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Spin-Specific Photoelectron Diffraction Using Magnetic X-Ray Circular Dichroism

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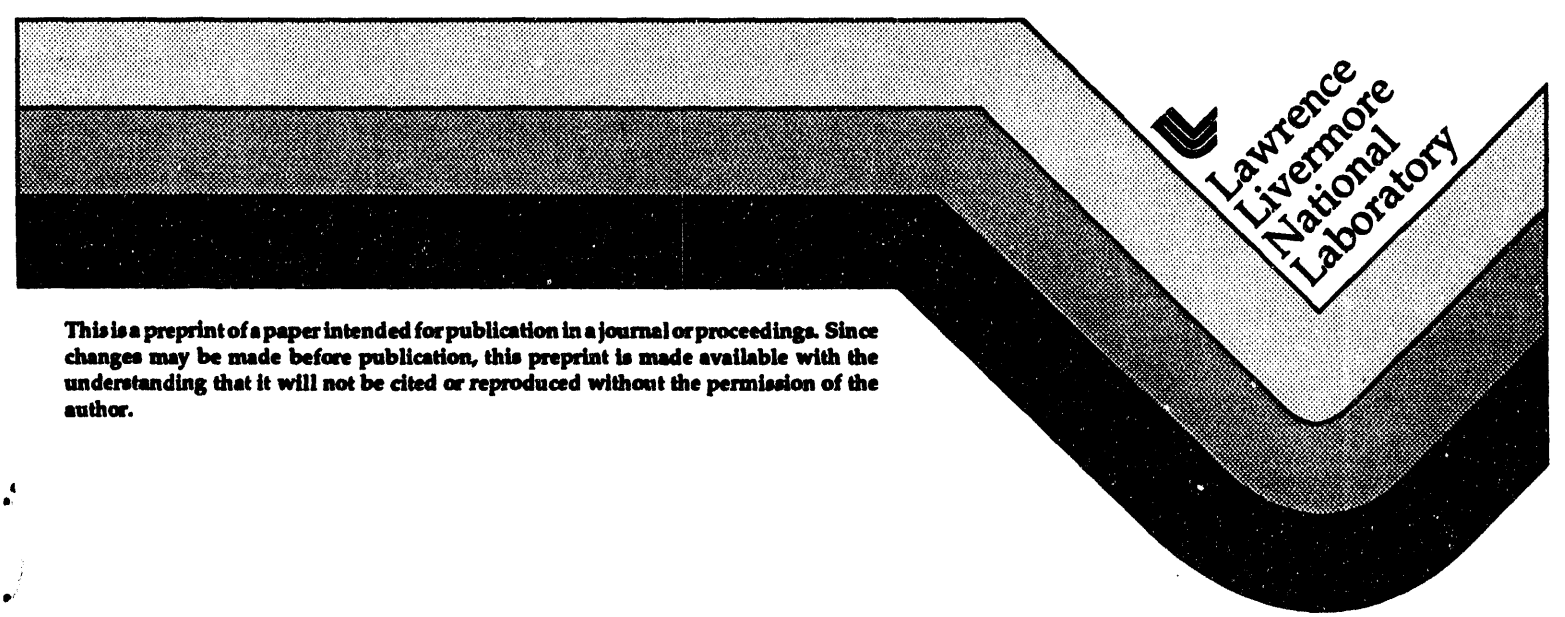

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SPIN-SPECIFIC PHOTOELECTRON DIFFRACTION USING MAGNETIC X-RAY CIRCULAR DICHROISM

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Abstract

The first observation of spin-dependent photoelectron diffraction using circularly-polarized x-rays is reported for monolayer ferromagnetic fcc Fe films on Cu(001). Circularly-polarized x-rays are used to produce spin-polarized photoelectrons from the Fe 2p spin-orbit split doublet, and intensity asymmetries in the 2p3/2 level of ≈ 3% are observed. The asymmetry is dependent on the relative orientation of the x-ray polarization vector and the Fe magnetic moment. This spin-dependent technique promises the direct, element-specific determination not only of local atomic structure, but magnetic structure as well.

