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Differential Projectile Energy Loss in Multiply Ionizing Collisions

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The momentum vectors of the recoil ion and up to three electrons were measured in coincidence with the projectiles which did not change charge state for 3.6 MeV/amu Au⁵³⁺ → Ne collisions. New techniques were applied to obtain differential energy loss spectra for multiple target ionization as a function of the recoil ion charge state. A resolution unprecedented for this energy regime was achieved. The data are in good agreement with a CTMC calculation. Our studies represent a first step for a method of modelling stopping powers on a microscopic level and are relevant for plasma applications.

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For decades it has been argued that data obtained from ion-atom collision research are highly relevant for applications e.g. in plasma physics, interactions of ions with solids and surfaces etc. Important parameters required to understand such interactions are, for example, the charge state distribution, the energy loss, or the intensity of radiation when ions pass through matter. These are quantities which, under certain conditions, are routinely obtained in ion-atom collision studies (e.g. 1-3). However, it is not always clear how to establish a connection between a quantity measured under relatively simple conditions controlled to a large extent by the investigator (e.g. collisions in dilute gases) and the same quantity one needs to know for the significantly more complex conditions encountered e.g. in a dense plasma (4,5) or in a solid (6).

One particularly important parameter for ions interacting with both plasmas or solids is the energy loss. The normal procedure for theoretical models to calculate, for example, the stopping power of solids is to first calculate the energy loss spectra for ions interacting with single atoms and then to convolute these energy-loss spectra over many collisions which the ions suffer in a solid (e.g. 7-12). Among others, there are three major challenges these calculations have to deal with: first, the equilibrium charge state of the ion resulting from the competition between capture and stripping processes is difficult to determine for solids. Second, in a solid the impact parameter range is significantly restricted compared to single collision conditions (to approximately half the lattice constant) because the atomic layer closest to the ion shields to a large extent the more distant layers. Therefore, in order to perform the convolution over many collisions, the impact parameter dependence of the energy loss spectra has to be known accurately. Third, in this impact parameter range multiple ionization in a single collision dominates over single ionization for highly charged, energetic ion impact. The energy loss spectra therefore need to be known as a function of the recoiling target ion charge state. Both sets of information cannot be obtained from stopping power or energy loss measurements in solids (e.g. 13-22). This makes a critical test of theoretical

models difficult, which, in turn, is important to thoroughly understand the underlying mechanisms on a microscopic level.

In a plasma the ion energy loss is important in order to determine the energy balance, i.e. the plasma temperature (23). However, the temperature is not only affected by the total energy loss, rather it could potentially depend sensitively on which process caused the energy loss. For example, if excitation followed by radiative decay occurs, the corresponding energy loss is transferred almost exclusively to the emitted photons which, in turn, interact comparatively weakly with matter and thus can escape the plasma with high probability. In an ionization process, in contrast, a significant fraction of the energy loss is transferred to the ionized electrons which interact much more strongly with matter than photons and are therefore less likely to escape the plasma. Consequently, one might expect a larger cooling effect due to excitation followed by radiative decay than due to ionization. This means that the relative contributions from ionization and excitation followed by radiative decay to the total energy-loss spectra need to be known accurately.

Numerous energy-loss measurements have been performed for dilute gases, i.e. under single collision conditions (e.g. 1,24-27). However, some of these experiments were restricted to excitation or single ionization (1,26,27), for others the accuracy of the data was significantly limited by the experimental energy-loss resolution (24 and 25). All of these studies were not kinematically complete. To the best of our knowledge, there were only two kinematically complete experiments reported on double ionization (28,29) for which, however, the projectile energy loss was not analyzed. No kinematically complete experiments were performed for a higher degree of ionization.

In this letter we report on the first kinematically complete experiment on triple ionization as well as the first measurements of electron momentum spectra coincident with the momentum analyzed recoil ions for charge states as high as $6+$. Furthermore, we present a new method to determine highly differential energy-loss spectra for multiply ionizing collisions, which

represents a first crucial step toward modelling stopping powers on a microscopic level. The second important step is to determine the impact parameter dependence of the energy-loss spectra which will be discussed in a forthcoming paper. At the same time the method allows one to separate the contributions from ionization and excitation followed by radiative decay to the total energy loss spectrum. Here, the method is demonstrated for heavy ion impact, however, it can also be applied to other projectiles like e.g. electrons.

