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Photoelectron and Auger electron asymmetries: Alignment of $\text{Xe}^+ 2D_{5/2}$ by photoionization

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Angular distributions of photoelectrons from the $\text{Xe } 4d$ subshell, and $N_{4,5}OO$ Auger electrons, have been measured using synchrotron radiation. The $4d$ asymmetry parameter exhibits strong oscillations with energy, in agreement with several theoretical calculations. The Auger electrons show large asymmetries due to alignment of Xe^+ by photoionization.

Flügge, Mehlhorn, and Schmidt¹ showed that photoionization of an atomic subshell with $j > \frac{1}{2}$ can produce an aligned ionic state, followed by an anisotropy in the angular distributions of Auger electrons. No experimental observation of this effect has been reported, but Berezhko *et al.*² have calculated a strong, energy-dependent alignment of $\text{Xe}^+ 2D_{5/2}$ following photoionization of the $\text{Xe } 4d_{5/2}$ subshell. The energy dependence is predicted to arise from a centrifugal barrier and a Cooper minimum in the $4d \rightarrow \epsilon f$ photoionization channel. The same factors were predicted by Manson³ to produce strong oscillations in the photoelectron asymmetry $\beta(\epsilon)$ as the photon energy is varied through the range 0–150 eV above threshold. These oscillations had also not been confirmed experimentally, although Torop *et al.*⁴ found that $\beta(\epsilon)$ increases with energy in the range $\epsilon = 20$ –100 eV, as predicted by Manson. Here we report the first results of a study based on synchrotron radiation in which we have measured the asymmetry parameters $\beta(\epsilon)$ for both photoelectrons and Auger electrons following photoionization of the $\text{Xe } 4d$ shell, over a photon energy range $h\nu = 70$ –330 eV. Good agreement was obtained with theoretical predictions.

Angular momentum and parity conservation restrict the form of the angular distribution.⁵ For plane-polarized incident radiation, the differential cross section for any photofragment produced by electric dipole excitation of a randomly oriented target is given by

$$\frac{d\sigma(\epsilon)}{d\Omega} = \frac{\sigma(\epsilon)}{4\pi} [1 + \beta(\epsilon)P_2(\cos\theta)] \quad (1)$$

where $P_2(\cos\theta) = \frac{1}{2}(3\cos^2\theta - 1)$, θ is the angle between the fragment propagation direction and the photon polarization vector, ϵ is the fragment kinetic energy, and the asymmetry parameter is restricted to values $-1 \leq \beta(\epsilon) \leq 2$. The $\beta(\epsilon)$ parameter is the object of our experimental measurement. Its functional form depends on the transition matrix elements and continuum-wave phases, which are in turn determined by electronic structure and photoionization dynamics.

As described previously,⁶ the time structure of the synchrotron radiation at the Stanford Synchrotron Radiation Laboratory was used to record time-of-flight (TOF) spectra of the ejected electrons. Two TOF detectors were operated simultaneously, at $\theta = 0^\circ$ and $\theta = 54.7^\circ$, the “magic angle” where $P_2(\cos\theta) = 0$. Equation (1) shows that $\beta(\epsilon)$ can be determined by measurement of the relative intensities of electrons ejected at two angles. This double-angle-TOF technique has certain advantages; notably a high collection efficiency, because all of the ejected electron peaks, at both angles, are recorded simultaneously, giving an enhanced signal-to-noise ratio, and minimization of systematic errors. Figure 1 shows the TOF spectra of Xe obtained at 70.9-eV photon energy. All spectra were recorded with the monochromator operated at a fixed bandpass $\Delta\lambda \approx 5 \text{ \AA}$ full width at half maximum (FWHM).

The ratio of collection efficiencies of the two detectors was determined as a function of electron energy by measurement of Ne $2p$ photoelectrons, for which $\beta(\epsilon)$ is accurately known.⁷ We estimate the incident

