# Inner-shell Two-photon Decay in Gold Atoms

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## Introduction

An inner-shell vacancy in an atom can decay by the simultaneous emission of two photons. Although this is a rare decay mode, it is important because it contributes to the continuum radiation on the low-energy side of the characteristic x-ray lines. The sum of the energies of the two photons is equal to the transition energy, but the energies of the individual photons vary continuously. Two-photon decay is also important from a theoretical standpoint, as it provides a unique way of testing atomic structure calculations. It is a second-order process, and the lowest-order calculation requires summation over a complete set of virtual states. Typical measurements provide information on the transition probabilities differential in the opening angle distribution and on the energy of the individual photons. Together these characteristics provide a wealth of information for testing the details of the calculations. Recent interest in theoretical atomic structure has focused on the high-Z region, where relativistic and quantum electrodynamics (QED) effects become more important.

The dominant mode of two-photon decay is the emission of two electric dipole photons (2E1), which involves transitions in which the initial and final states must have the same parity. The most important cases involving an initial K hole are the transitions ns $\rightarrow$ 1s and nd $\rightarrow$ 1s. Calculations of these processes have been made for only a few special cases that were motivated by existing experiments. Nonrelativistic self-consistent field calculations exist for Mo [1,2] and Xe [3], and relativistic self-consistent field calculations have been done for Mo, Ag, and Xe by Tong et al. [4] and Mu and Crasemann [5]. The spectral distribution for the 2s $\rightarrow$ 1s case has a broad maximum at half the transition energy, dropping to zero at either endpoint. The shapes for the ns $\rightarrow$ 1s transitions when n is $\geq$3 have broad local maxima at half the transition energy, but there are narrow resonances at energies corresponding to the n's $\rightarrow$ np $\rightarrow$ 1s cascades. The 3d $\rightarrow$ 1s, 4d $\rightarrow$ 1s, etc. transitions have local minima at half the transition energy and resonances at energies corresponding to the n"d $\rightarrow$ n'p $\rightarrow$ ns cascades.

Bannet and Freund made the first observation of two-photon decay in a heavy neutral atom [2]. They observed both the 2s $\rightarrow$ 1s and the 3d $\rightarrow$ 1s transitions following photoionization of Mo (Z = 42). Ilakovac and coworkers used radioactive sources in which the K vacancies were produced via electron capture by the nucleus. They observed back-to-back two-photon decays for the 2s $\rightarrow$ 1s, 3s $\rightarrow$ 1s, 3d $\rightarrow$ 1s transitions and for the (unresolved) 4s $\rightarrow$ 1s, 4d $\rightarrow$ 1s transitions in xenon (Z = 54), silver (Z = 47) and hafnium (Z = 72) [6-8]. They established the existence of all of these decay modes and verified the shapes of the continuum radiation, including the resonance effect in the 3d $\rightarrow$ 1s transitions. In another experiment with a radioactive source, Schäffer and collaborators [9, 10] observed the 2s $\rightarrow$ 1s, 3s $\rightarrow$ 1s, 3d $\rightarrow$ 1s, and 4s/d $\rightarrow$ 1s transitions in silver with better statistical accuracy. In addition, they probed the angular dependence of the photon opening angle $\theta$ verifying the ratio of transition probabilities between $\theta = \pi$ and $\theta = \pi/2$.

## Methods and Materials

The current effort in the area of innershell two-photon decay is to study this process in higher Z atoms to understand the physics of the combination of relativistic effects and electron-electron interactions. For these systems, it is more convenient to use photoionization to create the initial K-vacancy, particularly with the availability of third-generation synchrotron sources that provide tunable beams of high-energy x-rays with a small spot size. We have begun a program at the APS to study heavier systems and have already obtained data on a measurement of two-photon decay in gold atoms.

