RESONANCE EFFECT IN INNER-SHELL nd $\rightarrow$ 1s TWO-PHOTON DECAY*

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Emission of photon pairs by the two-photon decay process from silver and hafnium atoms with a vacancy in the K shell was investigated. Radioactive decay of ^{109}Cd and ^{179}Ta was used to generate the K-shell vacancy states in silver and hafnium atoms, respectively. The measuring system consisted of a pair of planar high-purity germanium detectors, a fast-slow coincidence system, and a $128 \times 512 \times 512$ channel pulse-height analyzer. Two-dimensional spectra of numbers of events as functions of amplitudes of coincident pulses from the two detectors were analyzed. The results on the 3d $\rightarrow$ 1s two-photon decay show the resonance behaviour, seen as a gradual increase of the differential transition probability as the partition of energy among the two photons of the pair deviates from equal-energy partition ($E_1 = E_2 = E_0/2$). The effect was predicted by the second-order perturbation theory of two-photon decay in which summation over intermediate states is carried out over occupied and unoccupied bound states, and over unbound states.

I. Introduction

Simultaneous emission of photon pairs that continuously share the transition energy, the two-photon decay (also named double-photon decay and xx de-

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cay) is a very rare process in transitions among atomic inner-shell vacancy states. Investigation of the two-photon decay process in many-electron atomic systems with a vacancy in the K-shell was initiated by Freund[1]. He made a nonrelativistic calculation based on second-order perturbation theory and independent-particle model. Frozen orbitals were assumed, the matrix elements were related to the photoelectric cross sections and oscillator strengths, and the sums were made over unoccupied bound and over unbound virtual intermediate states. From the calculated rate for 2s → 1s two-photon decay in copper atoms with a K-shell vacancy he predicted that detection of the process was feasible. Bannett and Freund[2] developed an improved theory and made calculations of 2s → 1s and 3s → 1s two-photon decay in molybdenum atoms. They used nonrelativistic electric-dipole approximation, tabulated wave functions of the Roothaan type and frozen orbitals. The sum over intermediate states was extended to include the occupied bound states of electrons in the atom. For 3s → 1s two-photon decay they calculated the energy distribution that showed the resonance effect. The effect is due to the energy denominator of the term of two-photon-decay matrix element which is due to the 3s → 2p → 1s sequence of states (i. e. the occupied 2p state acts as the virtual intermediate state).

Bannett and Freund[2] also reported the results of the measurements of 2s → 1s and higher shell → 1s two-photon decay in molybdenum atoms in which the K shell vacancies were produced by a beam of silver K x rays. Ilakovac et al.[3,4] made measurements of two-photon decay in xenon atoms. Nuclear electron capture decay in ^{131}Cs was used to generate xenon atoms with K-shell vacancies. They derived the results on 2s → 1s 3s → 1s 3d → 1s and 4sd → 1s two-photon decay.

The early experimental work on two-photon decay in many-electron systems prompted the development of detailed theories of the process. Theoretical investigation of two-photon transitions in many electron systems are very difficult. Second-order perturbation theory matrix element of the process is given by an infinite sum over discrete bound states and over states in the continuum of products of matrix elements of multipole-field operators of transitions between the initial state and a multitude of intermediate states, and between the intermediate states and the final state, each product being divided by an energy factor. To make comparison with the experimental data, calculations were made for medium-heavy and for heavy atoms. Therefore, relativistic self-consistent-field theory was required. As analytic solution of the problem is not expected to be found, numerical evaluations were made. Mu and Crasemann[5] made elaborate relativistic self-consistent-field calculations of two-photon transitions in xenon atoms. The differential transition probability of two-photon decay is given by[5,6]

$$\frac{\mathrm{d}^3 W}{\mathrm{d}E_1\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2} = \frac{\alpha^2}{8\pi^3}\,\omega_1\,\omega_2 |\,M_{fi}\,(\mathbf{k}_1, \alpha_1; \mathbf{k}_2, \alpha_2)|^2,$$

where $\alpha = 1/137$ is the fine structure constant, ω_1 and ω_2 are the frequencies of the two photons, and M_{fi} is the symmetrized matrix element of the transition:

$$M_{fi}\,(\mathbf{k}_1, \alpha_1; \mathbf{k}_2, \alpha_2) = M\,(\mathbf{k}_1, \alpha_1; \mathbf{k}_2, \alpha_2) + M\,(\mathbf{k}_2, \alpha_2; \mathbf{k}_1, \alpha_1).$$

