Increase of true double-electron-capture cross sections in slow Xe^{q+}-(Xe,He) collisions at very high q

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We have measured cross sections for true double-electron capture (DC) and transfer ionization (TI) in slow Xe^{q+}-(Xe,He) collisions in the charge-state regime $15 \leq q \leq 42$. We find that the probabilities for radiative stabilization $P_{rad} = \sigma_{DC}/(\sigma_{DC}+\sigma_{TI})$ increase rapidly with q for $q \geq 28$ and are very similar for two-electron transfer from Xe and He. This surprising similarity indicates that the core structure of the projectile, rather than the initially populated capture state, is decisive for the electronic relaxation process.

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Electron transfer in slow ($v \ll 1$ a.u.) collisions can be described in terms of well-localized transitions between adiabatic quasimolecular states of the same spin and symmetry. It has been demonstrated repeatedly [1–3] that his approach is fruitful when all the relevant quasimolecular states are taken into consideration. However, it becomes prohibitively difficult to include all possible reaction channels for the high densities of projectile capture states present at high projectile charge. Then the simplified approach of the extended classical over-the-barrier (ECB) model [4,5] becomes useful since it assumes that a projectile capture state always is available once a target electron is classically allowed to pass the internuclear potential barrier between the remaining target ion and the projectile. This assumption is more justified for higher projectile charge q, because the density of projectile capture states increases strongly with q. Charge transfer at very high q is expected to allow greatly simplified discussions of electron-transfer mechanisms, making different features of the collisions more regular and therefore easier to predict.

In the ECB model, two-electron transfer from the target B to a highly charged projectile A^{q+} populates double Rydberg states $(nl,n'l')$:

$$A^{q+}+B \xrightarrow[R_1]{} A^{(q-1)+}(nl)+B^{+} \xrightarrow[R_2]{} A^{(q-2)+}(nl,n'l')+B^{2+} , \quad (1)$$

where n (n') and l (l') are the principal and angular momentum quantum numbers of the electrons transferred at the internuclear distance R_1 (R_2). The predictions of this independent-electron model, which obviously excludes (dynamic) electron correlation, has been reasonably successful in predicting absolute cross sections for electron removal from the target [6], projectile-energy gain [7], and scattering angles [8]. It has also been shown experimentally that the principal quantum numbers n [9] and (n,n') [10] of the capture states are reasonably well accounted for. In contrast, the subsequent decay of the double Rydberg state $A^{(q-2)+}(nl,n'l')$, where the interaction between the two highly excited electrons (static correlation) plays an important role, is much less understood.

The probability P_{rad} that both electrons are attached to the projectile at large internuclear separations is defined by

$$A^{q+}+B \longrightarrow A^{(q-2)+}(nl,n'l')+B^{2+} \xrightarrow[P_{rad}]{} A^{(q-2)+}+B^{2+} \quad (2)$$

$$\xrightarrow[(1-P_{rad})]{} A^{(q-1)+}+B^{2+}+e^{-} . \quad (3)$$

contributions to (3) from independent capture and ionization can be disregarded, since they are small in the present range of collision velocities. P_{rad} can be expressed in terms of the cross sections for true double-electron capture (DC) (2) and transfer ionization (TI) (3):

$$P_{rad} = \sigma_{DC}/(\sigma_{DC}+\sigma_{TI}) . \quad (4)$$

Here we report a rapid increase for $q \geq 28$ in $P_{rad}(q)$ for double Rydberg states of $Xe^{(q-2)+}$ ($15 \leq q \leq 42$) populated in collisions with Xe and He, reaching $P_{rad} \sim 0.4$ at $q = 35$. This confirms an earlier tentative and indirect identification of radiative stabilization following two-electron transfer in Xe^{q+}-Xe collisions at very high q [11] and we present experimental evidence for a strong increase in true double-electron-capture cross sections at very high q. Furthermore, we find $P_{rad}(q)$ for Xe^{q+}-Xe and Xe^{q+}-He collisions to be very similar. This was completely unexpected since two-electron transfer from Xe and He populate quite different excited states of $Xe^{(q-2)+}$. We tentatively interpret this as an indication that the projectile core structure is important for the electronic relaxation process, while the detailed characteristics of the initial capture state seems to play a less important role. The concept of a point like highly charged projectile, which is appropriate for the initial stage of the charge-transfer process, thus seems to be inadequate for the subsequent decay process. This

surprising result suggests that different projectile species might give different values for P_{rad} even with the same q and target. For example, true double-electron capture from He by highly charged plasma-contaminating metal ions, expected to dissipate twice as much energy as transfer ionization, could be much more important than has been inferred earlier [12].

