Multiple Ionization Dynamics of Ar_2 by Slow Highly Charged Ions

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Abstract

Multiple ionization dynamics of rare gas dimers by slow highly charged ions is investigated with the three-center Coulombic over-barrier model. Quasi-molecule formation of an electron at the distant atomic site from the projectile is shown sensitive to partial screening of ion-core charge at the near atomic site due to electrons already forming quasi-molecules. It is found that the screening effect suppresses simultaneous chained electron removal from both sites and hence enhances charge-asymmetric ion pair formation. The ion pair distribution measured in $Ar^{9+} + Ar_2$ collisions (Matsumoto J *et al.* 2010 *Phys. Rev. Lett.* **105** 263202) is well reproduced by taking a screening factor of $s = 0.4$ in the model.

Keywords: highly charged ion, rare gas dimer, three-center over-barrier model, screening effect, dissociating ion pair distribution

1. Introduction

Much attention has been called to Coulomb explosion of molecules in collisions with slow (velocities of $v \ll 1$ au) highly charged ions. Recent progress of the experimental technique of recoil-ion momentum spectroscopy [1] permits us to measure the dissociating ion pair distribution in the collisions [2,3]. Such observations are of great interest from a viewpoint of multi-center multi-electron dynamics. In contrast with a covalent diatomic molecule, however, little effort has been devoted to a rare gas dimer. In the rare gas dimer, because of its large internuclear distance, a range of selectron motion is well localized in either atomic site in a time scale of the collision. It is expected that such a characteristic structure gives intriguing multiple ionization dynamics.

About ten years ago, we proposed a three-center Coulombic over-barrier model to describe multiple ionization of rare gas dimers with slow highly charged ions [4]. In a preceding work [5], stimulated by an experiment done at GANIL [3], we modified the model so as to incorporate a screening effect during a collision. The screening effect

was found crucial to explaining the population of charge-asymmetric ion pairs such as $(Q, Q') = (2, 0)$.

In the present work, we carry out systematic calculations based on the modified model to reveal how the screening affects the ion pair distribution and show how it explains the result of measurement in $Ar^{9+} + Ar_2$ collisions [3].

2. Three-center over-barrier model

We treat a process of multiple ionization (actually electron transfer) of a rare gas dimer BC with a slow highly charged ion A^{q+}. The outermost electrons at respective atomic sites, B and C, in the target dimer are supposed to sequentially form quasi-molecules with projectile A^{q+}. During this course, the target state is labelled by a pair of charge states (Q_B, Q_C) and evolves in respective steps either as $(Q_B, Q_C) \to (Q_B + 1, Q_C)$ or as $(Q_B, Q_C) \to (Q_B, Q_C + 1)$. Accordingly the active electron in the former process is taken as $t_B = Q_B + 1$ in site B, while that in the latter process is taken as $t_C = Q_C + 1$ in site C, where $t_{B,C}$ denotes the rank of an electron in each atom. It is assumed for simplicity that the electron never returns to the target once trapped in a quasi-molecule; we only consider the *way in* process to derive partial cross sections of ion pair (Q, Q') formation when $q \gg Q, Q'$.

Relevant one-electron dynamics is described by a three-center Coulomb potential,

$$U(\boldsymbol{r}) = -\frac{q_A}{|\boldsymbol{r} - \boldsymbol{R}_A|} - \frac{q_B}{|\boldsymbol{r} - \boldsymbol{R}_B|} - \frac{q_C}{|\boldsymbol{r} - \boldsymbol{R}_C|}, \tag{1}$$

