Double-Photon Decay in Xenon Atoms

K. Ilakovac

Faculty of Science and Mathematics, University of Zagreb, Zagreb, Yugoslavia, and Institute "Rudjer Bošković," 41000 Zagreb, Yugoslavia

J. Tudorić-Ghemo

Faculty of Electrical and Mechanical Engineering and Shipbuilding, University of Split, Split, Yugoslavia

and

B. Bušić and V. Horvat

Faculty of Science and Mathematics, University of Zagreb, Zagreb, Yugoslavia

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Decay of xenon atoms with a vacancy in the K shell by the emission of photon pairs which continuously share the transition energy was investigated. The results show the expected dominance of transitions of electrons from the $2s$, $3s$, and $3d$ states, for which the single–x-ray transitions are strongly forbidden. In the energy range of the present measurements the resonance effect due to the intermediate $2p$ (and $3p$) states for transitions of electrons from higher shells has not been observed.

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Since the original work of Goeppert-Mayer[1] on double-photon emission from excited states of atoms, and elaboration of the theory for the hydrogenlike and heliumlike systems,[2] many experimental investigations of double-photon decay of metastable states of one- and two-electron ions have been reported.[3] Theory of the double-photon decay of states of atoms which have all electrons in their normal states except for one electron missing from the K shell was developed by Freund.[4] In addition to a further elaboration of the theory, Bannett and Freund[5] reported the results of their measurements of double-photon decay in molybdenum atoms with a vacancy in the K shell. Energy resolution did not allow identification of the higher subshells involved in the double-photon transitions, but they stressed that the main contributions are expected to be due to the $2s \rightarrow 1s$, $3s \rightarrow 1s$, and $3d \rightarrow 1s$ double-photon transitions.

Florescue[6] developed a theory of double-photon transitions in hydrogenlike systems including the transitions from higher shells. According to her results the $3d \rightarrow 1s$ double-photon transition should be about 5 times stronger than the $2s \rightarrow 1s$ double-photon transition.

In the following the results of an experimental investigation of double-photon decay in xenon atoms are presented.

Sources of radioactive ^{131}Cs, decaying by electron capture only, were used as "generators" of xenon atoms with a vacancy in the K shell. A standard procedure of preparation of high-purity ^{131}Cs was applied: $BaCO_3$ was irradiated by slow neutrons in a nuclear reactor, a threefold precipitation of $BaCl_2$ was made to remove Cs, and ^{131}Cs was accumulated from the decay of ^{131}Ba and carefully separated by means of two ion-exchange columns. The initial radioactive impurity of the sources, as determined from the singles γ-ray spectra in a Ge(Li) detector, was about one decay per 10^8 decays of ^{131}Cs. Each source was about 0.4 mm in diameter and was placed in the center of a 0.7-mm-diam double-conical hole in a 2.6-mm-thick aluminum shield. The shield with the source was placed between two high-purity germanium detectors (supplied by EG and G Ortec) in a close 180° geometry. Pulses from the detectors were fed into a fast-slow coincidence system with a three-parameter (time, energy E_1, energy E_2) $(128 \times 512 \times 512)$-channel pulse-height analyzer. The initial singles counting rate in either detector in the $K\alpha$ peak of xenon was about 60 s^{-1}. Energy resolution of the detectors at 5.9 keV was 230 and 215 eV, respectively, time resolution was about 18 ns, and the coincidence efficiency was between 90% and 95%. The data were collected for 1757 h. Analysis of the recorded data was made off line in a UNIVAC 1110 computer.

