Low-Energy Electrons and Their Dynamical Correlation with Recoil Ions for Single Ionization of Helium by Fast, Heavy-Ion Impact

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(Received 22 July 1994)

Helium single ionization by 3.6 meV/u Ni²⁴⁺ impact was explored in a kinematically complete experiment by combining a high-resolution recoil-ion momentum spectrometer with a novel 4π low-energy electron analyzer. More than 90% of the "soft electrons" ($E_e \leq 50 \text{ eV}$) are ejected in the forward direction in agreement with classical-trajectory Monte Carlo predictions. The electron longitudinal momentum is not balanced by the longitudinal momentum change of the projectile but mainly by the backwards recoiling He¹⁺ ion. Energy losses of the 0.2 GeV projectiles as small as $\Delta E_P/E_P = 3.4 \times 10^{-7}$ are observable.

PACS numbers: 34.10.+x, 34.50.Fa

Until now the investigation of target single ionization by energetic ion impact has been mainly restricted to total cross section measurements and to studies differential in the momentum of only one of the outgoing particles (electron, recoil-ion, or projectile). No kinematically complete experiment that is comparable to so-called (e, 2e) experiments [1] for electron-atom collisions or $(\gamma, 2e)$ measurements [2] for photoionization has been reported for heavy projectiles.

This is partly due to the large momentum p_P carried by the heavy incoming projectile and its mostly undetectable tiny change Δp_P during the collision: For target (multiple) ionization $\Delta p_P/p_P$ typically ranges from 10^{-4} to even 10^{-9} . Only a few ion-beam facilities [3] or storage rings [4] deliver beams with a momentum spread as small as $\Delta p_P/p_P \approx 10^{-5}$. Thus, conventional energy loss or gain measurements have been limited to this level of precision. Moreover, this 10^{-5} accuracy so far has only been achieved for projectile energies below 400 keV [3].

A further serious complication arises since most electrons emitted in ion-atom collisions have energies well below 50 eV (so-called "soft electrons"). Conventional techniques to measure the angular and energy distribution of such electrons, especially for $E_e \leq 10$ eV, are faced with tremendous experimental difficulties [5,6]. Therefore, there are no systematic investigations for $E_e \leq 5$ eV in spite of the fact that these electrons considerably contribute to the total emission cross section. There exists only one recent experiment for $E_e \leq 1$ eV [6,7] where neither the final charge state of the recoiling target ion nor that of the projectile was controlled.

In order to circumvent these basic experimental problems and provide new, fundamental information on the dynamics of ionizing collisions, recoil-ion momentum spectroscopy (RIMS) has been developed over the last few years [8–10]. Using ultracold supersonic jet targets of subkelvin temperatures [cold-target recoil-ion momentum spectroscopy (COLTRIMS)], transverse and longitudinal momentum transfers to the recoiling target ion corresponding to energies as small as $\pm 30 \ \mu eV$ become observable with a solid angle of nearly 4π [10,11].

In this Letter we report both on the first kinematically complete experiment for single target ionization in an ionatom collision and on the measurement of all soft electrons down to zero emission velocities for experimentally controlled final charge states of both the projectile and the target ions. This has been accomplished by developing a high-resolution recoil-ion momentum spectrometer (longitudinal energy resolution $\Delta E_{R\parallel} \approx \pm 12 \ \mu eV$) with an integrated 4π electron analyzer ($\Delta E_{R\parallel} \approx \pm 130$ meV for $\Delta E_{e\parallel} \lesssim 10 \text{ eV}$). Six out of nine final momentum components of the particles emerging from the collision are determined. Moreover, the projectile energy loss is accessible with a resolution of $\Delta E_P/E_P = 3.4 \times 10^{-7}$ (FWHM) for the 0.2 GeV Ni projectiles. This is orders of magnitude better than achievable in any energy gain or loss measurement using conventional techniques. The position of the centroid of the projectile energy loss distribution corrected for the measured continuum energy of the emitted electron yields the difference in the binding energies between the initial and final electronic states of the target and the projectile. This centroid is determined with a precision of 10⁻⁸ corresponding to a measurement of electronic binding energies of 2 eV in the present reaction.

