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Electron impact double ionization of Ba and Ba^+

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Abstract. Electron impact double ionization cross-sections for Ba and Ba^+ have been calculated in the binary encounter approximation. Hartree-Fock velocity distribution has been used for the first ejected electron and a hydrogenic velocity distribution for the second. For Ba^+ the focusing effects of the target ion on the incident electron have been incorporated in the calculations. Contributions from ionization-autoionization to the ionization cross-sections, as observed in experiments, have been included in the present work. The calculated results show structures as observed in recent experiments.

Keywords. Electron impact; single ionization; double ionization; ionization-autoionization.

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1. Introduction

Electron impact single and multiple (particularly double) ionization processes of atoms and ions are of fundamental importance (see [1] and references quoted therein). In some experimental studies of electron impact ionization of heavy atoms and ions large anomalous features have been observed for double ionization of the systems close to $Z = 56$ (Ba) at electron energies less than two times the threshold [2]. These features have been interpreted as giant scattering resonances appearing in the scattered electron channel [3].

In many cases, besides direct ionization, electron impact double ionization of atoms and ions takes place due to several alternative mechanisms. In some cases these indirect ionization channels have been found to dominate over the direct ionization process. A single ionization event ejecting an inner shell electron can leave the product in an excited state which may subsequently cascade through several autoionizing levels resulting in relatively large net multiple (mainly double) ionization [4–6].

Due to complexities in quantal studies of direct double ionization the calculations are limited to a few lighter targets only [7–10]. Hence for heavy targets, the results of single ionization cross-sections for the inner shell involved are taken as a measure of total double ionization cross-sections. This cannot be strictly justified because the contributions from direct double ionization are small but not insignificant. On the other hand the modified binary encounter model proposed by Roy and Rai [11] based on Gryzinski's [12] double binary encounter model, has been successfully used for calculations of electron impact direct double ionization cross-sections of a number of atomic systems [13, 14]. Later on the contributions from inner shell ionization-autoionization to the electron impact double ionization of some atoms and ions were incorporated in the binary encounter calculations [1, 15, 16]. In the present work we extend the above calculations to more complex systems, namely Ba and Ba^+.

2. Calculation

The electron impact double ionization cross-section for an atomic system including the contribution from the inner shell ionization-autoionization process (Auger transition) is given by

$$Q^{ii}(T)=Q_{\rm D}^{ii}+Q_{\rm A}^{ii}$$

where $Q_{\rm D}^{ii}$ stands for the contribution to the double ionization cross-section from direct ejection of two electrons and $Q_{\rm A}^{ii}$ is that from the Auger emission. For Ba the expression for $Q_{\rm D}^{ii}$ is given by

$$Q_{\rm D}^{ii}=\frac{2n_e(n_e-1)}{4\pi\bar{r}^2}\int_{t=0}^{\infty}\int_{U_i}^{E_q-U_{ii}}\left\{\frac{2}{(s^2+t^2+1)U_i}\left[\left(\frac{1}{(\Delta E)^2}+\frac{4t^2U_i}{3(\Delta E)^3}\right)\right.\right.$$
$$\left.+\left(\frac{1}{(s^2U_i+U_i-\Delta E)^2}+\frac{4t^2U_i}{3(s^2U_i+U_i-\Delta E)^3}\right)-\frac{\phi}{\Delta E(s^2U_i+U_i-\Delta E)}\right]$$
$$\left.\alpha f(t)U_i^{1/2}\right\}{\rm d}(\Delta E){\rm d}t\times 8{\cdot}797\times 10^{-17}(\pi(a_0^2)) \quad (1)$$

and that of $Q_{\rm A}^{ii}$ is given by

$$Q_{\rm A}^{ii}=an_e\int_0^{\infty}\left\{\frac{4}{(s^2+t^2+1)U_i^2}\left[\frac{s^2-1}{s^2}+\frac{2t^2}{3}\left(\frac{s^4-1}{s^4}\right)-\frac{\phi\ln s^2}{(s^2+1)}\right]\right\}$$
$$f(t)U_i^{1/2}{\rm d}t \quad (2)$$

where the various symbols appearing in the above equations have already been defined by Roy and Rai [11] and Chatterjee and Roy [1].

