Two-photon decay of *K*-shell vacancies in silver atoms

P. H. Mokler,[1,2,*] H. W. Schäffer,[1,†] and R. W. Dunford[3]
[1]*GSI Darmstadt, Darmstadt, Germany*
[2]*University of Giessen, Giessen, Germany*
[3]*Argonne National Laboratory, Argonne, Illinois 60439, USA*
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The spectral distributions for the two-photon decay modes of singly *K*-shell ionized silver atoms are determined by x-ray–x-ray coincidence measurements. Ag *K*-shell vacancies were induced by nuclear electron capture decay of radioactive cadmium isotopes ^{109}Cd and two-photon coincidences were taken back to back $(180°)$ and at a 90° opening angle for the emission. Each of the two-photon transitions from the 2*s*, 3*s*, and 3*d* states exhibits unique angular and spectral distributions. The measurements agree nicely with relativistic self-consistent field calculations of Tong *et al.* Our results also confirm and extend the earlier experimental data of Ilakovac and co-workers with improved accuracy.

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I. INTRODUCTION

Two-photon decay—first discussed in 1929 by Göppert-Mayer [1,2]—gives access to the complete structure of an atomic system. For a recent overview of the field, see Mokler and Dunford [3] and the references cited there. Two-photon decay can be described analytically as a summation over all possible intermediate (n) states under the boundary condition of energy conservation, i.e., the energy sum of the two photons gives the difference in total binding energy of the initial (i) and final (f) states. As the photons are indistinguishable, the two-photon spectra are symmetric around the midpoint at half the total transition energy. The angular distribution for the opening angle between the two photons is given by the angular correlations of the cascades via the intermediate states.

Recently, emphasis was given to investigations of two-photon decay processes in heavy He-like systems [4–6]. These systems are simple and give access to true relativistic phenomena; the strength of the central force, i.e., the atomic number, influences considerably the spectral distribution for two-photon decay [7,8]. For many-electron systems—like true atoms—the influence of all the other atomic electrons on the central potential has to be taken into account [9–14]. In heavy atomic systems the two-photon decay is an extremely weak branch (typically 10^{-6}) compared to the standard $(E1)$ single-photon ground-state transitions [15]. The first observation of two-photon decay of heavy atoms with an initial *K*-shell vacancy was made by Bannett and Freund [15,16]. They irradiated Mo with Ag x rays from a sealed x-ray tube and measured two-photon coincidences between two solid state Si(Li) x-ray detectors. Ilakovac and co-workers [17–19] used radioactive nuclei to generate *K*-shell vacancies in Xe, Ag, and Hf atoms via nuclear electron capture. By observing two-photon coincidences with a pair of Ge(i) x-ray detectors they detected back-to-back emission of two-photon decay for the transitions $2s \rightarrow 1s$, $3s \rightarrow 1s$, $3d \rightarrow 1s$, and $4s$, $4d \rightarrow 1s$. In this paper we report on an advanced measurement for *K*-shell ionized Ag atoms using a similar experimental technique. We improved the experimental accuracy for the two-photon distributions, extended the accessible spectral range, and also measured two-photon coincidences at an opening angle of 90°. The results are compared with predictions [12] for the spectral shape of the different two-photon decay modes and with the expected angular distributions.

II. EXPERIMENTAL ARRANGEMENT

The radioactive cadmium isotope ^{109}Cd decays with a lifetime of 462.6 days via electron capture to an isomeric state in ^{109}Ag* atoms. An atomic *K*-shell vacancy is produced with a probability of 0.815 per nuclear decay. The isomeric ^{109}Ag* decays further with a lifetime of 39.6 s to the ground state by photon emission $(E3)$ or internal conversion, and for this decay branch an atomic *K*-shell vacancy is produced with a probability of 0.417. Hence, ample single *K*-shell vacancies will be created in this radioactive decay. The *K*-shell excited Ag atoms produced decay predominantly via characteristic *K*-shell x-ray emission (fluorescence yield $\omega = 0.834$), but a tiny fraction decays via two-photon emission.