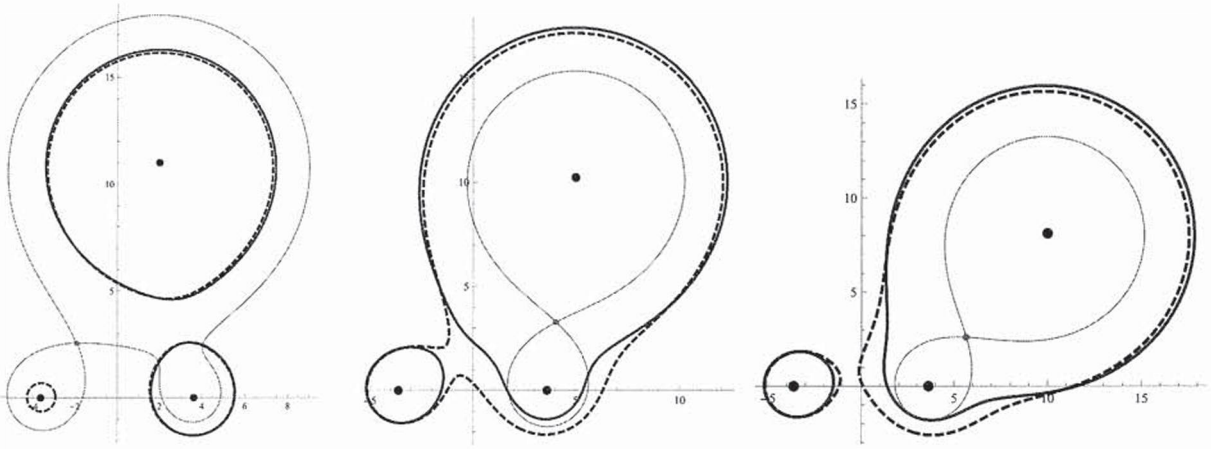
with three ion core charges q_A, q_B and q_C. Virtually in any case, this potential has two saddle points, lower one (U_{low}) and higher one (U_{high}). Stark-shifted energies of active electrons localized at atomic sites B and C are given by

$$E_B = -I(t_B) - \frac{q_C}{2d} - \frac{q_A}{|\boldsymbol{R} - \boldsymbol{d}|}, \tag{2}$$

$$E_C = -I(t_C) - \frac{q_B}{2d} - \frac{q_A}{|\boldsymbol{R} + \boldsymbol{d}|}, \tag{3}$$

where $I(t)$ denotes the atomic t-th multiple ionization potential; nuclear configuration is represented by a relative vector $\boldsymbol{R} = \boldsymbol{R}_A - (\boldsymbol{R}_B + \boldsymbol{R}_C)/2$, with intra-dimer vector $\boldsymbol{d} = (\boldsymbol{R}_B - \boldsymbol{R}_C)/2$ being fixed during a collision. Hence we can formulate the *over-barrier criterion* of quasi-molecule formation in such a way as either saddle height U_{low} or U_{high} gets lower than either electron energy E_B or E_C. A set of the criterions was generally given according to the potential topography with three centers and two saddle points [4,5]. The (Q_B, Q_C) evolution along a given collision trajectory is determined by applying the "rule

of first arrival" to the alternative of active site B or C. It should be noted that the over-barrier criterion may give two-electron *chained* processes (simultaneous electron removal from both sites) as $(Q_{\rm B}, Q_{\rm C}) \to (Q_{\rm B}+1, Q_{\rm C}+1)$ [4,5]. This mechanism is illustrated in figure 1.



