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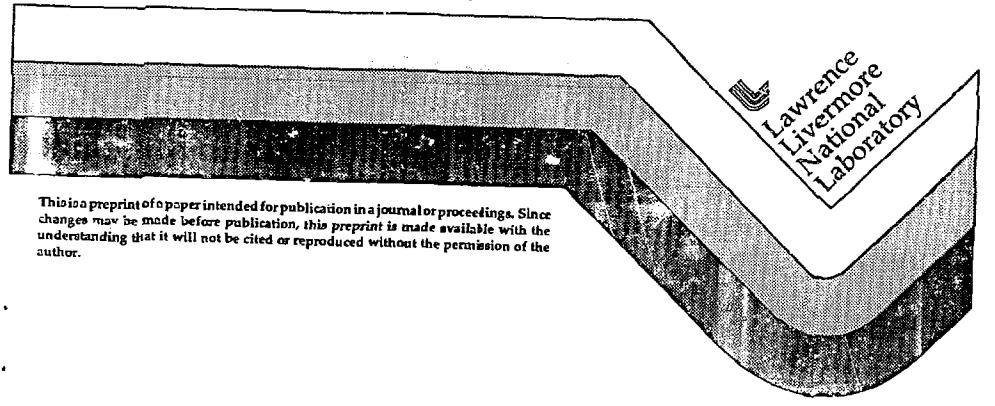

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Magnetic X-Ray Circular Dichroism in Spin-Polarized Photoelectron Diffraction

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ABSTRACT

The first structural determination with spin-polarized, energy-dependent photoelectron diffraction using circularly-polarized x-rays is reported for Fe films on Cu(001). Circularly-polarized x-rays produce spin-polarized photoelectrons from the Fe 2p doublet, and intensity asymmetries in the $2p_{3/2}$ level are observed. Fully spin-specific multiple scattering calculations reproduce the experimentally-determined energy and angular dependences. A new analytical procedure which focuses upon intensity variations due to spin-dependent diffraction is introduced. A sensitivity to local geometric and magnetic structure is demonstrated.

Keywords: MXCD, Photoelectron Diffraction, Magnetic Structure

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MAIN TEXT

Determining the nanoscale structure-property relationships of a magnetic surface, ultrathin film or interface remains a formidable but potentially extraordinarily rewarding task. For example, as studies of giant magneto resistive[1] and spin valve[2] systems have progressed, it has become clear that spin-specific interfacial scattering is a critical but poorly understood event. To properly address such problems, a probe which combines elemental specificity with a sensitivity to local order, both geometric and magnetic, is essential. Spin-polarized photoelectron diffraction (SPPD) is potentially an ideal candidate for studying nanoscale magnetic systems, with the above attributes plus the promise of an ultimate extension to element-and spin-specific imaging[3]. However, while the potential for SPPD is very high, particularly with the advent of third generation synchrotron radiation facilities such as the Advanced Light Source, until now a definitive demonstration has been lacking. This is primarily because of the relatively inefficient nature of spin-detectors and the relatively weak nature of magnetic scattering effects. Here we present a prototype study using magnetic x-ray circular dichroism[4], to permit a direct and unambiguous control of electron polarization without the necessity of the low-efficiency spin-detectors. This is coupled with a fully spin-specific, multiple scattering computational analysis. While we remained hampered by the limitations of existing sources of circularly-polarized x-rays and hence are constrained in terms of signal to noise, this is the first definitive demonstration of SPPD.

In some respects, this SPPD investigation is an independent verification and extension of the pioneering studies of G. Schütz, et al.[5], who used spin-polarized EXAFS to probe bulk magnetic systems. Consistent with non-spin PD and EXAFS studies, the SPPD shows a larger effect: the SPPD oscillations are on the order of 2% while the Gd metal SPEXAFS oscillations are $\leq 1/3$ %. Additionally, SPPD has the advantage of both energy and angular variations, which is essential to the extension to photoelectron diffraction imaging[3]. Both this work and the ground-breaking studies of Schütz, et al.[5] are predicated upon control of spin-polarization of ejected electrons via excitation with circularly-polarized x-rays. In a simplistic picture, 2p photoemission total cross sections from ferromagnetic materials will exhibit a polarized distribution of 62.5% (37.5%) minority spin electrons from the $2p_{3/2}$ and 25% (75%) minority spin electrons from the $2p_{1/2}$, when excited with right (left) circularly-polarized radiation that is collinear with the magnetic axis of the sample. These adjustably spin-polarized electrons can then scatter off of nearby neighbors, producing a sensitivity to both local geometric and magnetic ordering. (Although we have chosen to use a ferromagnetic system as a test case, these same selection rules will apply in general, e.g., to paramagnetic and anti-ferromagnetic ordering, and the multiple scattering analysis should be sensitive to differences in the local order of each

