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Doubly differential cross sections for single and multiple ionization of Ne by electron impact

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We present doubly differential cross sections for single and multiple ionization of the outer shell of neon by 750 eV electron impact. The distinction between single and multiple ionization was achieved by performing a charge state analysis of the recoil ions in coincidence with forward scattered, energy analyzed electrons. By a comparison to photon impact data, the contribution of the second-order double ionization mechanism is estimated and found to be negligible at this impact energy. Following a similar procedure adopted by J. A. R. Samson [Phys. Rev. Lett. 65, 2861 (1990)], the importance of the first-order TS-1 double ionization mechanism is also estimated. As a result it is found that for large energy losses shakeoff is the dominant double ionization mechanism.

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INTRODUCTION

Interactions between electrons are one of the most fundamental interactions in nature. For decades, a substantial effort was expended to study inelastic collisions involving electron impact. Most of the experiments found in the literature deal with total cross sections ([1–3] and references therein). On the other hand, differential measurements are quite rare in the literature, even though they provide more detailed information concerning the dynamics of the single and multiple ionization.

The theoretical description of multiple ionization is far from a simple task mainly due to the complexity of the many possible pathways involved. Double ionization of atoms by charged particles can result from either a single or a double interaction of the projectile with the target electrons. This constrasts with photoionization in which the incident photon interacts with only one of the target electrons after which a second electron is removed via either the so-called shake-off or TS-1 mechanism. In the shake-off mechanism [4], the ejected electron leaves the atom very fast. This instantly changes the field seen by the second electron and causes its ejection. The fast electron carries away almost all the transferred energy, while the energy of the second electron is on the order of the double ionization potential. In the TS-1 mechanism, the first electron is ejected due to the absorption of a photon. As it departs, it knocks out the second electron. Thus the second electron has an energy distribution similar to that found for impact ionization by electrons having energies given by the photon energy minus the first ionization potential. It is important to note, however, that these mechanisms are model dependent [5] and it is not possible to distinguish TS-1 from SO.

Most of the papers in the literature studying the dynamics of double ionization are devoted to the helium atom [5–11]. On the other hand, papers devoted to a quantified study of heavier targets are quite scarce. In this paper we present doubly differential measurements for electron impact ionization of neon valence shell electrons.

METHOD AND RESULTS

The experimental setup has been described in detail in previously published papers [12–15]. Briefly, an electron beam is produced via secondary emission from the surface of a tungsten moderator coupled to a $^{22}$Na source. The beam intersects a jet of neon gas emerging from a needle source. The forward-scattered projectiles are energy and angle analyzed by an electrostatic spectrometer and recorded by a microchannel plate position sensitive detector located at the focal plane of the analyzer. The ionized recoil ions are extracted from the collision region by a weak electric field (10 V/cm). They are separated according to their mass-to-charge ratio by a time-of-flight spectrometer and detected by another microchannel plate detector. The information about single and multiple ionization is obtained by coincidences between recoil ions and scattered electrons.

Doubly differential cross sections are shown in Fig. 1 for 750 eV electron impact on neon. The data are for electrons scattered into vertical and horizontal angles between 0 and $\pm 8^\circ$ and 0 and $\pm 6.5^\circ$, respectively. The single ionization cross section has a maximum at small energy loss and then decreases roughly as $\Delta E^{-2}$ for energy losses up to 150 eV. Higher energy losses, up to 460 eV, have roughly a $\Delta E^{-4}$ dependence. For energy losses above 460 eV, the dependence is flat. This is because at very large values of energy loss there are increasingly stronger contributions from ejected “target” electrons.

The fractions of single and multiple ionization of neon by 750 eV electron impact, corrected for the corresponding detection efficiencies, are presented in Fig. 2. As seen, at large projectile energy losses the fractions of single and multiple ionization remain fairly independent of the amount of energy lost by the projectile. For small energy loss, as one approaches the threshold, the fractions of double and triple ionization drop. By integrating the present doubly differential cross sections in Fig. 1, we find a quite good agreement with the fractions of total cross sections measured by Almeida et al. [1], indicated in Fig. 2 by the horizontal arrows ($F_1$, $F_2$, $F_3$), estimated and found to be negligible at this impact energy.
and \( F_3 \) (note that the major contributions to the total cross sections comes from the low energy loss part). Figure 2 also compares the present measured fractions with those from photoionization data. Due to the large discrepancies between the experimental data \([16–25]\), only the recomended data from Bizau and Wuilleumier \([16]\) for \( F_2 \) are shown in Fig. 2. The \( F_2 \) fractions for photons above 280 eV were extrapolated from Ref. \([16]\). For triple ionization, the data are from Refs. \([17,18]\).

Figure 3 shows the angular dependence of projectile electrons for different energy losses leading to single ionization of neon. Small energy losses lead to a maximum at 0°. For larger energy losses, the angular distributions become broader with the intensity at 0° decreasing rapidly until the distributions become isotropic in the angular range studied in this Brief Report.