In the present experiment—for more details see Ref. [20]—we used a standard radioactive ^{109}Cd calibration source with an activity of 75 kBq (a commercial source from Amersham) for the production of single atomic *K*-shell vacancies in Ag atoms. We measured the x-ray emission and registered photon coincidences using three Si(Li) x-ray detectors. In Fig. 1 a sketch of the simple experimental arrangement is shown. Two detectors (*A* and *B* with a sensitive area of 200 mm^2 each) are facing the source and the third one (*C* with 80 mm^2) is oriented perpendicular to the others. The detectors are carefully shielded by 3-mm-thick aluminum apertures to reduce cross talk due to photon scattering, i.e., each detector ideally reacts only to the source and to neither of the other two detectors. Estimates show that contributions from Compton scattering, *K*-escape radiation from a detec-

*Electronic address: P.Mokler@GSI.de
†Permanent address: IBM Germany, D-60528 Frankfurt, Germany.

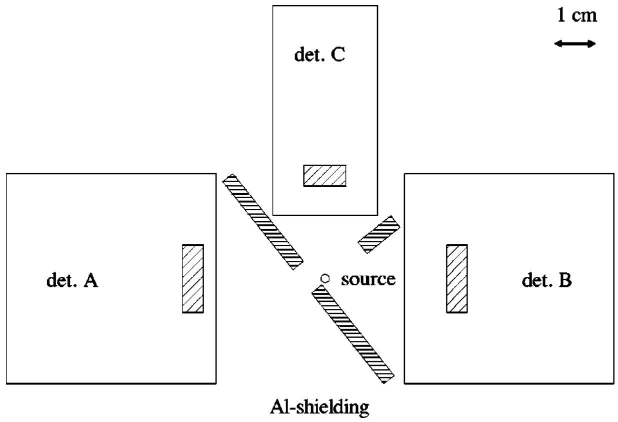

FIG. 1. Experimental arrangement.

tor, and bremsstrahlung will not disturb the determination of the coincident two-photon decay spectra.

Two-photon coincidences between the detectors were registered in event mode using standard electronic modules for signal processing and data acquisition. Standard slow-fast coincidence techniques similar to those described in Refs. [5,6,20] were used; special care was taken to get high electronic coincidence efficiencies especially at low x-ray energies (with a cut below 2 keV). The detector geometry (solid angles typically 0.015 sr), detector efficiencies, and photon absorption in the detector windows, the air, and the source were all carefully determined, as well as the different electronic coincidence efficiencies (about 100%). For details see Ref. [20]. As the strength of the source is known and was also independently measured, absolute numbers for the two-photon transition probabilities were determined. In the present experiment we integrated over 1.47×10^{11} single K-shell vacancy decays in the source.

In the data analysis, time spectra for the coincidences were created initially to enable subtraction of random coincidences from the prompt ones. In Fig. 2 a two-dimensional event plot for true two-photon coincidences is displayed for the back-to-back emission case; the photon energy registered in detector B is plotted against the sum energy of the two photons in detectors A and B. In this representation the two-photon decay events lie along the vertical lines indicated by the arrows at the top for the two-photon transitions from $2s$ to $1s$ and from $3s/3d$ to $1s$. The two-photon spectra are hidden in these vertical ridges. For clarity we take horizontal cuts at three different photon energies in detector B and show in Fig. 3 the corresponding sum x-ray spectra (for E_B=5.5, 10.5, and 15.5 keV). The two photon ridges for $2s\rightarrow1s$ and $3s/3d\rightarrow1s$ clearly show up and stay at the same sum energy. The line intensities along the ridges yield the two-photon spectra.

III. RESULTS AND DISCUSSION

In Fig. 4 the final results for the observed two-photon transitions $2s\rightarrow1s$, $3s\rightarrow1s$, and $3d\rightarrow1s$ are shown (full squares), on the left side for 180° (back-to-back) observation and on the right side for 90° emission. The two-photon spectra are already corrected for all efficiencies, and absolute differential transition probabilities are presented. For convenience, the photon energy is normalized to the total energy for each transition; hence, all spectra range up to scaled photon energies of 1. The spectral shapes for the different two-photon transitions are quite different. Also, the absolute values for 180° and 90° emission are partially at variance.

