Forward-backward asymmetries of atomic photoelectrons

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Introduction

The electric-dipole (E1) approximation to the photon-electron interaction is commonly used in theoretical treatments of photoionization and in interpreting experimental results. However, for experiments that measure or are sensitive to photoelectron angular distributions, the E1 approximation may be inadequate, because angular distributions are sensitive to interference between E1 transition amplitudes and amplitudes for higher-order interactions (e.g., the electric quadrupole, E2). The interference terms give rise to a forward-backward asymmetry (FBA) that is not included in theoretical treatments within the E1 approximation. Referring to the coordinate system in Figure 1, FBA means that angular distributions are asymmetric with respect to reflection in the plane normal to **k**. Angular distributions therefore depend on $\mathbf{k}\cdot\mathbf{p}$ and $\varepsilon\cdot\mathbf{p}$, while only $\varepsilon\cdot\mathbf{p}$ appears in the E1 approximation.

It was understood in early treatments of atomic photoionization that an FBA results unless the E1 approximation is adopted [1]. The early treatments qualitatively explained the FBAs observed in early photoemission experiments with high-energy x-rays [2] and more recent experiments using soft x-rays [3]. In the simplest modern treatments [4, 5], E1 and E2 transition amplitudes are calculated using nonrelativistic, independent-particle wavefunctions; the differential cross section can be expressed by Equation 1:

$$d\sigma/d\Omega = (\sigma/4\pi)[1 + (\beta/2)(3\cos^2\theta - 1) + (\delta + \gamma\cos^2\theta)\sin\theta\cos\varphi]. \quad (1)$$

In this expression, σ is the angle-integrated cross section, β is the pure-E1 anisotropy parameter, and δ and γ are parameters which characterize the FBA. Use of this expression assumes that the photon beam is 100% linearly polarized, but extension to arbitrary polarization is straightforward. In the special case of photoemission from an atomic s-subshell, $\beta = 2$, $\delta = 0$, and the FBA is characterized by γ (see Equation 2):

$$\gamma = 3k(Q_2/D_1)\cos(\delta_2 - \delta_1). \quad (2)$$

Here, k is the photon wave vector, D_1 and δ_1 are the dipole matrix element and phase shift, respectively, for the ns $\rightarrow$ εp transition, and Q_2 and δ_2 are the quadrupole matrix element and phase shift, respectively, for the ns $\rightarrow$ εd transition. These expressions indicate how angular distributions are parameterized and related to photoionization amplitudes.

Theoretical calculations predict interesting variations of γ and δ parameters with energy, atomic number, and subshell [4, 5]. To test these predictions, we made measurements on Ar 1s, Kr 2s, and Kr 2p photoelectrons using 2–5 keV x-rays on a bending-magnet beamline at the National Synchrotron Light Source [6]. The Ne 2s and 2p subshells have also been studied using soft x-rays at the Advanced Light Source [7]. Other recent experiments studied FBA effects in low-energy autoionization in atomic Cd [8] and in x-ray standing waves in surface-adsorbed atoms [9]. In the experiment described here, we measured FBAs for Ne 1s, Ar 1s, Xe $2p_{3/2}$ and Kr 1s photoelectrons using 4-19 keV x-rays on the BESSRC-CAT undulator beamline (12-ID). Preliminary results are presented for Kr 1s.

Methods

A diagram of the experimental system is shown in Figure 1. The x-ray beam passes through a Kr gas jet, and photoelectron spectra are recorded with a parallel-plate electron analyzer (PPA). The PPA is rotatable around the linear-polarization axis ε of the x-ray beam at fixed polar angle $\theta = \cos^{-1}(3^{-1/2}) \approx 54.7°$. This is the "magic angle," which (in the limit of a 100% linearly-polarized photon beam) eliminates terms in the photoelectron angular distribution involving the pure-E1 anisotropy parameter β. Hence, a cone of azimuthal angles ϕ is selected which emphasizes the dependence on γ and δ (Equation 3):

$$I(\phi) = 1 + (2/3)^{1/2}(\delta + \gamma/3)\cos\phi. \quad (3)$$

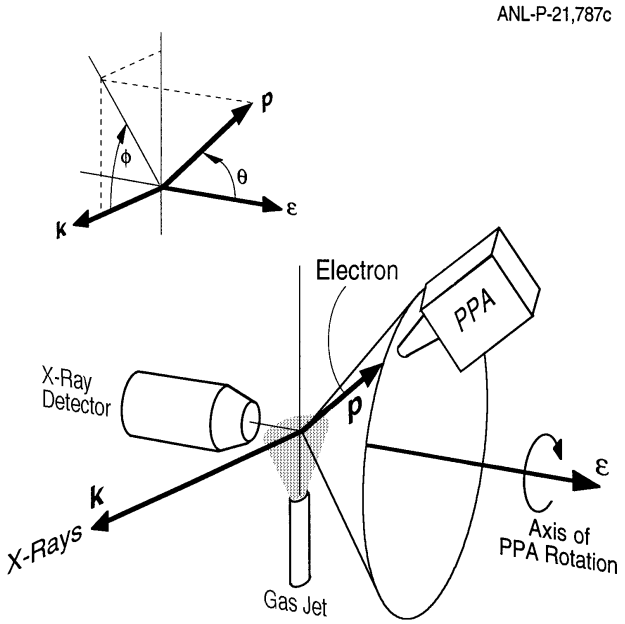

Figure 1: Diagram of system used to measure forward-backward photoelectron asymmetries. The parallel-plate electron analyzer (PPA) is rotatable about the linear-polarization vector ε of the x-ray beam. The photoelectron momentum p is measured relative to ε and the propagation