$\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors and α_1 and α_2 the polarization states of the two photons, and

$$M(\mathbf{k}, \alpha; \mathbf{k}', \alpha') = \sum_n \frac{\langle f \mid \alpha \cdot \varepsilon^{(\alpha')*} \mathrm{e}^{-\mathrm{i}\mathbf{k}' \cdot \mathbf{r}'} \mid n \rangle \langle n \mid \alpha \cdot \varepsilon^{(\alpha)*} \mathrm{e}^{-\mathrm{i}\mathbf{k} \cdot \mathbf{r}} \mid i \rangle}{E_n - E_i + \omega},$$

where i and f represent the initial and final states, the sum is carried over virtual intermediate states n, α is the Dirac matrix, $\varepsilon^{(\alpha)*}$ and $\varepsilon^{(\alpha')*}$ are the conjugates of the photon polarization vectors, E_i and E_n the energies of the initial and intermediate states, and ω is the energy of one of the photons.

In the following paper[6] they extended the calculations to two-photon decay in molybdenum and silver atoms. A nonrelativistic self-consistent-field calculation of the two-photon decay process in krypton, xenon, and radon atoms was made by Wu and Li[7]. Tong, Li, Kissel and Pratt[8] made elaborate relativistic self-consistent-field calculations of two-photon decay in molybdenum, silver, and xenon atoms.

The two-photon transitions among atomic inner-shell-vacancy states are closely related to the two-photon decay in hydrogen and one-electron ions. Generally, the single-vacancy states in many-particle systems and the states of one-particle systems have many common features. The role of occupied electron states in the calculation of the sum over intermediate states was at first uncertain. Guo[9] proved that occupied intermediate electron states are not excluded by the Pauli principle, and contribute the same way as the unoccupied intermediate states. This was previously assumed in calculations of two-photon decay in molybdenum[2] and xenon[5]. Therefore, theoretical results on two-photon decay in hydrogen and one-electron ions and in single-vacancy many-electron systems bear many similarities. The original work on two-photon transitions was made by M. Göppert-Mayer[10] in 1931. She developed the theory of 2s $\rightarrow$ 1s two-photon decay in hydrogen atoms. Reliable numerical results on transition probabilities were calculated by Shapiro and Breit[11]. The nonrelativistic problem of two photon decay in hydrogenic systems has been solved analytically, first for 2s $\rightarrow$ 1s decay[12,13] and more recently for other transitions[14–17]. All results of calculations on ns $\rightarrow$ $\rightarrow$ 1s and nd $\rightarrow$ 1s, $n \geq 3$, two photon decay for one-electron systems show the resonance effects.

We report here on our experimental results on two-photon decay in two many-electron system, the silver and hafnium atoms with a K-shell vacancy, which show the predicted rise of the differential probability for increasingly asymmetrical partition of transition energy between two photons. A report on the results of this work was given at a conference[18].

2. *The apparatus*

All inner-shell-vacancy states decay very fast by emission of characteristic radiation, Auger electrons, and by other processes. These transitions create new vacancies, which subsequently decay by emission of photons and electrons. Since these events are very fast and about six orders of magnitude more frequent than the searched-for two-photon decay events, care must be exercised to reduce excessive coincidence rates. Serious problems are caused also by the processes that closely mimic the two-photon decay events and by accidental coincidences. For these reasons great care must be exercised to eliminate or to reduce these effects to levels that allow clear observation of two-photon events.

A pair of planar high-purity germanium detectors (supplied by ORTEC, Oak Ridge, U. S. A.) was used for the detection of photons emitted from the sources. They were placed head-on in a close 180° geometry. Multilayer shields were made of aluminium with different inserts. In the centres of the shields 1 mm diameter double-taper holes were made. Small sources of ^{109}Cd or ^{179}Ta were placed between two foils of polyethylene and carefully centered into the hole of a shield. The detectors and the shield with the source were placed in a cylindrical lead shield of the 52 mm inner and 80 mm outer diameter.

