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LETTER TO THE EDITOR

Dependence of binary encounter electron production on the charge state of the recoil ion

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Abstract. We study the dependence of the production of binary encounter electrons on the charge state of recoil ions for 2.4 MeV u^{-1} Xe^{21+} on He and Ar. Doubly differential cross sections of electron emission are calculated with the nCTMC method. We find that the contributions to the binary electron cross section from various recoil ion charge states reach a maximum near Ar^{5+} for the Ar target. In the case of the He target, double ionization dominates over single ionization for all ejected electron energies above 100 eV. However, an unexpected local drop of double ionization in the binary peak region has been observed. This decrease is found to be related to a two-step, sequential removal of the two electrons.

In the past few years, substantial progress has been made in the understanding of ionizing collisions involving partially-stripped, highly-charged ions. Non-coincidence experimental measurements of the ejected electron spectra have been the primary source of information on the collision dynamics. However, due to the complexity of the problem, combined experimental and theoretical studies have for the most part focused only on a subset of the electron emission spectrum usually referred to as the binary encounter (BE) electrons (Wolff *et al* 1992).

The origin of the well known BE electron production can be traced to the simple classical head-on collision (McDowell and Coleman 1970) between the heavy projectile ion travelling at speed V_p and the target electron. Assuming a quasi-free electron at rest, the electron acquires an energy of $2V_p^2\cos^2\theta$, where θ is the electron ejection angle. In fast collisions ($V_p \gtrsim 5$ au), the BE electrons are well separated from the slow electrons in momentum space. Therefore, the study of the BE electrons serves as a good starting point in the delineation of the complex ionizing mechanisms for heavy ion impact. Although some unique features (such as the double peak structure) of BE electrons by partially-stripped, highly-charged ion impact have been understood qualitatively (Richard *et al* 1990, Reinhold *et al* 1991), recent systematic studies have revealed large discrepancies between experimental data and theoretical predictions, most notably in the position and width of the binary peak, and the relative height of the double peaks and the observed target dependence (Shinpaugh *et al* 1993, Wolff *et al* 1992). In addition to the internal structure of the projectile, many other factors are attributable to the observed disagreements between experiment and theory, including strong prior- and post-collision distortions, multiple ionization, and two-centre effects (Crothers and McCann 1983, Fainstein *et al* 1988, Want *et al* 1992).

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In this letter, we present a first theoretical investigation of the dependence of the binary encounter electron production on the charge states of the recoil ion. The ejected electron spectrum will be studied in coincidence with the recoil ion charge state in order to provide further insight on the relative importance of the aforementioned factors that determine the electron production mechanisms. As will be seen later, this approach can also be viewed as qualitatively equivalent to the determination of the impact parameter range of the collisions. Thus the information derived may be directly compared to experimentally measurable quantities.

We show that for a heavy target such as Ar, the contributions to the BE electron production cross section reach a maximum near a recoil ion charge state of +5, which is surprisingly low compared to the highest possible charge state of 18. In addition, we find evidence that the atomic shell structure of the Ar target atom has a profound effect on the charge state distribution. For the He target, an unexpected drop of double ionization and a corresponding local enhancement of single ionization are observed at the binary peak. This is attributed to the sequential removal of the target electrons. Although our study is motivated primarily toward a better understanding of the ionization mechanisms in heavy ion–atom collisions, it also provides a framework which can be adapted to the production of fast electrons in other areas including ion–solid interactions. Our results suggest interesting implications to the fundamental aspects of double ionization in the non-perturbative regime where a two-step independent collision mechanism is thought to be important (Tanis *et al* 1992).

When we consider target ionization by highly charged ion impact, the Massey parameter Z_p/V_p, a measure of the perturbation strength, is large. Clearly, a perturbative method such as a Born approximation is inapplicable. In addition, the many-body problem requires a theoretical description that can adequately take into account the evolution of the many particles interacting among themselves. We have used the many-body classical trajectory Monte Carlo (*n*CTMC) method, which is particularly suitable for the task at hand. Here, not only is the perturbation large, the momentum transfers are also large for the BE electrons under consideration. Those are the criteria under which a classical description remains valid (McDowell and Coleman 1970). We note, however, that although the quantal interference effect in the ejected electron spectrum is not present in a classical description, it appears only at a specific angle and is relatively weak in a fast collision (2.4 MeV u^{-1} in this case).

