Spin-polarized photoelectrons excited by circularly polarized radiation from a nonmagnetic solid

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We report a spin analysis of core-level photoelectrons excited by circularly polarized x rays from a nonmagnetic solid. In a combined experimental and theoretical study, we show that the spin-orbit-split W $4f_{7/2}$ and $4f_{5/2}$ photoemission lines from W(110) exhibit high spin polarizations of opposite sign that vary with energy and emission direction. These results suggest the study of the magnetic structure of nonmagnetic/ferromagnetic interfaces formed on high-atomic-number substrates by spin-polarized photoelectron diffraction.

Photoelectrons from *unpolarized* atoms can be highly spin polarized when ejected by circularly polarized (CP) light, due to the Fano effect:[1(a)] although the dipole operator does not act upon the electron spin explicitly, the photon angular momentum is partially transferred to the photoelectron spin by the spin-orbit (SO) interaction. Shortly after its prediction in 1969, this effect was observed in the *valence levels* of alkali-metal[1(b)] and rare-gas atoms[2] in gaseous and solid-state phases, and today it is well known as the basic mechanism behind the GaAs source for spin-polarized electrons.[3] In particular, photoelectrons excited by CP light from *p*, *d*, and *f* shells can acquire substantial spin polarization over a wide photon energy range.[4] Up to now, the spin analysis of photoelectrons from unpolarized targets has mostly served fundamental interests of (i) understanding photoemission (PE) dynamics through quantum mechanically ''complete'' experiments[5,6] and (ii) characterizing the symmetry of valence bands in nonmagnetic solids.[7]

However, spin-polarized photoelectrons from *core levels* also have an important potential application as internal sources of polarized electrons in spin-polarized photoelectron diffraction[8] (SPPD) and spin-dependent inelastic scattering experiments.[9] For example, it has been demonstrated that SPPD is capable of probing short-range magnetic order around a photoemitting atom when it is a constituent of the magnetic crystal lattice[8(a),8(b)] and it has also been suggested that direct imaging of magnetic order via holographic methods should be possible.[8(c)] Despite its potential, SPPD has so far been applied experimentally to only two cases of antiferromagnetic manganese compounds.[8(a),8(b)] Yet the performance of such experiments is simple, requiring only two core-level photoelectron peaks of significantly different spin polarization that are reasonably close in energy. Low kinetic energies of 100 eV or less are also required to yield large enough exchange scattering contributions to the diffraction patterns.[8(a)]

In the present paper, we report a spin analysis of core-level photoelectrons excited by circularly polarized soft x rays from a nonmagnetic solid, and show that all the conditions for performing SPPD and other magnetic scattering experiments can thus be met. The intense and well-resolved spin-orbit split W $4f_{7/2}$ and $4f_{5/2}$ photoemission lines are found to exhibit high spin polarizations of opposite sign over a wide photon energy region around the maximum of the $4f$ photoemission cross section. In addition, we compare the present experimental results on the $4f$ PE lines with calculations at both the free-atom level and with a full multiple-scattering treatment of photoelectron diffraction effects near the solid surface. We note here that Roth *et al.*[10] have recently made an observation of spin polarization in the Cu

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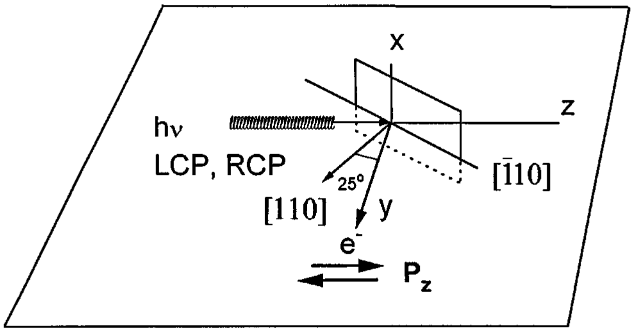

FIG. 1. Experimental geometry. Circularly polarized light is incident on the W(110) single crystal along the *z* axis. Electrons are detected normal to the light propagation direction along the *y* axis. The electron spin detector is sensitive to the in-plane polarization components P_z and P_y.