The experiments were performed using a 3.6 MeV/u stripped and charge state analyzed Ni²⁴⁺ beam from the UNILAC of GSI. Only a brief description of the apparatus is provided in this Letter; details will be

given in a following paper. The beam was collimated to a size of about $1 \text{ mm} \times 1 \text{ mm}$ in the target region, the charge state was analyzed after the collision, and Ni²⁴⁺ ions (no charge exchange) were recorded by a fast scintillation detector at a rate of up to 1 MHz. A single stage supersonic jet (see Fig. 1) provided a well localized helium target of 3 mm diameter and a density of about 3×10^{11} atoms/cm² at the intersection point with the ion beam. In order to improve the resolution the nozzle was cooled to about $T_0 = 30$ K (COLTRIMS [8,11]). From the jet operation parameters a longitudinal temperature (along the jet expansion and transverse to the ion beam) of 10 mK and a transverse temperature of 200 mK is expected. This results in a recoil-ion momentum resolution along the ion beam of $\Delta p_{R\parallel} \approx$ ± 0.05 a.u. $(\Delta E_{R\parallel} \approx \pm 5 \ \mu \text{eV}).$

Recoil ions created at the ion-beam jet intersection were extracted by a uniform electric field of 4.55 V/cm provided over 22 cm along the beam (total voltage applied $V_T = +100$ V; potential at the intersection point $V_I = 50$ V). After 11 cm of acceleration, the recoil ions drift over 22 cm to be focused in time. They are postaccelerated ($V_a \approx -2000$ V over 2 mm) and detected by a two-dimensional position sensitive (2D PS) channelplate detector (wedge and strip anode) of 40 mm active diameter mounted directly beneath the straight path of the projectile beam. The He1+ time of flight (TOF) in the entire apparatus was about 8 μ s measured by an electronrecoil-ion-projectile coincidence. The TOF provides the recoil-ion charge state and longitudinal momentum $p_{R\parallel}$, the position on the detector gives the transverse momentum $p_{R\perp}$ and the azimuthal emission angle.

Electrons emerging from the collision with $E_{e\parallel} \leq$ 50 eV are all ($\Delta \Omega = 4\pi$) accelerated into the opposite direction. They hit another 2D PS microchannel plate mounted at about the end of the acceleration field (12 cm flight path; this detector is horizontally displaced by 3 cm to provide space for the incoming ion beam). The electrons are postaccelerated ($V_a \approx +200$ V over 2 mm) to guarantee optimum and energy-independent detection efficiency. The total TOF is about 50 ns for $v_{e\parallel} = 0$



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

(time resolution $\Delta t \approx 1$ ns). In addition to the electric field, a nearly parallel solenoidal magnetic field of 12 G is generated by two Helmholtz coils forcing electrons of nonzero transverse energies to spiral trajectories. The electron longitudinal momentum is obtained from the TOF, the position gives the transverse momentum along with the azimuthal emission angle. The magnetic field is slightly tilted with respect to the direction of the ion beam to guide electrons of zero transverse energy out of the ion path onto the detector.

The longitudinal sum momentum of the recoil ion and the electron is well defined from energy and momentum conservation [see one of the following paragraphs and Eq. (1)]. Therefore, it can be controlled experimentally, and a value of $(\Delta p_{e\parallel}^2 + \Delta p_{R\parallel}^2)^{1/2} = \pm 0.11$ a.u. was achieved (see Fig. 3). The longitudinal recoil-ion energy resolution is $\Delta E_{R\parallel} \leq \pm 12 \ \mu \text{eV} \ (\Delta p_{R\parallel} \leq \pm 0.08 \ \text{a.u.})$, the lowest value ever reported. The transverse resolution is about $\Delta p_{R\perp} = \pm 0.25$ a.u. and is limited by the size of the spatial ion-beam-target-beam overlap of $1 \times$ 1 mm². The electron energy resolution in the longitudinal direction is $\Delta E_{e\parallel} \leq \pm 130 \text{ meV} (\Delta p_{e\parallel} \approx \pm 0.1 \text{ a.u.})$. In the transverse direction the resolution is $\Delta E_{e\perp}/E_{e\perp} =$ $2\Delta x/R$ for electrons being on the maximum distance away from the center (electrons with $E_{e\perp} = 0$) when hitting the detector (R is the diameter of the cyclotron motion; Δx is an extension of the ion beam of 1 mm). Thus, an energydependent accuracy of $\Delta E_{e\perp}/E_{e\perp} = [(0.13 \text{ eV})/E_{e\perp}]^{1/2}$ is obtained ($E_{e\perp}$ in eV). The electric and magnetic fields were chosen in such a way that more than 90% of all electrons hit the detector between two focus points of the cyclotron motion guaranteeing a total resolution of $\Delta E_{e\perp} \lesssim 400 \text{ meV}.$

This technique removes many of the experimental difficulties of conventional electron spectrometers: First, the target extension is well defined by the supersonic jet.



