PHYSICAL REVIEW A VOLUME 53, NUMBER 4 APRIL 1996

Time for the empty L shell of a hollow atom to be filled

J.-P. Briand and B. d'Etat-Ban
Laboratoire de Physique Atomique et Nucléaire-Institut du Radium, Unité associée au CNRS No. 771, Université Pierre et Marie Curie,
4 Place Jussieu, 75252 Paris Cedex 05, France

D. Schneider, M. A. Briere, V. Decaux, J. W. McDonald, and S. Bardin
Lawrence Livermore National Laboratory, L-421, P.O. Box 808, Livermore, California 94550

(Received 15 September 1995)

The dynamics of the first capture and decay processes occurring during the interaction of slow highly charged ions below a surface has been studied in looking at the x rays emitted directly, or in coincidence, by impinging bare or hydrogenlike ions of various atomic numbers on solid targets. Some results on the decay processes of these hollow atoms, mainly formed below the surface, for argon, iron, and krypton ions are presented. By measuring the changes of the number of electrons in the L and M shells of the ions, compared to the lifetime of the K shell, it has been possible to evaluate the mean time for the filling of the L and M shells. These measurements are compared with a model of interaction of the ions with the surface.

PACS number(s): 31.50.+w, 32.30.Rj, 79.20.Rf

INTRODUCTION

When a slow, highly charged ion approaches, touches, or penetrates a surface many electrons of the solid are captured in highly excited states of the ion, leading to the formation of hollow atoms [1]. These very excited states then rapidly decay via a cascade of Auger transitions that reionizes the ion, which in turn may recapture new electrons and so on. At the end of the decay process, the filling of the K and L shells leads to the emission of a complex array of x-ray satellite lines and one observes the signature of all stationary states with any number of $L,M,N, \ldots$ electrons.

At high-resolution (crystal) spectroscopy the energy of the $K\alpha L^x$ satellite lines gives the exact number of L electrons and the mean number of M electrons at the time of the filling of the K hole. At low resolution (SiLi detectors) the relative intensity of the $\alpha,\beta,\gamma, \ldots$ lines gives the mean number of L, M, or N electrons present at the time of the decay. It is thus possible to deduce from the time evolution of these ion configurations some information about the interaction of the ion with the solid and the decay of the hollow atom.

Below the surface the hollow atoms of high Z atomic numbers ($Z\geqslant 18$) are formed by very fast capture processes of the target electrons into the M and N shells of the ions. These highly excited electrons then fill the empty K and L shells mainly through a cascade of Auger transitions. The L shell is therefore quickly and completely stepwise filled via LMM, LNN, or LNM transitions, and at a given time of this cascade the K hole(s) is (are) filled and a characteristic $K\alpha$ transition is observed. The main interest in the field today [2–6] is to study this stepwise filling of the eight holes of the L shell, which constitutes some atomic clock, by the observation at a given time of the $K\alpha L^x$ line(s) filling the K shell.

This paper presents some experiments carried out to study the time evolution between the intermediate states of the cascade. The principle of the first experiment was to vary, for similar initial configurations of the ions, the K-shell lifetimes of the ions τ_K by changing either the atomic number of the projectiles ($\tau_K \sim Z^{-4}$) or in a given ion the number of K holes (τ_K is roughly divided by a factor of 2 [7] when the projectile is a bare nucleus—two K holes—with respect to the lifetime of a hydrogenlike ion: one hole instead of two). In the second part of the experiment, we studied directly and in coincidence the actual configurations of the ions when the two K holes of a bare ion are stepwise filled. In this case there is a change in the number of L electrons between the time of the filling of the first K hole and that of the second one (two snapshots instead of one). We shall deduce from these experiments the time evolution of the L- and M-shell fillings and get information on the processes of interaction [3].

