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Double ionization of noble gases by positron impact

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The cross sections for double ionization and the ratios of double to single ionization, including Ps formation for Ne, Ar, Kr, and Xe are presented from threshold to 100 eV. Within the energy region 6.8 eV below the second ionization potential, i.e., the second Ore gap, we find Ne to have a double-ionization cross section indistinguishable from zero, which is consistent with prior measurements. However, the total double-ionization cross sections for Ar, Kr, and Xe just below the thresholds for direct double ionization are around 7–24 % of the maximum cross-section value for each atom, all of which are of the order of $10^{-21}\,\mathrm{m}^2$ and occur around 70–90 eV. In contrast to what has previously been found for He and Ne, this is direct evidence of a significant amount of transfer ionization for the three heavier noble gases in the second Ore gap, which is consistent with previous measurements for Ar and Xe. [S1050-2947(99)00310-8]

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INTRODUCTION

Ionization of atoms by positron impact is a fundamental process, and studies have permitted comparisons to be made with collisions involving electrons and other charged particles, e.g., Refs. [1–3]. Ionization of a neutral atom (A) by positron impact can take place by the following reactions:

$$e^{+}+A\rightarrow e^{+}+A^{n+}+ne^{-}, \tag{1}$$

$$e^{+}+A\rightarrow \mathrm{Ps}+A^{n+}+(n-1)e^{-}. \tag{2}$$

The first process is analogous to ionization by electron impact and is called direct ionization (DI). In Eq. (2) a positronium atom is formed and this process is called Ps formation if $n=1$, or transfer ionization (TI) if $n=2$. For the same n value the threshold for Eq. (2) is 6.8 eV below that for Eq. (1), due to the binding energy of Ps. By analogy to single ionization, the energy range between the thresholds for Eqs. (1) and (2) with $n=2$ will be called the second Ore gap.

Charlton *et al.* [4,5] were able to distinguish between processes (1) and (2) by detecting final-state positron-ion coincidences. They compared the ratios of double to single direct ionization by positrons [$n=2$ and $n=1$ in process (1)] with that for electrons, protons, and antiprotons for noble-gas targets. Process (1) was also studied by measuring positron-ion coincidences by Kara *et al.* [6] for Ne, Kr, and Xe, and Kruse *et al.* [7] for Xe. Helms *et al.* [8] measured total ionization cross-section ratios (i.e., the ratios of the sum of Eqs. (1) and (2) with $n=2$ to the sum with $n=1$) for Ar, Kr, and Xe, and Bluhme *et al.* [9] reported total double-ionization cross sections for He and Ne, noting that the yield of doubly charged ions in the second Ore Gap was zero within the experimental uncertainties, indicating an unexpected suppression of TI at these energies. They further deduced that the cross sections for TI were also suppressed at higher energies. The aim of the present paper is to extend these data and determine whether this phenomenon is true for the other noble gases.

EXPERIMENTAL APPARATUS AND TECHNIQUES

These measurements are carried out at ORELA, the LINAC-based positron beam facility at Oak Ridge National Laboratory. Positrons are produced by pair production when a 175-MeV electron beam strikes a water cooled Ta target and bremsstrahlung gamma rays are intercepted by a W vane converter. Some of these positrons are moderated to thermal energies in the W converter and escape into the vacuum due to the negative positron work function of W. They are accelerated to around 3 keV and magnetically guided to the experimental station approximately 10 m from the ORELA target. This system has been used to produce up to 10^8 positrons s^{-1} [10]. During the present work the accelerator is operated at lower repetition rates and output powers and, due to this and moderator degradation, the primary beam intensities are of the order of $10^6\,\mathrm{s}^{-1}$.