Details of the *n*CTMC method can be found elsewhere (Olson *et al* 1989). Briefly, the initial conditions of the collision system are sampled at random from an ensemble approximating the phase space distribution of the quantum-mechanical initial state. The system is subsequently evolved according to its many-body classical Hamiltonian. Except for the adjustments of the ionization potentials of the target electrons, the collision process is treated within the independent electron model (McGuire and Weaver 1977). At the end of the evolution when the free particles are sufficiently far apart, the reaction is determined and analysed.

We present in the following our study for collisions of 2.4 MeV u^{-1} ($V_p = 9.8$ au) Xe^{21+} on Ar and He targets, respectively. Before proceeding to the numerical results, we note that the partially stripped projectile Xe^{21+} is structured. In the *n*CTMC calculation, the structured projectile is regarded as a structureless particle with a frozen core. The projectile electrons have only passive screening effects which can be expressed in terms of an effective potential (Garvey *et al* 1975). Clearly, effects due to explicit electron–electron interaction (correlation or anti-screening) which can cause simultaneous excitation of the projectile electrons are neglected in the independent electron model (Wang *et al* 1992, McGuire *et al* 1981). Since the projectile electrons of Xe^{21+} in the ground state are tightly bound, the

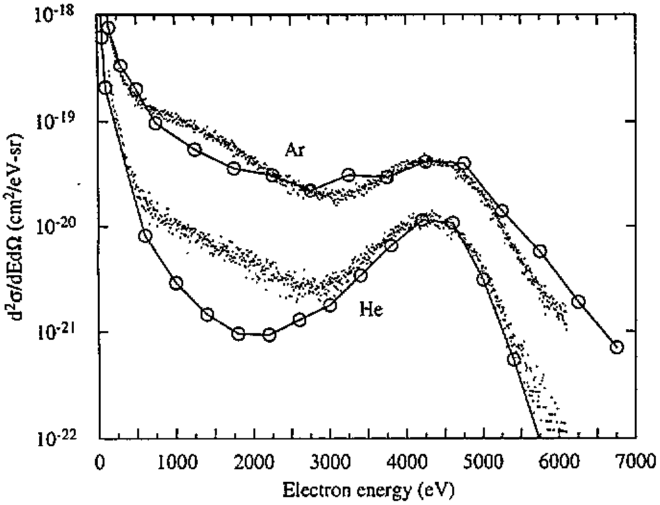

Figure 1. The total doubly differential cross section of electron emission summed over all possible recoil ion charge states for 2.4 MeV u^{-1} Xe^{21+} on Ar and He, respectively, at 20°. Full curve, theory; dots, experiment (non-coincidence data).

omission of correlation effects is not expected to cause substantial difference in the electron emission spectrum near the binary peak.

The doubly differential cross sections (DDCS) of electron emission are displayed in figure 1 for 2.4 MeV u^{-1} Xe^{21+} on Ar and He, respectively, at 20°. Both the *n*CTMC results and the experimental data represent the total DDCS summed over all possible recoil ion charge states. The experimental set-up and non-coincidence measurements are described elsewhere (Shinpaugh *et al* 1993). The relative experimental data have been normalized to the theoretical values at the binary peak with the same normalization factor used for both Ar and He. There is reasonably good agreement between theory and experiment around the binary peak (electron energy $\gtrsim$ 3 keV). The stripping of projectile electrons has not been included in the calculations so the theoretical results underestimate the electron production in the region of the electron loss to continuum peak of ~ 1300 eV.

In figure 2 we show the dependence of binary encounter electron production on the charge state of the recoil ion for 2.4 MeV u^{-1} Xe^{21+} on Ar. The contributions to the production cross section are expressed as fractional percentages for each recoil ion charge state evaluated right at the binary peak of the electron spectra at three ejection angles 10°, 20° and 40°. The corresponding peak positions at the three angles are 4750 eV, 4250 eV and 2750 eV, respectively. The peak energies follow approximately the classical $2V_p \cos^2\theta$ law. Due to the binding energy and two-centre effects, the exact peak positions are slightly below the classical values for a free electron at rest.