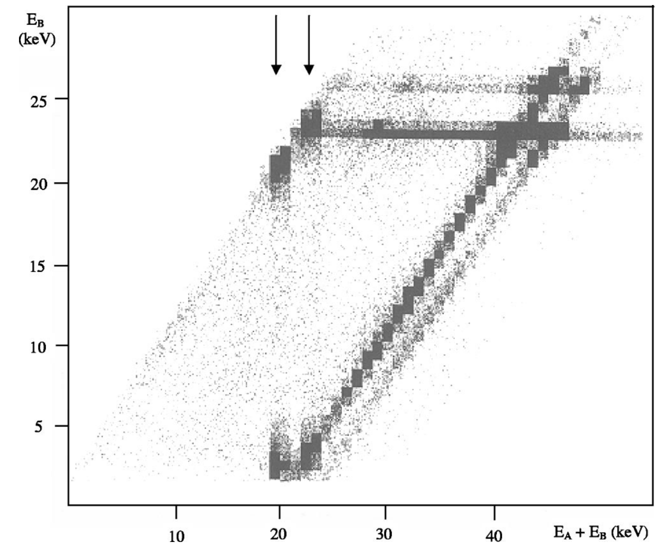

FIG. 2. Two-dimensional event plot for true two-photon coincidences. X-ray events registered in detector B are plotted against the sum energy of the two photons coincidently registered in detectors A and B (cf. Fig. 1). The arrows point to the two-photon ridges.

The 180° data can be compared to earlier results of Ilakovac *et al.* [19] (see open circles). The spectral range covered by Ilacovac *et al.* is somewhat narrower and the statistical uncertainty larger. However, there is a general good agreement between the measurements; the present data maybe give smaller absolute values for the weakest transition $3d\rightarrow1s$. Moreover, all the results are compared to theoretical predictions from Tong *et al.* [12]. Our measurements are in excellent agreement with these relativistic self-consistent-field calculations both in spectral shape and in absolute value.

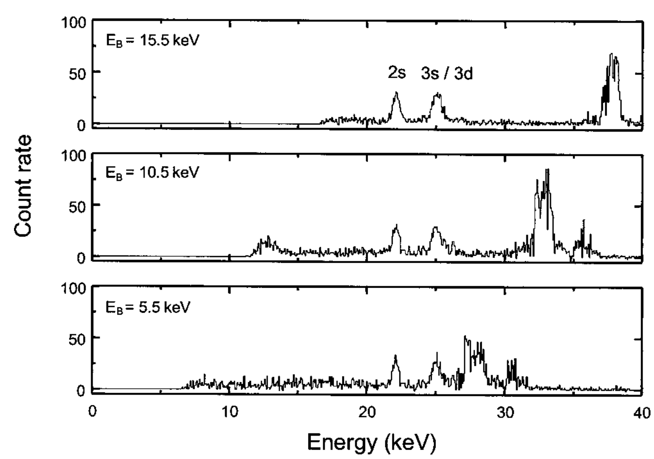

FIG. 3. Sum x-ray spectra at 5.5, 1.5, and 15.5 keV (horizontal cuts in Fig. 2).