The experimental results presented here can also give input to the presently very vivid discussions of formation and stabilization of multiple Rydberg states (hollow atoms) populated in slow ion-surface interactions [14,15]. The understanding of the decay of double Rydberg states (hollow ions) is a first step towards a correct modeling of vacancy-filling processes in hollow atoms. In the following paragraphs, we will briefly describe the experimental technique and finally we will attempt a qualitative explanation for the findings reported here.

A 90% enriched ^{136}Xe gas was injected into the cryogenic electron-beam ion source at the Manne Siegbahn Institute of Physics in Stockholm [16] and Xe^{q+} beams ($15 \leq q \leq 42$) were extracted at an energy of $\sim 4.5q$ keV. A position-sensitive detector was used for registration of highly charged Xe ions after their passage through the collision cell and a 160° electrostatic hemispherical energy analyzer (cf. Fig. 1). Recoil ions were extracted from the gas cell and a time-of-flight spectrometer was used to determine the charge state of the recoil ions. The Ceratron recoil ion detector provided the start pulse to the time-to-amplitude converter, stopped by a signal from the back of the second channel plate in the projectile detector. The length of the beam pulse was set to ~ 50 ms and Xe^{q+} intensities were limited to $\sim 100-200$ per pulse.

A projection of the two-dimensional detector image on the axis of charge-state dispersion is shown in Fig. 2 for Xe^{37+}-Xe collisions. In Fig. 2(a) coincidences with Xe^{+} recoil ions are shown. The 36+ peak is thus due to single-electron capture,while the 35+ peak exclusively is due to double collisions. In Fig. 2(b) coincidences with Xe^{2+} target ions are shown. This 35+ peak results mainly from true double-capture events, with a minor contribution from double collisions. We estimate this double-collision contribution by multiplying the intensity of the transfer-ionization peak [coincidences between Xe^{36+} and Xe^{2+} in Fig. 2(b)] with the ratio of $(q-2)$ to $(q-1)$ events in the Xe^{+} coincidence spectrum [Fig. 2(a)].

In earlier investigations at more moderate q it has been found that true double-electron capture dominates transfer ionization at the lowest q while the opposite relation prevails at somewhat higher q [17]. This behavior is well accounted for by the fact that doubly excited projectile states of excitation high enough for autoionization are created only at sufficiently high q. An assumption about unit probability for electron emission [i.e., $P_{rad}=0$ in (2) and (3)] has in general been sufficient to explain experimental data in the q range between about 8 and 20. It has, however, also been anticipated [18] that the strong q scaling of the radiative decay may favor the charge stabilization process (2) at still higher q. For specific state, $\Delta n \neq 0$ electronic dipole transition rates scale as q^4, while Coulombic autoionization is independent of q. Against this it has been argued [19] that double Rydberg states formed after transfer of two electrons (1) should autoionize rapidly due to the energy proximity of $Xe^{(q-2)+}(nl,n'l')$ states to highly excited states of $Xe^{(q-1)+}$.