FIG. 2. Sum of the electron and the recoil-ion longitudinal momentum which is minus the projectile momentum change $\Delta p_{P\parallel}$: $p_{e\parallel} + p_{R\parallel} = (Q + E_e)/v_P = -\Delta p_{P\parallel}$. Upper scale: $\Delta p_{P\parallel}$ relative to the incoming projectile momentum p_P . Histogram: experiment. Full line: theory (CTMC; arbitrarily normalized).

Second, electrons from rest-gas ionization are completely suppressed. Third, as for the recoil ions, the influence of electric fringe fields and magnetic distortions is drastically reduced by extracting the electrons. The energy resolution is excellent and nearly independent of the solid angle. Moreover, due to the simultaneous recoil-ion detection, the coincident final target charge state is measured, the longitudinal momentum resolution for the electrons and the recoil-ions is precisely controlled, and the spectra are free of background on the level of 10^{-4} . We emphasize that the resolution and the energy acceptance of such a soft-electron spectrometer can be significantly and easily enhanced in the future by elongation of the electron flight path, the use of time-focusing geometry in the direction of extraction, and implementation of large active-diameter 2D PS electron detectors.

Absolute cross sections are easily obtained since practically all reaction products are simutaneously detected. Thus, the sum of all events can be normalized on the total single ionization cross section of $\sigma^{1+} = (2.4 \pm 0.5) \times 10^{-15} \text{ cm}^2$ measured for the 3.6 MeV/u Au²⁴⁺ impact [12]. Theoretical cross sections are $\sigma^{1+} = 1.6 \times 10^{-15}$ cm² [classical-trajectory Monte Carlo (CTMC) calculations] and $\sigma^{1+} = 2.6 \times 10^{-15} \text{ cm}^2$ [continuum-distorted-wave-eikonal initial state (CDW-EIS) calculations; [12])]. The single differential electron emission cross sections (integrated over all emission angles) for helium single ionization $(d\sigma^{1+}/dEe)$ are displayed in Fig. 4 (full circles) in comparison with theoretical results (full line). A possible maximum is observed at electron energies of about 2 eV. Above 4 eV at the differential cross section decreases rapidly and falls off by a factor of 10 at $E_e \approx 40$ eV. Our data cannot be directly compared to results of any other measurements, since in previous investigations at low electron energies neither the final charge state of the projectile nor that of the target was controlled. A similar dependence has been observed for 1 MeV/u O⁸⁺ on He collisions [13,14] for $E_e > 4 \text{ eV}.$



FIG. 3. Differential electron emission cross sections (DCS) for helium single ionization $d\sigma^{1+}/dEe$ in collisions with 3.6 MeV/u Ni²⁴⁺ as a function of the electron energy.

In Fig. 2 the longitudinal electron momentum distribution (full circles) is shown, and a strong asymmetry along the forward-backward direction is found. About 93% of all soft electrons are emitted in the direction of the outgoing projectile. This behavior is correctly predicted by the theory (full line; to enable a better comparison the theoretical data are multiplied by a factor of 1.6). Inspection of the projectile longitudinal momentum change $\Delta p_{P\parallel} = -(p_{R\parallel} + p_{e\parallel})$ yields a sharp peak with its center close to zero (see Fig. 3). Thus, $p_{e\parallel}$ is not balanced, as might be expected, by the projectile momentum transfer, but is nearly completely compensated by the recoil ion (open circles in Fig 2): They are emitted backwards with a most probable energy of 300 μ eV. Surprisingly, the longitudinal momenta of the helium-atom "fragments" (the electron and the He¹⁺ recoil ion) are considerably larger than the net-momentum transfer to the target atom by the projectile: The atom seems to "dissociate" in the strong, long-ranging projectile potential. In order to elucidate this behavior in more detail CTMC calculations have been performed using antinickel \overline{Ni}^{-24} as a projectile: In this case electrons are preferentially ejected backwards, whereas the recoil ion is found in the forward direction.