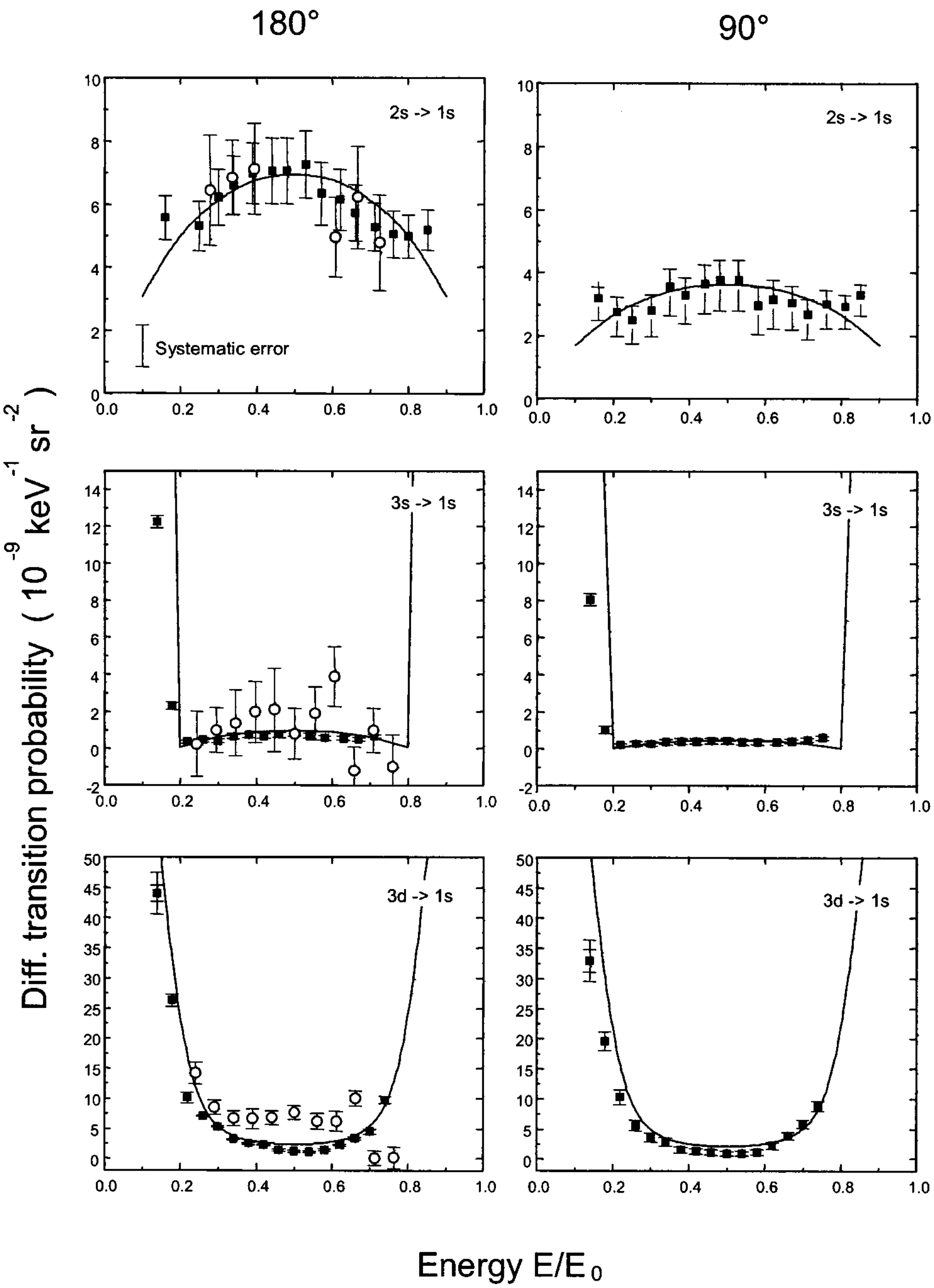

FIG. 4. Differential two-photon transition probabilities for back to back and 90° emission of the two photons. The full squares are the present results, the open circles the results of Ilakovac *et al.* [19], and the full lines give the prediction of Tong *et al.* [12].