Nominal size of the detectors was 200 sq. mm $\times$ 7 mm thick. Energy resolution of the two detectors for 5.9 keV photons was about 230 and 240 eV. The electronic system consisted of a fast-slow coincidence unit and a three-parameter 128 $\times$ 512 $\times$ 512 channel analyzer. The three analog-to-digital converters (ADCs) were used to measure the time difference (k_0 channel, the 128-channel ADC) and pulse heights of pulses from the two detectors (k_1 and k_2 channels, two 512-channel ADCs) for each coincidence event. The experimental setup was the same as in the previous measurements of two-photon decay in xenon, except for different electronic adjustments (see Ref. 4 for a more detailed description). For each series of measurements several test runs were made to check the system and to make adjustments. Before the start and after the end of each measurement the system was checked for peak positions and counting rates in the singles spectra, and calibrated with a high precision mercury-relay generator.

3. *Measurements and analysis of data on two-photon decay in silver*

Silver atoms with a vacancy in the K shell were generated in the decay of ^{109}Cd. Radioactive ^{109}Cd in a 1N HCl solution was acquired from New England Nuclear (Boston, Mass., USA). In order to prepare a source of very small dimensions, a 0.4 mm diam. cutting from pure cellulose paper was immersed into a drop of the ^{109}Cd solution and the drop was dried. The small piece of radioactive paper was placed between two foils of polyethylene and centered in the hole of the shield. The shield with the source was placed between the germanium detectors.

Two series of measurements were made, the first of 21 and the second of 12 measurements. Initial strength of the source of the first 4 measurements of the first series was about 0.8 kBq and of the remaining 17 measurements about 2.2 kBq, the gain in either energy branch was close to 60 eV per channel and the shield was made of aluminium with brass inserts. In the second series of measurements the initial strength of the source was about 1.3 kBq, the gain 70 eV per channel and the shield was made of brass with gold inserts.

Stability of the measuring system was checked for each measurement by making projections of the recorded data onto the k_0, k_1 and k_2 axes. The criterion for acceptance was that shifts of characteristic peaks amount to less than one channel. One measurement of the first series was rejected. All data from the 20 accepted measurements of the first series were formed into one set of data and all data from the 12 measurements of the second series into the second set. Each set was analyzed separately. The total times of collection of data were 1121 and 2118 hours for the first and second set, respectively.

A detailed description of the methods of analysis of data on two-photon decay in xenon atoms was given in a previous publication (Ref. 4). The same methods were adapted for the analysis of data on two-photon decay in silver. They are only briefly described here. As the measurements lasted for several months, great care was taken to achieve stable operation of the apparatus. In order to avoid effects of small shifts, the analysis of data relied entirely on the complete sets of records of the three-parameter data.

Projection of the three-parameter data onto the k_0 axis for various sections of the $k_1 - k_2$ plane yielded the time spectra. They were analyzed with the aim to determine the intervals of k_0 channels which included about 95% of in-coincidence events. The data in the chosen time intervals were projected onto the $k_1 - k_2$ plane. In this way the tables of in-coincidence numbers of events, the $E_1 - E_2$ energy spectra (one for each of the two sets of data) were obtained. The $E_1 - E_2$ energy spectra were the basis of all subsequent analyses.

Due to the 180° geometry, cross talk among the two detectors via germanium K x rays caused prominent two-dimensional peaks in the $E_1 - E_2$ spectra. Absorption of a silver K x ray in the sensitive volume of one detector accompanied by the emission of a K x ray from a germanium atom, escape of the x ray from the detector, its passage through the hole in the shield between the detectors, and its absorption in the second detector caused the cross-talk events[19]. Energy deposited in the first detector equals E (Ag Kx) $-$ E (Ge Kx) and in the second detector E (Ge Kx). The cross-talk peaks allowed a very reliable energy calibration of the energy axes. Fixed energy scales allowed the use of the linear fitting routines in the analyses of the $E_1 - E_2$ spectra.

An improvement in the measurements of two-photon decay in silver, in comparison to the study of two-photon decay in xenon atoms[3,4], was the use of lower discriminator thresholds. Reliable determination of numbers of events due to two-photon decay below the energy of germanium K x rays was possible.