(a) $(1,0) \to (1,1)$ (b) $(1,0) \to (2,1)$ (c) $(1,0) \to (2,0)$

Figure 1: Competing equipotential surfaces $U_{\rm B}(\boldsymbol{r}) = E_{\rm B}$ and $U_{\rm C}(\boldsymbol{r}) = E_{\rm C}$ at critical nuclear configurations for the second electron removal following $(0,0) \to (1,0)$ in the $(Q_{\rm B}, Q_{\rm C})$ evolution. The surfaces are calculated for $A^{9+} + Ar_2$ with a screening parameter of $s = 0.4$ and plotted in the $x-y$ plane ($x \equiv \boldsymbol{r} \cdot \hat{\boldsymbol{d}}$) with the origin taken at the midpoint of sites B and C. Thin solid lines represent the surfaces of just forming relevant quasi-molecules for $t_{\rm C} = 1$ electron in panel (a) and for $t_{\rm B} = 2$ in (b) and (c). Thick solid lines indicate the surfaces for alternative electrons, $t_{\rm B} = 2$ in (a) and $t_{\rm C} = 1$ in (b) and (c), just *before* the quasi-molecule formation, with dashed lines just *after* that. It is found in panel (b) that, resultantly, the $t_{\rm C} = 1$ electron also forms a quasi-molecule at identical nuclear configuration; a simultaneous two-electron chained process $(1,0) \to (2,1)$ occurs at a configuration taken in panel (b), while it does not occur in (a) and (c).

The ion core charges in equations (1—3) are effectively taken as

$$q_{\rm A} = q, \qquad q_{\rm B} = Q_{\rm B} + 1, \qquad q_{\rm C} = (1-s) \times Q_{\rm C},$$

for active site B, and

$$q_{\rm A} = q, \qquad q_{\rm B} = (1-s) \times Q_{\rm B}, \qquad q_{\rm C} = Q_{\rm C} + 1,$$

for active site C [5]. Here is introduced a *screening* factor s ($0 \le s \le 1$) as a parameter. The screening effect during a collision comes from a situation peculiar to a rare gas dimer

that the electron cloud at site B and that at C are well separated from each other; the electrons already forming quasi-molecules from the non-active site (supposing B) still partially screen their original ion core when seen from the active site (C) and also from the active saddle point. Note that electron removal from B and that from C are *not* correlated at all when $s = 1$ (*i.e.*, the independent event model).

3. Results and Discussion

The model is applied to $A^{9+} + Ar_2$ collisions with active electrons $1 \leq t_B, t_C \leq 4$.

Critical nuclear configurations for ion-pair (Q, Q') formation are plotted in figure 2 for a screening factor of $s = 0.4$ as an example. As seen from the figure, the critical configurations for quasi-molecule formation from site B (near site to the projectile) compose concentric circles around site B, while those from site C (far site) compose fragmented segments around site C. A pair of the former and latter curves merge into a curve composed of critical configurations of two-electron chained processes. This pattern of the critical curves varies sensitively with the parameter s, especially in a region of $0.3 \lesssim s \lesssim 0.5$. As the screening factor s increases, the simultaneous two-electron chained processes such as $(1,0) \rightarrow (2,1)$ and $(2,1) \rightarrow (3,2)$ are suppressed; hence the populations of charge-asymmetric ion pairs such as $(2,0)$ and $(3,1)$ are enhanced.

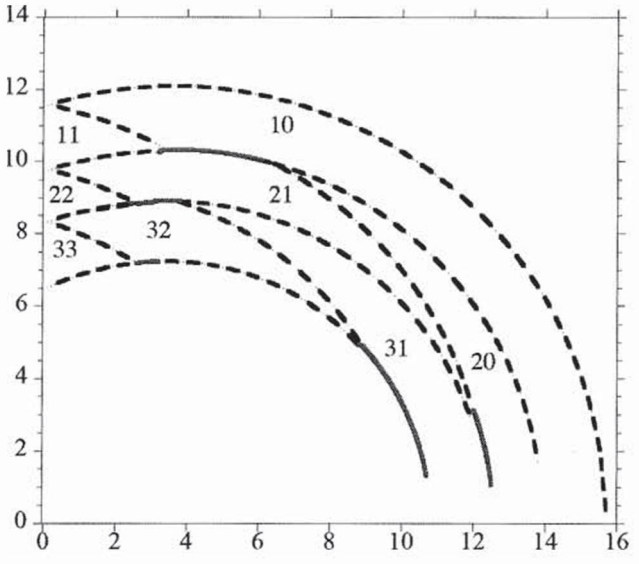


Figure 2: Critical curves for ion-pair (Q_B, Q_C) formation in the first quadrant of nuclear configuration $X - Y$ plane, calculated for $A^{9+} + Ar_2$ collisions with screening parameters of 0.4. The critical curves shown include the two-electron chained processes (solid lines) and unchained processes (dashed lines).

