Double Rydberg States Formed in Stabilized Triple Electron Capture

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Double Rydberg states were populated by triple electron capture in collisions of highly charged ions with atoms. The highest stabilization of states produced by triple capture at $v \cong 0.2$ a.u. was found for binding energies of the projectile electrons near the limits of the $(2, n, n')$ and $(3, n, n')$ series. Population of triply excited double Rydberg states was confirmed by the emission of Rydberg transitions. Results are interpreted by two successive autotransfer into Rydberg states for $(3, n, n')$ states and of one-step two-electron autotransfer for $(2, n, n')$ states. [S0031-9007(96)01692-4]

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In slow ($v < 1$ a.u.) collisions of highly charged ions with atoms, electrons are generally captured from the target into highly excited states of the projectile which decay radiatively or by autoionization [1]. Substantial progress has been achieved in the understanding of two-electron capture processes by using electron, photon, and energy gain spectroscopies. $(nl, n'l')$ symmetrical or quasisymmetrical ($n' \approx n$) and asymmetrical ($n' \gg n$) states can be populated in double electron capture depending on collisional systems. Elaborate quantum multistate close coupling calculations [2] give precise cross sections for production of either configurations. Contributions of single capture followed by transfer excitation and of other dielectronic interactions for populating asymmetrical configurations [3–5] depend on collision systems and impact energies. In the case of primary population into (n, n') symmetrical configurations which are strongly mixed with (low n_1, high n_2) Rydberg states, the transfer from the former to the latter by autotransfer to Rydberg states (ATR) is very efficient [6,7]. In a recent paper, we showed that the radiative stabilization of both electrons in double capture is the highest when the asymmetrical Rydberg series $(3, n)$, $(4, n)$, and $(5, n)$ are populated [8]. Here, the radiative stabilization of three electrons in triple electron capture is considered. To our knowledge, true triple capture cross sections have only been measured for collisions of Ar^{q+} ($q = 4$–11) on Ar [9,10]. The classical barrier model gives predictions [10] of the true triple capture cross sections which are relatively good for $q < 5$ and too low for $q > 6$. Autoionization yields of $3l3l'nl''$ ($n = 4$–9) triply excited states of N V, calculated in SCA (single configuration average) approximation [11], are so high that radiative stabilization can be neglected. More experiments using new techniques are needed for understanding the true triple electron capture processes and the stabilization schemes of triply excited states, mainly in highly charged ions, for which theory and first experimental results disagree and have left puzzles.

This Letter reports the measurements of relative cross sections for true (radiatively stabilized) triple capture (σ_{TTC}) and for triple capture followed by autoionization of one electron (σ_{1ATC}) and two electrons (σ_{2ATC}), in collisions of bare C^{6+}, O^{8+}, and Ne^{10+} ions on Ne, Ar, Kr, and Xe at an acceleration voltage of 2 kV ($v \cong 0.2$ a.u.). The variations of the stabilization ratio $R_s^3 = \sigma_{\mathrm{TTC}}/(\sigma_{\mathrm{TTC}} + \sigma_{\mathrm{1ATC}} + \sigma_{\mathrm{2ATC}})$ and the ratio of the total triple electron capture to the single electron capture $\sigma_{\mathrm{TEC}}/\sigma_{\mathrm{SEC}}$ have been studied in terms of the three-electron capture binding energies estimated from the classical barrier model [12]. Radiative Rydberg transitions emitted in the visible range have been analyzed in the case of the Ne^{10+} + Ar collision. Results show that double Rydberg series play an important role for the stabilization of three electrons in triple electron capture.