The primary positron beam is implanted into a 3000-Å single crystal tungsten foil and remoderated to the desired energies (E) by biasing the foil. Pulses of remoderated positrons then enter a 10-cm-long Penning trap where they are confined for 50 μs, thus effectively multiplying the interaction length by many orders of magnitude. At 30 eV, up to 25×10^3 positrons can be stored in the Penning trap. Gas is admitted to the center of the trap through a 1-cm-wide microchannel capillary array to form a target jet perpendicular to the beam axis. After confinement, the positrons are expelled from the trap and the ions are accelerated towards a microchannel plate detector by a quadratic potential that is applied to the interaction region and spectrometer flight tube, causing similar ions to travel to the detector with flight times that are independent of their starting position in the Penning trap [11]. Data are accumulated in the form of ion time-of-flight spectra at different positron energies, and data sets are gathered automatically as the positron beam energy is ramped under the control of the data acquisition computer. More details of the apparatus are given elsewhere [12].

The energy distribution of the positrons entering the Penning trap is measured using a retarding field analyzer. The energy spread is approximately 1.3-eV full width at half

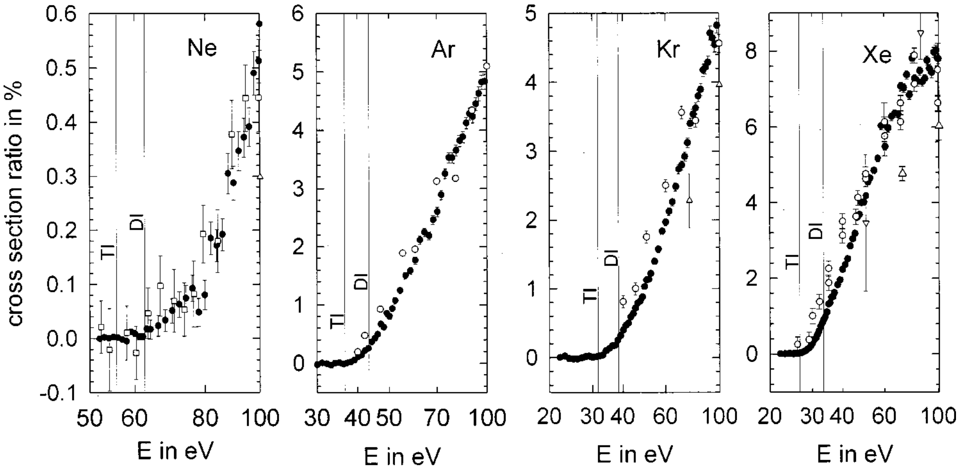

FIG. 1. Ratios of double- to single-ionization cross sections. Ne: ●, present results; □, [9]; △, [6]. Ar: ●, present results; ○, [8]. Kr: ●, present results; ○, [8]; △, [6]. Xe: ●, present results; ○, [8]; △, [6]; ▽, [7].

maximum and, ignoring contact potential effects, the mean energy is known to within ±0.25 eV. A pressure study shows that, within the statistical uncertainties of the present data, the relative yields of double and singly charged ions are independent of target density, under the conditions used to collect data.

All the data are measured with a mixture of the test gas and He. Since the total single-ionization cross section for He is well known, e.g., Refs. [13–15], the relative double-ionization cross-section data are derived from the ratio of the doubly charged ion yield from the test gas to the He^+ yield, thus avoiding systematic effects due to beam intensity, target thickness, and the effective length of the interaction region, the latter of which depends on positron energy and confinement time. The same is true for the relative cross-section ratios, which are obtained from the numbers of doubly and singly charged ions measured in the spectra. The relative confinement and detection efficiencies may vary significantly for ions of different species, and so all the yields are relative at present and have been normalized to existing data.

RESULTS AND DISCUSSION

The present results for the double to single ionization ratios are presented in Fig. 1. For Ne these indicate that within statistical uncertainty there is no evidence of double ionization below the threshold for DI, in accord with the findings of Bluhme *et al.* [9]. The ratio rises monotonically from the background somewhere around the threshold for DI at 62.6 eV and increases steadily up to 100 eV. The present data for Ne are normalized to those of Bluhme *et al.* [9] by weighted least-squares fit at all energies in the range of overlap above the threshold for DI. Within the statistical uncertainties there is good agreement between the two data sets. The data of Kara *et al.* [6] which do not include contributions from Ps formation, is around 30% lower than that reported by Bluhme *et al.* [9] at 100 eV.