Text

The last several years have witnessed a massive growth in the research and development of nanoscale magnetic materials. Perhaps the best review is provided by the Falicov Report[1] on "Surface, Interface, and Thin-Film Magnetism." Three general lessons can be derived from this report: (1) Magnetism is one of those special cases where fundamental research can directly lead to technological applications; (2) The key to understanding and manipulation of magnetic properties is the subtle yet overwhelming interplay of atomic geometric structure and local magnetic properties. For example, the giant magneto-resistance effect (GMR), which is already being explored for technological exploitation[2,3,4], appears to be intimately coupled to interfacial and thin film effects and probably will require elementally-specific probes for an explicit determination of the underlying causes[5,6,7]. This also appears to be the case for spin valves[8,9,10], another source of device miniaturization in read heads and magnetic sensors. [While it may eventually be found that these two effects are fundamentally connected, for now it appears that the GMR effect (up to 60%) is dependent upon an anti-ferromagnetic coupling through a non-ferromagnetic layer while the spin valve effect ($\leq$ 10%) is associated with an uncoupled ferromagnetic layer[9], which can be controlled externally.] (3) The importance of probes with a direct spin-dependence. A very recent illustration of this is the development of the magnetic x-ray circular dichroism (MXCD) using x-ray absorption[11-15] and photoemission[16,17] as a probe of surface, monolayer, and multilayer magnetism. It is this advantage that we propose to utilize, as described in the next section. However, before beginning that discussion, it is useful to summarize the state of of core-level photoelectron spectroscopy and diffraction at this point.

In the case of photoelectron spectroscopy and diffraction, there have been some significant strides recently. High resolution core-level spectroscopy has been demonstrated by Himpsel[18], to be a sensitive measure of oxidation state of surface silicon. Photoelectron diffraction[19-21] has been shown to be a powerful probe of metal overlayer structure. Using the internal spin polarization of the 3s state of Mn, Fadley, et. al, have reported observation of spin-specific photoelectron diffraction in bulk systems[22], and used it to study short range magnetic order. In attempting to extend such measurements to metal overlayer systems[23], limitations to this method became apparent. The small 3s cross section, the overlap of the split peaks, the large background on which the peaks rode, and the generally unknown spin-composition of the peaks all militate for a better defined approach. This approach must possess some sort of independent spin sensitivity or selectivity, such as that shown in Figure 1, and a more rigorous analysis based upon multiple scattering theory. One avenue to better spin-sensitivity is the utilization of spin detectors, which unfortunately carry with them a concomitant loss. (Efficiencies of 10^{-2} to 10^{-4} are common, relative to unpolarized detection.) Despite this handicap to spin-polarized, core-level, photoelectron spectroscopy, the first results were reported by Kisker, et. al, and Carbone, et. al, looking at the shallow 3p [24] and 3s [25,26] levels of bulk Fe. Subsequently, the measurements have been extended to include magnetic overlayers, demonstrating effects such as anti-ferromagnetic coupling between substrate and overlayer[27]. [In parallel with this, spin-polarized photoemission studies of valence band structure have also been pursued. Recent examples include the investigations of quantum well states, by Johnson, et. al[28], and Carbone, et. al[29], which suggest that these states are connected to oscillatory interlayer coupling. Johnson has also led the effort at NSLS to extend their spin-resolved measurements to include shallow core levels[30].] Finally, Roth, et. al[31], reported the observation of strong dichroism effects in the Fe3p spectra using linear polarization, with and without spin detection, by using specific high

symmetry geometries. These first studies were invariably done using linearly-polarized soft x-rays, and the spin-sensitivity was provided by electron spin-polarizers coupled to energy analyzers. An alternative means to extract spin-specific information from core-levels is to use circularly-polarized x-rays and the strong dipole selection rules that govern these transitions. The observation of photoemission circular dichroism was first demonstrated using the Fe2p states of bulk Fe by Baumgarten, et. al[16], and then ultrathin films of Fe/Cu(001) by Waddill, et. al[17]. Subsequently, Kaindl et. al. extended this work to rare-earth systems, with the observation of very strong effects[32]. More recently, a large circular dichroism in the Fe3p emission from Fe/Cu(001) has been observed and quantitatively simulated[33], using a spin-specific, fully relativistic, multiple scattering theory that can also explain the large linear dichroism that was previously observed[31].It is this spin selectivity, based upon circular polarization of soft x-rays, that we have used to perform spin-specific photoelectron diffraction.

In photoelectron diffraction, an electron is ejected from a core level and can scatter or diffract off of its nearest neighbors. In the usual experiments a small solid angle of electrons is collected and linearly-polarized x-rays are used as the excitation. From the energy or angular variations of the partial cross-section, the local geometrical structure can be obtained. To gain sensitivity to local magnetic structure, the spin of the electron must come into play (Figure 2). One way to do this is to use circularly-polarized x-rays as the excitation: In this case the 2p peaks will be intrinsically spin polarized. The $2p_{3/2}$ will be $\pm$ 25% polarized and the $2p_{1/2}$ will be $\mp$ 50% polarized. These spin-polarized electrons can then be used to determine local atomic magnetic structure, for example, to distinguish local anti-ferromagnetic ordering versus local ferromagnetic ordering.