The two measurements at 180° and 90° give the possibility of checking experimentally the angular correlation for two-photon emission. Nonrelativistic calculations of Florescu [21] give for $ns \rightarrow 1s$ transitions a $1+\cos^2\theta$ distribution and for $nd \rightarrow 1s$ transitions a $1+1/13\cos^2\theta$ correlation (θ is the opening angle between the two photons). Similar results are predicted by relativistic calculations [12,13]. Accordingly, we expect intensity ratios for $2s$, $3s$, and $3d \rightarrow 1s$ transitions of 2.00, 2.00, and 1.08 for the photons emitted with an opening angle of 180° to those with 90°. Our experimental values of 1.90 ± 0.27, 1.49 ± 0.34, and 1.22 ± 0.25, respectively, are in reasonable agreement, confirming the stated angular correlations. These intensity ratios should be independent of the spectral region, i.e., of the photon energy. In Fig. 5 we show for completeness the ratio as a function of the reduced total transition energy. The systematic error is small compared to the statistical uncertainty. The data are, within statistical accuracy, energy independent and agree with the intensity ratios deduced from Tong *et al.* [12]. The possible small variance for the $3s \rightarrow 1s$ transitions has to be checked with higher experimental accuracy.

The shapes for the two-photon spectra vary considerably with the transition considered (see Fig. 4). The $ns \rightarrow 1s$ transitions show in principle a parabolalike shape with a maximum at half the total transition energy. This is especially demonstrated by the $2s \rightarrow 1s$ case. For the $3s \rightarrow 1s$ case,

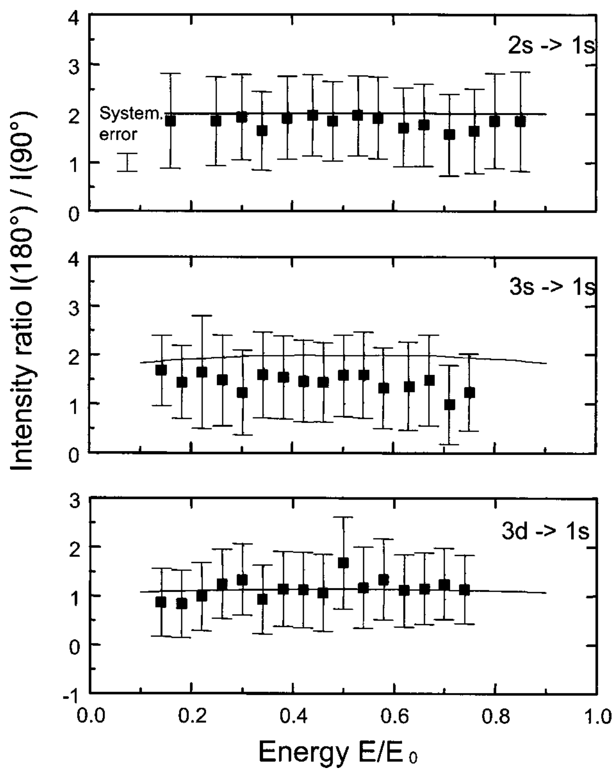

FIG. 5. Intensity ratio for 180°:90° two-photon emission: the lines are calculations according to Tong *et al.* [12].

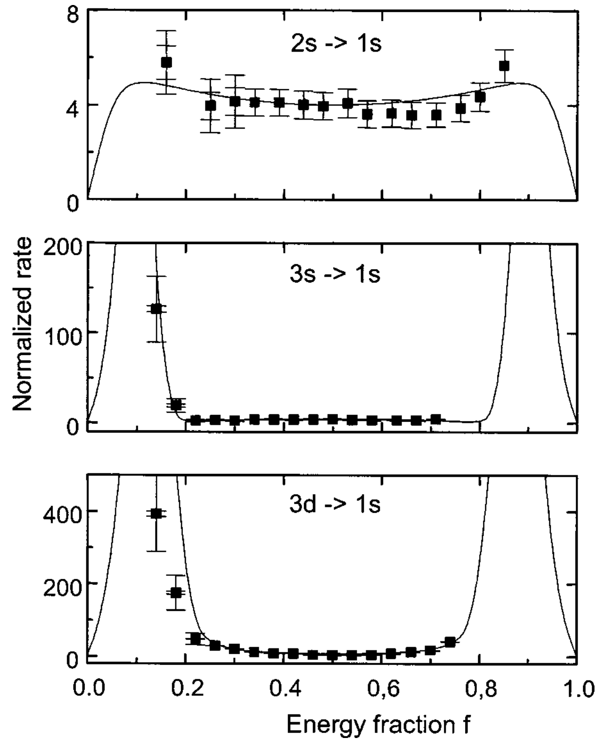

FIG. 6. Normalized rates (square of the reduced matrix elements) for two-photon emission; present results (full squares) compared with theory (solid lines) of Tong *et al.* [12].