The experimental setup has already been described [13,14]. The ions were extracted from the electron cyclotron resonance (ECR) ion source of the AIM (accélérateur d'ions multichargés), a CEA/CNRS facility at Grenoble. Beam currents of magnetically analyzed C^{6+}, O^{8+}, or Ne^{10+} ions onto the target were typically 1 nA when recording photon spectra and were reduced to less than 1 pA for coincidence measurements. The rare gas target was furnished by an effusive gas jet. The effective density was adjusted between 2 and 5×10^{-5} mbar depending on the kind of measurements, either coincidence or spectroscopy. The residual pressure was 2×10^{-8} mbar in the accelerator beam line and 2×10^{-7} mbar in the collision chamber. The scattered ions, detected by a channeltron electron multiplier (CEM), were analyzed by a cylindrical electrostatic analyzer. Intensities have been corrected for CEM efficiency [9]. The rare gas recoil ions were extracted by an electric field of 10 V/cm perpendicular to the ion beam. Their charge state was determined by time of flight up to multichannel plates whose efficiency has been assumed to only depend on ion energies [9]. A 2π parabolic mirror collected visible photons emitted in the interaction region and sent them into a spectrometer equipped with a 1200 lines/mm grating blazed at 500 nm. Photons were detected by a cooled selected EMI 6256 photomultiplier with 0.1 dark count per second. A positive voltage has been applied to the collision region in order to discriminate collisions

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occurring inside the gas jet from collisions occurring before it. The single, double, and triple electron capture processes have been analyzed by scanning the voltage applied to the electrostatic analyzer (vertical scale) and measuring the time of flight of the recoil ions (horizontal scale) leading to 2D spectra.

Horizontal projections of 2D spectra for Ne^{10+} + Ar collisions at 20 keV impact energy are presented in Fig. 1. They are time of flight spectra of recoil ions recorded in coincidence with successive charge states of the outgoing projectiles. The Ar^{+}, Ar^{2+}, and Ar^{3+} peaks corresponding to Ne^{9+} outgoing ions [Fig. 1(a)] are due to single electron capture (SEC), autoionizing double capture, and 2ATC, respectively. Corresponding peaks of Fig. 1(b) (Ne^{8+}) result from double collision, true double capture, and 1ATC. In Fig. 1(c) (Ne^{7+}), the Ar^{+} and Ar^{2+} peaks are due to double collisions and the Ar^{3+} one to TTC. The signals were corrected for the low contributions (about 1%) of double collisions using the method described in Ref. [14]. The stabilization ratio R_s^3 has been measured for collision systems combining C^{6+}, O^{8+}, Ne^{10+} projectiles and Ne, Ar, Kr, Xe targets. Relative cross sections deduced from peak intensities are given in Table I. In Fig. 2, the measured R_s^3 are plotted as a function of $E(10/Z)^2$ which is the normalized three-electron binding energy on the projectile of incident charge state Z^{+}. This E energy, determined by the electron independent extended classical barrier (ECB) model, is the sum $E_1 + E_2 + E_3$ of binding energies on the projectile of the three external electrons which had I_1, I_2, I_3 ionization energies on the target [12]. For instance, for Ne^{10+} + Ar collision, the binding energies on the projectile calculated in the ECB model for $I_1 = 15.76$ eV, $I_2 = 27.63$ eV, and $I_3 = 40.74$ eV are $E_1 = 31.9$ eV, $E_2 = 45.79$ eV, and $E_3 = 61.17$ eV. This calculated binding energy agrees relatively well with energy gain measurements [1]. Relative error on R_s^3 is estimated at about 20% from statistics and uncertainties on double collision corrections. The double ionization thresholds of Ne X are indicated by vertical lines along the abscissa axis. The $(2, n, n')$, $(3, n, n')$, and $(4, n, n')$ double Rydberg series of Ne VIII extend to the higher values of energy than these limits. Figure 2 exhibits clearly large variations of the stabilization when three-electron binding energies vary along and above double Rydberg series. R_s^3 grows from 0.4 up to 4.5% at the vicinity of the $n_{\text{in}} = 2$ limit and from 0.2 to 2.4% around the $n_{\text{in}} = 3$ limit. These enhancements of radiative stabilization strongly suggest that true triple capture is due to the population of highly asymmetrical triply excited states (n_{in}, n, n') where n and $n' \gg n_{\text{in}}$.