EXPERIMENTAL RESULTS

We prepared at the Lawrence Livermore National Laboratory (EBIT) source beams of Fe^{25+}, Fe^{26+}, Kr^{35+}, and Kr^{36+} ions, at 7 kV/q kinetic energy [8] that we sent onto metallic targets. The x rays emitted in flight by these ions were observed with intrinsic germanium detectors of 200 mm^2 area and 180 eV resolution at 6 keV. The spectra are presented in Figs. 1–4. The $K\alpha$ lines observed are made of a

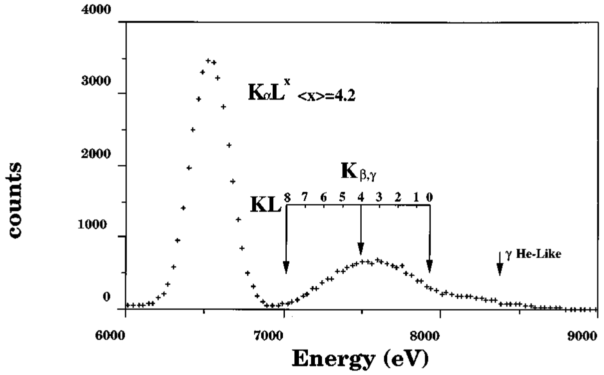

FIG. 1. K x-ray spectrum observed with Fe^{25+} ions.

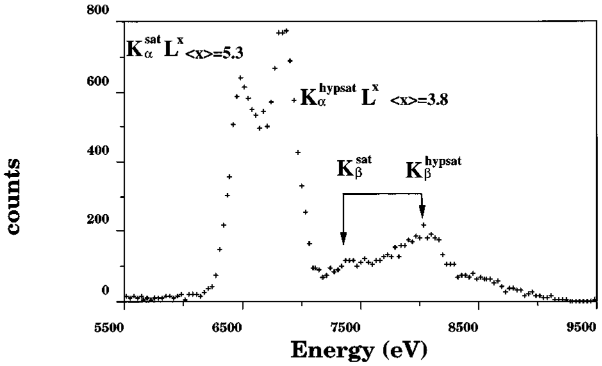

FIG. 2. *K* x-ray spectrum observed with Fe^{26+} ions.

complex array of KL^x satellite lines corresponding to all the transitions of an *L* electron to the *K* shell in the presence of any number of *L* spectator electrons. These satellites cannot be separated with a germanium detector, but the mean energy of the line measured gives the mean number of *L* electrons present at the time of the decay (Tables I and II), as we demonstrated [1] by comparing the same spectra recorded with a SiLi detector and a crystal spectrometer. As shown in Fig. 1 and Table II, the energy $K\alpha$ line emitted by Fe^{25+} ions is centered on the KL^4 satellite (four *L* spectator electrons, on an average, instead of, e.g., five, as observed [1] for argon). This result is in agreement with the observed shape of the $K\beta$ line (whose KL^x components are more widely separated than for the $K\alpha$ line), which shows that most of the eight satellite lines (except maybe the last KL^8 and KL^7 transitions) are present. In the case of Kr^{35+} (Fig. 3) the $K\alpha$ line is centered on a KL^x satellite, whose *x* mean value is 2.7. It clearly appears from the $K\beta$ spectrum presented in Fig. 3 that only the (first) few KL^x lines are emitted.

One of the most visible features of these spectra is the very large (unusual) intensity of the $K\beta$ lines as compared to the $K\alpha$ lines: 38% and 42% for iron and krypton, respectively (Table II) (i.e., more than for the argon value). Since the transition rates for the $K\beta$ lines compared to the $K\alpha$ lines are for an equal number of *p* electrons in the $n=2$ and 3 shells, for, e.g., krypton between 15% (six electrons in each shell: neutral atom [13]) and 26% (one electron in each shell: hydrogenlike ions [14]), these results mean that the mean number of *M* electrons (of the $3p$ state at least) is always about two times as large as the *L* electron. Like in the case of Ar, the *M* shell is then roughly closed when the *L* shell is empty (a similar conclusion may also be drawn for the *N* shell: $K\gamma$ lines). These results also mean, like in the case of argon, that the ion is quickly fed to reach its equilibrium charge state, which corresponds at the actual kinetic energy to charges $\sim 3+$, before or at the beginning of its radiative decay.

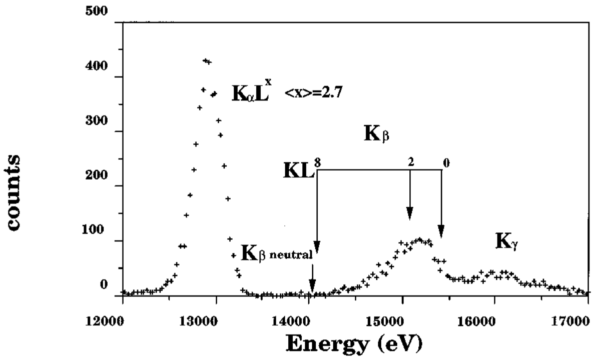

FIG. 3. *K* x-ray spectrum observed with Kr^{35+} ions.