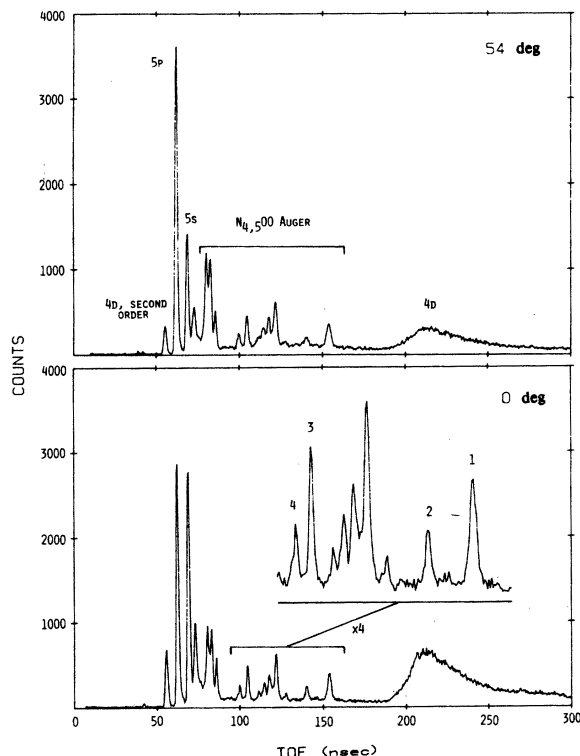


FIG. 1. Time-of-flight electron spectra from photoionization of xenon by 70.9-eV photons, at $\theta = 0^\circ$ and $\theta = 54.7^\circ$, relative to the polarization direction. Photoelectron peaks are present from both first- and second-order light (at $h\nu = 141.8$ eV), bracketing the $N_{4,500}$ Auger peaks.

radiation to be 98% plane polarized, based on the very high asymmetry ($\beta = 1.94$) measured for $Ag 4s \rightarrow \epsilon p$ photoelectrons with a rotatable analyzer.⁸ The β values reported here were corrected, typically by <0.05 β unit, for this and geometric effects. We estimate residual systematic errors as ≤ 0.05 β unit.

Photoelectrons from the spin-orbit split $4d_{5/2}$ and $4d_{3/2}$ subshells were not resolved in the present measurements. Figure 2 shows the measured β values for the Xe $4d$ subshell along with the measurements of Torop *et al.*,⁴ the theoretical results of Kennedy and Manson⁹ using Hartree-Fock wave functions, the relativistic Dirac-Fock (DF) calculations of Ong and Manson,¹⁰ and the many-electron relativistic random-phase approximation (RRPA) theory of Johnson and Huang.¹¹ Our main result is to confirm experimentally the oscillatory nature of $\beta(\epsilon)$, which is primarily a nonrelativistic, one-electron effect.^{3,9} The sharp drop near threshold is due to rapid variation of the Coulomb phase shifts, while the oscillation across $10 \leq \epsilon \leq 100$ eV is produced by variation of the non-Coulombic phase of the f wave due to a centrifugal barrier¹² in the scattering potential. The final oscillation in $\beta(\epsilon)$ occurs about the Cooper minimum where the f -wave radial matrix element

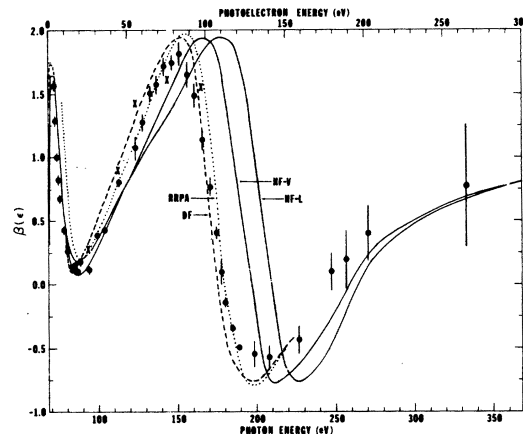
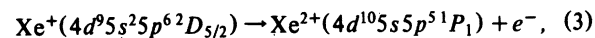
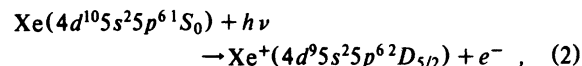


FIG. 2. Oscillatory variation of the xenon $4d$ photoelectron asymmetry with energy, compared with the data (\times 's) of Torop *et al.* (Ref. 4) and with theory: Hartree-Fock length (HF-L) and velocity (HF-V) calculations, Ref. 9; Dirac-Fock (DF), Ref. 10; relativistic random-phase approximation (RRPA), Ref. 11. The spin-orbit splitting was unresolved in these measurements.

changes sign. We note that the DF and RRPA theories predict the position of the Cooper minimum more accurately.

Either of two approaches can be used to show that Auger electron angular distributions obey Eq. (1). In a very general way, Auger electrons can be regarded as "photofragments." We shall, however, follow Berezhko *et al.*² in using the more heuristic two-step model.

In the two-step model of the Auger process the decay of the excited ion state is considered independent of the excitation process. In the present example these steps are described by the processes



where the Xe^{2+} final state, assigned from the work of Werme *et al.*,¹³ is Auger peak number 3 in Fig. 1.