In the two $E_1 - E_2$ energy spectra derived from the recorded three-parameter data with the ^{109}Cd sources, the events due to two-photon decay were observed as continuous bands along the lines of constant sum energy corresponding to the 2s $\rightarrow$ 1s, 3s $\rightarrow$ 1s, 3d $\rightarrow$ 1s and 4sd $\rightarrow$ 1s transition energies. Identification of data due to two-photon decay is based on energy arguments. Transition energy of the 3d $\rightarrow$ 1s two-photon decay is larger than the energy of $K\beta_1$ x rays (which are due to the 3p $\rightarrow$ 1s transitions) by the difference of binding energies of 3p and 3d electrons. The difference amounts to 210 eV in silver. The sum spectra of sections of the $E_1 - E_2$ spectrum containing the cross-talk peaks due to the $K\beta$ x rays of silver, and of sections containing the nearby ridges clearly shows the shift of about 210 eV (see Fig. 1).

Two methods were applied to determine the numbers of events. In one complex surfaces were fitted to the sections of each of the two $E_1 - E_2$ spectra. Regions around the cross-talk peaks were omitted. In the second method projections of data onto the $k_1 + k_2$ axis were made for several intervals of the ratio of energy of one photon and of the transition energy of two-photon decay, $x = E/E_0$, omitting again the regions around the cros-stalk peaks. The one-dimensional spectra

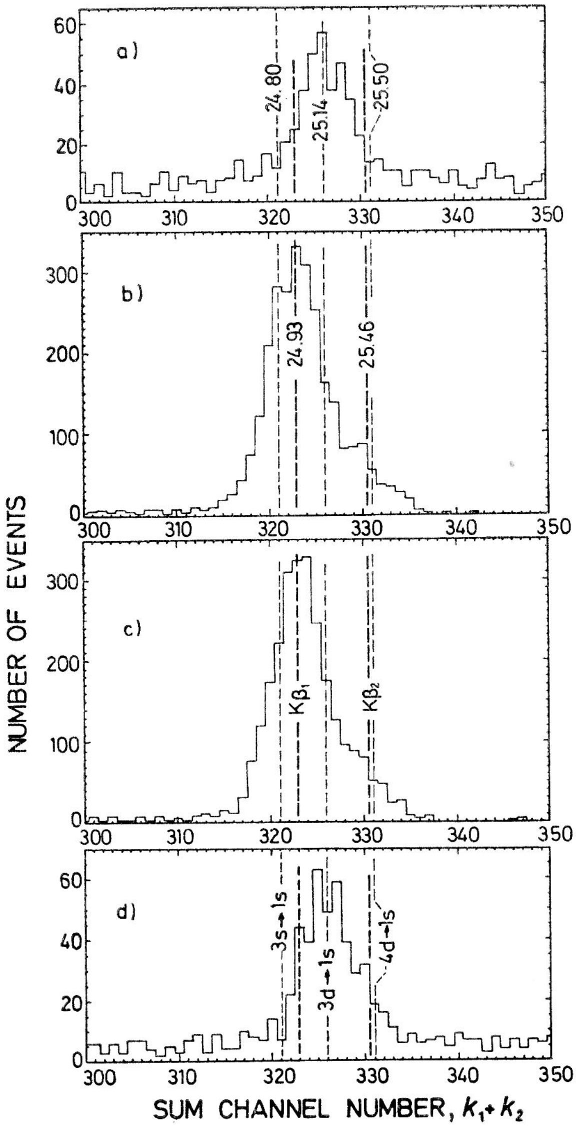

Fig. 1. Sum spectra of sections of the two-dimensional $E_1 - E_2$ spectrum. The sum spectra shown in a) and d) were obtained by projection of data in section containing the ridges, i. e. of regions where continuous partition of transition energy occurs, while the sum spectra shown in b) and c) were obtained by projection of data in the strong two-dimensional peaks due to the detection of $K\beta_1$ (E = 24.93 keV) or $K\beta_2$ (E = 25.46 keV) x rays of silver in one detector accompanied by escape and absorption od a germanium K x ray in the other detector. Sum energy of events in the ridge is seen to be larger than the energy of the $K\beta_1$ x rays. The events in the ridge are interpreted to be mainly due to the 3d → 1s two-photon decay because the energy difference amounts to the difference of binding energies of the 3p and 3d electrons in silver (about 210 eV).

were fitted by curves. In either case linear fitting routines were applied, because the energy scales were fixed. The numbers of events were derived using equal weights and the errors using $1/n$ weights (one for zero count[20]). The methods are described in more detail in Ref. 4.