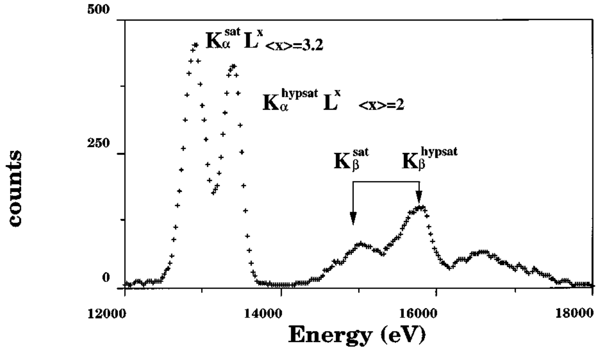

FIG. 4. *K* x-ray spectrum observed with Kr^{36+} ions.

DISCUSSION OF THE EXPERIMENTAL RESULTS

We present in Tables I and II the energy of the $K\alpha$ lines observed for Fe^{25+} and Kr^{35+}. The first interesting result of these tables, compared to previous ones on argon ions [1], is the decrease of the number of *L* spectator electrons present at the time of the *K* hole filling as a function of the atomic number of the ion: 5, 4.2, and 2.7 for argon, iron, and krypton hydrogenlike ions, respectively. In any case, the ions are quasineutralized, i.e., have comparable *L* shell filling rates for an equal number of electrons (the Auger rate, e.g., for the *LMM* transitions does not scale or vary slowly with *Z*). As discussed below, the *L* shell filling rate is, however, also faster, owing to the larger number of outermost shell electrons for the ions of larger *Z* atomic numbers (a factor of approximately 2 between Ar and Kr). This result shows clearly the effect of the fast decrease of the *K* shell lifetime as a function of *Z* (the radiative lifetime $\tau_K \sim Z^{-4}$), which leads to a relatively faster filling rate for the *K* shell than for the *L* shell for increasing atomic numbers.

The second interesting result is obtained by comparing the numbers of *L* spectator electrons for, e.g., Fe^{25+} and the first *K* transition (hypersatellite) for Fe^{26+}, which is found to be 3.8 for Fe^{26+} and 4.2 for Fe^{25+} (Tables I and II). This result can be explained by considering that the lifetime of the *K* hole is about two times shorter when two *K* holes are present instead of one [7]. Since the *L* shell filling rate in this case is roughly the same for Fe^{26+} and Fe^{25+}, this means that the $K\alpha$ line is emitted in a shorter time for Fe^{26+} than for Fe^{25+} and then for fewer *L* electrons. A similar effect is also observed with Kr^{35+} and Kr^{36+} ions ($0.7e_L$ for Kr^{35+} than for Kr^{36+}).

A third result that can be extracted from the data presented in Table II is the mean time for the filling of an *L* hole, which can be deduced from the energies of the satellite and the hypersatellite lines for a given element. In this case we used bare ions Fe^{26+} and Kr^{36+} whose two *K* vacancies are sequentially filled through the hypersatellite-satellite cascade [15]. By looking at the electronic configuration of the

TABLE I. (a) Experimental energies of the KL^x satellite lines for $\mathrm{Fe}^{25+,26+}$ and $\mathrm{Kr}^{35+,36+}$ [9–12] and (b) theoretical values.

Line	Fe^{25+}	Fe^{26+}	Kr^{35+}	Kr^{36+}
		(a) Expt. energy (eV)		
$K\alpha^s$	6555	6535	12 935.5	12 917.4
$K\alpha^h$		6867		13 389
		(b) Theor. energy (eV)		
Line	Fe		Kr	
$K\alpha_2^s$ neutral $K\alpha_1^S$ neutral	6392 6404	$\langle E\rangle$ $K^1L^8M^x=6397.5$	12 605 12 657	$\langle E\rangle$ $K^1L^8M^x=12\,631$
$K\alpha_2^h$ neutral $K\alpha_1^h$ neutral	6558 6678	$\langle E\rangle$ $K^0L^8M^x=6668$	12 990 13047	$\langle E\rangle$ $K^0L^8M^x=13\,018$
He-like $^3P_1$ He-like $^1P_1$	6667 6701	$\langle E\rangle$ $K^1L^1M^0=6684$	13 023 13 114	$\langle E\rangle$ $K^1L^1M^0=13\,068$
H-like Ly-α_2 H-like Lyα_1	6952.5 6971.2	$\langle E\rangle$ $K^0L^1M^0=6965$	13 431 13 509	$\langle E\rangle$ $K^0L^1M^0=13\,483$

