PHYSICAL REVIEW A VOLUME 43, NUMBER 1 1 JANUARY 1991

Subfemtosecond study of the hypersatellite-satellite cascade in "hollow" atoms

J. P. Briand, L. de Billy, P. Charles, and S. Essabaa
Laboratoire de Physique Atomique et Nucléaire–Institut du Radium, Université Pierre et Marie Curie,
4 place Jussieu F-75252 Paris CEDEX 05, France

P. Briand, R. Geller, J. P. Desclaux, S. Bliman, and C. Ristori
Service de Physique Atomique, Département de Recherche Fondamentale, Centre d'Etudes Nucléaires de Grenoble,
85X, F-38041 Grenoble CEDEX, France

(Received 23 July 1990)

The behavior of slow, fully stripped Ar^{18+} ions near metallic surfaces has been studied. These ions capture, while touching the surface, a large number of electrons in outermost shells, leading to the formation of "hollow" atoms. The timing of the hypersatellite-satellite cascade filling the double K-vacancy states is studied in this paper by using the internal clock constituted by the "tops" of the temporal sequence filling stepwise the L shell of the ion.

In a previous paper[1] we reported on experimental studies of the interaction of slow, highly charged Ar^{17+} ions, with metallic surfaces. In that paper we showed that the capture of a large number of target electrons into the excited $M, N, \ldots,$ states of the ion, on the very first monolayers of the surface, i.e., in a very short time, a few 10^{-16} s, leads to the excitation of very unusual atomic species where most of the electrons are located in the outermost shells ("hollow" atoms). The relaxation of these very excited ions with many electrons in the $M, N, \ldots,$ shells, and none in the L shell, is much slower and was found to proceed via two main, competing decay channels: (i) the filling of the L holes via LMM or LMN Auger transitions, and (ii) the filling of the K shell by L electrons (radiative decay, KLL Auger).

As shown in Ref. 1, the stepwise filling up of the L shell ($L^1 \to L^2 \to L^3 \to \cdots \to L^8$), via a cascade of LMM (LMN) Auger transitions, is, at each step, more probable than the filling of the K vacancy. The emission of a $K\alpha$ line by the ion then extends over a relatively long period of time. All states with any number $1 \leq n \leq 8$ of L spectator electrons are observed in Fig. 3(b) (KL^n satellites of the $L \to K$ transition). A simple model, using Larkins's statistical procedure,[2] led to the conclusion that each step of the cascade has roughly the same lifetime, of the order of $(4-8) \times 10^{-16}$ s, and that the eight observed lines may be used as time markers of the process or boundary marks along the ion trajectory.[1]

We report here on a similar experiment carried out with fully stripped Ar^{18+} ions traveling at low velocity (0.60 a.u.) toward a metallic surface. The filling in two steps of the two K vacancies, by the well-known hypersatellite ($K^{-2} \to K^{-1}L^{-1}$)-satellite ($K^{-1}L^{-1} \to L^{-2}$) cascade,[3] introduces some time delay in the intermediate state, which we study in this paper, by using the "tops" given by these ~ 0.5-fs time markers described in Ref. 1, i.e., the lifetimes of each of the levels of the cascade filling the L shell.

The fully stripped argon ions were produced by the 16-GHz ECR source developed in Grenoble by R. Geller. The ionic current was of the order of few nA. The x rays emitted in flight by the ions impinging a silver surface at normal incidence were studied first with a Si(Li) detector and second with a crystal spectrometer. The spectrometer was made of a flat mosaic graphite crystal ($\Delta\alpha = 0.4°$) focusing the x rays into a position-sensitive detector (Fig. 1). The typical resolution of this spectrometer was of the order of 5 eV for takeoff angles of a few degrees and the transmission of the order of a few 10^{-6}. This very high transmission allows the observation of the x-ray spectra emitted by these very weak Ar^{18+} beams, in runs lasting 6 to 8 h. The energy calibration (± 1.5 eV) of the spectrometer was achieved by looking at the L x-ray spectrum of silver irradiated by a 5-keV electron beam.

We present in Fig. 2 the $K\alpha$ hypersatellite spectrum ($K^{-2} \to K^{-1}$) obtained with Ar^{18+} beams, near the target surface, using a crystal spectrometer. This spectrum shows eight well-separated lines labeled K^0L^n and corresponding to all states with n L spectator electrons from $n=1$ up to $n=8$. The precise energy calibration of the spectrum (± 1.5 eV) enables the identification of the ionic configurations at the time of the $K\alpha$ line emission.

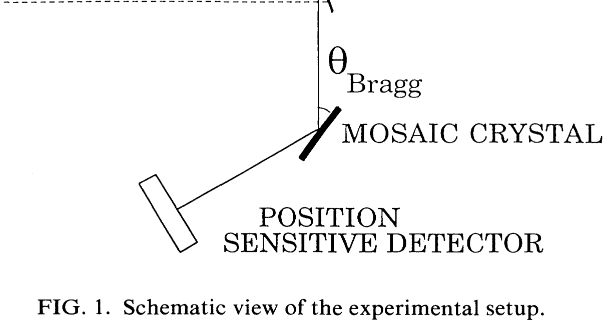

FIG. 1. Schematic view of the experimental setup.