structure.) To avoid extraneous effects and to allow internal cross-checking of data, measurements were performed only in mirror planes, where only the relative alignment of the photon helicity and magnetization is crucial. Thus reversing the absolute value of these quantities, while maintaining the same relative spin orientation, serves as a convenient but absolutely essential consistency test to determine if the observed asymmetry is due to spin-dependent diffraction. It is the absence of such polarization control or electron spin-detection, plus the ill-defined nature of the intrinsic 3s polarization, that has hampered previous attempts at SPPD using the 3s level of 3d transition metals[6-8].

Because the calculated spin-dependent intensities are for an entire $2p_{3/2}$ or $2p_{1/2}$ manifold, the best comparison with experiment uses an integrated spin-dependent intensity asymmetry (or integrated SIA) defined as follows:

$$\text{Integrated SIA}(h\nu) = \frac{\left[\frac{I^{+}_{3/2}(h\nu)}{I^{+}_{1/2}(h\nu)} - \frac{I^{-}_{3/2}(h\nu)}{I^{-}_{1/2}(h\nu)}\right]}{\left[\frac{I^{+}_{3/2}(h\nu)}{I^{+}_{1/2}(h\nu)} + \frac{I^{-}_{3/2}(h\nu)}{I^{-}_{1/2}(h\nu)}\right]} \qquad (1)$$

where, for example, $I^{+}_{3/2}$ is the parallel intensity integrated over the $2p_{3/2}$ manifold. In Figure 1, we show the calculated asymmetry (solid curve) along the [111] direction for a model with fcc-Fe spacings (d_{12}=1.8Å, d_{23}=1.8Å) [8] as a function of photon energy compared to the asymmetry extracted from the experimental data. The integrated SIA gives a good quantitative agreement with the calculations and even reproduces the sign change in the asymmetry *ca.* hν = 840 eV. The energy oscillations due to spin-dependent photoelectron diffraction are fundamental and are present even in the peak height intensities. Furthermore, variations of the structural parameters by ±0.2Å destroys the agreement between the measurements and simulations. In fact, we have performed a detailed structural determination by varying the spatial parameters in the model and performing a quantitative r factor analysis of calculated and measured values. Our best fit occurs for d_{12}=1.9Å and d_{23}=1.7Å (dashed curve), with a Pendry r factor (r_p) of 0.17, which is 9% better than the unrelaxed structure, a measurable difference. This is consistent with our previous determination[9] but exceeds those earlier results with a clear improvement in sensitivity to the small surface relaxations. Removal of sample remanent magnetization obliterated any effect, driving ISIA to zero. Thus, the agreement between theory and experiment indicates that the intensity asymmetry arises from spin-dependent diffraction and demonstrates a sensitivity to both local geometric and magnetic structure.

In summary, we have for the first time measured spin-dependent photoelectron diffraction using circularly-polarized x-rays. The resultant spin-polarized 2p photoemission peaks exhibit angle and energy dependent intensity variations due to spin-dependent final state diffraction. The results are well described by spin-polarized photoemission, multiple scattering calculations.

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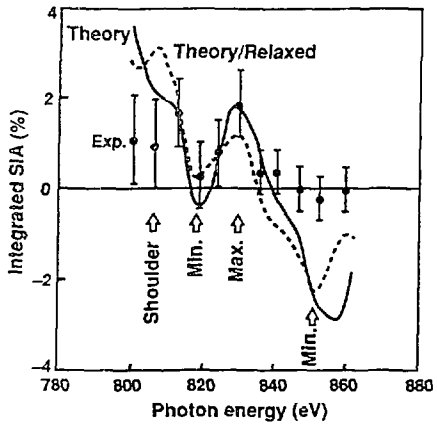
Figure 1. MXCD–Photoelectron Diffraction:
Comparison of Experiment and Theory for 4 ML Fe/Cu(001)



Calculated (solid curve: $d_{12} = d_{23} = 1.8$Å, $r_p = .19$; dashed curve: $d_{12} = 1.9$Å, $d_{23} = 1.7$Å, $r_p = 0.17$) and measured intensity asymmetries along the [111] direction as a function of photon energy. The oscillatory behavior in the curves is due to spin-dependent photoelectron diffraction. Representative error bars are included with the experimental data, shown as discrete values (triangles). See text for details. It is the positions of the minima and maxima that is the crucial variation in photoelectron diffraction. Exact quantitative agreement will require higher signal to noise and refinement of the model structure. [Note match of zero crossings.]