4. *Measurements and analysis of data on two-photon decay in hafnium*

Radioactive ^{179}Ta was produced in the cyclotron of the R. Bošković Institute. A rotating copper target, onto which small pieces of high-purity hafnium metal plates were hard soldered, was bombarded by the 14-MeV deuteron beam. The essential steps in separation and purification of ^{179}Ta were dissolving of filings of the irradiated hafnium metal in a mixture of HF and HCl acids, preparation of the solution, solvent extraction from the solution with the diisobutil ketone equilibrated with a mixture of 12 M HCl and 0.4 M HF acids, and aqueous extraction from that solution. The aqueous solution was dried and the traces of substance were collected using a minute speck of glue. When dry, a small spherically-shaped source of a diameter of about 0.4 mm was obtained.

Three measurements were made with a ^{179}Ta source about 3.4 kBq strong. Projections of the three-parameter data onto the k_0, k_1 and k_2 axes were analyzed to check for the consistency of the data. Negligible shifts were found, so the complete records of the three measurements were treated as one set of data. Total time of collection of data of the three measurements was about 426 hours.

As in the analysis of data on two-photon decay in silver atoms, the time spectra were made for several sections of the $k_1 - k_2$ field. A large central region showed negligible shifts of the coincidence peaks. The analysis was limited to the central region, corresponding to the photon energies in either detector above about 17 keV. An about ± 2 standard deviations wide interval of time (k_0) channels was selected for the subsequent analyses.

All three parameter data in the accepted interval of time (k_0) channels were projected onto the $k_1 - k_2$ plane yielding the two-dimensional $E_1 - E_2$ energy spectrum. All the subsequent analyses were based on the $E_1 - E_2$ energy spectrum.

The discriminator thresholds were set above the hafnium L x ray peaks to avoid a high coincidence rate. Therefore, coincidences due to the cross talk among the detectors via germanium K x rays were also eliminated. Since no isolated two-dimensional peaks were observed in the analyzed region of data, energy calibration was based on the analysis of the sum spectra. Namely, the two-photon decay events appear in the $E_1 - E_2$ spectrum as ridges at constant sum energy that project into peaks when sum spectra are made. The sum spectra were analyzed assuming the presence of peaks due to the 2s $\rightarrow$ 1s, 3s $\rightarrow$ 1s, 3d $\rightarrow$ 1s and 4sd $\rightarrow$ 1s two-photon decay, and due to the cross talk among the germanium detectors via Compton effect. Two separate measurements were made, one before and the other after the above described measurements, with a weaker source and lower discriminator thresholds. Since the pulses from the absorption of L x rays triggered the discriminators, multiple two-dimensional peaks were observed due to the coincidences of K and L x rays of hafnium. Two-dimensional fitting routines were applied to determine the positions of the peaks and to calibrate the energy scales. The energy scales on the k_1 and k_2 axes thus determined were in a very good agreement, and when recalculated to the sum-energy scale, a very good agreement was obtained with the scale derived from the sum spectra. In this way stability of the system and the close equality of the energy scales on the k_1 and k_2 axes were checked.

Sum spectrum for a broad section of the $k_1 - k_2$ field, for a range of the parameter $x = E/E_0$ from about 0.35 to about 0.65 was analyzed by a minimum χ^2 routine with the aim to determine the sum-energy scale and the peak widths. The component functions in the sum spectrum were assumed to be the peaks due to the 2s $\to$ 1s, 3s $\to$ 1s, 3d $\to$ 1s and 4sd $\to$ 1s two-photon decay and the peaks due to cross talk via backscattered Compton radiation from the $K\alpha_2$, $K\alpha_1$, $K\beta_1$ and $K\beta_2$ x rays of hafnium.

