Double Rydberg States of High Angular Momentum ($l = 6$–8) Produced in Ar VIII by Ar^{9+}-Cs Collisions

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Doubly excited states have been produced in Ar VIII by double electron capture from cesium atoms by 90-keV Ar^{9+} ions. Transitions between $1s^22s^22p^55g7l$ ($l=3,\ldots,6$), $1s^22s^22p^55g8l$ ($l=4,\ldots,7$), and $1s^22s^22p^55g9l$ ($l=5,\ldots,8$) double Rydberg states have been observed by photon decays. The spectra have been analyzed using an independent-particle framework with a frozen-core approximation leading to a good agreement for transition energies.

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Doubly excited $(nl,n'l')$ Rydberg atoms are of considerable interest as being three-body quantum-mechanical systems. Theories[1] of highly excited two-electron ions generally center on two extreme situations. When $n' \gg n$, the outermost electron can be considered as attracted in a Rydberg orbital to the parent ion built up of the core and the other excited electron. Properties of these states can be analyzed in terms of independent-electron models and quantum-defect theory. When n' is comparable to n, the above expectations will no longer be valid and correlation with be significant between the pair attached to the grandparent ion. Initial experimental investigations have concentrated on the alkaline-earth atoms (mainly Ba) with two easily excited electrons outside an inert core. Spectroscopic studies of barium atoms have used multistep laser excitation[2-4] because single-photon excitation will not efficiently excite these states. This method has the advantage of high spectral resolution. However, only states with $n' \gg n$ and low l have so far been observed using this technique. The double Rydberg states we have produced belong to an intermediate class of such states where n' is not so much higher than n, and moreover, larger values of l and l' have been observed.

Our experiments on doubly excited Rydberg states have been conducted by means of a completely different excitation technique. We have used the efficiency of collisions between multiply charged ions and multielectron neutral atoms to produce doubly excited states by double electron capture.[5] In such multi-ionized atoms, doubly excited states with both electrons in low principal quantum numbers have been analyzed by low-resolution electron spectroscopy,[6] showing that autoionizing levels are strongly populated. Photon spectroscopy is only applicable to levels whose radiative rates are not negligible compared to autoionization rates. Because of their metastability against Coulomb autoionization the core-excited quartet levels of alkali-metal ions are good candidates for optical investigations. In a recent paper, referred hereafter as I, we have presented results on $1s^22s^22p^53snl\,{}^4L_J$-$1s^22s^22p^53sn'l'\,{}^4L_{J'}$ doubly excited transitions of Ar VIII with $n=7$–12 and $l=6$–11.[7,8]

In this Letter we report spectroscopic studies in the visible range of the sodiumlike $2p^55g\,7l$ ($l=3$–6), $8l$ ($l=4$–7), and $9l$ ($l=5$–8) quartet terms of Ar^{7+} observed in double capture between Ar^{9+} and cesium. Such doubly excited Rydberg states are currently called valley states.[9] We have used uncorrelated wave functions to calculate the screening constants as a starting approximation to analyze the spectra.

An Ar^{9+} beam was sent at 90 keV energy through an atomic cesium vapor produced in a cell maintained at a constant temperature of 70 and 90 °C corresponding to pressures of about 8×10^{-5} and 3×10^{-4} mbar, respectively. Photons originating from Ar^{9+}-Cs collisions were observed at an angle of 90° to the Ar^{9+} beam axis using a 0.6-m Czerny-Turner spectrometer, equipped with a 1200-lines/mm grating blazed at 300 nm and a photomultiplier covering the spectral range 200–650 nm.

In paper I we had presented typical spectra obtained from Ar^{8+} and Ar^{9+} beams. We are able to resolve peaks in the low-wavelength wings of intense Ar VIII $8k$-$9l$ and $7i$-$8k$ lines and to identify them with the $3s8k\,{}^4L_{19/2}$-$3s9l\,{}^4M_{21/2}$ and $3s7i\,{}^4K_{17/2}$-$3s8k\,{}^4L_{19/2}$ transitions, respectively. Using better statistics, at the sacrifice of spectral resolution, other peaks also related to Ar VIII $7i$-$8k$ and $8k$-$9l$ lines have been observed at still shorter wavelengths (Fig. 1). These new lines were not observed when using an incident Ar^{8+} beam. They were present in Ar^{9+} beams colliding with low-temperature cesium vapor (70 °C). Thus, they are interpreted as being produced from a double-electron-capture process to $1s^22s^22p^5nln'l'\,{}^4L$ terms of Ar VIII.