which also has an intermediate resonance state, the 2*p* level, the spectrum increases dramatically toward this resonance, symmetric at low and high energies. This clearly demonstrates that occupied intermediate states contribute to the two-photon transitions in accordance with the fact that the Pauli exclusion principle does not prohibit within the uncertainty principle contributions from occupied intermediate states [14]. Alternatively, one may argue that the 2*p* electron decays first to the 1*s* level and then the 3*s* electron fills the 2*p* vacancy; in this time-backward case [see Fig. 1(b) in [14]], the 2*p* resonance does not involve a violation of the Pauli exclusion principle.

For the $3d \rightarrow 1s$ two-photon case we have the 2*p* level also as intermediate-state resonance and the spectrum increases correspondingly toward this resonance. However, for this case we have a local minimum at half the transition energy, in contrast to the central local maximum for $ns \rightarrow 1s$ transitions.

The spectral emission probability for the two-photon decay can be factorized into a more trivial phase space factor $(1-f)f$ (with f the fractional photon energy of the transition) and the square of a reduced matrix element $|R_{fi}|^2$ containing the physics of the two-photon transition (cf. Refs. [3,22]). Dividing out the phase space factor from the spectral distributions, we get access to the reduced matrix elements for the transitions. The results—normalized to the midpoint of the original distribution and then divided by the phase space factor—are given in Fig. 6. Statistical errors are given; for the three leftmost points in each plot, the systematic uncertainties are additionally inserted (larger error bars). The data are compared to relativistic self-consistent-field calculations from [12] using the same normalization procedure. A generally good agreement can be stated. For $ns \rightarrow 1s$ two-photon transitions the reduced matrix element is relatively energy independent, at least far from intermediate resonances. The slight deviation from a constant value is determined by the structure of the individual atom under consideration. Here, a comparison with the corresponding He-like system is highly desirable.

IV. SUMMARY

The results of present measurements of the two-photon decay in singly *K*-shell ionized Ag atoms are in good agreement with relativistic self-consistent-field calculations [12]. The measurements confirm and substantially extend earlier measurements by Ilakovac *et al.* [19]. The distributions of the two-photon spectra gave access to the reduced matrix elements for the various possible transitions, confirming the theoretical approach up to $Z=47$, i.e., in the soft relativistic region (cf. Fig. 6). Moreover, coincidence measurements at different opening angles of the two photons represent a step toward angular correlations in the atomic two-photon decay of a heavy atom. Here also, reasonable agreement with theory can be stated. Depending on the transition, $ns \rightarrow 1s$ or $nd \rightarrow 1s$, the angular correlations as well as the spectral distributions vary (cf. Fig. 4). The angular distribution is a lot more pronounced for $ns \rightarrow 1s$ transitions. Furthermore, these transitions show a local maximum at half the transition energy, whereas the $nd \rightarrow 1s$ transitions have a local minimum in the spectrum there. Finally, we would like to emphasize that intermediate-state resonances $(2p)$ lead to prominent resonance features in the corresponding spectra despite the fact that the intermediate states are occupied.

The present experiment with Ag penetrates into the soft relativistic area; here the available calculations are still in agreement with experiment. It is of special interest to extend these measurements into the strong relativistic regime, i.e., to heavier atomic species. For this strong field case higher-multipole and spin-flip transitions may contribute to the transitions and influence the emission probabilities. There a comparison of corresponding atomic and ionic systems may be of special interest. Some measurements for heavy atomic systems using radioactive sources have been reported for Xe and Hf [17,18]. In particular the Hf system is especially promising for future investigations. Recently, an experiment was performed for Au atoms using photoionization by polarized synchrotron radiation at the Advanced Photon Source at ANL [23]. Here a comparison of atomic and He-like [6] species seems to be possible in future.

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