Two methods of analysis of the $E_1 - E_2$ spectrum were applied: fitting of a surface to the central region that was subdivided into several sections, and fitting of curves to the sum spectra that were made by projecting the data in the $E_1 - E_2$ spectrum onto the $k_1 + k_2$ axis. Since the energy scales and the widths of the peaks were fixed, linear routines were applied. The two methods of analysis are described in more detail in Ref. 4. The fitting function was different in that the ridges (in surface fitting) and the peaks (in curve fitting) in addition to the 2s $\to$ 1s, 3s $\to$ 1s, 3d $\to$ 1s and 4sd $\to$ 1s lines, included also the cross-talk lines due to Compton backscattering of K x rays of hafnium. The numbers of events were derived using equal weights and the errors using $1/n$ weights (1 for zero count[20]). The analysis yielded the numbers of events, Δn, due to the 2s $\to$ 1s, 3s $\to$ 1s, 3d $\to$ 1s and 4sd $\to$ 1s two-photon decay in hafnium for a number of energies of one photon of the pair. A good agreement between the two sets of results was obtained. The results of the curve fits seem to be more reliable. We consider here only the data on 3d $\to$ 1s two-photon decay.

5. *Results and discussion*

The differential transition probabilities of the two-photon decay per decay of a K-shell vacancy in the atoms of silver and hafnium were calculated from the formula[4]

$$\frac{1}{W_K}\left(\frac{\mathrm{d}W}{\mathrm{d}E\,\mathrm{d}\Omega_1\,\mathrm{d}\Omega_2}\right)_{\Theta=\pi} = \frac{\Delta n/\Delta E}{n_K\,\varepsilon_1\,\varepsilon_2\,\varepsilon_C\,\Delta\Omega_1\,\Delta\Omega_2\,F}$$

where Δn is the number of events due to two-photon decay in an energy interval ΔE, n_K is the number of K-shell vacancies generated in the source during the measurement, ε_1 and ε_2 are the peak efficiencies of the detectors, ε_C is the coincidence efficiency, $\Delta\Omega_1$ and $\Delta\Omega_2$ are the solid angles of the detectors subtended from the source, and F is a correction factor (equal to the average value over solid angles of the two detectors of the product of the angular correlation function, of the photon-attenuation factors, and of the probabilities of total energy absorption in germanium[4]).

In the measurement of two-photon decay in silver atoms the solid angles of the detector sensitive volumes were $\Delta\Omega_1 = 1.61$ sr and $\Delta\Omega_2 = 1.57$ sr, the coincidence efficiency was $\varepsilon_C = 0.95$, and the peak efficiencies of the detectors were $\varepsilon_1 = \varepsilon_2 = 0.95$.

The results on the relative differential transition probabilities of 3d → 1s two-photon decay in silver atoms are given in Table 1 and are shown in Fig. 2. The dashed curve shows the results of nonrelativistic electric-dipole approximation for one-electron silver ions that was calculated from the results of Tung, Ye, Salamo, and Chan[14] and of Florescu[15] (which closely agree). The solid curves in Fig.

TABLE 1.

First measurement		Second measurement	
$x = E/E_0$	$\frac{1}{W_K}\frac{d^3W}{dE\,d\Omega_1\,d\Omega_2}$	$x = E/E_0$	$\frac{1}{W_K}\frac{d^3W}{dE\,d\Omega_1\,d\Omega_2}$
0.261	16.08 ± 2.54	0.241	14.20 ± 1.78
0.309	9.36 ± 1.98	0.291	8.48 ± 1.22
0.357	7.88 ± 1.34	0.341	6.66 ± 1.22
0.406	5.98 ± 3.76	0.391	6.58 ± 1.60
0.454	10.50 ± 3.84	0.441	6.74 ± 1.14
0.500	6.54 ± 1.06	0.500	7.64 ± 1.14
0.548	5.48 ± 3.80	0.561	6.18 ± 1.30
0.596	9.50 ± 3.64	0.611	6.20 ± 1.62
0.644	6.94 ± 1.30	0.661	9.96 ± 1.28
0.692	10.68 ± 1.36	0.712	10.28 ± 1.20
0.740	15.44 ± 1.68	0.762	15.16 ± 1.74

Relative differential transition probabilities od 3d → 1s two-photon decay in silver in units of 10^{-9} keV^{-1} sr^{-2} at $\Theta = 180°$. The transition energy is $E_0 = 25.144$ keV.