We have determined the reaction window for double-electron capture in Ar^{9+}-Cs collisions using the extended classical over-barrier model of Niehaus.[10] Results presented in Fig. 2 show that $5ln'l'$ ($n' \geq 7$), $6ln'l'$ ($n' \geq 6$), $7l7l'$, and $8l8l'$ doubly excited states are possibly populated. Here, we limit our study to $5ln'l'$ states because they are more easily calculated than the other ones. More comprehensive results will be published later.

The l distributions of doubly excited and singly excited

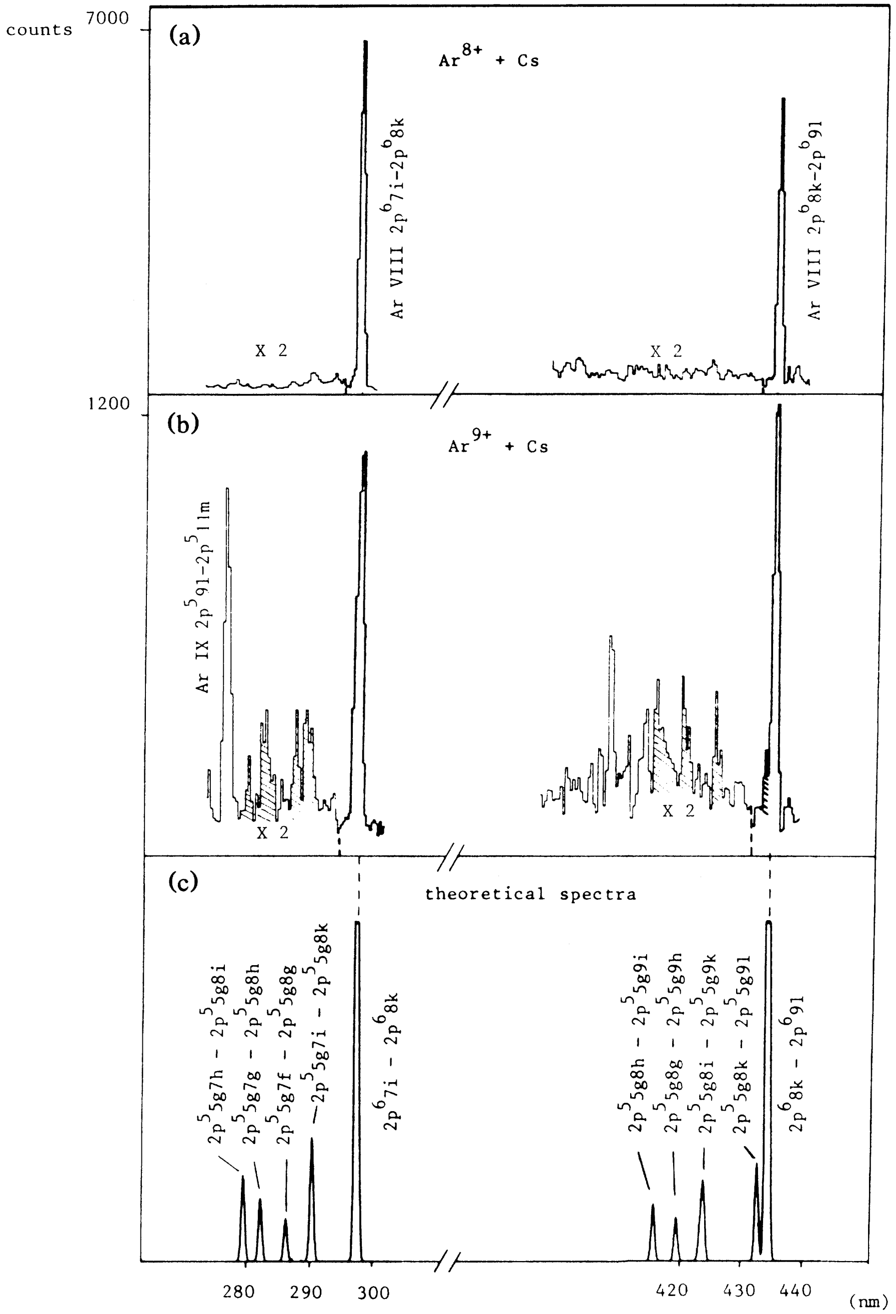

FIG. 1. Short-wavelength spectra of Ar VIII $7i$-$8k$ and $8k$-$9l$ lines: (a) Observed in Ar^{8+}-Cs collisions (80 keV) after single electron capture. (b) Observed in Ar^{9+}-Cs collisions (90 keV). Ar VIII ($2p^67i$-$2p^68k$) and ($2p^68k$-$2p^69l$) lines are broadened by Ar VIII ($2p^53s7i$-$2p^53s8k$) and ($2p^53s8k$-$2p^53s9l$) lines, respectively. Transitions between double Rydberg states of Ar VIII are observed in short-wavelength parts of the spectra. (c) Calculated for $2p^55g7l$-$2p^55g8(l+1)$ and $2p^55g8l$- $2p^55g9(l+1)$ transitions of Ar VIII. Amplitudes of $2p^67i$- $2p^68k$ and $2p^68k$-$2p^69l$ are only indicative. They depend on the double-collision rates, i.e., the vapor pressure in the cell. A better agreement is obtained for $2p^55g8l$-$2p^55g9(l+1)$ than for $2p^55g7l$-$2p^55g8(l+1)$ doubly excited transitions.