The time spectra for the various regions of the E_1-E_2 plane were carefully analyzed to ascertain the proper choice of time intervals and to determine the efficiency of the coincidence. The in-coincidence peaks in the E_1-E_2 plane due to the crosstalk of $K\alpha$ and $K\beta$ x rays of germanium, and the accidental coincidence peaks of the $K\alpha$, $K\beta_1$, and $K\beta_2$ x rays of xenon, allowed an accurate calibration of the energy scales.[7] The sum spectrum shown in Fig. 1(a) is due to the absorption of $K\alpha_2$ and $K\alpha_1$ x rays of xenon (29 458 and 29 779 eV, respectively)[8] by photoelectric effect in one detector, followed by the escape of K x rays of germanium from the detector, passage of the x rays through the hole in the shield, and their absorption in the other detector. The sum spectrum in Fig. 1(j) is

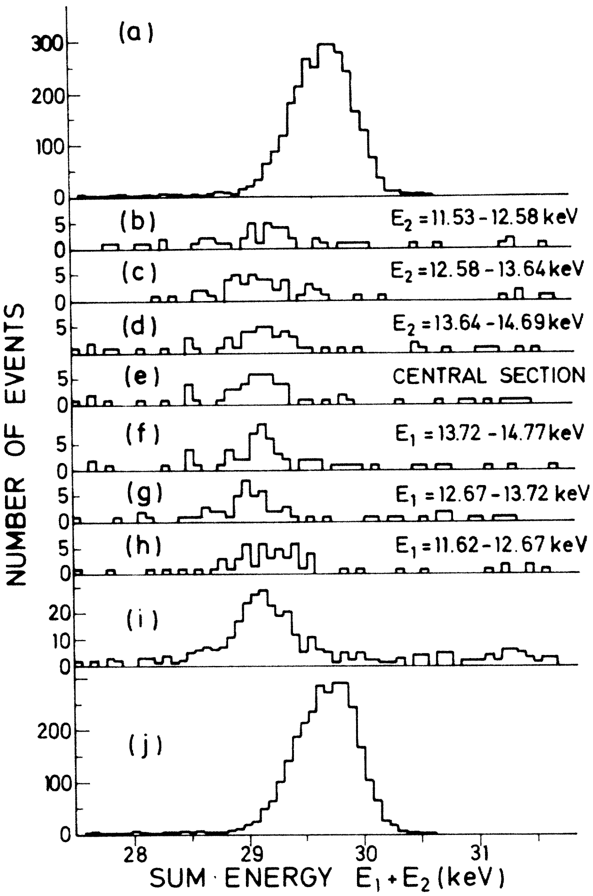

FIG. 1. (a),(j) Sum spectra of the peaks of the $K\alpha_{1,2}$ x rays of xenon (real coincidences are due to the crosstalk via germanium K x rays). (i) Sum spectrum of the continuous distribution due to the double-photon decay. (b)–(h) Sum spectra of the sections of the continuous distribution.