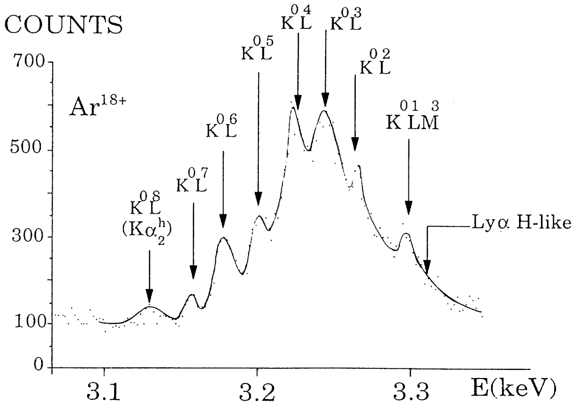

FIG. 2. Lyman-α spectrum ($K^0L^n \rightarrow K^1L^{n-1}$ hypersatellites) with Ar^{18+} beams observed with a crystal spectrometer. The solid line has been hand drawn to guide the eye. Arrows indicate theoretical positions of the lines.

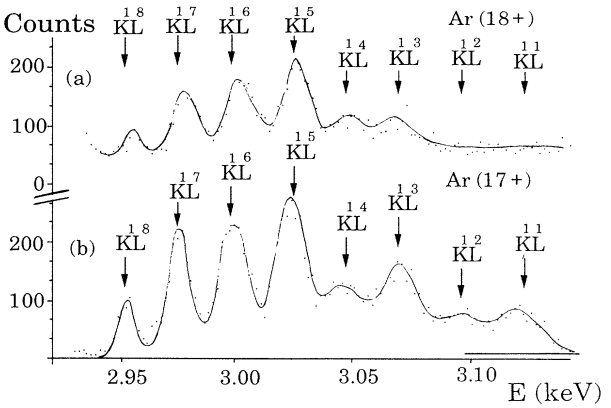

FIG. 3. Comparison of the Ar satellite lines ($K^1L^n \rightarrow K^2L^{n-1}$ spectrum), observed with (a) Ar^{18+} impinging atom (second step of radiative decay), and (b) Ar^{17+}. The background value is shown on the high-energy side as a flat solid curve. The solid line has been hand drawn to guide the eye. Arrows indicate theoretical positions of the lines.

The energy of the last line emitted exactly fits the one calculated[4] for $K\alpha_2{}^h$ in argon atoms with $K^0L^8M^8$ configuration. (As previously demonstrated,[5] the $K\alpha_1{}^h$ line of the spin-orbit doublet is very weak for $Z=18$ in comparison to $K\alpha_2{}^h$, owing to its triplet character in the *LS* coupling scheme). One must notice that, in this extreme case, the two *K* vacancies survive long enough for the outermost electrons to fill up the *L* shell. The first line of the cascade filling the *L* shell K^0L^1 is about 20 eV lower in energy than the Ly-$\alpha_{1,2}$ line of hydrogenlike argon. It corresponds, as in the case of Ar^{17+},[1] to configurations with a mean number of three *M* electrons and an unknown but large[1] number of *N* and *O* electrons $[K^0L^1M^3(N,O)^z]$. This fact can be explained easily: the cross sections for multiple capture (three *M* electrons plus an unknown number of *N* and *O* electrons) in the first monolayer of the target are very close to each other for Ar^{17+} and Ar^{18+} ions.

This spectrum is compared with the one recorded in the same conditions with Ar^{17+} beams and presented in Fig. 3(b). Two major differences appear in the shapes of these spectra [Figs. 2 and 3(b)].

(i) The hypersatellite distribution is mainly concentrated on the KL^2, KL^3, and KL^4 lines instead of being quasiuniform as in the case of Ar^{17+}.

(ii) The K^0L^8 and K^0L^7 hypersatellites are very weak.

These observations are explained by considering the time scale of the relaxation of the ion. It is well known that the transition probability of the hypersatellite lines in atoms with *closed shells* is roughly twice that of the diagram lines[6] and that the hypersatellite lines' natural widths are roughly three times larger than the usual $K\alpha$ diagram line. This is due to the fact that with two *K* holes the probability of the decay has roughly doubled, and that the intermediate state is still ionized once in the *K* shell. By contrast, when there are many holes in the *L* shell, the lifetime of the states is dominated by the *LMM* Auger channel and is not very dependent on the number of *K* holes. In this case, only the probability of decay by a $K\alpha$ line changes with the number of *K* vacancies. The probability of the first *K* hole filling is roughly two times larger than in the case of a single *K* vacancy; therefore more hypersatellite lines must be emitted at the early beginning of the *L* shell temporal sequence filling than in the Ar^{17+} case, i.e., with few *L*-shell electrons (first time markers: $KL^1 \rightarrow KL^4$), in agreement with the above results. The intensity of the K^0L^1 line, which cannot decay via a *KLL* Auger transition (the dominant mode of decay of the *L* shell), is smaller than for the other lines, as shown in Fig. 2. The decay of the *K* holes will then mainly take place when more than one *L* electron is present.