Our measurements were done by using x-rays from BESSRC bending magnet beamline 12-BM at the APS. The beamline monochomator used Si(333), and the resulting x-ray beam was delivered to the experimental hutch where a Ge(220) crystal was utilized to select the third order (85.5 keV) and suppress all other orders. In addition, a 2.5-cm-thick Al plate was used upstream of the Ge crystal to further reduce the first-order radiation (28.4 keV). The x-rays were incident on a 2 mg/cm$^2$ Au target, and two shielded Ge detectors arranged at 135° to the beam direction and 90° to each other detected x-rays from the target. Ta shields located between the two detectors suppressed "cross-talk." Two-photon decays are indicated by coincident events in which the sum-energy is equal to the transition energy.

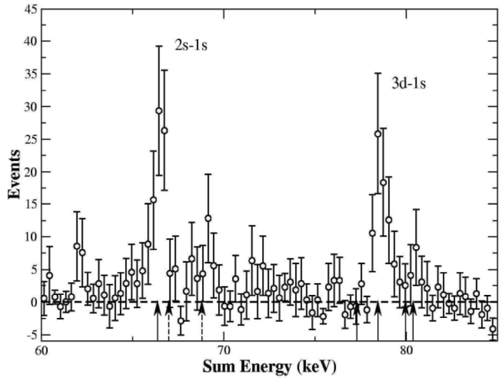


*FIG. 1. Sum energy spectrum for coincident events. Two peaks are seen at the 2s-1s and 3d-1s two-photon transition energies.*

## Results

For the initial experiment, we analyzed events in which the two photons shared the energy equally. Results were obtained for the 3d $\rightarrow$ 1s and 2s $\rightarrow$ 1s two-photon transitions (Fig. 1). We measured two-photon decay of single K-vacancies in gold atoms following photoionization with synchrotron radiation. Our results determined differential transition probabilities for the 2s $\rightarrow$ 1s, 3d $\rightarrow$ 1s, and 4sd $\rightarrow$ 1s two-photon decays for events in which the two photons share the transition energy equally and have opening angles near $\theta = \pi/2$. This is the highest-Z measurement of inner-shell, two-photon decay and the first to be done at a synchrotron light source.

Following the initial observation of two-photon decay in Au, the goal has been to measure the spectral shape of the two-photon continuum for this system. The drastically different continuum spectral shapes for the 2s $\rightarrow$ 1s, 3s $\rightarrow$ 1s, 3d $\rightarrow$ 1s, and 4s/d $\rightarrow$ 1s two-photon transitions will be studied. Improvements were made to the experimental setup, including better shielding from cross-talk and scattered x-ray background and improved statistics. An initial run has already been completed, and data analysis is in progress.

Although there have been no theoretical calculations for two-photon decay of inner-shell vacancies in Au, we compared our results with existing calculations of the differential cross sections for lower-Z neutral atoms and found that the cross section in the equal-energy-sharing case was lower than what would be expected from an extrapolation of the existing calculations to higher Z. Clearly, new calculations of two-photon decay in neutral atoms at higher Z are needed to compare with our measurements.

## Discussion

When the APS is used, all heavy atomic systems are accessible to structure investigations that use the two-photon coincidence technique. We intend to follow the Au work with measurements at the highest Z, and we plan to study two-photon decay of innershell vacancies in uranium. The required x-rays (about 120 keV) for the creation of the innershell vacancies are readily available from the wiggler beamline at BESSRC. The germanium detectors used in the Au measurement are also suitable for the proposed uranium measurement. The potential problem with cross-talk becomes more serious with the higher-energy photons, so additional shielding between the two detectors will be needed.

We will study the spectral shape of the photon energy distributions in uranium as a means of testing theory at very high Z. Of interest is the behavior of the atoms under the influence of the combined effects of relativistic effects and electron-electron interactions. Detailed measurements of the spectral distributions will also shed light on the question of the contribution of virtual electron-positron pairs, which should contribute to the two-photon emission process at the highest Z.

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