**COMPARISON TO PHOTOIONIZATION**

The connection between inelastic interactions induced by charged particle impact and photon impact is well known. The connection exists because the perturbation experienced by the target due to the interaction with an incident charged particle may be regarded as tantamount to a photon pulse, with the Fourier transform of the electric field providing the frequency components. However, in order to compare charged particle and photon impact data, information about the projectile energy loss is required since the energy loss is equivalent to the energy of the absorbed photon. For a charged particle with velocity \( v \) and impact parameter \( b \), the sharply pulsed electric field in the time domain \( \Delta t \sim b/v \) is equivalent to a flat continuum in the frequency domain. Also, it is important to remember that such comparisons are most valid when the energy loss is small compared to the initial energy due to the high frequency cutoff to the virtual photon field \( \omega_{\text{max}} \sim \Delta \nu \) in and the limit of small momentum transfer (i.e., small scattering angle) \([26,27]\). The minimal impact parameter \( b_{\text{min}} \) for distant collisions can be estimated \([28]\) as \( b_{\text{min}} = \hbar(2mI)^{-1/2} \), where \( I \) is ionization energy and \( m \) the electron mass. \( b_{\text{min}} \) is of the order of the Ne radius. Using those assumptions, the energy loss range where the virtual photon field is valid is up to 250 eV. The reader should keep this in mind when referring to Fig. 4 where we extend the comparison past this limit.

As stated earlier, double ionization of atoms by photons is frequently considered in terms of two first-order mecha-
nisms. These mechanisms are shake-off (SO) where the first electron is suddenly ejected which dramatically changes the field seen by the second electron causing it also to be ejected and the two-step-one (TS-1) where the first electron is ejected due to the absorption of a photon and subsequently collides with and knocks out the second electron [29]. For large energy losses, other rearrangement processes like the Auger process also contribute to the double ionization of atoms. Hence, the single and double ionization cross sections by photon impact can be written as

\[ \sigma_{2s}^{2s} = R_2(h\nu)\sigma_{1s}^{1s}, \]  

where \( R_2(h\nu) \) is the measured ratio of double to single ionization by photon impact and is a measure of the relative importance of the sum of the first-order processes, shake-off and TS-1. Note that \( R_2(h\nu) \) is dependent on the photon energy but independent of the projectile, i.e., should be the same for photons and electrons. Also note that the TS-1 process should be maximum when the velocity of the first ejected electron matches the orbital velocity of the second electron that is removed. On the other hand, SO takes place when the first electron is ejected rapidly thus causing a sudden change in the Coulomb potential seen by the second electron. Hence, the SO mechanism is expected to dominate at high photon energies.

For charged particle impact, besides those above mentioned processes the projectile can interact independently with two target electrons in the so-called two-step 2 (TS-2) mechanism. Then for electron impact, considering only outer shell ionization, the double ionization cross section can be written as

\[ \sigma_{2s}^{2s} = \sigma_{1s}^{1s} + \sigma_{2s}^{2s} + \sigma_{int}^{2s}. \]  

where \( \sigma_{1s}^{1s} \) is the first-order contribution which includes the shake-off and the TS-1 mechanisms, \( \sigma_{2s}^{2s} \) is the second-order TS-2 mechanism, and \( \sigma_{int}^{2s} \) is the term due to the interference between the first- and second-order mechanisms [29].

It is possible to estimate the relative importance of the various terms by dividing Eq. (2) by the single ionization cross section. Doing so yields

\[ R_{2}(e^{-}) = R_{2,1st-order} + R_{2,2nd-order} + R_{2,int}. \]

where \( R_{2}(e^{-}) \) is our measured double ionization ratio for electron impact. Next, we use the well established fact that for 1st-order interactions, the double ionization cross section is proportional to the single ionization cross section and the constant of proportionality is essentially the same for both charged particle and photon impact. Thus Eq. (3) can be rewritten as

\[ R_{2}(e^{-}) = R_{2}(h\nu) + R_{2,2nd-order} + R_{2,int}. \]

where \( R_{2}(h\nu) \) is the double ionization ratio measured for photon impact. Keep in mind that in Eq. (4) the ratios are for particular energy losses which are related to the photon energy by \( h\nu = \Delta E \). Thus, using Eq. (4) and taking differences between the double ionization ratios measured for electron and photon impact we can estimate the relative importance of 2nd-order interactions plus any interference terms to the electron impact double ionization cross section.

This is done in Fig. 4 where the present electron impact double-to-single ionization ratios are compared to similar data for photon impact taken from Ref. [16]. The relatively little difference between those two sets of data implies that the TS-2 mechanism plus interference effects are minimal for 750 eV electron impact. In addition, one can also estimate the TS-1 double ionization contribution by normalizing electron impact single ionization cross sections of Ne+ to photoionization double ionization fractions at low photon energies where the TS-1 mechanism is expected to dominate (see Ref. [30]). This is also shown in Fig. 4 where the single ionization cross sections of Ne+ taken from Ref. [25] are normalized to the double-to-single photoionization cross sections ratio at 30 eV. For neon, the normalizing constant \( k \) was set equal to \((1142 \text{ Mb})^{-1}\). By setting \( k = (\pi b^2)^{-1} \), one finds \( b \approx 2 \text{ a.u.} \), which is twice the Ne+ average radius [14]. Comparing the recommended data from Ref. [16] with the present double-to-single ionization data, as seen, at higher energy losses both the TS-1 and the TS-2 mechanisms are negligible and hence shake-off is the dominant mechanism for producing doubly ionized neon.

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