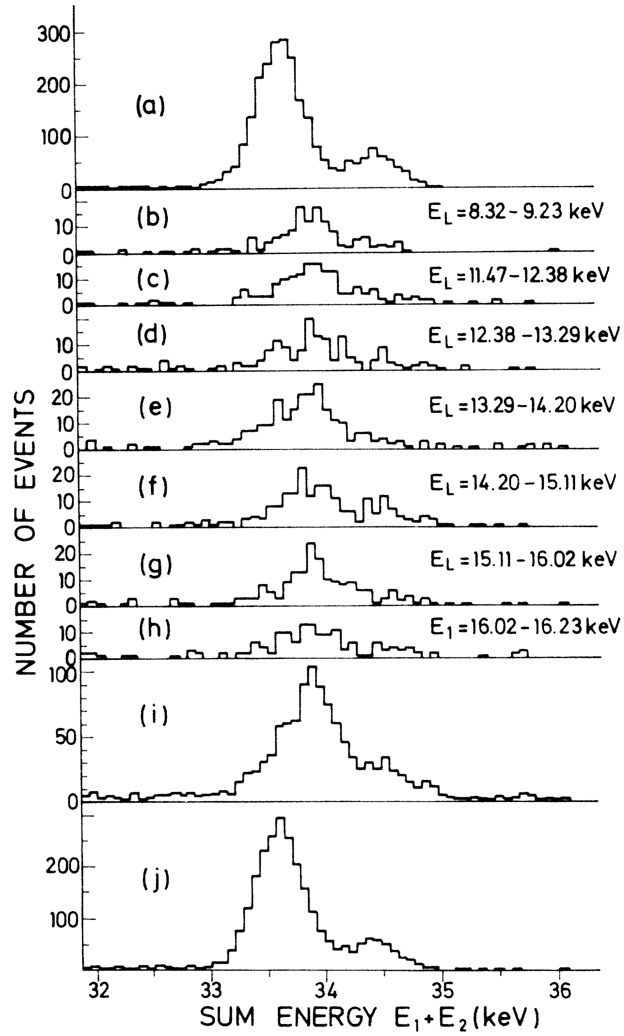

FIG. 2. (a),(j) Sum spectra of the peaks of the $K\beta_1$ and $K\beta_2$ x rays of xenon (real coincidences are due to the crosstalk via germanium K x rays). (i) Sum spectrum of the continuous distribution due to the double-photon decay. (b)–(g) Sum spectra of the sections of the continuous distribution. Data in the symmetrical sections were added (E_L is the lower value of E_1,E_2). (h) Sum spectrum of the central section.

due to the same processes, but for the reversed order of the detectors. The peaks in these and the corresponding crosstalk peaks due to the initial $K\beta_1$ and $K\beta_2$ x rays of xenon [Figs. 2(a) and 2(j)] allowed a very reliable determination of the energy scales and of the energy resolution of the system.

In addition to the well defined peaks described above, the in-coincidence data have shown continuous distributions in the E_1-E_2 plane along lines of constant sum energy. Crosstalk between the detectors via bremsstrahlung of photoelectrons ejected by xenon K x rays in one detector, escape of the bremsstrahlung and its absorption in the other detector was calculated and found to be less than 1% of the observed numbers of events.[7] Other processes which could contribute to the counting rates in the energy region studied, e.g., atomic cascade transitions, double Compton scattering, and pileup of pulses were considered, but because of the energy discrimination all were found to yield no or negligible contributions to the observed numbers of counts. The sum spectrum of the whole continuous distribution which was observed between the above described crosstalk peaks is shown in Fig. 1(i). The peak in this spectrum is shifted relative to the positions of the xenon $K\alpha$ x-ray crosstalk peaks [Figs. 1(a) and 1(j)]. The shift relative to the $K\alpha_1$ peak is -642 ± 36 eV. This value is in a very good agreement with the known difference of 666 eV of binding energies of the $2p_{3/2}$ and $2s_{1/2}$ electrons in xenon atoms.[8] For this reason the continuous distribution along the sum-energy line $E_1 + E_2 = 29\,113$ eV is considered to be due to the $2s_{1/2} \rightarrow 1s_{1/2}$ double-photon emission.

The spectrum in Fig. 1(i) was analyzed on the assumption of the contributions of peaks due to the $2s_{1/2} \rightarrow 1s_{1/2}$, $2p_{1/2} \rightarrow 2s_{1/2}$, and $2p_{3/2} \rightarrow 1s_{1/2}$ double-photon transitions. The latter two transition were found to be strongly forbidden. The results for the intensity ratios at the 95% confidence level are

$$\frac{I(2p_{1/2} \rightarrow 1s_{1/2})}{I(2s_{1/2} \rightarrow 1s_{1/2})} \leq 0.22,$$

and

$$\frac{I(2p_{3/2} \rightarrow 1s_{1/2})}{I(2s_{1/2} \rightarrow 1s_{1/2})} \leq 0.095$$

for the emission in opposite directions and E_1 in the range 11.53 to 17.58 keV.

The $2p \rightarrow 1s$ double-photon transitions were expected to be much weaker than the $2s \rightarrow 1s$ double-photon transitions because the electric dipole–electric dipole ($E1E1$) transitions, which require no parity change, have largest probability.