The most general formulation of angular correlations in scattering experiments is obtained using the density matrix and statistical tensor algebra.¹⁴ The statistical tensors describe the angular symmetry properties of the atomic system at each stage of a scattering process. In particular, the statistical tensors $\rho_{KQ}(J, J)$, with $J = 5/2$, describe the distribution of magnetic substates $|JM\rangle$ for the ensemble of $Xe^{+2}D_{5/2}$ ions produced in the photoionization step, Eq. (2). The density matrix (ρ) for randomly oriented target atoms is spherically symmetric, so that electric dipole photoexcitation⁵ introduces statistical tensors of rank $K \leq 2$.

Plane-polarized incident radiation produces an axially symmetric¹⁵ angular-momentum distribution with

respect to a quantization axis parallel to the polarization vector. All statistical tensors vanish except $\rho_{00}(J,J)$ and $\rho_{20}(J,J)$. Berezhko *et al.*² employ the relative alignment tensor

$$A_{20}(J) \equiv \rho_{20}(J,J)/\rho_{00}(J,J) . \quad (4)$$

In the present case of $\text{Xe}^{+2}D_{5/2}$ the alignment can be expressed as

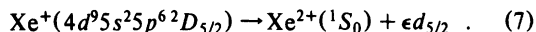
$$A_{20}(5/2) = \frac{1}{\sqrt{14}} \left[\frac{5\sigma(|5/2|) - \sigma(|3/2|) - 4\sigma(|1/2|)}{\sigma(|5/2|) + \sigma(|3/2|) + \sigma(|1/2|)} \right] , \quad (5)$$

where $\sigma(|M|)$ is the partial photoionization cross section for magnetic substate M or $-M$. Equation (5) shows that an alignment results when the magnetic sublevels are unequally populated.

The asymmetry parameter for the Auger electron is proportional² to the alignment tensor

$$\beta(h\nu) = \alpha A_{20}(h\nu) , \quad (6)$$

where α depends on transition matrix elements of the Coulomb operator and on the phases for the allowed¹⁶ continuum waves, which are $\epsilon p_{3/2}$, $\epsilon f_{5/2}$, and $\epsilon f_{7/2}$ for the Auger transition of Eq. (3). However, for the case of a Xe^{2+} final state having total angular momentum $J_f=0$, there is a single continuum wave, and Cleff and Mehlhorn¹⁶ have shown that the Auger angular distribution is then independent of the Coulomb matrix element and continuum phase shift. The coefficient α in Eq. (6) is then given by angular momentum coupling coefficients.¹⁷ Figure 3 shows the β values measured for the Auger transition of Eq. (3) along with a theoretical curve for an Auger transition of the type



The theoretical curve was obtained using the $\text{Xe}^{+2}D_{5/2}$ alignment tensor calculated² using Hartree-Fock wave functions. The data points are not expected to fall on this curve, because it is based on α for Eq. (7). Since the α parameters are independent of photon energy, however, the curve appropriate for these data should differ only by a constant scale factor. Berezhko *et al.*² have shown that the alignment is strong, so that the Auger asymmetry is large, when the $4d \rightarrow \epsilon f$ photoemission transition strength is rela-

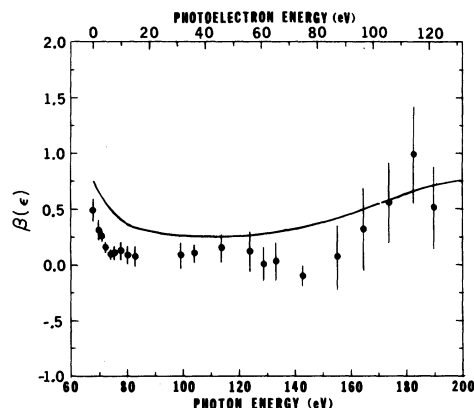


FIG. 3. Auger electron asymmetry, for the Xe NOO transition $N_5O_1O_{23}({}^1P_1)$. Theoretical curve is for the transition $N_5OO({}^1S_0)$, to show the expected energy dependence predicted by Berezhko *et al.* (Ref. 2). The energy of the preceding photoelectron is also shown.

tively small. Thus, the large Auger asymmetry observed near the $4d$ ionization threshold is due to suppression of the f -wave channel at photoelectron energies below the centrifugal barrier, while the large Auger asymmetry at high photon energy occurs near the f -wave Cooper minimum. Similar energy dependences of $\beta(h\nu)$ were observed for the four $N_{4,5}OO$ Auger peaks indicated in Fig. 1, as expected.

In summary, the first results described here have established the oscillatory behavior of $\beta(\epsilon)$ for the Xe $4d$ subshell. Alignment of the Xe^+ ion was confirmed by observation of Auger asymmetries following photoionization. Further results will be published in a more detailed report later.

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