Above results deduced from the comparative study of charge stabilization in a large number of collision systems have been complemented by optical measurements. As an example, the visible spectrum emitted by the Ne^{10+} + Ar collisions is displayed in Fig. 3. Ion beam intensity was

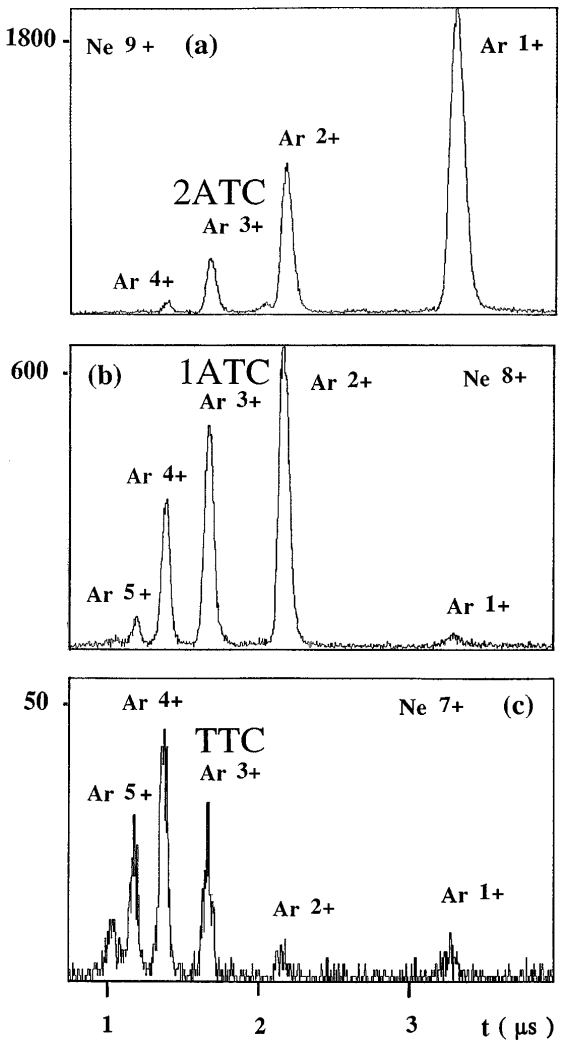

FIG. 1. Recoil ion spectra for Ne^{10+} + Ar collisions in coincidence with stabilized projectiles Ne^{9+} (a), Ne^{8+} (b), and Ne^{7+} (c). Triple captures followed by different decaying processes are responsible for Ar^{3+} peaks: 2ATC in (a), 1ATC in (b), and TTC in (c).

weak, so that photon spectra could be obtained only at low resolution. Attribution and intensity estimation of the lines were made easier by comparing several spectra recorded with various positions of the gas injection needle. When the ion beam volume observed by the spectrometer was downstream the gas jet, only the lines emitted by the projectile with a slow decay were observed, whereas the lines emitted by the target were also present when the observed volume was inside the gas jet. So, besides Ar II lines [17] emitted by the target, the Ne VIII 7-8, Ne IX 8-9, and Ne IX 9-10 lines have been clearly identified. The Ne IX 8-9 line at 343 nm was blended with Ar II lines. The contribution of argon lines was only a few percent of the total intensity. Ar II lines are due to double captures into asymmetrical configurations followed by recaptures of a Rydberg electron by the target [6] as already observed [15]. The weak Ne VIII 7-8 line

TABLE I. Stabilization ratio R_s^3 $(=\sigma_{\mathrm{TTC}}/\sigma_{\mathrm{TEC}})$ and relative cross sections for bare ions on rare gas atoms.

Collision	R_s^3 (%)	$\sigma_{1\mathrm{ATC}}/\sigma_{\mathrm{TEC}}$ (%)	$\sigma_{2\mathrm{ATC}}/\sigma_{\mathrm{TEC}}$ (%)	$\sigma_{\mathrm{TEC}}/\sigma_{\mathrm{SEC}}$ (%)	$E(10/Z)^2$ (eV)
C^{6+} + Ne	0.4	81.8	17.8	53	471.1
C^{6+} + Ar	4.5	70.0	25.5	17	315.9
C^{6+} + Kr	1.4	32.0	66.6	43	282.8
C^{6+} + Xe	0.1	62.7	37.2	65	245.5
O^{8+} + Ar	0.8	71.3	27.9	11	199.0
O^{8+} + Kr	1.0	62.2	36.8	29	178.0
O^{8+} + Xe	1.7	60.1	38.2	26	154.6
Ne^{10+} + Ar	2.4	56	41.6	15	138.9
Ne^{10+} + Kr	1.8	45.3	53.4	23	124.2
Ne^{10+} + Xe	1.1	54.6	44.3	18	107.9