In Fig. 3 we show absolute cross sections for transfer ionization and true double-electron capture for Xe^{q+}-Xe collisions. The latter cross sections increase by about one

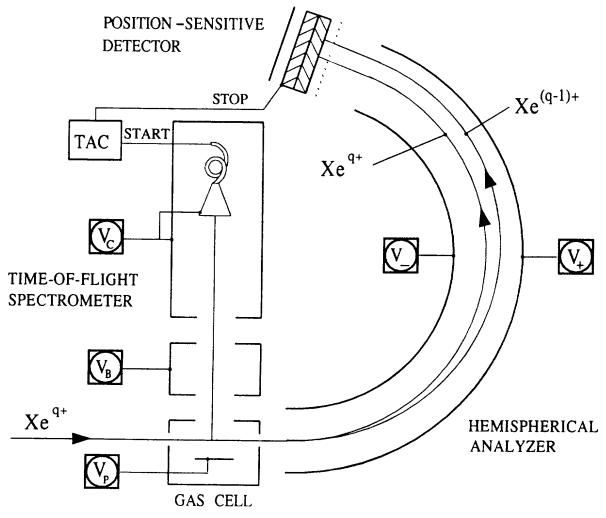

FIG. 1. Schematic of the experimental setup. Highly charged Xe^{q+} ions ($15 \leq q \leq 42$) entered the collision gas cell. The voltage between the analyzer spheres $V_+ - V_-$ was constant during a measurement and we have indicated a situation where the direct beam Xe^{q+} and the charge-exchanged beam $Xe^{(q-1)+}$ hit the detector. Recoil ions, produced in the gas cell, were extracted by a positive pusher voltage V_P further accelerated by V_B and the Ceratron potential V_C (cf text).

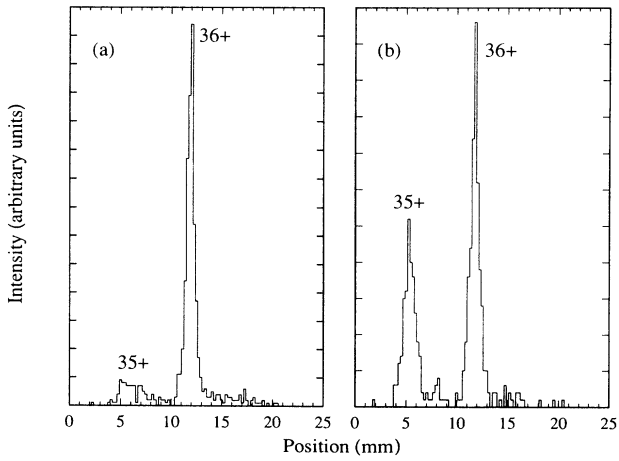

FIG. 2. The intensities of the 25-mm-diam two-dimensional position-sensitive detector projected on the axis of energy dispersion in coincidence with (a) Xe^{+} and (b) Xe^{2+} recoils for Xe^{37+}-Xe collisions. In both spectra, the peaks around 12 mm are due to Xe^{36+}, while the peaks at about 6 mm are due to Xe^{35+}. The small peak produced by the Xe^{35+}-Xe^{+} coincidences in (a) is due to double collisions (cf text).

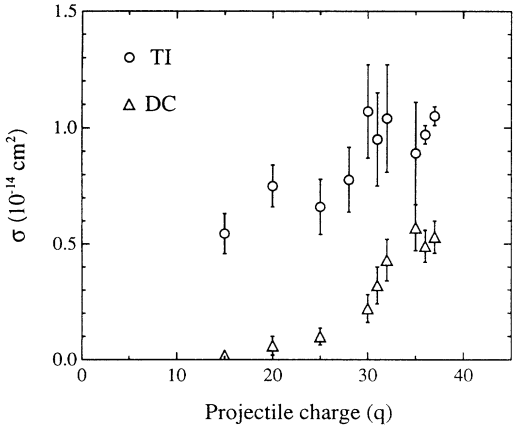

FIG. 3. Absolute cross sections for transfer ionization (○) and true double-electron capture (△) for Xe^{q+}-Xe collisions as a function of the projectile charge state q. The error bars are due to statistical errors and uncertainties in double-collision corrections.

order of magnitude between $q=25$ and 35, while the former ones increase only slightly throughout the same q region. This is the first experimental manifestation of the effect that true double-electron capture becomes comparable with transfer ionization at very high q.