levels are very different. By single capture in Ar^{9+}-Cs collisions, $n=11$ and 12 levels are populated. Observed 7-8 and 8-9 transitions are due to cascades and then only $\Delta n=1$ yrast (maximum l) transitions are intense. Intensities of the $2p^68k$-$2p^67i$ and $2p^69l$-$2p^68k$ lines have been calculated using a cascade program assuming a $2l+1$ distribution of populations for initial levels $n=11$ and 12. For example, one obtains $I(7h-8i)/I(7i-8k)=0.22$ and

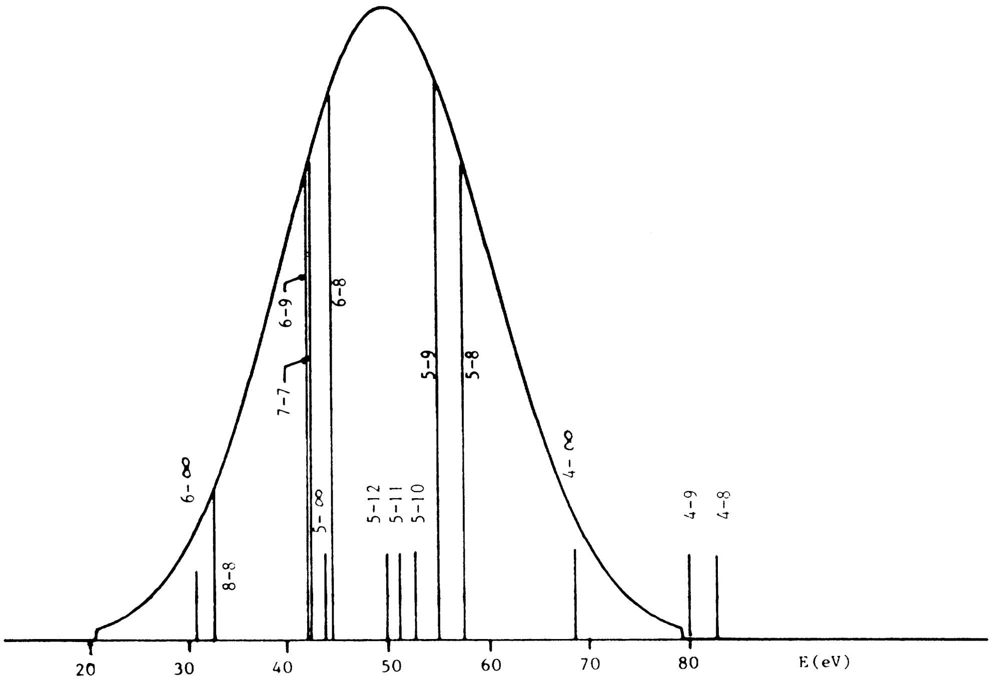

FIG. 2. Reaction window for double electron capture in Ar^{9+} from Cs at 90 keV energy calculated using the Niehaus model. In $2p^5nln'l'$ double Rydberg states, nl and $n'l'$ are the quantum numbers of inner and outer electrons, respectively. For 7-7 and 8-8 transitions, the screening constant σ is taken equal to 0.25 (Ref. 11).

$I(7g-8h)/I(7i-8k)=0.02$. Other population models lead to the same conclusion. On the contrary the 7-8 and 8-9 transitions emitted during double electron capture are the consequence of a direct population of various l angular momenta as for $5g8l$ and $5g9l$.

Our calculations of level energies are based, as a first approximation, on an independent-particle model because the wave functions $n=5$ and $n'=7,8,9$ are still relatively well separated and the exchange effects can then be neglected. We have used a modified Rydberg formula for expressing the energy of an electron n',l',

$$E_{nln'l'}=-\frac{R(Z-\sigma_{nln'l'})^2}{n^2}\,.$$

The screening constant $\sigma_{nln'l'}$ depends on the quantum numbers nl, $n'l'$ of both the inner and outer electrons. Quantum defects of the $n'l'$ electron have been neglected because the $1s^22s^22p^5$ core size is very small compared to the Rydberg size.