2*p* and 3*p* PE lines, as excited by *linearly* polarized (LP) light. However, this case does not provide as beneficial conditions for spin-dependent scattering experiments since the Cu 2*p* lines show only about 15% polarization and require high-energy photons to excite them, while the Cu 3*p* lines cannot be resolved in energy due to their intrinsic width and small spin-orbit splitting.

The PE experiments were performed with circularly polarized soft x rays from the AT&T Bell Laboratories Dragon Beamline at the National Synchrotron Light Source.[11] By collecting radiation between 0.43 and 0.87 mrad above or below the storage ring plane we achieved a degree of circular polarization $S_3 \approx 86\%$ in the 80- to 250-eV photon energy interval used here and the residual linear polarization in the reaction plane is $S_1 \approx 51\%$, with the values weakly depending on the photon energy. S_1, S_2, and S_3 denote the usual Stokes parameters defining the experimental elliptically polarized light. The sample was a W(110) single crystal prepared by a standard oxidation-annealing procedure.[12] Chemical cleanliness was checked by C 1*s* and O 1*s* PE and found to be below our detection limit (a few percent of a monolayer). The experimental geometry is shown in Fig. 1. Photons were incident along the *z* axis and photoelectrons were detected along the *y* axis normal to the light propagation direction. The normal of the single-crystal sample was oriented 65° from the light incidence, such that the [110] direction coincided to within ±10° with the *y*-*z* interaction plane. Spin-resolved photoemission was accomplished using a hemispherical analyzer with 0.6 eV energy resolution and an acceptance cone of ±3°, backed by a low-energy spin detector.[13] The spin detector was oriented such that both components of the spin-polarization vector within the interaction plane, P_z and P_y, could be measured simultaneously. The experimental geometry was chosen to allow a more straightforward comparison with theory. Indeed, since the dominant photoelectron paths through the crystal are along the {011} mirror plane, P_z should not be significantly affected by final-state effects associated with traveling through the surface barrier.[14] From symmetry considerations, the component perpendicular to the light propagation, P_y, is also expected to vanish identically for free atoms[4] and solids.[15] This component was indeed found to vanish experimentally, serving as a useful check to rule out possible apparatus asymmetries.

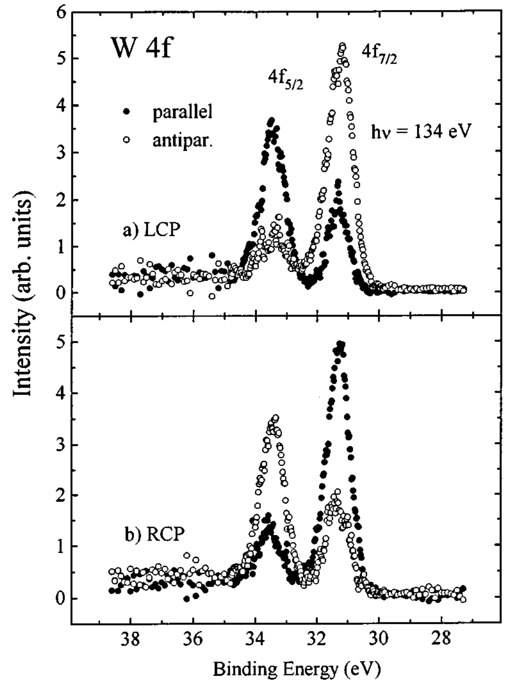

FIG. 2. Spin-resolved PE spectra of the W 4*f* spin-orbit doublet excited at $h\nu = 134$ eV with (a) left circularly polarized light and (b) right circularly polarized light. Solid and open symbols give the intensities for electrons with spin parallel and antiparallel to the light propagation direction (positive *z* direction in Fig. 1).