The stepwise filling of the two *K* holes introduces some delay corresponding to the lifetime of the second (satellite) transition. One must then observe after this delay a change in the filling of the *L* holes as shown in Figs. 3(a) and 3(b). In these figures, the K^1L^n Lyman-α spectrum observed with Ar^{17+} projectiles[1] and the corresponding spectrum, taken at the second step of the decay of K^{-2} states of Ar^{18+} ions ($K^1L^n \rightarrow K^2L^{n-1}$), are compared. The main difference between the two spectra is the change in relative intensity of the lines: the decay of K^1L^n states with Ar^{18+} projectiles occurs with more *L* electrons $3 < n \leq 8$ than in the case of Ar^{17+}. [The two K^1L^1 and K^1L^2 lines completely vanish in the spectrum (a), and the K^1L^3 and K^1L^4 are relatively weaker than in the spectrum (b).]. This easily could be explained by the "delayed" emission: the first *K* hole is mainly filled in the presence of few *L* electrons, as shown above, whereas the second one is filled, later, in presence of more *L* electrons. One then observes through this hypersatellite-satellite cascade some snapshots of the *L*-shell filling (an average of four electrons fill the *L* shell between the emission of the hypersatellite and satellite lines).

We present in Fig. 4 the Lyman spectrum observed

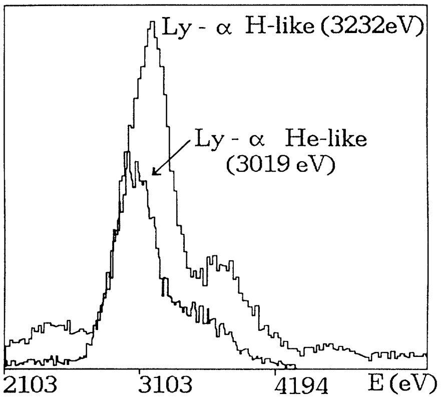

FIG. 4. K x-ray spectrum observed with a Si(Li) detector.

with a Si(Li) detector at a resolution of 180 eV (full width at half maximum). With such a resolution, it is possible to separate roughly the two main components in the decay of the hypersatellite-satellite cascade $K^{-2} \rightarrow K^{-1}L^{-1}$ and $K^{-1}L^{-1} \rightarrow L^{-2}$ (irrespective of the number of L spectator electrons). The energy of the hypersatellite lines $K^{-2} \rightarrow K^{-1}L^{-1}$ lies between the hydrogenlike Lyman-α line and the $K\alpha^h$ line in atoms. In a similar way, the satellite line energy lies between those of the heliumlike $(1s)(2p)^1P_1 \rightarrow (1s)^2\,{}^1S_0$ line and of the $K\alpha$ line in atoms with closed outermost shells, in agreement with previously described results. The main result deduced from this spectrum is that the satellite line is roughly half as intense as the hypersatellite one in contrast with what is observed with neutral atoms.[3] The previously described model may easily explain why this satellite line is weaker ($\sim$50%) than the hypersatellite. The filling of the first K hole, which happens in the presence of a small number of L electrons, mainly favors the radiative decay with respect to the Auger transitions—mainly KLL [this transition being impossible for instance in K^0L^1 states ($\omega=1$) and weak in K^0L^2 states]. The second K transition occurs with more L electrons and may then proceed through KLL Auger emission (the KLL Auger rate in a neutral atom is of the order of 90% of the total K decay rate).

The Laboratoire de Physique Atomique et Nucléaire-Institut du Radium is Unité associée No. 771 au Centre National de la Recherche Scientifique.

---

[1] J. P. Briand, L. de Billy, P. Charles, S. Essabaa, P. Briand, R. Geller, J. P. Desclaux, S. Bliman, and C. Ristori, Phys. Rev. Lett. **65**, 159 (1990).

[2] F. Larkins, J. Phys. B **9**, L29 (1971).

[3] J. P. Briand, P. Chevallier, M. Tavernier, and J. P. Rozet, Phys. Rev. Lett. **27**, 777 (1971).

[4] M. Suvanen, T. Åberg, Helsinki University of Technology Research Report No. 121, 1977 (unpublished).

[5] J. P. Briand, A. Touati, M. Frilley, P. Chevallier, A. Johnson, J. P. Rozet, M. Tavernier, S. Shafroth, and M. O. Krause, J. Phys. B **9**, 1055 (1976).

[6] J. P. Mossé, P. Chevallier, and J. P. Briand, Z. Phys. A **322**, 207 (1985).