ion at the times of the filling of the two K holes (the KL^x satellite structure of the $K\alpha$ hypersatellite *and* of the satellite) it is possible to observe separately two steps of the time evolution of the electronic configuration of the hollow atoms, the two lines being emitted one after the other. A completely different KL^x distribution was observed, where more L electrons are present on the satellite spectrum than on the hypersatellite one, which demonstrates clearly the stepwise filling of the L shell [3].

The lifetimes of the states during the cascade of LMM (plus LNN) Auger transitions filling the L shell ($KL^1 \rightarrow KL^2 \rightarrow KL^3 \cdots$) vary slowly, as discussed in Ref. [1], and as it has been recently found [16], it is of the order of 5×10^{-16} s. The stepwise filling of the L shell of the ion may be described in the same way as the decay of a radioactive series by the Bateman's equations, as discussed in more details in Ref. [17]. To illustrate this stepwise filling of the L shell, we present in Fig. 5 the array of curves giving the probability, as a function of time to find the ion in a given KL^x state. In these calculations, based on the spectroscopic data of Refs. [1] and [16], the individual lifetimes take into account the two decay channels of any KL^x state: the filling of the K and L shells. If we multiply each of these curves by the K to L branching ratio and the fluorescence yields calculated by Bhalla [18] (Table III), we can directly obtain the probability to observe, at a given time, a $K\alpha$ line with a given number of L spectator electrons. If the $K\alpha$ line is emitted at a certain time t_1 one observes a given KL^x array of satellites, as shown in Fig. 5 (which must be different at a decay time t_2), this KL^x array of satellites then provides some photographs (snapshots) of the configuration of the ion at this time. When two successive $K\alpha$ lines are emitted in ions having two K holes (hypersatellite-satellite cascade [3]), one can then obtain two photographs at the electronic con-

TABLE II. Mean numbers of L spectator electrons for Ar^{17+}, Fe^{25+}, and Kr^{35+}; $K\alpha$, $K\beta$, and γ line relative intensities (values for argon are corrected from fluorescence yields); and changes in mean numbers of L spectator electrons during the hypersatellite-satellite cascade.

Ions	Ar^{17+}	Fe^{25+}	Kr^{35+}
$K\alpha L^{\langle x\rangle}$	$K\alpha L^{\langle 5\rangle}$	$K\alpha L^{\langle 4.2\rangle}$	$K\alpha L^{\langle 2.7\rangle}$
$I(K\beta)/I(K\alpha)$	0.3	0.38 (No. $e^-M>$ No. e^-L)	0.42 (No. $e^-M>$ No. e^-L)
$I(K\gamma)/I(K\alpha)$			(No. $e^-N>$ No. e^-L)
Ions	Ar^{18+}	Fe^{26+}	Kr^{36+}
$K\alpha_{\mathrm{sat}}L^{\langle x\rangle}$	$K\alpha K^{\langle 5.4\rangle}$	$K\alpha L^{\langle 5.3\rangle}$	$K\alpha L^{\langle 3.2\rangle}$
$K\alpha_{\mathrm{hypersat}}L^{\langle x\rangle}$	$K\alpha L^{\langle 3.8\rangle}$	$K\alpha L^{\langle 3.8\rangle}$	$K\alpha L^{\langle 2\rangle}$
No. of additional Le^-	2.6	2.5	2.2

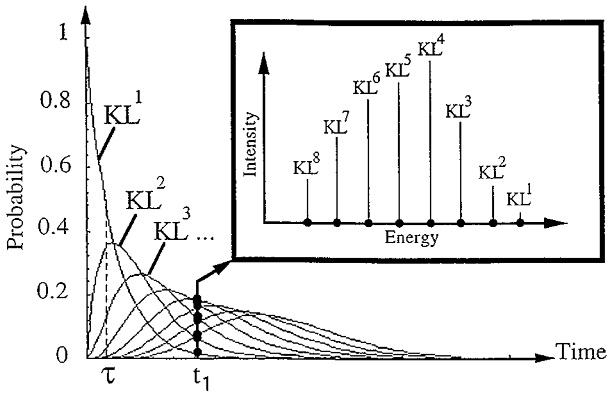

FIG. 5. Cascade of Auger transitions filling the empty L shell: probability to find the ion in a KL^x state as a function of time.

figuration of two different times.