For the completely uncorrelated wave functions for the two Rydberg electrons the screening constant is given by[12]

$$\sigma=\int_{r'=0}^{\infty}\int_{\Omega'}\left[\int_{r=0}^{r'}\int_{\Omega}|\varphi_{nl}(\mathbf{r})|^2d\mathbf{r}\right]|\varphi_{n'l'}(\mathbf{r}')|^2d\mathbf{r}'\,,$$

where φ_{nl} and $\varphi_{n'l'}$ are the hydrogenic wave functions with $Z=9$ and $Z=8$ effective charges, respectively. We have calculated the screening constant for the $5g7l$ ($l=3,\ldots,6$), $5g8l$ ($l=4,\ldots,7$), and $5g9l$ ($l=5,\ldots,8$) in that approximation.

The intensity of the $5g7l$-$5g8(l+1)$ and $5g8l$-$5g9(l+1)$ lines depend on level population distributions and on branching ratios for autoionization and radiative decays. We have taken into account the radiative branching ratios but not the relative value for autoionization and radiative rates because of the metastability of quartet states against autoionization. The $5gnl$ level decays to $4fnl$ and $5gn'(l-1)$ levels. The hydrogenic transition probabilities have been calculated using the effective charges $Z=9$ for the $5g$ level and $Z=8$ for the $n'l'$ level. Branching ratios 0.05, 0.04, and 0.03 have been deduced for the $5g8k$-$5g7i$, $5g8i$-$5g7h$, and $5g8h$-$5g7g$ transitions, respectively. Assuming a $2l+1$ initial population, the line intensities are found to be proportional to 7.0, 4.5, and 2.7 for the $5g8k$-$5g7i$, $5g8i$-$5g7h$, and $5g8h$-$5g7g$ transitions, respectively. The $5ln'l'$-$5l(n'-1)(l'-1)$ transitions have a lower calculated branching ratio for $l<4$ and are expected to be weaker. For example, the intensities of the $5f8k$-$5f7i$, $5f8i$-$5f7h$, $5f8h$-$5f7g$, and $5d8k$-$5d7i$ transitions are proportional to 3.0, 2.1, 1.3, and 1.2, respectively.

Theoretical spectra have been constructed using these rough calculations for energies and intensities [Fig. 1(c)]. A fair agreement with experimental spectra has been found for the $5g8l$-$5g9(l+1)$ and $5g7l$-$5g8(l+1)$ transitions. Tentative attribution of the $5gnl$-$5gn'(l+1)$

TABLE I. Single and double Rydberg transitions of Ar VIII observed in Ar^{9+}+Cs.

	$\lambda_{\rm expt}$ (nm)	$\lambda_{\rm theor}$ (nm)
$2p^{6}7i-2p^{6}8k$	297.58 ± 0.15	297.68
$2p^{5}5g7i-2p^{5}5g8k$	288.9 ± 0.5	290.5
$2p^{5}5g7h-2p^{5}5g8i$	279.3 ± 0.5	279.4
$2p^{5}5g7g-2p^{5}5g8h$	282.0 ± 0.5	282.2
$2p^{5}5g7f-2p^{5}5g8g$	286.9 ± 0.5	286.9
$2p^{6}8k-2p^{6}9l$	434.16 ± 0.15	434.19
$2p^{5}5g8k-2p^{5}5g9l$	432.1 ± 0.2	432.5
$2p^{5}5g8i-2p^{5}5g9k$	425.0 ± 0.5	423.8
$2p^{5}5g8h-2p^{5}5g9i$	415.0 ± 0.5	415.7
$2p^{5}5g8g-2p^{5}5g9h$	419.0 ± 0.5	419.5

transitions is given in Table I. Agreement seems to be better for 8-9 than for 7-8 transitions than could be explained by a significant effect of correlation between $5g$ and $7l$ levels (close values of n and n'). The $6h8l-6h9(l+1)$ and $6h7l-6h8(l+1)$ transitions are shifted to shorter wavelengths than the $5gnl-5gn'l'$ ones because the screening constants are stronger for a $6h$ than for a $5g$ intern electron. These transitions seem to be weaker, maybe because Coulomb ionization rates are higher for a $6h$ than for a $5g$ intern electron.

High-n and -l doubly excited states have been observed by radiative decay for the first time in this experiment. Precise calculations of energy levels would require more complex theories.[11,13] Work for further identifications is in progress. Generally, double electron capture by multicharged ions at low energy in alkali-metal vapors has proved to be very promising for producing double Rydberg states, circular states, and maybe "ridge" states also.[9]

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