For collisions with small energy and momentum transfer (perfectly fulfilled here at a level below 10^{-7}), it follows from momentum and energy conservation for the longitudinal momentum balance (all in atomic units)

$$p_{R\parallel} = (Q + E_e)/v_p - \sum p_{e\parallel}.$$
 (1)

Q is the total change in potential energy, which for single ionization of He(1s²) simply is the binding energy of 0.903 a.u.; the continuum energy of the emitted electron E_e is typically about 1 a.u.; and v_P is the incoming projectile velocity of 12 a.u.



FIG. 4. Longitudinal momentum distribution $d\sigma^{1+}/dp_{\parallel}$ of soft electrons (full circles) and recoil ions (open circles) for single ionization of helium in collisions with 3.6 MeV/u Ni²⁴⁺. Full and dashed lines: results of CTMC calculations multiplied by a factor of 1.6.

Thus, the $(Q + E_e)/v_P$ term in Eq. (1) is only on the order of 0.2 a.u., showing that the longitudinal recoil-ion momentum mirrors the (sum) momentum of the emitted electron(s) for single (multiple) ionization at large v_p : $p_{R\parallel} \approx -\sum p_{e\parallel}$. This provides a convenient, sensitive, and novel experimental tool for the investigation of electron-electron interactions in multiple ionization, a subject of considerable current interest. In the present experiment He²⁺ production has been measured simultaneously, the results will be reported elsewhere.

The center of the sum-momentum peak is slightly shifted from zero by $Q/v_P = 0.075$ a.u. Its asymmetry which is clearly visible only in the theoretical distribution (full line, arbitrarily normalized) is a result of the energy distribution of the continuum electrons (E_e/v_P) . Since the electron energy is measured simultaneously, this term is known and can be subtracted for a precise determination of Q. Thus, with the present setup, electronic binding energies are detectable with a FWHM of $0.22 v_P$ a.u. This corresponds to an energy resolution of 72 eV for this collision system and to an energy loss measurement of the projectile with a relative accuracy of $\Delta E_P/E_P \approx 3.4 \times 10^{-7}$ (FWHM). Using a colder supersonic jet or an ultracold laser produced atomic target $(T \leq 100 \ \mu \text{K})$, an improvement by a factor of 10 seems to be realistic. Then, for instance, electronic binding energies of heavy few-electron systems (U⁹¹⁺, Pb⁸¹⁺, etc.), which are available at the GSI experimental storage ring (ESR) at energies as low as 10 MeV/u can be measured with a resolution of 10 eV (FWHM).

In conclusion, we have performed the first kinematically complete experiment on single ionization by heavyion impact combining high-resolution COLTRIMS with a novel low-energy electron spectrometer. The main findings of our work are the dramatic forward emission of the soft electrons, the nearly complete compensation of the electron longitudinal momentum by the recoil ion with a comparably small momentum change of the projectile. Moreover, a possible maximum in the soft electron energy distribution is observed at about 2 eV, realized in the coincident measurement of the charges of both heavy particles. The longitudinal recoil-ion momentum mirrors the (sum) momentum of the emitted electron(s) for single (multiple) ionization at large projectile velocities: $p_{R\parallel} \approx -\sum p_{e\parallel}$. This provides a convenient experimental method for future investigations of electron-electron correlations in multiple ionization events. It was demonstrated that the technique provides the possibility of energy-loss (gain) measurements of atomic reactions in heavy-ion atom collisions with so-farunachieved resolution: State-selective studies of electron capture, the kinematically complete investigation of resonant transfer and excitation or radiative electron capture, and the spectroscopy of electronic states in heavy fewelectron ions can be envisaged in the near future.

Finally, we want to emphasize that the coincident, precise, and complete measurement of the momenta of both the electron and recoil ion using advanced ultracold targets provides a realistic experimental approach to determine angular correlations and even the neutrino mass in β -decay experiments [8].

One of us (P. J.) would like to thank GSI for generous hospitality and the European Community for financial support. The support of NSF INT-9112815 is gratefully acknowledged (R.E.O.). We want to thank C. L. Cocke for many contributions to the design of the spectrometer and M. Horbatsch for many fruitful discussions. The help of W. Enders during the installation of the apparatus and the help of the UNILAC crew for providing excellent beams is gratefully acknowledged. This work was supported by Gesellschaft für Schwerionen Forschung, Bundesminister für Forschung und Technologie, and Deutsche Forschungsgemeinschaft.

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