Figure 2 displays spin-resolved PE spectra of the W 4*f* spin-orbit doublet excited with left-hand (LCP) and right-hand (RCP) circularly polarized light at 134 eV. The $4f_{7/2}$ and $4f_{5/2}$ PE lines exhibit high spin polarizations P_z of opposite sign along the light propagation axis. For LCP [Fig. 2(a)] the spin polarization is about +55% for the $f_{5/2}$ line and about −40% for the $f_{7/2}$ line; upon reversal of the light helicity [Fig. 2(b)] the polarization of both lines changes sign but remains constant in magnitude. Furthermore, when using linearly polarized light with the electrical-field vector in the interaction plane along the *y* axis (cf. Fig. 1), we find that the spectra (not shown here) yield vanishing spin polarizations for both lines. These experimental findings are in agreement with the nonrelativistic theory of spin-polarized PE from isolated atoms developed by Cherepkov.[4,5] For CP excitation, this theory predicts opposite polarization along the *z* axis and, in particular, a polarization ratio reciprocal to the statistical weights $(2J+1)$ of the two lines, i.e., $P_{5/2}:P_{7/2} = -1.33:1$; for linearly polarized light, the in-plane spin-polarization components (P_z and P_y) are expected to vanish. We show below that, for CP excitation, the statistical ratio is predicted to hold also for the solid-state case where the photoelectrons are in principle subject to scattering from the nonmagnetic W atoms. The spin-resolved spectra in Fig. 2 yield, after subtracting a linear background in the peak region, a polarization ratio of $-(1.4 \pm 0.1)$ that is very close to the expected statistical value. In contrast to the spin polarization of the Cu 3*p* PE lines observed previously with LP excitation,[10] the W 4*f* lines are well resolved, show high spin polarizations of opposite sign when excited with CP light and, in addition, reside on a low background. The peak polarizations can thus be obtained easily, with little uncer-