In the case of Ba^+, as the electron reaches the neighbourhood of the positive ion its kinetic energy is increased and impact parameter is reduced due to Coulombic attraction. Thomas and Garcia [17] have shown that the effects of these two physical processes can be approximately incorporated in the ionization cross-section through the relation

$$\sigma_{\rm I}(E_1)=\sigma_{\rm I}(E_1')\left[\frac{1}{2}+\frac{1}{2}\left(1+\frac{Ze^2}{E_1\xi}\right)^{1/2}\right]^2$$

where E_1 is the incident energy, Ze is the charge on the target ion, $\sigma_{\rm I}(E_1')$ is the ionization cross-section at the increased electron energy E_1 and ξ is the collision radius whose value depends upon the ionic radius and electron–electron separation in the shell. Following the above prescription the expressions for $Q_{\rm D}^{ii}$ and $Q_{\rm A}^{ii}$ for Ba^+ are obtained after replacing s by s' in (1) and (2) and multiplying each of them by $(s+s')^2/4s^2$ where $s'^2=s^2+(2Z/\xi U_i)$.

In the derivation of $Q_{\rm D}^{ii}$ an accurate Hartree-Fock (HF) velocity distribution and a hydrogenic velocity distribution have been used while considering the ejection of the first and the second electron, respectively. The use of a HF velocity distribution in both cases would have been appropriate but this would give rise to complex numerical integration in the evaluation of $Q_{\rm D}^{ii}$. Contributions of ionization from inner shells have also been included in the direct double ionization cross-section.

In the present work we have used the HF binding energy of shells as reported by Mclean and Mclean [18]. For shell radii of Ba the quantum mechanical values of points of maximum radial probability density reported by Desclaux [19] have been used. For Ba^+ the HF value of $\langle r \rangle$ reported by Mclean and Mclean [18] have been used for shell radii. Momentum distribution function for Ba and Ba^+ have been constructed using HF radial distribution reported in [18].

Here we point out that after the ejection of one electron the target becomes Ba (for Ba) and Ba^{2+} (for Ba^+). Hence one would expect the use of the binding energies of Ba^+ and Ba^{2+} while considering the ejection of the second electron. However, we have used the binding energies of Ba and Ba^+ while considering the ejection of both the electrons. This consideration is physically justified at high impact energies so that the ejection of two electrons may be supposed to be simultaneous, leaving no time for the target to relax after the ejection of the first electron. This choice is expected to overestimate the cross-section close to the threshold but would be more appropriate with increase in impact energy.

At this stage we mention that correlation plays an important role in the double ionization process. In the direct double ionization calculations one should consider correlation of events called dynamic correlation as well as electron–electron correlation [10]. The present calculations, based on Gryzinski's [12] double binary encounter model, consider the correlation of events. Moreover, one of the two binary encounter processes suggested by Gryzinski in which the first ejected electron knocks out the second electron partly takes into account electron–electron correlation [20]. The electron–electron correlation is taken into account in the theoretical studies of double ionization process by the use of correlated wave functions for the target electrons e.g. configuration interaction approach based on Slater–Condon theory, multi-configuration Hartree Fock (HF) approach used by Fischer (see Griffin and Pindzola [21]). Calculations using these wave functions are difficult to perform, particularly for heavier targets, as they require extensive computational work. HF wave functions consider the electron-electron correlation to some extent through antisymmetrization [21]. Moreover, for fast projectiles the effects of electron-electron correlation may not be significant [10]. Therefore, the use of HF wave function in the studies of double ionization processes using the BEA is reasonable.

3. Results and discussion

Our results for barium have been presented in figure 1 along with the recent experimental observations of Dettmann and Karstensen [22] and calculation of McGuire [23]. Calculated results of McGuire are singly peaked and are lower than the experimental cross-sections throughout the energy range.

The maxima obtained in the experimental observations [22] have been attributed to the ionization processes involving O shell and N_4 or N_5 shell, respectively. The present direct double ionization cross-sections include, besides the valence shell ionization the contributions from the process in which one electron is ejected from the valence shell and the other from inner shell (5s or 5p). Besides this we have also included the contributions of ionization-autoionization from 4d subshell. In the latter calculations we have evaluated the electron impact single ionization cross-sections for the 4d subshell of Ba and taken the same as the contribution from ionization-autoionization to the double ionization cross-section assuming the branching ratio for the process equal to unity, due to non-availability of the same in literature.

