Observation of K Hypersatellites and KL Satellites in the X-Ray Spectrum of Doubly K-Ionized Gallium

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The K x-ray spectrum arising from doubly K-ionized gallium atoms has been studied by means of a coincidence experiment with two solid-state detectors. A new energy range in K x-ray spectra (hypersatellite range) is observed. Hypersatellite lines are shifted about 15 times more than the usual satellite lines. The K-satellite spectrum following KL-ionized states has also been studied.

In a previous paper,[1] we reported the existence of a new energy range in L x-ray spectra. We observed the correlation between the creation of a double vacancy in the L shell, following KLL Auger electron emission, and some new x rays in a coincidence experiment with two solid-state detectors (Si-Li). These new x rays are strongly energy shifted from single-ionization rays, about 10 times more than ordinary satellites of L x rays, and were thus named "hypersatellites." Hypersatellites arise when the initial state of the atom is doubly ionized in the *same* shell. This study was completed by a second set of coincidence experiments with KLX Auger lines, in which we observe the ordinary x-ray satellite spectrum, thus proving that ordinary XL satellites are also due to doubly ionized atoms but in two *different* shells. In the usual experiments in x-ray spectrometry, LL-ionization states are certainly more seldom created than LX states; this remark explains why these hypersatellites had not been previously observed.

Our purpose now is to describe an experiment in which we observe K hypersatellites from doubly K-ionized atoms. The double vacancy may be created by K atomic or nuclear shakeoff during K ionization. Although K shakeoff has not yet been proved in K photoionization, it has been observed by several authors[2-6] in radioactive atoms, namely, in K-electron–capture radioactivity. The probability of this effect is very small according to the ordinary theory of autoionization of Migdal and Feinberg.[7,8] It decreases following a $1/Z^2$ law and thus it seems more interesting to use a low-Z emitter. As the K fluorescence yield increases with atomic number Z, we chose to study gallium ($Z=31$, $\omega_k=0.49$). The initial K hole in gallium comes from the favored K-electron capture of ^{71}Ge. The probability of double K ionization in ^{71}Ge was determined by several authors to be 1.2×10^{-4} per disintegration.[6,9] The de-excitation of doubly K-ionized atoms is mainly a two-step process (the simultaneous emission of two $E1$ photons, for example, is about 10^5 times smaller than the two-step mode). Assuming that the fluorescence yield for doubly ionized atoms is the same as for singly ionized atoms (although this is not yet proved, it is a crude estimation to describe qualitatively the simplest mode of de-excitation), that is, $\omega_k\cong0.5$, then there are in this case four equally probable modes of de-excitation: two K x rays, one x ray and one Auger electron, one Auger electron and one x ray, or two Auger electrons. The aim of this experiment is to establish a time correlation (coincidence) between the two K x rays which can be emitted after a double K ionization (first case mentioned above).

The first x ray to be emitted (K^2 holes$\rightarrow KL$ holes) is obviously much more shifted in energy than the second one ($KL\rightarrow L^2$ or LX). The first x ray is a hypersatellite, which we shall denote by K^h. The second one will be called a KL satellite. The initial state (K^2 holes) is a unique $^1S_0$ state. The second one (KL or KX) is defined by appropriate selection rules for transitions in doubly ionized atoms. For KL-ionization states it is only restricted to the $^1P_0$ state (KL_{II} or KL_{III}) since $^1S_0\rightarrow{}^1S_0$ and $^1S_0\rightarrow{}^3S_1$ (KL_I) transitions are either forbidden or very much hindered. Both K x rays are detected by two solid-state detectors (Si-Li) from Kevex Corporation (195 and 320 eV full width at half-maximum for 6-keV photons). With these detectors the $\alpha_{1,2}$ components of $K\alpha$ rays and the β_1, β_3, and β_5 components of $K\beta$ rays cannot be resolved; the same will be true for hypersatellites lines ($K\alpha^h$ and $K\beta^h$). The second transition to be emitted (KL satellites) has a much more complicated spectrum which we shall discuss later. The time correlation of these events is established using a