at 296.3 nm is attributed to the decay of the Rydberg electron $n' \gg n_{\mathrm{in}}, n$ of triply excited (n_{in}, n, n') states produced by TTC and by partly autoionizing multielectron captures. This transition can also be due to double collision events where true double electron capture on Ne^{9+} previously produced by single capture collision could populate Ne VIII Rydberg states. The contribution of double collisions has been roughly evaluated to 40% of total intensity. A precise determination of contributions of different processes requiring a coincidence measurement of Ar^{3+} recoil ions and Ne VIII photons has not been performed because the Ne VIII line intensity was too weak. The Ne IX lines at 343.7 and 478.6 nm correspond to radiative transitions following two-electron [16], three-electron, or multielectron captures. The high intensity of Ne IX 8-9 line at 343.7 nm allowed us to detect it in coincidence with recoil ions and to assign the primary capture processes involved. The coincidence spectrum (Fig. 4) displays an intense Ar^{3+} peak showing that a large part of this line is due to TEC which populates primarily $Ne^{7+}(4,5,6)$ states. The Ne IX 8-9 transition is essentially attributed to the 1ATC channel and to the decay of $Ne^{8+}(3, n \geq 9)$ states which can be populated from the $(4,5,6)$ states after two successive ATR-AI (autoionization) or AI-ATR processes passing through $(4,4,n \geq 9)$ or $(4,4)$ intermediate states. This line can also be related to the TTC channel and to the decay of the intermediate electron n of triply excited double Rydberg states $(3,n,n')$ of Ne^{7+} with $9 \leq n \ll n'$. The wavelength of such a decay is about the same as for the external n electron of Ne^{8+}, because the extra $n' \gg 9$ electron is loosely bound. The population of $(3,n,n')$ could be interpreted by two successive ATR processes from primary $(4,5,6)$ states via $(4,4,n' \gg 6)$ states. The two ATR processes are made possible by the overlap of the double Rydberg series $(3,n,n')$ and an intermediate single Rydberg series $(4,4,n')$. An estimation of each contribution would ask for triple coincidence

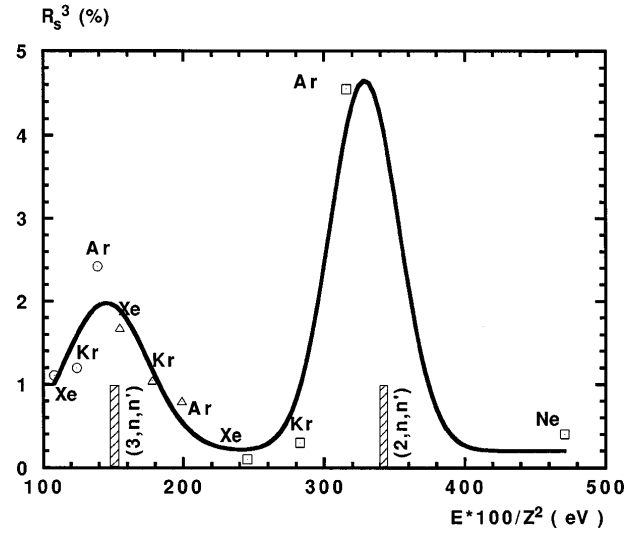

FIG. 2. Stabilization ratio of triple electron capture of C^{6+} (□), O^{8+} (△), and Ne^{10+} (o) from rare gas atoms as a function of the triple electron binding energy E.

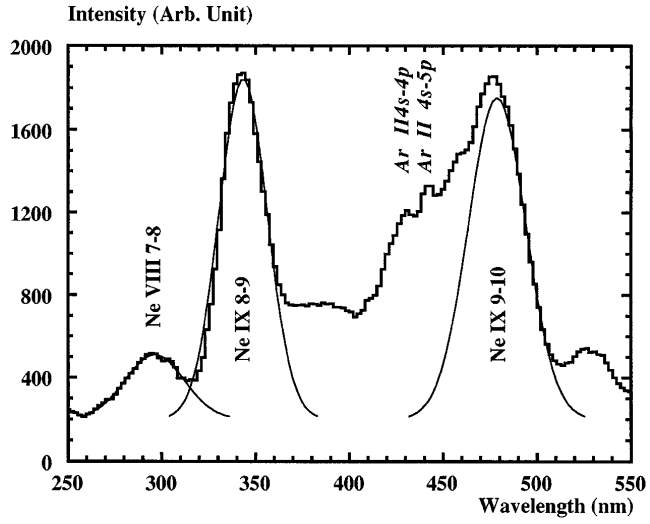

FIG. 3. Visible spectrum for Ne^{10+} + Ar collisions at 50 keV. The Ne VIII (7-8) line at 296 nm is due to the population of (n_{in}, n, n') triply excited states with $n' \gg n_{\mathrm{in}}, n$ and also, partly, to double collisions.