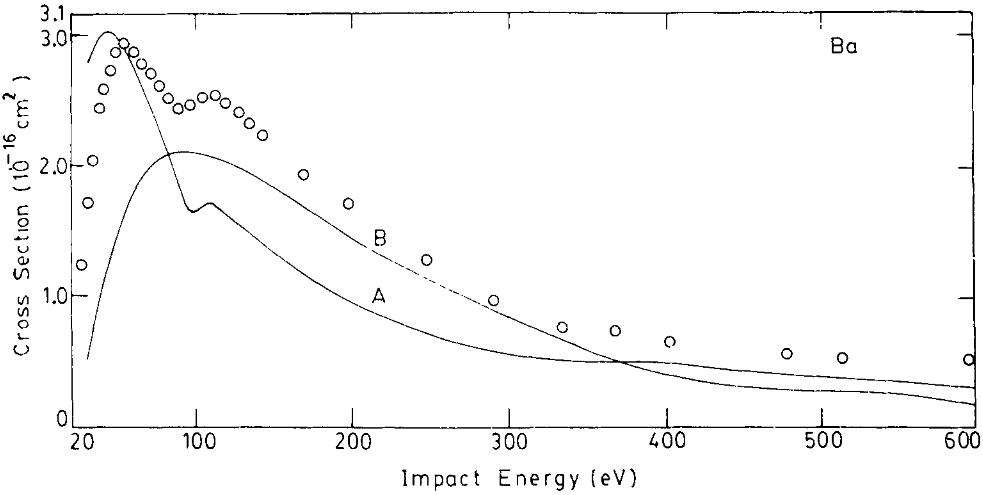

Figure 1. Results for Ba. A: Present results; B: Calculations of McGuire [23]; O: Experimental observations of Dettmann and Karstensen [22].

The present calculations also show two peaks at impact energies of 45 eV and 108 eV respectively. Positions of these peaks are close to those observed in experiments. The calculated cross-section (about $3{\cdot}0 \times 10^{-16}\,cm^2$) at the first peak is slightly higher than its experimental counterpart (about $2{\cdot}85 \times 10^{-16}\,cm^2$) but the present value of the second peak (about $1{\cdot}7 \times 10^{-16}\,cm^2$) is lower than the corresponding measured value (about $2{\cdot}5 \times 10^{-16}\,cm^2$). Moreover, below 50 eV impact energy calculated results are higher than the experimental observations whereas beyond this the present values are lower. The overestimation at low impact energies might be due to the use of binding energy of neutral barium for the ejection of both the electrons while evaluating direct double ionization cross-sections and use of a hydrogenic velocity distribution for the second ejected electron. In spite of these discrepancies the present results are always within a factor of 2 from the experimental observations. In the energy region of 80 eV to 370 eV the calculated results of McGuire are closer to experiment as compared to the present cross-sections. However, the calculations of McGuire fail to exhibit the structure in cross-section curve found in experiment as well as in the present study.

Figure 2 shows the results of Ba^+. Calculated cross-sections have been compared with the experimental measurements of Hirayama *et al* [24]. Experimental cross-section curve has a broad rise at about 50 eV and shows a steep rise around 85 eV impact energy.The cross-section has a sharp maximum at about 115 eV and exhibits a small but distinct shoulder around 200 eV. The structure in the ionization function above 85 eV has been attributed to 4d ionization followed by autoionization of the residual ion expressed as Ba^+ $(4d^{10}5s^2 5p^6 6s) \rightarrow Ba^{2+}(4d^9 5s^2 5p^6 6s) + e \rightarrow Ba^{3+}$ $(4d^{10} 5s^2 5p^5) + 2e$. The shoulder-like structure observed around 200 eV is considered to be due to the second maximum in the cross-section function for the 4d ionization (ionization energy of 114 eV). The broad rise ranging from 50 eV to 90 eV impact energy is a specific feature of Ba^+ ion. This structure is attributed to the ionization of the 5s-electron followed by autoionization expressed as Ba^+ $(5s^2 5p^6 6s) \rightarrow Ba^{2+}$ $(5s\,5p^6 6s) + e \rightarrow Ba^{3+}(5s^2 5p^5) + 2e$ considering that the binding energy (46·3 eV) of