vector k. The x-ray detector records scattered and fluorescent x-rays for normalization of the photoelectron intensities. The parameter γ (for s-subshells) or $\delta + \gamma/3$ (for other subshells) can be determined from the variation of photoelectron intensity vs. ϕ. An x-ray detector positioned opposite the PPA records scattered and fluorescent x-rays from the gas jet for use in normalizing the photoelectron intensities. Small instrumental asymmetries are determined by measurements on Auger electrons. Except near threshold, it can usually be assumed that the two-step model of Auger emission is valid, and Auger electrons have no FBA in this model [10].

Results and Discussion

Figure 2 shows a Kr 1s photoelectron and Auger-electron spectrum. Data were taken from near threshold to ≈ 5000 eV kinetic energy. Based on data analysis methods developed for our first experiments [6] and general considerations of the dependence of angular distributions on photon beam polarization [11], we determine γ parameters from electron intensities measured at four angles, ϕ = 45°, 135°, 225°, and 315°. These angles are "magic" with respect to both ε and **k**, and the intensities are independent of the degree of polarization. Figure 3 shows relative intensities measured at the four angles for selected kinetic energies of Kr 1s photoelectrons and for Auger electrons.

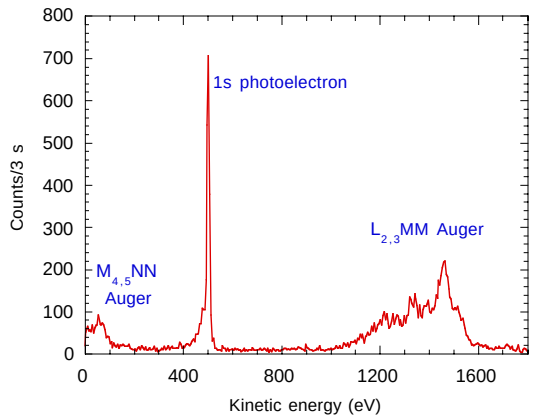

Figure 2: Kr 1s photoelectron and Auger electron spectrum recorded at 14.826 keV, which is 500 eV above threshold.

The Auger electron intensities should be isotropic, but show a small up-down instrumental asymmetry which is accounted for in deriving γ values from the photoelectron intensities. The Kr 1s intensities at 32 eV kinetic energy are larger at 135° and 225° (backward) than at 45° and 315° (forward), indicating $\gamma < 0$. At 1217 eV, the Kr 1s intensities are approximately the same, indicating $\gamma \approx 0$. At 5017 eV, the forward intensities are larger than the backward intensities, indicating $\gamma > 0$. These results are consistent with the calculated γ values [4]. The Kr 1s data, along with data on Ne 1s, Ar 1s, and Xe $2p_{3/2}$, are presently being analyzed to determine sets of γ parameters vs. kinetic energy for comparison with theory. We are particularly interested in the variations predicted near threshold [4], as well as the significance of electric-octupole (E3) amplitudes at high energy [12].

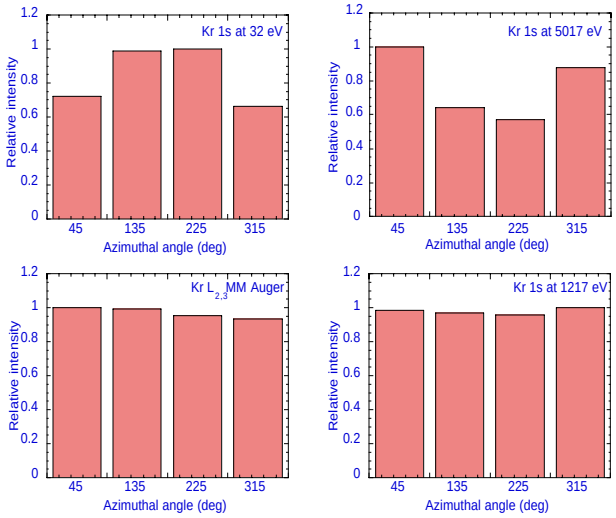

Figure 3: Angular dependence of the relative intensities of Kr 1s photoelectrons measured at 32 eV, 1217 eV, and 5017 eV kinetic energies and for Kr $L_{2,3}$-MM Auger electrons.

Acknowledgments

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