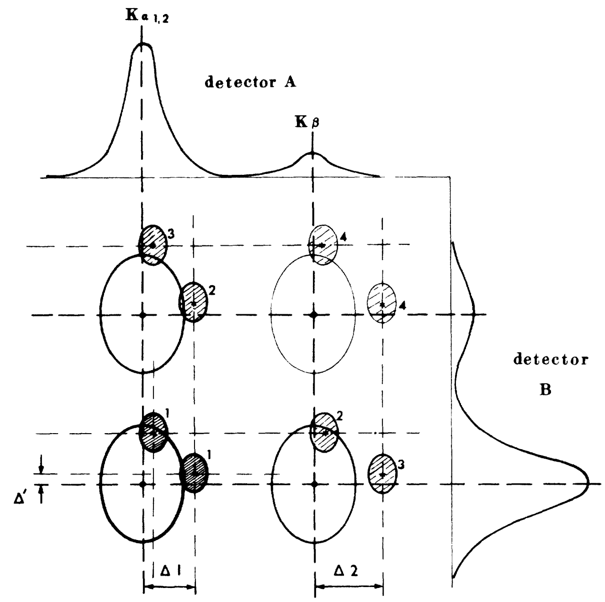

FIG. 1. Schematic cross section of the experimental coincidence plane. Shaded areas are true coincidences between hypersatellites and KL satellites; empty areas, random coincidences.

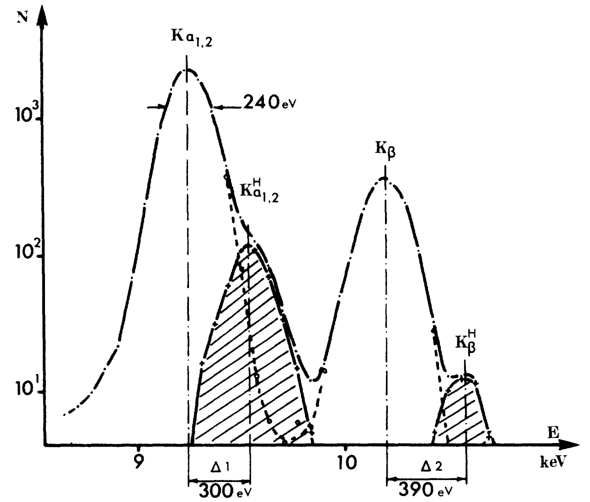

FIG. 2. X-ray spectrum in coincidence with an energy band corresponding to $K\alpha_{1,2}$ of gallium (summation of corresponding lines in two-dimensional analysis). Shaded areas are true coincidences, i.e., $K\alpha_{1,2}$ and $K\beta$ hypersatellites; empty ones are random coincidences.

fast coincidence amplifier ($2\tau = 35$ nsec). A 1-μCi sample of ^{71}Ge, deposited on a thin Mylar film, is placed between the two detectors. A major difficulty in this experiment is the very high rate of random coincidences with respect to the true ones (a factor N_t/N_r of $\frac{1}{30}$ was found in most of our runs), but previous experiments on L hypersatellites seem to prove that hypersatellite lines are so strongly shifted in energy that we may expect to be able to separate random-coincidence peaks from the true ones. Moreover, the use of two-dimensional analysis (64×64) in a 4096-channel analyzer (Nuclear Data) made this separation easier and also yielded much more information about the whole phenomenon.

The results of our work are presented in Figs. 1 and 2. Figure 1 is a schematic cross section of the coincidence plane. We represent as shaded areas the true coincidences; the other peaks are random-coincidence peaks. It is interesting to note that these random-coincidence peaks are an exact picture of the single-ionization spectra (diagram rays) and thus allow useful comparisons between x-ray spectra arising from singly and doubly ionized atoms under identical experimental conditions. In Fig. 2 we present the spectrum observed with detector A in coincidence with the entire $K\alpha_{1,2}$ ray seen in detector B. We observe two new x rays, $K\alpha^h$ and $K\beta^h$. A similar spectrum is observed in coincidence with the $K\beta$ line in detector B. $K\alpha^h$ and $K\beta^h$ are, respectively, shifted in energy by 300 and 390 eV with respect to the corresponding diagram rays. In Fig. 1 we observe a shift $\Delta' \simeq 30$ eV for the coincidence peak along the axis of the detector B. The Δ' shift indicates a coincidence between K hypersatellites ($K\alpha^h$ and $K\beta^h$: $K^2 \rightarrow KL$ and $K^2 \rightarrow KM$) and KL satellites which then appears to be shifted by approximately 30 eV. The ordinary satellite spectrum is known to be shifted by 10 to 20 eV. The KL satellite spectrum thus seems to be in the high-energy range of the ordinary satellite spectrum.