The present results for Ar, Kr, and Xe are normalized to the data of Helms *et al.* [8] by least-squares fits at energies between the threshold for TI and 100 eV. When normalized in this way, the present data for these three gases are in reasonable accord with those of Helms at the higher energies, but are systematically smaller at lower energies. The data of Kara *et al.* [6] for Kr and Xe that, as in the case of Ne do not include Ps formation, are significantly smaller than the present results. Like Ne, the cross-section ratios for Ar, Kr, and Xe, all increase monotonically over the energy range studied. In contrast, the ratios and hence the double-ionization cross sections rise from the background well below the threshold for DI due to TI. In this respect the present data agree with those of Helms *et al.* [8] for Ar and Xe, and provide direct evidence of TI in the second Ore gap.

After obtaining the yield of doubly charged ions, the present data are normalized using values of the double-ionization cross sections obtained by other workers. For Ne the data are normalized at 100 eV to the product of the cross-section ratio at this energy (see Fig. 1) and the total single-ionization cross section [16]. The data for Kr and Xe are also normalized at 100 eV using the sum of the direct single-ionization [6] and Ps-formation [17,18] cross sections. The Ar results are normalized at 75 eV using the direct single [19] and Ps-formation cross section [20] values reported at this energy. The absolute uncertainties in the cross sections resulting from normalization are estimated to be around 20–30 %.

The cross sections for double ionization are shown in Fig. 2. The data are plotted as a function of E', where

$$E' = (E - E_{th})/E_{th}, \qquad (3)$$

and E_{th} is the threshold for TI. Also plotted in Fig. 2 are total single ionization data [21] and in this case E_{th} is the threshold for Ps formation.

For Ne, the double-ionization cross section rises from the background around the threshold for DI and then increases monotonically up to the highest energy studied. As noted above, there is no evidence of TI in the second Ore gap in agreement with the findings of Bluhme *et al.* [9]. The mean value of the cross section for TI in the second Ore gap is $2.1 \pm 4.7 \times 10^{-25}\,\mathrm{m}^2$; i.e., it is indistinguishable from zero. For the other gases, the cross sections rise from zero around the thresholds for TI to maxima around $E' = 1.4$ for Ar, 1.8 for Kr, and 1.7 for Xe, corresponding to impact energies of approximately 87, 73, and 68 eV, respectively. For Ar the data of Hippler *et al.* [22] agree reasonably well below the maximum in the present results, but reach their maximum around 110 eV, approximately 20 eV higher than the present results. In Kr, our data are higher than those of Kara *et al.* [6]

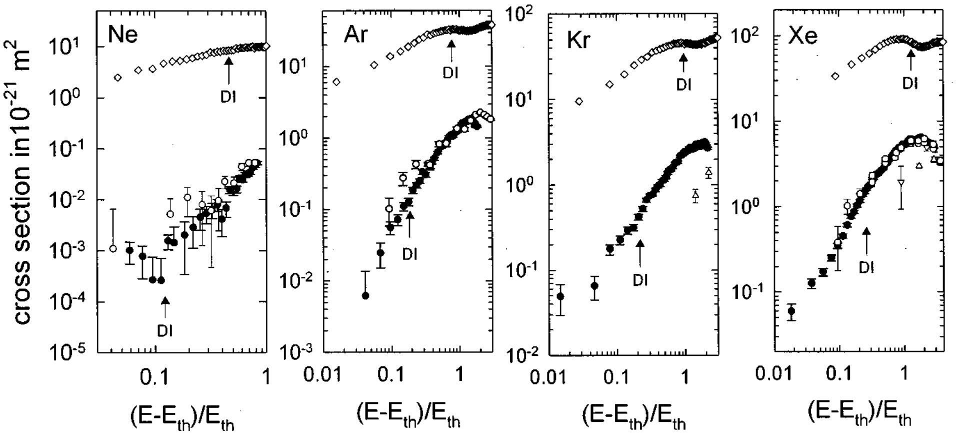

FIG. 2. Double- and single-ionization cross sections. Ne: double ionization ●, present results; □, [9]; △, [6]; and single ionization ◇, [21]. Ar: double ionization ●, present results; ○, [22]; and single ionization ◇, [21]. Kr: double ionization ●, present results; △, [6]; and single ionization ◇, [21]. Xe: double ionization ●, present results; ○, derived from [8] (see text); △, [6]; ▽, derived from [7] (see text); and single ionization ◇, [21].

