Double K-photoionization of Heavy Atoms

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INTRODUCTION

With the advent of modern synchrotron radiation sources providing intense, collimated beams of tunable monochromatic x-rays, there has been increased interest in the investigation of multielectron processes [1]. Beyond the importance of such processes in understanding electron-electron correlations, they have also been implicated in the production of satellite structures in extended x-ray absorption fine structure (EXAFS) and x-ray absorption near-edge structure (XANES) studies of materials [2,3]. The most basic multielectron process is the complete emptying of an atomic K shell in photoabsorption. The effect of electron-electron correlations on such processes has been found to be *constant* with increasing Z [4] and consequently is challenging to treat in high-Z systems where relativistic effects simultaneously become important [5]. In this work [6] we used a tunable synchrotron x-ray source and investigated that process by detecting double K-vacancy production in molybdenum using incident photon energies just below and nearly 10 keV above the double-K ionization threshold.

EXPERIMENTAL PROCEDURE

The experiment was performed on the 12BM bending magnet beamline. Photon beams of 40.2 and 50 keV were obtained using 3rd order reflections from a Si(111) double-crystal monochromator and then collimated with a set of slits to produce a 1×1 mm^2 beamspot at the target. The lower energy used (40.2 keV) is below the double-K ionization threshold of 40.654 keV. However, since the K binding energy is 20 keV, this photon energy is 200 eV above the 40-keV threshold for the background process of photabsorption on one atom followed by electron-impact ionization of another atom by the resulting photoelectron. At the higher energy (50 keV), both processes can contribute. Furthermore, this energy is close to that corresponding to the predicted maximum ($\sim$ 53 keV) in the double photoionization cross section for He-like Mo [7].

The target consisted of a 25-μg/cm^2 film of natural molybdenum deposited on a 5-μg/cm^2 carbon foil and oriented with the surface at an angle of 30° with respect to the incident beam. Two Si(Li) detectors faced each other and were normal to the beam, lying in the same plane as the beam polarization and the target normal. With this geometry, Compton and Rayleigh scattering were suppressed. The crystal of one detector subtended an angle of $\sim$300 msr while the other crystal subtended $\sim$200 msr for a combined angular efficiency of $\sim$4%. Each detector was covered with a 127-μ Kapton filter to suppress high energy photoelectrons produced by the energetic higher-order components of the beam while transmitting the $\sim$ 17–20 keV Mo $K_{\alpha,\beta}$ fluorescence with $>$ 99% probability.

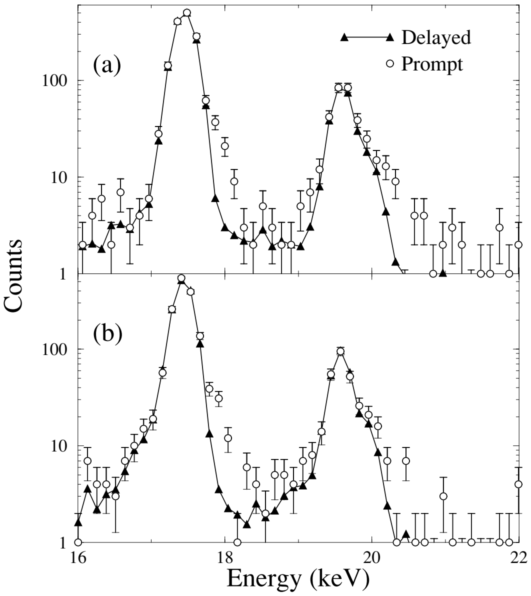

FIG. 1. Energy spectra in each detector for coincidences with $K_{\alpha,\beta}$ x-rays in the other detector. Spectrum for detector "1" is in (a) while that for "2" is in (b). Incident photon energy was 50 keV. Data points (open circles) are for prompt coincidences while the filled triangles connected with the solid line correspond to delayed coincidences.

RESULTS AND DISCUSSION

The region of the energy spectrum in each detector corresponding to K x-rays is shown in Fig. 1 for those x-rays detected in coincidence with K x-rays in the other detector. The open circles in each spectrum correspond to "prompt" coincidences. The "delayed" accidental coincidences are shown as filled triangles (connected by solid lines).