In the case of argon hollow atoms the KL^x satellite distribution has already been observed in crystal spectrometry for the first (hypersatellite) and the second (satellite) K transitions [3]. These distributions, corrected for the fluorescence yields calculated by Bhalla [18], are presented in Fig. 6 and Table III. As discussed in Ref. [3], the first (hypersatellite) $K\alpha$ transition is emitted when only few L electrons are present ($\langle x \rangle = 3.8$) and the second one with a larger number ($\langle x \rangle = 5.4$) of L electrons (as seen in this figure, one does not observe any satellite—second $K\alpha$ transition—having one and two L electrons).

The mean number of L electrons present at the time of the K x-ray emission is then, for argon, increased by 1.6 (+1 owing to the fact that one L electron is removed at each $K\alpha$ transition) during the time that separates the emission of the first (hypersatellite) from the second (satellite) K transitions. This time is, for the $K^1L^5M^8$ state, of the order of 10^{-15} s [16,18]. The mean time for the filling of an L hole, around a mean value of 5.4 L spectator electrons, would then be $[10^{-15}\ (\mathrm{s})]/2.6 = 4\times10^{-16}$ s, i.e., in agreement with the theoretical estimates for the partial lifetime of the LMM (plus LNN, . . .) Auger transition: 4×10^{-16} s [16,19,20]. Then it is possible by measuring the increase of the number of L electrons between the filling of the first K hole and the second one to estimate the mean time for the filling of an L hole in a given KL^x configuration, which is found, in the present case, to be equal to the calculated LMM (+LNN, . . .) Auger rate.

TABLE III. Calculated fluorescence yields of argon [18].

Line	Fluorescent yield (%)
KL^8	13
KL^7	14–15
KL^6	15–17
KL^5	18–20
KL^4	18–25
KL^3	20 ± 5
KL^2	50 ± 30
KL^1	100

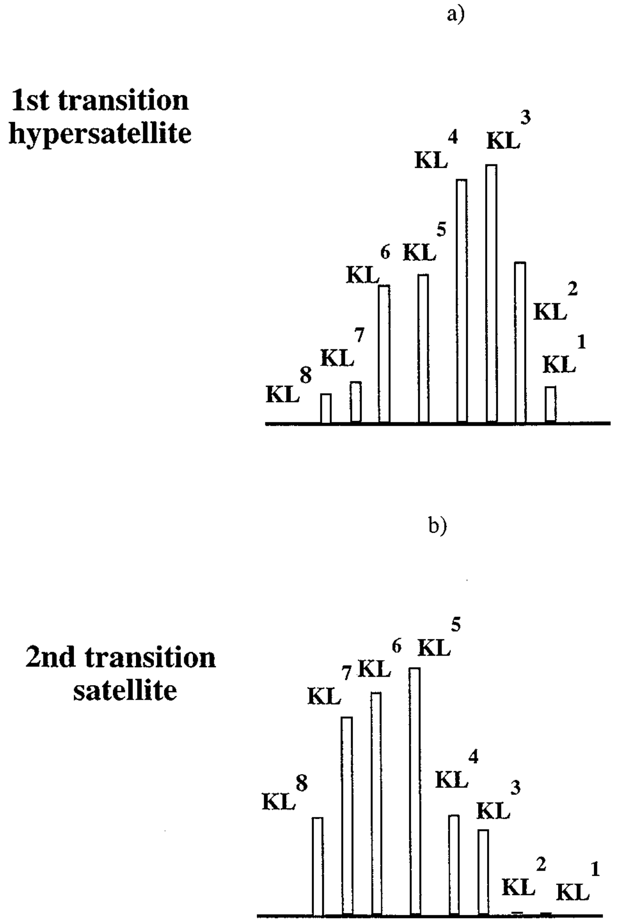

FIG. 6. KL^x distribution corrected of the fluorescence yields for the Ar (a) hypersatellite and (b) satellite $\mathrm{Ar}^{18+}\rightarrow\mathrm{Ag}$.