The present coincidence data give a direct measurement of $P_{rad}(q)$ (4). These data are shown in Fig. 4 for Xe^{q+}-Xe and Xe^{q+}-He collisions. We want to stress two features exhibited in Fig. 4. First, the rapid increase in $P_{rad}(q)$ for both targets with an onset at $q=28$ reaching P_{rad} values of about 0.4 close to $q=35$, shows that radiative stabilization indeed is an important process at very high q. Second, there has been a general expectation that the projectile can be treated as a point charge without internal structure in collisions between projectile ions of very high charge and neutral atoms. This has been motivated by the fact that electron transfer to projectiles

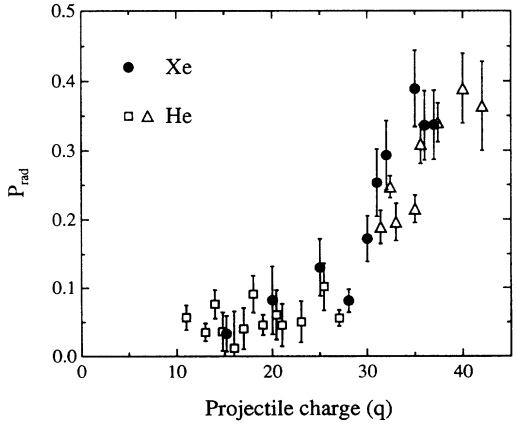

FIG. 4. The probabilities for radiative stabilization, P_{rad}, as functions of the projectile charge state q for double Rydberg states of $Xe^{(q-2)+}$ after transfer of two electrons from Xe (●) and from He (□, from Ref. [13]; △, present measurement). The error bars represent statistical errors and uncertainties in double-collision corrections.

of very high charge take place at internuclear distances in excess of 10 a.u., while the size of the core is much smaller than 1 a.u. Nothing contradicts this reasoning for the initial part of the electron-transfer process. However, close similarities of $P_{rad}(q)$ for double Rydberg states of $Xe^{(q-2)+}(nl,n'l')$, populated in collisions with Xe and He, suggest that the electronic structure of the incoming projectile rather than the exact nature of the initial capture state (which depends on the initial binding of target electrons) is decisive for the balance between the transfer ionization and true double-electron-capture processes.

We note that the increase in $P_{rad}(q)$ occurs within the q region where the number of vacancies in the Xe^{q+}-projectile $3d$ shell increases. In the following, we will attempt a qualitative explanation for the rise in $P_{rad}(q)$ in the $26<q<36$ region. The main components in the argument are (i) the initial capture process and the subsequent radiative cascade will favor population of $4fn''l''$ states; and (ii) the rates for radiative decay for $4fn''l''$, but not for autoionization, will be strongly affected by the number of holes in the $3d$ shell.

We assume that the experimentally corroborated [10] ECB prediction about population of double Rydberg projectile states (n,n') is valid. Some uncertainty still remains about a possible l (l') state selectivity in the present ($v=0.1$–0.2 a.u.) velocity regime. A preference for high l has been demonstrated for collision velocities