Each curve in figure 2 peaks at 5, indicating that the largest contribution to the BE electron cross section occurs at a recoil ion charge state of +5. This value is so remarkably low that it is even less than $\frac{1}{3}$ of the largest possible charge state 18. However, figure 2 also indicates that when a binary electron is created, the target is mostly multiply ionized, as the fraction for singly charged recoil ion is rather small. This is to be expected for two reasons: the projectile Xe^{21+} exerts a strong perturbation capable of knocking many electrons off the target; and that binary electron production is most effective at small impact

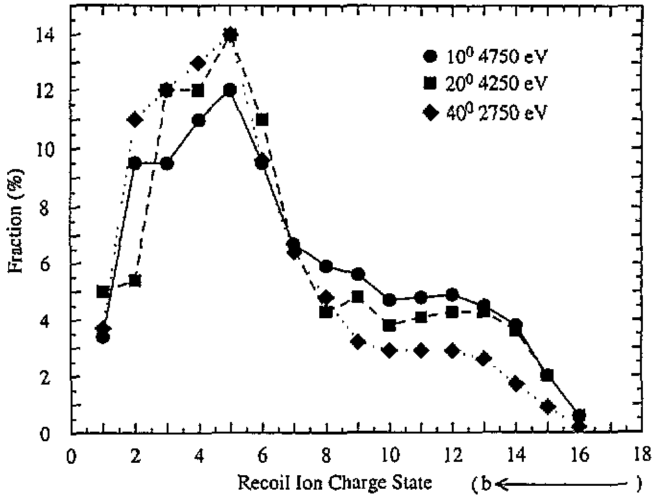

Figure 2. Fractional distribution of the binary encounter electron cross section as a function of the charge state of the recoil ion for 2.4 MeV u^{-1} Xe^{21+} on Ar at three ejection angles. Full circle, 10°; full square, 20°; full diamond, 40°. The energies indicated in the figure are the binary peak positions at the three angles, respectively, where the fractions are evaluated.

parameters when the projectile penetrates deeply into the target charge cloud. Both factors favour multiple ionization events.

For a given projectile, the structures seen in figure 2 as a function of the recoil ion charge state are best understood in terms of the latter reasoning. At large impact parameters, the momentum transfers are mediated by distant 'soft' collisions resulting in slow electrons. As the impact parameter decreases, the probability for ejecting a BE electron increases, as the collision becomes more violent (Rudd and Macek 1972). When the impact parameter decreases further such that it is comparable to the radius of the outermost shell (M shell) of the Ar atom, a burst of binary electrons is expected to be released. As figure 2 shows, there is a very rapid increase of the fraction at a peak charge state of +5. The peak value is less than the charge state 8 which would be obtained by a complete removal of the M-shell electrons. This is due to the fact that as M-shell electrons are being removed, the remaining electrons in the shell shift to lower energy states (with higher ionization potentials) that are harder to be ejected. If the impact parameter is comparable to the radius of the L shell, one would again predict an increase in the fractions. Obviously, figure 2 does not show a peak corresponding to the L shell. But it does display a shoulder structure near 12. This is because L-shell electrons are more deeply bound and are more difficult to knock out than those of the M shell. Furthermore, because of the stronger binding, the momentum spread of these electrons is large. These two factors combine to yield a broad shoulder structure rather than a peak.

From these discussions, it is clear that the recoil ion charge states correspond directly to the impact parameter range of the collision. By measuring the binary electron in coincidence with the recoil ion, one gains direct information on the impact parameter being probed. It affords a much more sensitive test on the current understanding of ionization involving highly charged ions. Unfortunately, no experimental data on the coincidence measurements are currently available for a direct comparison between theory and experiment.

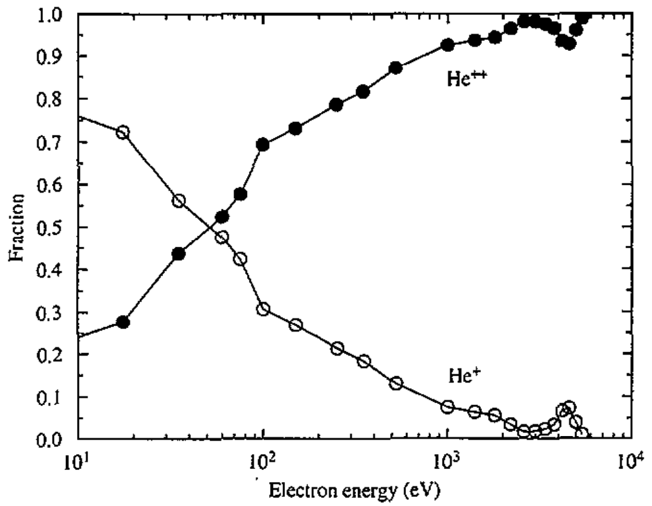

Figure 3. Fractional distribution of the binary encounter electron cross section as a function of the ejected electron energy for 2.4 MeV u^{-1} Xe^{21+} on He at 20°. Open circle, single ionization He^{+}; full circle, double ionization He^{2+}.