The changes in energy of the iron and krypton $K\alpha$ hypersatellite and satellite lines observed with the germanium detector (Table I) can be converted into an approximate mean number of L spectator electrons at the time of the decay (Table II). The changes of the number of L spectator electrons during the period of time between the emission of the hypersatellite and the satellite have been found, without any fluorescence yield corrections, to be $\Delta n = 1.5\ (+1)$ and 1.2 (+1) for iron and krypton, respectively (still one must add one electron because one L electron disappears during the filling of the first K hole). However, the relative intensity of the eight KL^x lines has to be corrected to take into account the fluorescence yield ω of each of the KL^x states. In the case of argon ω varies over a large range: $0.1<\omega<1$ (0.1 for KL^8, 1 for KL^1) [18]. For iron and krypton, the fluorescence yields are larger: $0.3<\omega<1$ and $0.6<\omega<1$, respectively [21]. These changes around a mean number of 5 ± 1 L electrons (iron) and 3 ± 1 (krypton) are, however, very small for iron ($\sim$15%) and for krypton ($\sim$10%) and can be neglected.

The mean time between the emission of the satellite and the hypersatellite τ_K, i.e., the inverse of the total transition rate for the filling of the K shell (K x rays plus KLL and KLM Auger), can be estimated by using the Larkins statistical procedure [19], which for argon fits reasonably with Hartree-Fock calculations [18]. It is found, by using the Bhalla or Vaeck KLL transition rates [18,16] and scaling the radiative rates with the Z^4 law, to be approximately 3×10^{-16} s for the KL^5 state of iron and 1.5×10^{-16} s for the KL^3 state of krypton. The experimental mean times for the filling of one L hole are thus about $3\times10^{-16}/2.5=1.2\times10^{-16}$ s for iron and $1.5\times10^{-16}/2.2=0.7\times10^{-16}$ s for krypton. The experimental value of the mean time for the filling of one L hole of iron is roughly equal to the LMM (plus $LNN, \dots$) Auger partial lifetime for an ion having 16 electrons in the M and N shells [19,16] $\tau\sim1.2\times10^{-16}$ s (a similar conclusion can also be drawn for Kr).

Since the M shell is filled roughly two or three times faster than the L shells, as discussed in Ref. [1], it is now possible to give an estimate of the mean time for the filling of the whole M shell in the case of argon, which has to be roughly $\frac{8}{3}$ the time that for the filling of an L hole at the beginning of the interaction (KL^1 or KL^2 state), i.e., of the order of 10^{-15} s. At velocities of the order of 1.2×10^6 m s^{-1} (20 kV/q) such a filling process would occur along a mean range of 12 Å, i.e., about three or four atomic monolayers. If we do not take into account the slower depopulation of the M shell via LMM Auger cascades, this means, roughly speaking, that a mean number of about 2 or 3 electrons is captured in the M shell through an atomic row in argon, plus an unknown number of N electrons (maybe of the same order of magnitude). At this level one must point out that *these numbers are mean numbers* and that the rate of capture varies obviously in time, i.e., along the ion path below the surface (it decreases with time and then charge, leading to an asymptotic number of M electrons). The M shell is then filled up relatively quickly in a shorter time. For iron and krypton ions the filling rate is faster, but many more electrons have to be captured to reach the equilibrium charge state and one can expect that the full neutralization still holds along few atomic monolayers.

COINCIDENCE STUDY OF THE HYPERSATELLITE-SATELLITE CASCADE

We have studied for iron the K hypersatellite-satellite cascade in biparametric coincidences in order to know more about the filling of the L shell and obtain at least qualitative information on the individual correlation between a given KL^x state of the hypersatellite and of the satellite. The design of the experiment is presented in Fig. 7. The $K\alpha$, $K\beta$, and $K\gamma$ lines were detected and analyzed by two intrinsic germanium detectors of 200 mm^2 area located on both sides of an iron target. A general view of the hypersatellite-satellite coincidence plane is shown in Fig. 8. In the case of iron the energy separation of the KL^x satellite ($\sim$40 eV) is much lower than the resolution of the Ge detector (180 eV) and one can only observe some asymmetries on the $K\alpha^s$ and $K\alpha^h$ sections of the coincidence plane or some correlations between the $K\beta$ and $K\gamma$ lines. One sees clearly, for example, the presence of a $K\gamma$ line for the hypersatellite and its vanishing for the satellite (the N electrons have already moved to lower states at the time of the emission of the second transition). This experiment, which demonstrates the existence of a cascading process, cannot provide detailed information on the individual coincidence events, but illustrates only the general trends of the cascade filling the L shell. More detailed information could be obtained in a similar way for more highly charged ions such as krypton or xenon.