Sum spectra of sections of the continuous distribution are shown in Figs. 1(b)–1(h). They were analyzed on the assumption of the contributions of the three double-photon decay components and a constant background. The differential probabilities of the $2s_{1/2} \rightarrow 1s_{1/2}$ double-photon decay per K-shell vacancy in xenon atoms are shown in Fig. 3(a). In the calculations the $1+\cos^2\theta$ angular distribution of the photons, the absorption in the source, supporting foil, air, and beryllium window, and the losses of counts due to the escape of germanium K x rays from the detectors were taken into account.[7]

The same measurement yielded a second, more intensive, continuous distribution at the sum energy close to that of the crosstalk peaks due to the $K\beta_1$ and $K\beta_2$ x rays of xenon [whose sum spectra are shown in Figs. 2(a) and 2(j)]. The sum spectrum of the whole distribution between the two double peaks is shown in Fig. 2(i). Again a shift with respect to the single-photon transitions is seen, and the spectrum shows some structure. An analysis of the spectrum was made on the assumption of double-photon transitions of electrons from the $3s_{1/2}$, $3p_{1/2,3/2}$, $3d_{3/2,5/2}$, $4s_{1/2}$, $4p_{1/2,3/2}$, and $4d_{3/2,5/2}$ states. From the fits the upper limits of the $3p \rightarrow 1s$ and $4p \rightarrow 1s$ double-photon decay probabilities were derived:

$$\frac{I(3p_{1/2,3/2} \rightarrow 1s_{1/2})}{I(3d_{3/2,5/2} \rightarrow 1s_{1/2})} \leq 0.027,$$

and

$$\frac{I(4p_{1/2,3/2} \rightarrow 1s_{1/2})}{I(3d_{3/2,5/2} \rightarrow 1s_{1/2})} \leq 0.26,$$

both at the 95% confidence level. As in the case of the $2p \rightarrow 1s$ double-photon transitions, we find, as expected, that the states dominating in the single-photon transitions do not contribute appreciably to the double-photon decay.

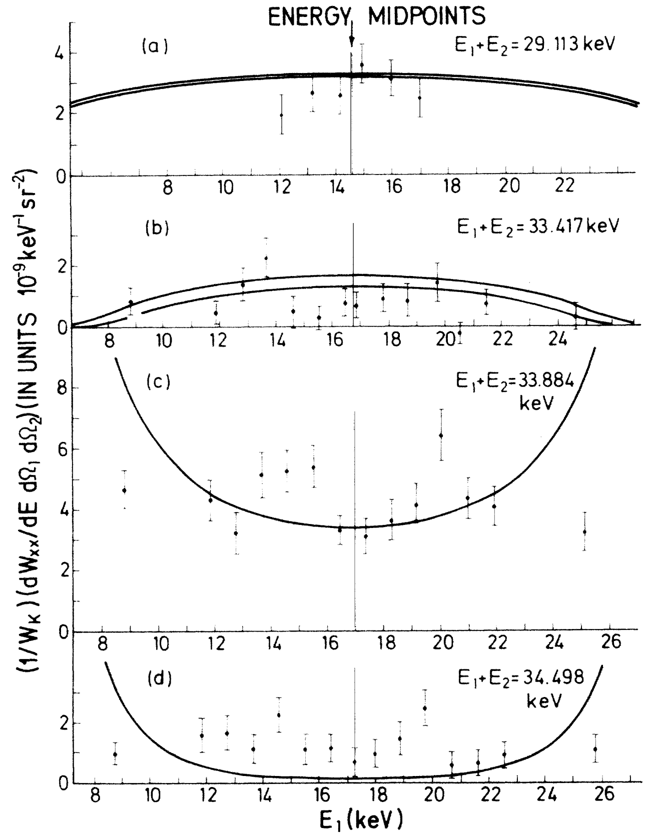

FIG. 3. The differential probabilities per K-shell vacancy of the (a) $2s \rightarrow 1s$, (b) $3s \rightarrow 1s$, (c) $3d \rightarrow 1s$, and (d) $4s,d \rightarrow 1s$ double-photon decay in xenon atoms. The lower curve in (a), the lower curve in (b), and the curve in (d) show the results calculated from the theory of Freund (Ref. 4) and Bannett and Freund (Ref. 5), multiplied by factors 1/2.1, 4.1, and 50, respectively. The theoretical results in (d) do not include the contribution of the $4d \rightarrow 1s$ double-photon decay, which is expected to be larger than that of the $4s \rightarrow 1s$ decay. The upper curve in (a) shows the values calculated from the theory of Shapiro and Breit (Ref. 2) multiplied by factor 1/3, and the upper curve in (b) and the curve in (c) show the results calculated from the theory of Florescu (Ref. 6) multiplied by factors 1/1.25 and 1/8, respectively.