The experimental techniques have been reported previously (30) and only the salient points are repeated here. A Au^{53+} beam from the UNILAC at GSI Darmstadt collided with very cold ($< 1\text{K}$) Ne gas target atoms from a supersonic jet. The initial projectile energy of 3.6 MeV/amu was chosen to fall within a regime, where stopping powers usually maximize. The recoiling target ions were extracted by a weak electric field (1.36 V/cm) and their charge state and complete momentum vector were determined using positron sensitive detectors and time of flight techniques. With the same field the ionized electrons were extracted in the opposite direction: a uniform magnetic field of 20 G confined the transverse electron motion. Up to three electrons from the same collision were detected simultaneously with a single detector using a multi-hit technique. For each electron the complete momentum vector was obtained. The projectiles which did not change charge state in the collision were measured in coincidence with both the recoil ions and all detected electrons.

In the following we will discuss projectile energy loss due to multiple ionization only and compare it with the total projectile energy loss. Since for each detected electron the complete momentum vector was recorded, the continuum energy of up to three ionized electrons were obtained simultaneously for each collision. This information is sufficient to determine the projectile energy loss due to single, double, or triple ionization. Energy conservation yields

$$K_0 = K_f + K_{\text{rec}} + \Sigma K_e - \Sigma I_e \quad (1)$$

where K_0 and K_f are the initial and final projectile kinetic energies, ΣK_e is the summed kinetic energy of all ionized electrons and ΣI_e is the ionization potential for the removal of Q

electrons from the target. Even though the recoil energy K_{rec} has been measured as well, it can safely be neglected even for close collisions. The projectile energy-loss due to Q -fold ionization $\Delta E_p^i = K_0 - K_f$ is thus simply given by the sum of ΣK_e , which was measured for up to $Q=3+$, and ΣI_e for the corresponding Q . It should be emphasized that the energy loss due to excitation of either the target or the projectile followed by radiative decay is not included in ΔE_p^i .

In Fig. 1 we show the measured spectra of ΔE_p^i for double (upper part) and triple ionization (lower part) as open symbols. For comparison, the crosses represent an independent electron model. Here, ΣK_e was obtained by convoluting the measured electron energy spectrum for single ionization Q times with itself, however, for ΣI_e the correct value of the corresponding Q was used. Not surprisingly, the independent electron model is in poor agreement with the directly measured data. Not only are possible electron correlation effects ignored in the independent electron model, it also does not account for closer collisions required in multiple compared to single ionization.

The closed symbols in Fig. 1 show the ΔE_p^i obtained from a similar convolution procedure as was used in the independent electron model. However, here the electron energy spectrum of a single electron measured in coincidence with the recoil ions with corresponding charge state $Q+$ instead of the one for single ionization was convoluted Q times with itself. It should be noted that this model accounts at least partly for both electron correlation effects and the requirement of closer collisions with increasing degree of ionization since the measured spectrum of a single electron for Q -fold ionization is affected by both factors. For simplicity, in the following we refer to this model as the average electron model. As can be seen from Fig. 1 the average electron model is in excellent agreement with the directly measured data for double and triple ionization.

Since the average electron model appears to work so well for double and triple ionization, it is reasonable to assume that it also provides an adequate description for a higher degree of ionization. It was therefore applied to create the spectra for ΔE_p^i for charge states 1+ to 6+, which are shown in Fig. 2. The data were normalized to measured total cross sections summed over all charge states for 5.9 MeV/amu $U^{65+} \rightarrow Ne$ (31), which corresponds to the same perturbation Q/v_0 within better than 5%. For the sake of a clear presentation, the data were divided by 10^9 . An additional test of the average electron model was performed for Ne^{6+} . Here, the summed kinetic energy spectrum of three detected electrons was convoluted once with itself. The spectrum of ΔE_p^i obtained from this procedure is in perfect agreement with the average electron model.

The curves in Fig. 2 show n-body Classical Trajectory Monte Carlo (CTMC) calculations (9,32). The calculated magnitudes are absolute, however, in Fig. 2 they were multiplied by a factor of 3.5 for a comparison in shape with the data. Even though the calculation is too low in magnitude, it is in reasonable agreement with the shape of the measured spectra. For low charge states the calculated spectra fall off somewhat steeper with increasing energy loss and for high charge states somewhat shallower than the experimental spectra. For $Q=3,4+$ the agreement is nearly perfect.

