron therefore contributing not at all to the observed backward scattering of the recoil ion. In order to explain the experimental results assuming one "hot" electron, the ratio of longitudinal to transverse momentum of this electron must be larger than for a binary encounter electron which seems unlikely. Therefore, independent of any theory, our experimental results provide clear evidence that most of the continuum electrons from a multiple ionization event by fast highly charged ion impact emerge collectively into the forward hemisphere.

In conclusion, we have measured the recoil-ion momentum distributions for multiple ionization of Ne by 5.9 MeV/u U⁶⁵⁺ impact using high-resolution recoil-ion momentum spectroscopy. Our results provide a first and unique test for theories calculating differential manyelectron emission cross sections. The main finding of our study is a large longitudinal electron sum momentum pointing into the forward hemisphere for all electron multiplicities with a maximum of $\sum_{n=1}^{6} p_{e||}^{n} \approx +9$ a.u. for sixfold target ionization. The data provide clear evidence that most of the electrons are emitted collectively into the forward direction as predicted theoretically. The experimental momentum distributions in longitudinal as well as in transverse direction are in excellent agreement with the theoretical results of nCTMC calculations proofing the ability of classical many-particle models to describe accurately many-electron processes in the regime of strong perturbations.

Future experimental effort will concentrate on an increase of the recoil-ion momentum resolution and on the implementation of three independent multihit capable electron detectors [23]: This way complete experiments for multiple target ionization can be envisaged in the near future. One particular goal is the experimental verification of negative projectile scattering angles predicted for the major part of multiply ionizing collisions and possible rainbow scattering of the heavy projectiles.

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