Sum spectra of the sections of the second continuous distribution are shown in Figs. 2(b)–2(g) (with symmetrical spectra added) and 2(h) (central section). Sum spectra of the sections were analyzed with the assumption only of the peaks due to the transitions of electrons from the $3s$, $3d$, and $4sd$ states and a constant background. Similar calculations to those applied in the analysis of the $2s,p \rightarrow 1s$ double-photon decay yielded the results on differential probabilities shown in Figs. 3(b)–3(d).

From the data shown in Figs. 3(a)–3(d) the average

TABLE I. Average values of the differential probabilities per K-shell vacancy of the double-photon decay in xenon atoms for the central 6-keV-wide intervals of photon energy and emission of photons in the opposite directions.

Double-photon transition	Energy interval (keV)	$\frac{1}{W_K}\left(\frac{d^2W_{xx}}{dE\,d\Omega_1\,d\Omega_2}\right)$ (10^{-9} keV^{-1} sr^{-2}) Theory	This measurement
$2s \rightarrow 1s$	14.56 ± 3.0	6.68[a] 9.91[b]	2.50 ± 0.24
$3s \rightarrow 1s$	16.71 ± 3.0	0.31[a] 2.01[c]	0.89 ± 0.20
$3d \rightarrow 1s$	16.94 ± 3.0	28.04[c]	4.72 ± 0.28
$4sd \rightarrow 1s$	17.25 ± 3.0	0.004[a,d]	1.32 ± 0.18

[a]Reference 5.
[b]Reference 2.
[c]Reference 6.
[d]Only $4s \rightarrow 1s$ double-photon transition is included.

differential probabilities in 6-keV-wide intervals of energy about the central point, $\frac{1}{2}(E_1+E_2)$, were calculated. The results are shown in Table I. Since the differential probabilities for these transitions are expected to be almost constant in their midenergy ranges, these results represent the differential probabilities at photon energies $E_1 = E_2$.

Figures 3(a)–3(d) also show the differential probabilities for double-photon decay in xenon atoms calculated from the theories of Shapiro and Breit[2] and Florescu[6] for hydrogenlike systems, and from the theory of Freund[4] and Bannett and Freund.[5] For the $2s \rightarrow 1s$ double-photon decay in molybdenum Bannett and Freund[5] reported an excellent agreement between their theoretical and experimental results. For the same transition in xenon atoms [Fig. 3(a)] we find the values calculated from their theory about 2.1 times larger than our experimental results. The values calculated from the theory of Shapiro and Breit[2] show a larger discrepancy, but they apply to a Xe^{53+} ion. Large discrepancies were also found for other transitions [see Figs. 3(b)–3(d)].

The $3s \rightarrow 1s$ and $3d \rightarrow 1s$ (and $4sd \rightarrow 1s$) double-photon transitions are expected to show strong resonance behavior as a result of the real $2p$ (and $3p$) intermediate state.[4] In the calculated $3s \rightarrow 1s$ spectrum the resonance causes a gradual decrease of the differential probability as the energy is lowered (or increased) from the energy midpoint, at about 7 keV it reaches a zero value, and at still lower energy a very rapid rise is obtained. The range of energy accepted in the measurement was too narrow to observe the rise. The resonance effect in the calculated $3d \rightarrow 1s$ and $4s \rightarrow 1s$ spectra causes a gradually increasing rise of the differential probability as the energy is lowered (or increased) from the energy midpoint. Our results, in the energy range accepted in the measurements, do not support that prediction.

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