As mentioned above, the energy loss obtained from the summed electron energies does not include excitation followed by radiative decay. To determine the total energy-loss spectra (electron capture is negligible for this collision system) requires an analysis of the momentum balance of the collision in the longitudinal direction (defined by the initial projectile beam direction). Momentum conservation yields:

$$\Delta p_{pl} = -p_{rec} - \Sigma p_{el} \quad (2)$$

where Δp_{pl} , p_{rec} , and Σp_{el} are the longitudinal component of the momentum difference vector of the projectile, the recoil ion, and the summed electrons, respectively. Furthermore, for small projectile scattering angles (which is always fulfilled for heavy ion impact), the total

projectile energy loss ΔE_p^i can be expressed in terms of the change in longitudinal momentum:

$$\Delta E_p^i = K_0 - K_f = (p_{p0}^2 - p_{pf}^2)/2M \approx -\Delta p_{pl} v_0 \quad (3)$$

Where M is the projectile mass and v_0 the initial projectile speed. Finally, substituting (2) in (3) yields:

$$\Delta E_p^i = (p_{recl} + \Sigma p_{eti}) v_0 \quad (4)$$

Since the complete momentum vectors of the recoil ion and up to three electrons were measured simultaneously, the total energy loss could be determined up to $Q=3$ and is shown for Ne^{2+} (open squares) and Ne^{3+} (open circles) in Fig. 3, respectively. For comparison, the ΔE_p^i are shown again as closed symbols for both cases. If the cross sections would exhibit a step function at the threshold for Q -fold ionization (62.7 eV for $Q=2$ and 126.7 eV for $Q=3$), then the shape of the spectra at the threshold would be a direct measure of the energy-loss resolution. Since the cross sections do not have this step function characteristic in a strict sense, the shape of the spectra near the threshold provides an upper limit for the energy-loss resolution. For ΔE_p^i we find a resolution of less than 150 eV full width at half maximum (FWHM), which is significantly worse than the estimated value of 20 eV FWHM for ΔE_p^l . However, even the resolution in ΔE_p^l is an order of magnitude better than in previous studies (24,25) of multiple ionization in the MeV/amu energy regime.

The difference in resolution is mainly responsible for the different shapes of the spectra for ΔE_p^i and ΔE_p^l near the threshold. For a sufficiently large energy loss above the threshold, in contrast, the shape of the continuous energy-loss spectrum for ionization should no longer be affected by the resolution. Therefore, if excitation followed by radiative decay is negligible, the spectra for ΔE_p^i and ΔE_p^l should become identical above a certain energy loss. Indeed, these spectra are in very good agreement after approximately 150 to 200 eV above threshold

for both charge states. This may be taken as an indication that excitation followed by radiative decay does not significantly contribute to the total energy-loss spectrum.

The present resolution in $\Delta E_p^{\dagger}$ is not sufficient to allow for more quantitative conclusions about the contributions from excitation $\Delta E_p^{\dagger}$, which can in principle be determined from the difference $\Delta E_p^{\dagger} - \Delta E_p^{\ddagger}$. The accuracy in $\Delta E_p^{\dagger}$ is limited by the recoil ion momentum resolution according to $d(\Delta E_p^{\dagger}) = v_0 d(p_{\text{rl}})$, which currently is approximately 0.4 a.u. for Ne. In future, the experimental set-up can be further optimized so that a resolution of about 0.1 to 0.2 a.u. should be achievable. Therefore, the experimental techniques discussed in this Letter offer a promising and possibly the only method to study energy losses separately for various collision processes.

In summary, we have performed the first kinematically complete experiment on triple ionization. Furthermore, we have determined energy-loss spectra for multiple ionization with a resolution, which is unprecedented in the MeV/amu energy regime. This energy regime, in turn, is particularly important for stopping powers of solids. Determining the energy loss spectra as a function of charge state is one important step in modelling stopping powers. The second important step is to study the impact parameter dependence of the energy-loss spectra, which will be discussed in a forthcoming paper.

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Figure Captions

Fig. 1 : Projectile energy-loss spectra for Ne^{2+} (upper part) and Ne^{3+} (lower part) due to multiple ionization only. The open symbols represent the directly measured data, the crosses the independent electron model, and the closed symbols the average electron model (see text).

Fig. 2: Projectile energy-loss spectra (due to multiple ionization only) obtained from the average electron model (see text) for Ne^{Q+} . The curves show a CTMC calculation, which was multiplied by a factor of 3.5.

Fig. 3: Total projectile energy-loss spectra (open symbols) for Ne^{2+} (upper part) and Ne^{3+} (lower part) in comparison with the energy-loss spectra due to multiple ionization only (closed symbols).

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