The $K\beta^h$ energy shift with respect to the $K\beta$ diagram ray gives a good approximation of the energy of doubly K-ionized atoms: $2B_K + \delta$ (B_K is the ordinary single-ionization energy). The final state of the $K\beta^h$ transition (KM holes) must be very near in energy to the sum of the two single-ionization energies $B_K + B_M$. We can assume with confidence ($\Delta Z = 1$) that the energies of $KM_{\mathrm{IV,V}}$ states do not differ from those of K singly ionized states by more than $B_{M_{\mathrm{IV,V}}} + 20$ eV. The δ value of K^2 states is then roughly equal to $390 + 20 = 410$ eV. The ordinary screening constant ΔZ of Bergströem and Hill thus becomes $\Delta Z = 0.56 \pm 0.04$ for K^2 states.

The $K\alpha^h$ shift in energy with respect to the $K\alpha$ diagram ray gives the energy of $KL_{\mathrm{II,III}}$ ionization states, $\delta = 410 - 300 = 110$ eV. This brings about a ΔZ screening constant nearly equal to 1. This result is not surprising because the lack of one supplementary K electron (in addition to the L hole) gives rise to a spherically symetric change in the effective Coulomb field for an L electron.

Finally, the Δ' shift of Fig. 1, which is the mean shift of the KL satellites ($K^2 \to KL$), is now in good agreement with the energy values of L^2 ionized states which may be deduced from the calculations of Hörnfeld[10] and from various experiments on the KLL Auger spectrum. (The δ value for L^2 states is roughly equal to 75 eV or $\Delta Z = 0.78$.) From the value we observe for KL ionized states ($\delta \simeq 110$ eV), the Δ' shift must be equal to $110 - 75 = 35$ eV, which is in good agreement with our experimental results.

We arrive at the following conclusions: There exists a new energy range in the K x-ray spectrum corresponding to the doubly K-ionized atoms. These new x rays (hypersatellites) are much more shifted with respect to diagram lines than is the ordinary satellite spectrum (about 15 to 20 times) and cannot be confused with it. The K hypersatellite lines are approximately (Fig. 2) the same as the diagram rays in nature of transitions and in relative intensity. The intensity of this hypersatellite spectrum is 1.2×10^{-4} times that of the diagram-ray spectrum in our experiment (it must be twice this intensity in direct spectrometry). This value is in very good agreement with previous experiments[6] and theoretical predictions.[11, 12]

The ΔZ screening constant for Ga ($Z = 31$) is equal to 0.56. This value should be compared with the ΔZ value for the two K-electron atoms, helium ($Z = 2$) and helium-like argon ($Z = 18$),[13] for which one finds, respectively, $\Delta Z = 0.33$ and 0.5.

The KL satellite spectrum which may be observed in direct crystal spectrometry is very complicated. It originates mainly from direct KL ionization (L shakeoff) and with a low probability from direct K^2 ionization (K shakeoff). The K^2 initial states give two distinct contributions to this KL satellite spectrum:

(1) KL satellites following a first transition of the Auger type; this spectrum is very intricate because it arises from a three-hole initial state. The special, unique satellite observed by Von Oertzen[9] in the direct spectrum of ^{71}Ge with a curved crystal, and which is shifted by about 90 eV, is probably a KL^2 satellite of this type. This value should be in good agreement with the KL^n shifts recently observed.[14-16]

(2) KL satellites following hypersatellite emission; this spectrum is a good fingerprint of the KLL Auger spectrum with only those transitions which are authorized by the appropriate selection rules. We have only observed this part of the satellite spectrum. It is slightly shifted (by ~30 eV) and situated in the high-energy part of the ordinary satellite spectrum.

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