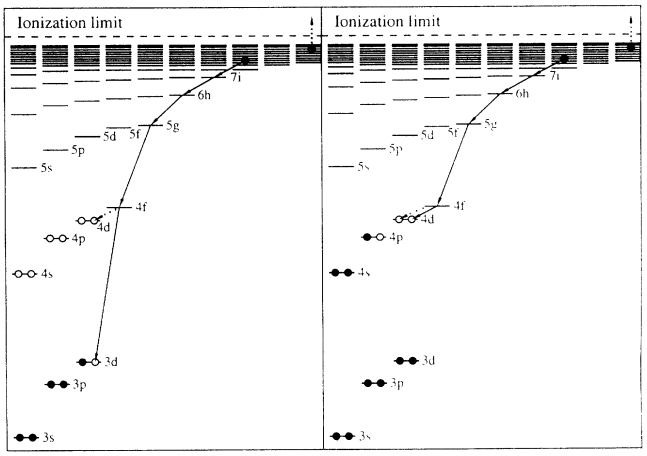

FIG. 5. Schematic diagrams of photon cascade paths (full lines) from double Rydberg states populated after transfer of two electrons to Xe^{q+} projectiles with (left) and without (right) holes in the $3d$ shell. Filled core shells are indicated by two filled circles, while vacancies in the shells close to the core are indicated by one or two open circles. The $4fn''l''$-$3dn''l''$ transition is expected to be fast and consequently the radiative lifetime of the $4fn''l''$ state will be short, when there are vacancies in the $3d$ shell (left figure). When the $3d$ and $4s$ shells are filled and the $4p$ shell is partly filled (right figure) a $4fn''l''$-$4dn''l''$ transition must be completed before stabilization can occur giving long radiative relaxation times of $4fn''l''$ states (cf text). Autoionization processes (dotted lines) favor emission of slow electrons strongly and their rates are much less affected by the introduction of $3d$ holes.

approaching 1 a.u. [20], while collisions below 0.01 a.u. show a complete lack of l selectivity [21]. Here we assume that double Rydberg states (n,n') to a fair extent will have l and l' values of $(l,l')>(3,3)$. In Fig. 5 we indicate one-electron energy levels schematically and show competing radiative decay (full lines) and autoionization (dotted lines) for $Xe^{(q-2)+}$ double Rydberg states with (left) and without (right) vacancies in the $3d$ shell. In order to complete the radiative stabilization process each intermediate doubly excited state of $Xe^{(q-2)+}$ above the lowest ionization limit of $Xe^{(q-1)+}$ must survive from autoionization. We assume that the excitation energy is sufficient for autoionization as long as the inner of the excited electrons lies above the outermost partly filled (or empty) (nl) level of the core. The probability of an electric dipole transition increases strongly with the energy difference between the initial and final states. The radiative decay will thus favor cascading to states with angular momentum approaching their maximum values, so that the inner electron $(n'l')$ will favor decay to an $l'-1$ state with a principal quantum number as small as possible, i.e., equal to l'. After following the yrast cascade chain it will then eventually end up in a $4fn''l''$ state. This reasoning is supported by observations of the yrast transitions only in photon cascades of multiply excited states created in Kr^{18+}-Ar, Kr collisions [22].

The strong influence of the electron emission energy on the probability for autoionization implies that electron emission from the $Xe^{(q-2)+}(4fn''l'')$ state preferably will leave $Xe^{(q-1)+}$ in a $4d$ state irrespective of the existence of a hole in the $3d$ shell. When, e.g., the $3d$ but not the $4p$ and $4d$ shells are filled the radiative decay of the $4fn''l''$ state must pass a $4dn''l''$ state before it can stabilize through a transition to a singly excited $4pn''l''$ configuration. This means that a slow $\Delta n=0,\Delta l=-1$ electric dipole transition (which only increases linearly with q) must be completed before stabilization and thus autoionization dominates. When the Xe^{q+} projectile has a hole in the $3d$ shell the $4f$ state can decay radiatively to a singly excited configuration of $Xe^{(q-2)+}$ by a fast $\Delta n=1,\Delta l=1$ electric dipole transition. Then the radiative process can compete with autoionization. An increase in $P_{\rm rad}(q)$ can thus be expected for an increasing number of $3d$ holes occurring in the $26<q<36$ region.

The explanation we have suggested relies on an assumption that the two excited electrons to a fair extent survive from autoionization until states with one electron close to the core are reached. This accounts for the observed increase of $P_{\rm rad}(q)$ in the region $26<q<36$ and close similarities of $P_{\rm rad}(q)$ resulting from Xe^{q+}-Xe and Xe^{q+}-He collisions. Note that $P_{\rm rad}$ has not reached 0.5 even at the highest q and that there seems to be very little variation in $P_{\rm rad}$ between $q=35$ and 42. Extending the argument, we expect the next strong increase in $P_{\rm rad}$ for Xe^{q+} projectiles when $2p$ vacancies $(q\geq 45)$ are introduced.

In summary, we have shown experimentally that the cross sections for true double-electron capture increase rapidly with q for very high q. We have also argued that these findings imply a breakdown of the basic model concept of a pointlike highly charged projectile in slow collisions between very highly charged ions and atoms.

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