The main features in this region of the spectrum are the K_α and K_β normal diagram lines in Mo at 17.5 and 19.6 keV respectively. The prompt spectra show a clear excess on the high-energy sides of each peak corresponding to the hypersatellite transitions which, because of the reduced screening in the hollow atom, are systematically shifted up in energy. At the lower subthreshold energy, there was no discernible difference between the prompt and delayed spectra.

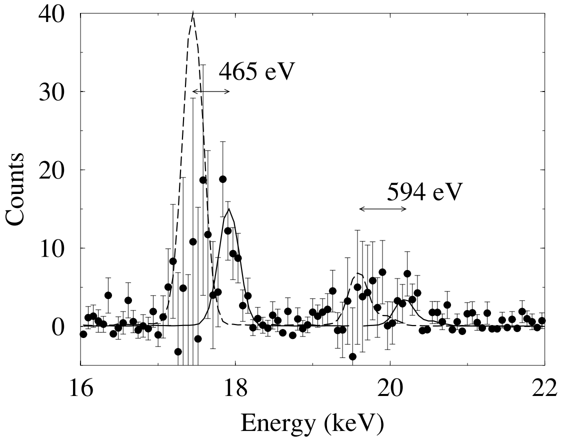

FIG. 2. Energy in detector "1" coincident with K x-rays in detector "2" for true coincidences (i.e. prompt-delayed from Fig. 1a). The delayed peaks have been arbitrarily scaled and superimposed (dashed lines). They have also been shifted up in energy by the theoretical hypersatellite shifts (solid lines) and rescaled to guide the eye.

The true coincidences were obtained by subtracting the prompt and delayed spectra and one such spectrum is shown in Fig. 2. Combining the measured yields with the fluorescence yields, detector efficiencies, and filter transmissions, we find the ratio of double/single K-ionization to be $R = 3.4(6) \times 10^{-4}$ for 50 keV incident photon energy.

At higher energies, the double photoionization of the K-shell has been discussed extensively in a 2-step picture where a fast photoelectron leaves with most of the available energy and then as the core-excited atom relaxes the second electron is shaken up/off leading to the concept of an energy-independent asymptotic limit of the ratio R [8]. We have performed such an estimate using relativistic wave functions and find this asymptotic ratio to be 3.18×10^{-5} in this case. Because of the relative isolation of K-shell electrons in heavy atoms, this result is not affected very much by the population of outer shell electrons and this value is in good agreement with the Z-scaling laws which have been given by several authors for He-like systems. Using the scaling law suggested by Forrey et al. [9] we find a value of 5.1×10^{-5} while the formula suggested by Kornberg and Miraglia [7] yields a value of 4.2×10^{-5}.

It is not surprising that all of these estimates are much smaller (by an order of magnitude) than the experimental result. As discussed earlier, the energies employed in this experiment were well below the asymptotic regime and in fact, using the scaled double photoionization cross sections [7], are very near the peak of that cross section. Hence it is crucial to treat the energy-dependent dynamical double ionization process in a rigorous fashion and this asymptotic value should only be viewed as a lower limit in this regime.

Recently, we were able to extend these measurements to the more interesting case of Ag (Z=47). Because of extensive studies of this system produced by the electron capture (EC) decay of ^{109}Cd, the shakeoff contribution is well-known experimentally for the single-electron final state produced in EC. Thus, our photoionization measurements will conclusively isolate the dynamic electron-electron scattering (TS1) term. Measurements were carried out at three different energies from the double K-ionization threshold to the region of the expected maximum in the cross-section and analysis is currently underway.

With the higher-energy beams which will become available at the BESSRC wiggler next year, we expect to extend these measurements to much heavier atoms such as Au (Z=79) and Pb (Z=82) which can provide a new probe of the effects of relativity on inner-shell wave functions and can be compared directly with the time-reversed double K-REC (Radiative Electron Capture) experiments. Some effort will also be devoted toward employing higher-resolution, large acceptance detectors (such as x-ray calorimeters) to improve the precision of these experiments.

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