at 75 eV ($E' = 1.2$) but are in closer agreement at the higher energy shown. For Xe, the double-ionization cross sections have been derived by multiplying the cross-section ratios [7,8] with the sum of the single direct ionization cross section [6] and the Ps-formation cross section [17]. The data derived from that of Helms *et al.* [8] are in very good agreement with the present data. Particularly at lower energies, these are higher than the results for double ionization [6,7] not including Ps formation.

At the thresholds for DI, the cross sections for TI are 0.14 ± 0.02 for Ar, 0.45 ± 0.03 for Kr, and 1.56 ± 0.05 for Xe, in units of $10^{-21}\,\mathrm{m}^2$. These are about 7.6%, 14%, and 24%, respectively, of the maximum values of the double-ionization cross sections at higher energies. Other than these features, little structure is seen in our results, although this might be masked by the large energy width of our positron beam.

Within the second Ore gap, the energy dependence of the cross sections approximately follow a power law in terms of E', with powers 1.59 ± 0.14 for Ar, 0.98 ± 0.10 for Kr, and 1.50 ± 0.03 for Xe. The cross sections, therefore, rise gradually from threshold, as can be seen from the ratios in Fig. 1, and not abruptly as is expected for single ionization by Ps formation [21].

For single ionization, the cross sections for Ps formation have been found to peak at around twice E_{Ps}, where E_{Ps} is the threshold for Ps formation, e.g., Ref. [20]. This is thought to be related to velocity matching between the projectile and the captured electron. The ratios between the direct single-ionization threshold and E_{Ps} gradually increase from 1.4 for He to 2.3 for Xe, so the Ps-formation cross section in Xe peaks close to the threshold for direct ionization. This implies a relatively large cross section for single ionization (by Ps formation) in the Ore gap. In contrast, the corresponding ratio for double ionization, i.e., the ratio between the thresholds for Eqs. (1) and (2) with $n = 2$, vary from 1.09 for He to 1.26 for Xe, suggesting that the positron may be too slow to transfer ionize effectively in the second Ore gap resulting in the comparatively small amount of TI at these energies.

At higher energies double ionization has been described as a two-step interaction with contributions from three mechanisms: ejection of one electron that liberates a second electron, interaction with the projectile and two electrons, and shake off, in which the projectile interacts with one electron followed by ionic relaxation leading to double ionization [1]. The amplitudes for these mechanisms are thought to interfere, resulting in charge-dependent differences in the cross sections for different projectiles [1]. At lower energies strong interference may obscure distinctions between these mechanisms. However, in considering what factors may tend to reduce the probability of TI, we note that the first is likely to be weighted by the probability of Ps impact target ionization to that for Ps breakup, the latter being expected to dominate at low energies. Also, the third mechanism is probably not very likely because electron capture is generally a soft collision process.

CONCLUSIONS

Relative cross sections for double ionization and double-to single-ionization ratios are determined for Ne, Ar, Kr, and Xe. Absolute values are assigned by normalization to existing data.

Positronium formation has previously been found to be suppressed for He and Ne [9] when accompanied by double ionization, and this is confirmed for Ne by the present results. Although it is an interesting puzzle, the present results show that the Ps-formation suppression phenomenon is limited to He and Ne among the noble gases, and is therefore probably not of fundamental significance.

The energy dependencies of the cross sections in the second Ore Gap for Ar, Kr, and Xe, follow approximate power laws with exponents of 1.59, 0.98, and 1.50, respectively. This is probably due to the three-body nature of the final state, which would not be expected to give rise to the sharp threshold effect found in single ionization (Ps formation) [21].

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