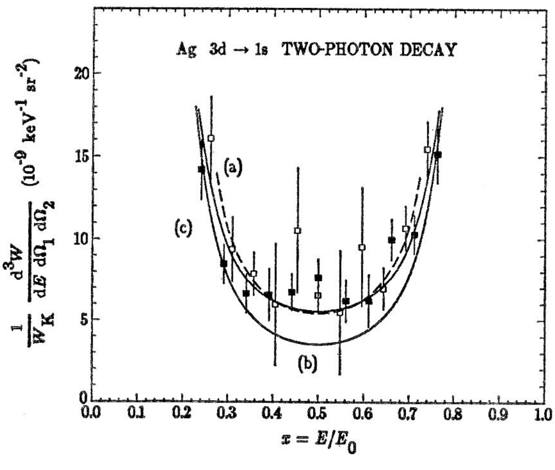

Fig. 2. Relative differential transition probabilities of 3d → 1s two-photon decay in silver at 180°. Experimental results from the first and second measurement are shown by hollow and full squares, respectively. The dashed curve (a) shows the results derived from expressions of Florescu[15] for one-electron silver ions. The solid curves show the results of relativistic self-consistent-field calculations of Mu and Crasemann[6] [curve (b)] and of Tong, Li, Kissel and Pratt[8] [curve (c)].

2 show the results of relativistic self-consistent-field calculations of Mu and Crasemann[6] and of Tong, Li, Kissel and Pratt[8]. Their results for 3d → 1s two-photon decay show a considerable difference [curves (b) and (c)]. In the derivation of all theoretical results the decay rate of the K-shell vacancy in silver atoms $W_K = 9.80\ 10^{15}\ s^{-1}$ was assumed.

In the measurement of the two-photon decay in hafnium atoms the solid angles of the sensitive volumes of the detectors were $\Delta\Omega_1 = 1.41$ sr and $\Delta\Omega_2 = 1.45$ sr, the coincidence efficiency was $\varepsilon_C = 0.95$, and the peak efficiencies of the detectors were $\varepsilon_1 = \varepsilon_2 = 0.90$.

The results on relative differential transition probabilities of 3d → 1s two photon decay in hafnium atoms are given in Table 2 and are shown in Fig. 3.

TABLE 2.

$x = E/E_0$	$\frac{1}{W_K} \frac{d^3W}{dE\, d\Omega_1\, d\Omega_2}$
0.355	10.16 ± 1.17
0.396	6.48 ± 0.88
0.437	5.23 ± 0.79
0.478	4.90 ± 0.86
0.519	6.56 ± 0.94
0.560	6.38 ± 0.97
0.601	6.38 ± 0.95
0.642	7.61 ± 1.01

Relative differential transition probabilities of 3d → 1s two-photon decay in hafnium atoms in units of 10^{-9} keV^{-1} sr^{-2} at $\Theta = 180°$. The transition energy is $E_0 = 63.667$ keV.

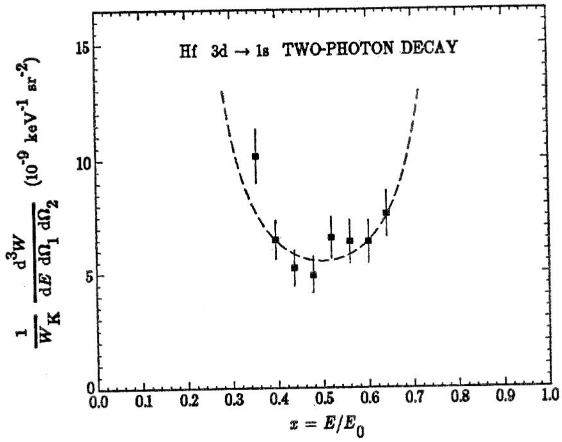

Fig. 3. Relative differential transition probabilities of 3d → 1s two-photon decay in hafnium at 180°. Experimental results are shown by full squares. The dashed curve shows the results derived from the expressions of Florescu[15,17] and from the results of Tung, Ye, Salamo and Chan[14] for one-electron hafnium ions.

The total transition probability of decay of a K-shell vacancy in hafnium atoms was assumed to have the value of $W_K = 5.24\ 10^{16}\ s^{-1}$. The dashed curve in Fig. 3. shows the results of our calculation of the differential transition probabilities of two-photon decay from the expressions of Florescu[15,77] for one-electron hafnium ions.