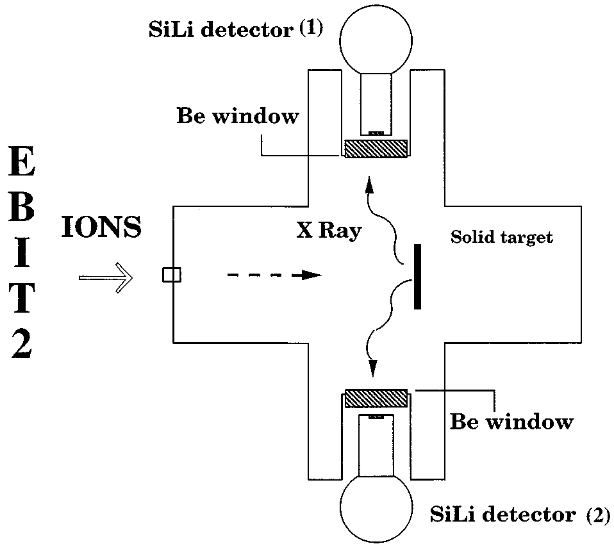

FIG. 7. Schematic view of the hypersatellite-satellite coincidence setup.

CONCLUSION

We have studied the dynamics of the filling of the empty L shell of hollow atoms of various atomic numbers by looking at the K hypersatellite-satellite cascade, which provides two snapshots of the ion electronic configuration. The measurements of the increase of the number of L electrons during the mean time between the filling of the two K holes of the ion allowed us to have some mean times for the filling of the L and M shells, which fit consistently with the models and the Auger rate calculations. The behavior of these hollow atoms below a surface proceeds through a cascade of Auger transitions, each lasting at least 10^{-16} s, interrupted sometimes by capture processes whose mean times are of the order of magnitude of at least 10^{-17} s (a few periods of rotation of a bound electron). Then, during a very short time

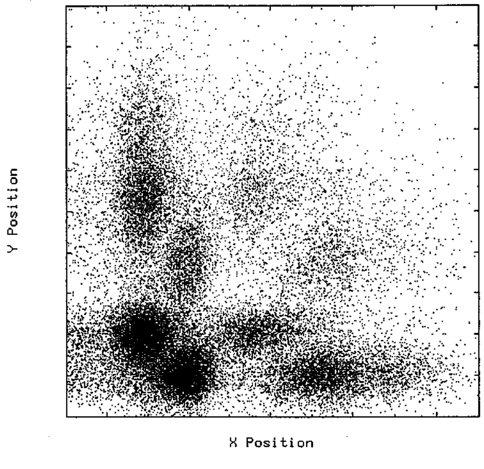

FIG. 8. Biparametric coincidence plane for the hypersatellite-satellite cascade for Fe^{26+} ions.

(about 8 times the mean value for one step, as shown in Fig. 5) there is a long series of very quick captures and spontaneous decays. The neutralization range we found is, however, slightly larger than that estimated by Herrmann *et al.* [22] (one monolayer) by extrapolating the energy losses of argon ions.

These findings are in agreement with the observation of true stationary states [1,3] for most of the KL^x states. However, one may remember that at the beginning of the interaction many electrons have to be captured in a very short time in order for the ion to reach its equilibrium charge states (2–4+ in the considered cases). The large number of very fast processes may thus block at the beginning of the decay the first KL^1 state, which could, as observed for Ar^{18+} [3] emit a very broad $K\alpha$ line signing the presence of a quasicontinuum state.

ACKNOWLEDGMENTS

J.-P.B. and B.d.-B. would like to express their gratitude to D. Schneider and the Lawrence Livermore National Laboratory for their kind welcome to carry out this experiment on the EBIT ion source.

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