We now turn our attention to the case of the light He target. Figure 3 displays the fractional distribution of single and double ionization as a function of the ejected electron energy for 2.4 MeV u^{-1} Xe^{21+} on He at 20°. At low electron energies, single ionization dominates over double ionization, as expected. As the electron energy increases, the situation is reversed. For electron energies greater than 1000 eV, double ionization is dominant. The same reason which caused the charge state of Ar to increase (see figure 2) is responsible for the dominance of double ionization as the impact parameter decreases. However, as the electron energy approaches the binary peak energy, the double ionization fraction starts to drop unexpectedly, reaching a local minimum right at the binary peak. It eventually recovers and levels off at large energies. Accordingly, the fraction for single ionization has a local enhancement at the binary peak.

The decrease in double ionization near the binary peak (figure 3) can be explained in terms of a two-step, sequential removal mechanism. When one electron is removed from the He target, the remaining electron is suddenly left in the field of the He^{2+} nucleus. The increased binding makes the second electron somewhat more difficult to be ionized. The momentum distribution of this electron, characterized by its Compton profile, also has a broader width of 2 (au), instead of 1.7 as determined from a simple variational principle calculation for the first electron (Bethe and Salpeter 1957). Since at the binary peak, the portion of the electron momentum distribution being probed is at the centre of the Compton profile (~ stationary electrons), the intensity for double ionization must decrease because of its broader distribution. Moreover, because of this effect the absolute magnitude of the binary peak cross section is found to decrease by ~ 10%, making quantitative comparisons to absolute measurements a very severe test of theory.

To confirm this idea, an estimate has been carried out in a quantal impulse approximation (Wang *et al* 1991). It is assumed that the two target electrons are removed sequentially. Except for the adjustment of the width in the Compton profile of the second electron (2 instead of 1.7), the two target electrons are assumed to be independent. Figure 4 shows the

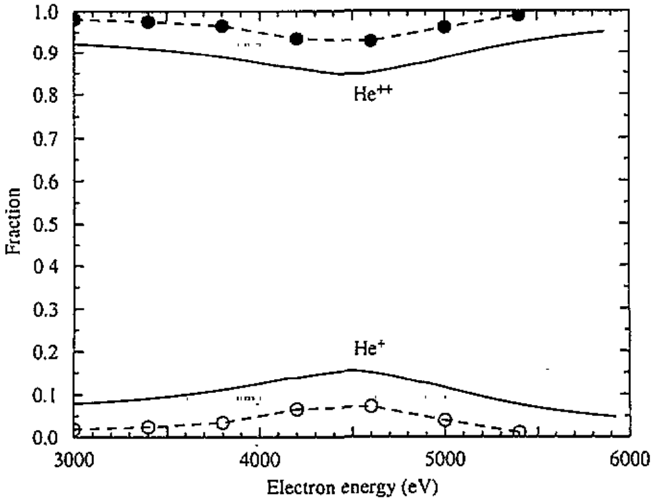

Figure 4. Same as in figure 2, except the fractions are calculated from the impulse approximation (IA). Full curve, IA (see text); broken curve, *n*CTMC, taken from figure 3.

fractional ratio obtained from the impulse approximation for the same collision system as in figure 3. The local decrease of double ionization and its corresponding local enhancement of single ionization are indeed present and are in qualitative agreement with the *n*CTMC results. We stress that our impulse approximation calculation is only for the purpose of showing qualitatively the effects of change in the width of the Compton profile.

Both the *n*CTMC and the impulse approximation calculations provide strong and clear evidence that double ionization by highly charged ions occurs through a two-step, independent collision mechanism (Tanis *et al* 1992). It suggests that future investigations about the fundamental aspects of double ionization should emphasize how to improve this mechanism when the perturbation is large.

In conclusion, the production of binary encounter electrons by impact of highly charged ions has been studied in terms of the charge states of the recoil ion. It is shown that the recoil ion charge state distribution serves as a direct probe into the impact parameter. The shell structure of the target atom plays an important role in the collision process. We find clear evidence of the two-step mechanism responsible for double ionization in the non-perturbative regime. The sequential removal process is expected to yield a slightly lower cross section at the binary peak than two completely independent collisions. It is hoped that in the near future experimental data from coincidence measurements will be available that will serve as a sensitive test on our current understanding of this complex problem.

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