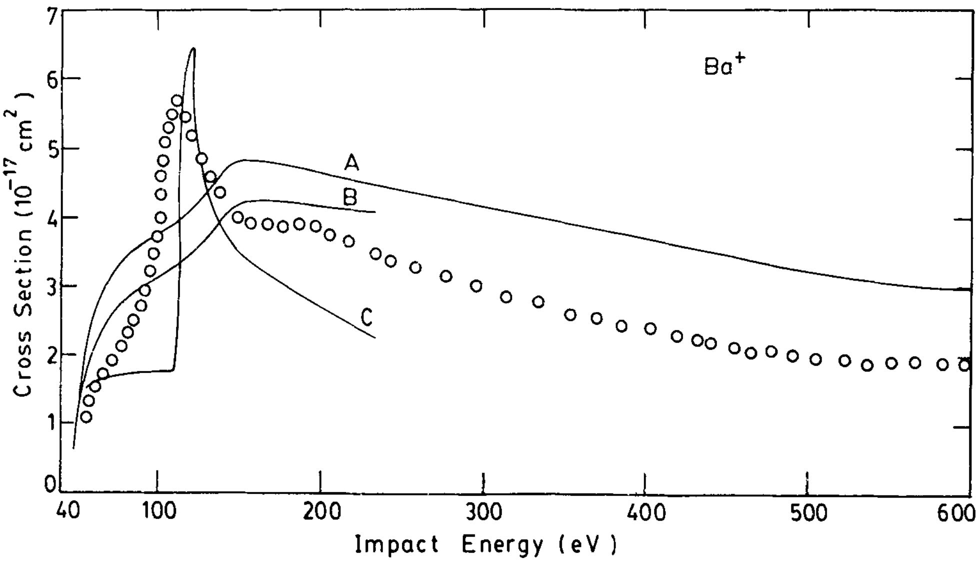

Figure 2. Results for Ba$^+$. A: Present results; B: Sum of the present values of single ionization cross-sections for 5s and 4d shells; C: The same as curve B calculated by Younger [25]; O: Experimental results of Hirayama *et al* [24].

5s electron in Ba$^+$ is close to threshold energy (about 45 eV as quoted by Hirayama *et al*) of double ionization of Ba$^+$.

In the present calculations of direct double ionization we have included the contribution from the processes in which one electron is ejected from the valence shell and the other from an inner shell (5p or 5s or 4d). We have also calculated the single ionization cross-sections for 5s and 4d and have taken the values obtained as the contributions of inner shell ionization-autoionization to electron impact double ionization cross-section. The present cross-section curve also shows two shoulder-like structures at impact energies of about 85 eV and 147 eV but it does not show the sharp peak at about 115 eV observed in experiment. The experimental peak has been attributed to the excitation of 4d-electron to states connected to the double autoionization e.g. $4d^{10}\,5s^2\,5p^6\,6s \rightarrow 4d^9\,5s^2\,5p^6\,6s\,nl \rightarrow 4d^{10}\,5s\,5p^6\,6s + e \rightarrow 4d^{10}\,5s^2\,5p^5 + 2e$. This process being quantum mechanical phenomenon, its contribution could not be included in the present work. Except in the energy range of 100 eV–135 eV the present cross-sections are higher than the experimental observations. The reasons for the discrepancies are similar to those discussed for Ba. Apart from the discrepancies mentioned above the calculated results are in reasonably good agreement with experiments, always being within a factor of 2 as compared with the measured cross-sections. We have also plotted the sum of the present single ionization cross-section for 5s and 4d subshell as well as the same calculated by Younger [25] in distorted wave Born exchange approximation. These plots indicate that inner shell ionization-autoionization dominates over direct double ionization for Ba$^+$.

4. Conclusions

From a close examination of our calculated results it is concluded that the present method gives reasonably accurate values of electron impact double ionization cross-sections for Ba and Ba^+. The use of binding energy of Ba and Ba^+ respectively, while evaluating direct double ionization cross-section is reasonable at high impact energies. At low impact energy there is a partial rearrangement. If the effect of partial re-arrangement could be incorporated, the present results would be lowered at low impact energies leading to a better agreement with experiments. In the case of heavy particle impact the concept of partial rearrangement of target has been incorporated by Chatterjee and Roy [26] but unfortunately, it is difficult to consider this for electron impact studies.

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