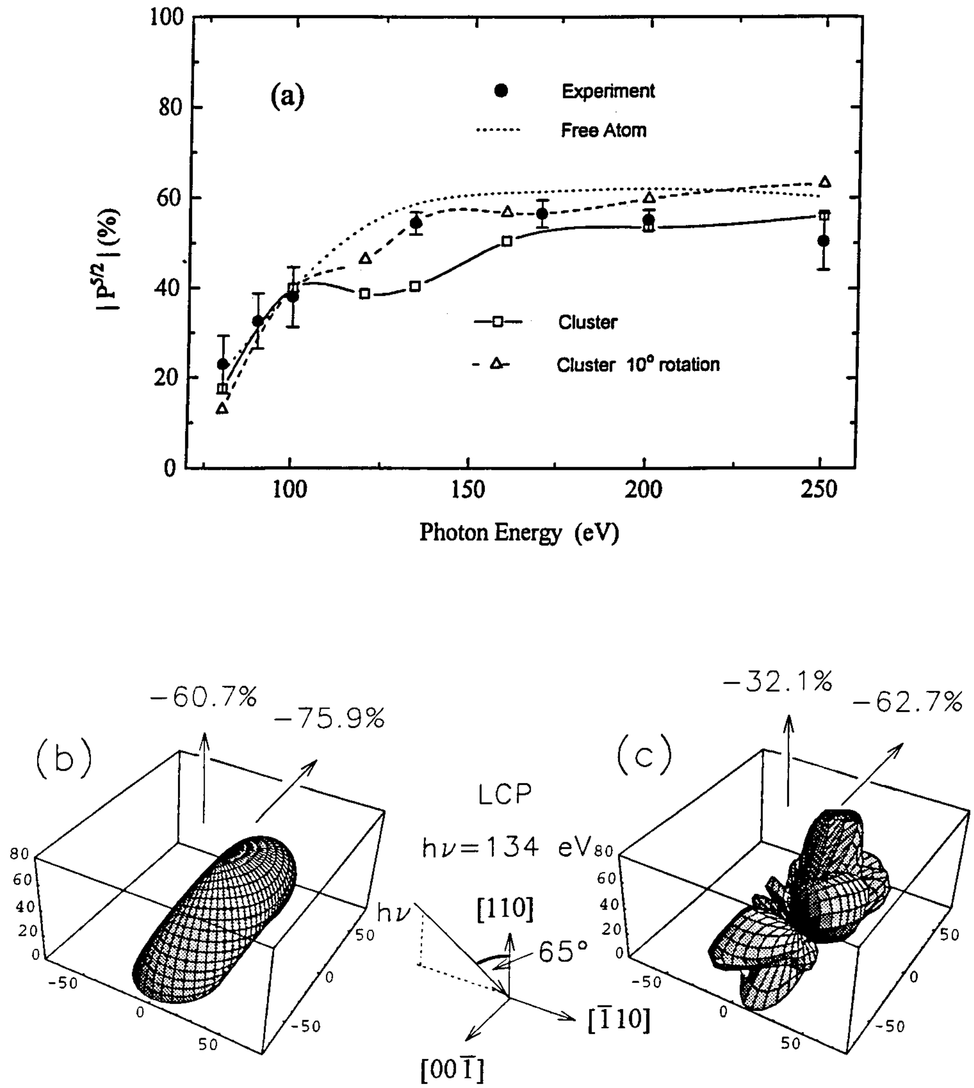

FIG. 3. (a) Energy dependence of the W $4f_{5/2}$ spin polarization. The experimental data (solid circles) are compared to free-atom theory based on Eq. (1) (dotted curve), and large-cluster multiple-scattering PD calculations for the geometry of Fig. 1 (solid curve and open squares), and with the crystal rotated 10° about the surface normal (dashed curve and open triangles). (b) Three-dimensional representation of the magnitude of the W $4f_{5/2}$ polarization with LCP excitation at 134 eV as a function of emission direction for a free atom. (c) As (b), but for a five-atom cluster of W with multiple scattering PD effects included.

tainty due to line-shape analyses.

We have further explored the energy dependence of the W $4f$ spin polarization in the low kinetic energy region; from 48 to 218 eV (photon energies between $h\nu=80$ and 250 eV). The $4f_{5/2}$ spin polarization in particular was determined as the fraction 4/7 of the *polarization difference* between both lines, assuming the validity of the statistical ratio of −4:3 given above; this scaled difference is insensitive to drifts in sensitivity of the spin analyzer. The experimental data are presented in Fig. 3(a). They show a ''plateau'' of high polarization (55%) above $h\nu=150$ eV in the region of the W $4f$ cross-section maximum.[16] Towards lower energies the polarization reduces monotonically; however, even at $h\nu=80$ eV it is still substantial (20%).

We now compare the energy-dependent experimental data in Fig. 3(a) with theoretical predictions. From the free-atom viewpoint, a $4f$ level is particularly suitable for a comparison with theory since there are no Cooper minima in the PE cross section and the photoelectron spin polarization originates from the core-level spin-orbit splitting. According to free-atom theory,[4] the spin polarization is then governed by the radial dipole matrix elements $R_{l\pm1}$ for excitation into $l\pm1$ (g or d) continuum states and the phase shifts $\delta_{l\pm1}$ that control the interference between the outgoing $l+1$ and $l-1$ waves. In particular, the polarization P_z^J of a given J manifold was calculated using[5]

$$P_z^J=\frac{S_3(A^J+\frac{1}{2}\gamma^J)+S_2\eta^J}{1+\frac{1}{4}\beta(1+3S_1)}, \quad (1)$$