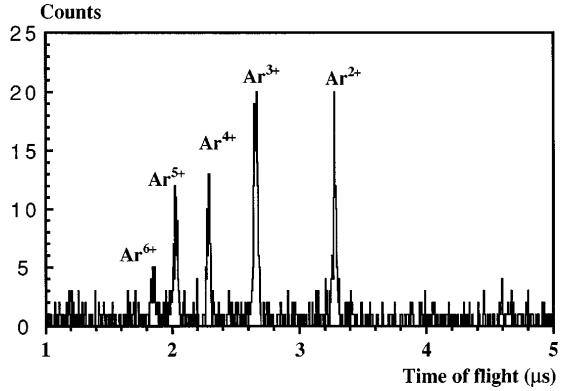

FIG. 4. Charge spectrum of recoil ions in coincidence with Ne IX 8-9 photons for Ne^{10+} + Ar collisions. The Ar^{3+} peak is due to triple capture followed by one-electron autoionization and photon emission from $(n, n' \geq 9)$ states and to triple capture into $(n_{\text{in}}, n \geq 9, n' \gg n)$ states.

measurements between Ne IX 8-9 photons, Ar^{3+} recoil ions, and Ne^{7+} stabilized projectiles.

The stabilization ratio R_s^3 of the C^{6+} + Ar collision is the highest that we found. It is related to the population of the double Rydberg $(2, n, n')$ states (Fig. 3). However, here, contrary to the Ne^{10+}-Ar system, the $(3, 3, 4)$ and $(3, 3, 5)$ states primarily populated by triple capture cannot be transferred to $(2, n, n')$ configurations via two successive ATR processes, because there is no intermediate C IV single Rydberg series in the vicinity. The only possible transfer from primary states to asymmetrical $(2, n, n')$ is through a direct three-electron process. The interaction between the two inner shell $(3, 3)$ electrons leads in general to the decay onto $n = 2$ of one of the electrons and the autoionization of the other one. The outgoing electron could also share the released energy with the third external electron ($n = 4$ or 5), resulting in a two Rydberg electron bound state with high n, n'. Similar simultaneous two electron excitation has been observed in double photoionization experiments [18]. However, in present collisions, the excitation is in bound states instead of in the continuum. When the interaction between the outgoing and external electrons is less important, the Auger process is completed by the autoionization of one electron and the excitation of the other one to a higher state ($n > 4$ or 5). This process is favorable for the 1ATC channel and could be compared with the shakeup of the external electron in interelectron photoionization experiment [19]. That the stabilization ratio R_s^3 has been found higher for $(2, n, n')$ configurations than for $(3, n, n')$ ones can be attributed to lower autoionization probabilities for lower n_{in} values of the internal electron. The stabilization ratios $R_s^3 = 2.4\%$ and 1.2% we have measured for Ne^{10+} + Ar collisions at 20 and 190 keV have to be compared to the value $R_s^3 = 0.5\%$ found for Ar^{10+} + Ar collisions at 400 keV [10]. The difference can be explained by R_s^3 velocity dependence and by core effects [8].

In conclusion, relative cross sections for true triple capture and for triple capture followed by one- or two-electron autoionization have been measured in C^{6+}, O^{8+}, Ne^{10+} + He, Ar, Kr, and Xe collisions. High values of the deduced stabilization ratio R_s^3 has been found when (n_{in}, n, n') asymmetrical $(n_{\text{in}} \ll n, n')$ states were populated. The observation of Rydberg transitions from n and n' electrons has confirmed the population of asymmetrical configurations. A detailed study of Rydberg transitions, including their radiative decays, would be tentatively effected for a determination of the initial population of double Rydberg states produced.

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