The rise of the differential transition probability of the 3d → 1s two-photon decay in silver and hafnium atoms for increasingly asymmetric partition of energy among the two photons (i. e. for increasing values of $|x - 0.5|$) is considered to be the experimental proof of the predicted resonance effect[2,5]. If the occupied 2p state of atoms with a K-shell vacancy did not take part as an intermediate state in the two-photon decay process, the differential transition probability would have a maximum at $x = 0.5$. Therefore, we may conclude, as predicted by theory, the occupied as well as unoccupied electron states take part as intermediate states in the two-photon decay process.

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References

1) I. Freund, Phys. Rev. **A 7** (1973) 1849;
2) Y. Bannett and I. Freund, Phys. Rev. Lett. **49** (1982) 539; Phys. Rev. **A 30** (1984) 299;
3) K. Ilakovac, J. Tudorić-Ghemo, B. Bušić and V. Horvat, Phys. Rev. Lett. **56** (1986) 2469;
4) K. Ilakovac, J. Tudorić-Ghemo and V. Horvat, Phys. Rev. **A 44** (1991) 7392;
5) X. Mu and B. Crasemann, Phys. Rev. Lett. **57** (1986) 3039;
6) X. Mu and B. Crasemann, Phys. Rev. **A 38** (1988) 4585;
7) Y.-J. Wu and J.-M. Li, J. Phys. **B 21** (1988) 1509;
8) X.-M. Tong, J.-M. Li, L. Kissel and R. H. Pratt, Phys. Rev. **A 42** (1990) 1442;
9) D. S. Guo, Phys. Rev. **A 36** (1987) 4267;
10) M. Göppert-Mayer, Ann. Phys. (Leipzig) **9** (1931) 273;
11) J. Shapiro and G. Breit, Phys. Rev. **113** (1959) 179;
12) B. A. Zon and L. P. Rapoport, Pis'ma Zh. Eksp. Teor. Fiz. **7** (1968) 70 [JETP Lett. **7** (1968)52];
13) S. Klarsfeld, Phys. Lett. **30 A** (1969) 382;
14) H. Tung, X. M. Ye, G. J. Salamo and F. T. Chan, Phys. Rev. **A 30** (1984) 1175;

15) V. Florescu, Phys. Rev. **A 30** (1984) 2441;

16) A. Costescu, I. Brândus, and N. Mezinescu J. Phys. **B 18** (1985) L11;

17) V. Florescu, S. Patrascu and O. Stoican, Phys. Rev. **A 36** (1987) 2155;

18) K. Ilakovac, in *X-Ray and Inner-Shell Processes*, Proceedings of the fifteenth international conference on x-ray and inner-shell processes, Knoxville, Tennessee, 1990. (Ed. T. A. Carlson, M. O. Krause and S. T. Manson). AIP Conf. Proc. No. 215, American Institute of Physics, New York, 1990, p. 498;

19) K. Ilakovac, J. Tudorić-Ghemo, V. Horvat, N. Ilakovac, S. Kaučić and M. Vesković, Nucl. Instr. and Methods **A 245** (1986) 467;

20) P. R. Bevington, *Data Reduction and Error Analysis for the Physical Sciences*, McGraw-Hill, New York, 1969.

REZONANTNI EFEKT U 3d → 1s DVOFOTONSKOM RASPADU MEĐU UNUTARNJIM ATOMSKIM LJUSKAMA

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Istraživano je zračenje parova fotona procesom dvojnog fotonskog raspada u atomima srebra i hafnija s K šupljinom. Radioaktivni raspad ^{109}Cd i ^{179}Ta služio je za tvorbu K šupljinskih stanja u atomima srebra odnosno hafnija. Mjerni uređaj sastojao se od para planarnih germanijskih detektora visoke čistoće, brzo-sporog sudesnog sustava, i $128 \times 512 \times 512$ kanalnog amplitudnog analizatora. Analizirani su dvodimenzijski spektri brojeva događaja kao funkcije amplituda sudesnih impulsa iz dvaju detektora. Rezultati za 3d → 1s dvojni fotonski raspad pokazuju rezonantni efekt, koji se opaža kao postepen porast diferencijalne prijelazne vjerojatnosti kako razdjela energije među dvama fotonima para odstupa od jednake razdjele ($E_1 = E_2 = E_0/2$). Taj je efekt predskazan u perturbacijskoj teoriji dvojnog fotonskog raspada drugog reda u kojoj se zbrajanje preko virtualnih međustanja provodi preko zauzetih i slobodnih vezanih stanja i preko nevezanih stanja elektrona.