by using tabulated values for the radial matrix elements and phases[16] and the known Stokes parameters S_1, S_2, S_3 for the incident light. β is the normal asymmetry parameter and the spin-polarization parameters A^J, γ^J, η^J are adopted from Ref. 4. As shown in Fig. 3(a), both characteristic features of the experimental data, i.e., the high-energy ''plateau'' and the reduction towards lower energies, are reflected in the free-atom calculation, although theory gives slightly larger values at high energies. The increase from low energies followed by a plateau is found by calculation to be a general property of $4f$ PE lines. For example, applying Eq. (1) to Au, we predict a significant reduction of the $4f$ polarization below $h\nu=200$ eV and a zero crossing at about 135 eV (equivalent in kinetic energy to $h\nu=147$ and 82 eV for W $4f$). To simulate possible solid-state effects on the $4f$ polarization we have also carried out fully converged multiple-scattering photoelectron diffraction (PD) calculations, using the Rehr-Albers formalism.[17] The top four layers of the W(110) surface, which should dominate the PE spectra in the energy range studied, were represented by a 145-atom cluster with photoelectron emitters placed on each layer. The inelastic attenuation length was varied from ~3 Å at $h\nu=80$ eV to ~6 Å at $h\nu=250$ eV and the inner potential was set to 13.75 eV, values consistent with recent experimental findings.[18] The same radial matrix elements and phase shifts as for the free-atom calculation were used. Spin flips due to spin-orbit scattering have been neglected and this is expected to be a good approximation in this energy range.

For the crystal orientation indicated in Fig. 1, the PD calculation yields the same overall trend with energy as the experiment and gives slightly lower polarization values than in the free-atom case. It also predicts a polarization ''dip'' between 120 and 150 eV photon energy which is not found experimentally. As a possible reason for this discrepancy, we have checked the influence of a slight azimuthal rotation of the crystal. The dashed curve in Fig. 3(a) represents the results for a cluster rotated by 10° about the surface normal with respect to the geometry in Fig. 1; it now reproduces the observed values excellently in the medium energy range.

The two cluster calculations taken together show that, even in a nonmagnetic material, photoelectron diffraction effects can cause the polarization to change significantly with direction, relative to the predictions of the free-atom model. Such PD effects on spin polarization are of obvious interest for future experiments. To provide a first indication of their overall form, we show in Figs. 3(b) and 3(c) calculated three-dimensional polarization plots for the W $4f_{5/2}$ peak excited at LCP at 134 eV. In the free-atom case, Fig. 3(b), the polarization as a function of direction is ''donut''-shaped with a maximum magnitude of −60.7% along directions perpendicular to the light incidence. In Fig. 3(c) the same polarization is shown, but for a five-atom cluster with a second-layer emitter below four surface scatterers. With PD effects thus included, dramatic variations in the directional dependence of the polarization are found. These variations are due to the different ''dumbbell'' and ''donut'' free-atom emission patterns of the two spins, as can be derived from Eq. (1). However, we still find that, even with diffraction included and regardless of direction, the ratio of the polarizations of the $4f_{7/2}$ and $4f_{5/2}$ peaks is −4:3, in agreement with free-atom theory.

In summary, we have shown that excitation of an energy-resolved spin-orbit core level in a nonmagnetic solid by circularly polarized x rays yields highly spin-polarized photoelectrons with net polarizations of opposite sign along the light propagation axis. In the energy region of the maximum W $4f$ photoemission cross section, spin polarizations of above 50% are obtained even with incompletely polarized light, and at low energies they are still substantial. In view of the frequent use of W and other crystals with high atomic number (Pt, Au, . . .) as substrates for ferromagnetic rare-earth and transition-metal overlayers, the present observations suggest the use of CP-excited SPPD and spin-dependent inelastic scattering for studying magnetic order near magnetic/nonmagnetic interfaces, with such experiments being sensitive to the magnetic near neighbors of the photoemitting substrate atoms. In potential experiments making use of this effect, the photon energy could either be tuned to the cross-section maxima for maximum intensity, or reduced so as to achieve higher exchange-scattering effects at lower kinetic energies.

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