Ion pair (Q, Q') formation cross sections are calculated for randomly oriented target dimers using the critical configurations obtained above. The result with different values of the screening factor $0 \leq s \leq 1$ are shown in figure 3, where comparison with experiment [3] is also made. As clearly seen from the figure, the (Q, Q') distribution is sensitive to the parameter s in the calculation. A partial screening model with $s = 0.4$ considerably well reproduces the overall distribution observed in the experiment. The value of $s = 0.4$ has been confirmed the best choice throughout the domain of $0 \leq s \leq 1$. In contrast, the model without screening ($s = 0$) and that with full screening ($s = 1$) indicate tremendous deviations from the experiment in particular in charge-asymmetric channels (2,0) and (3,1), although giving a reasonable agreement in symmetric channels (1,1) and (2,2). It is noted that these two models show opposite directions of deviation from the experiment in (2,0) and (2,1) channels. It is also found from the figure that the calculated distribution varies with the screening parameter drastically around $s \sim 0.4$.

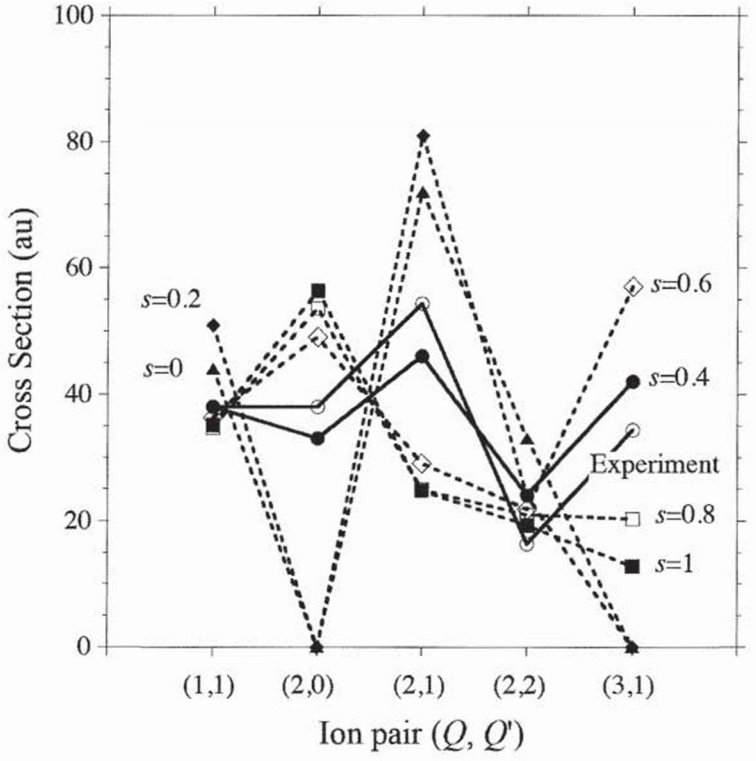


Figure 3: Screening parameter dependence of the ion-pair (Q, Q') formation cross sections in $A^{9+} + Ar_2$ collisions: $s = 0.0$ (full triangles), $s = 0.2$ (full diamonds), $s = 0.4$ (full circles), $s = 0.6$ (open diamonds), $s = 0.8$ (open squares), and $s = 1.0$ (full squares). The experimental result [3] (open circles) is also shown and normalized to the model result with $s = 0.4$ at $(Q, Q') = (1, 1)$.

In the present paper, we have described the multiple ionization dynamics of rare gas dimers by slow highly charged ions with the three-center Coulombic over-barrier model. The screening effect during a collision is analyzed in detail and found to alter the ion pair distributions crucially. The distribution observed in a recent experiment for $Ar^{9+} + Ar_2$ collisions is best reproduced by taking a screening factor of $s = 0.4$.

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