In fact, we have already done the first such experiment. In Figure 3 is an example of our data collected using ferromagnetic Fe/Cu(001) and circularly-polarized x-rays without spin-detection. It should be noted that our measurements were done in mirror planes and with variation of both the magnetization and helicity, to remove other extraneous effects and as a cross check upon our analysis. It is obvious from our spectra that there is a fundamental intensity asymmetry that is independent of exchange-induced peak shifting. A more thorough discussion, including a full multiple scattering analysis, is provided elseqhere[34]. Thus, spin-dependent photoelectron diffraction can provide a sensitivity to local magnetic order, similar to that demonstrated for spin-polarized EXAFS[35].

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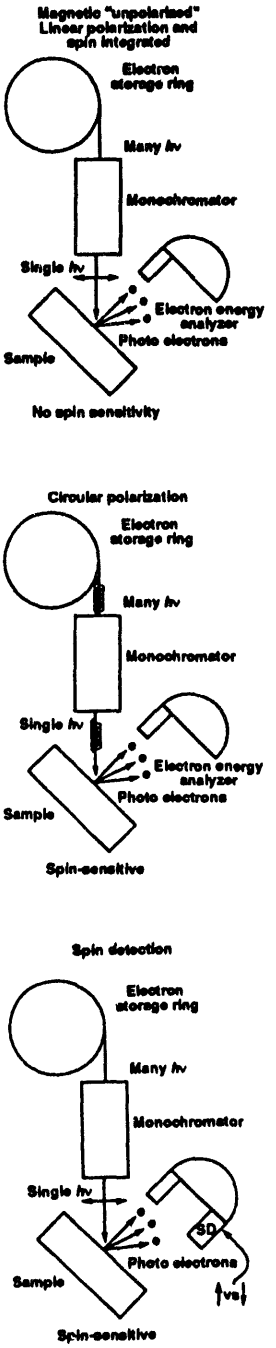
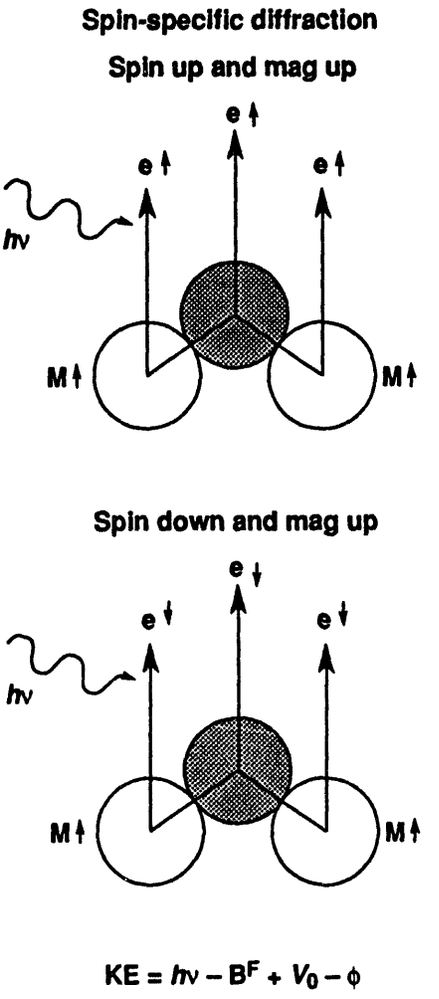


Figure 1. This figure shows schematically the experimental setup. A single energy (hv) of electromagnetic radiation is selected from the broad continuum of synchrotron radiation using a monochromator. The photons cause the ejection of photoelectron, which are then collected by the angle (± 3°) and energy resolving detector. The photons can be linearly or circularly polarized. The electron energy analyzer can be coupled to a spin-detector (SD).

Figure 2 This schematic illustrates the underlying cause of photoelectron diffraction: Interference between the direct and scattered waves. The interference is dependent upon the details of the local geometry and the emission angles, the kinetic energy (KE) and the spin of the outgoing electron, as well as the sample magnetization The kinetic energy is varied by scanning the photon energy, hν. The binding energy (B^F), the work function (Ø), and inner potential (Vo) are constant for a given state and material system.



$$KE = h\nu - B^F + V_0 - \phi$$

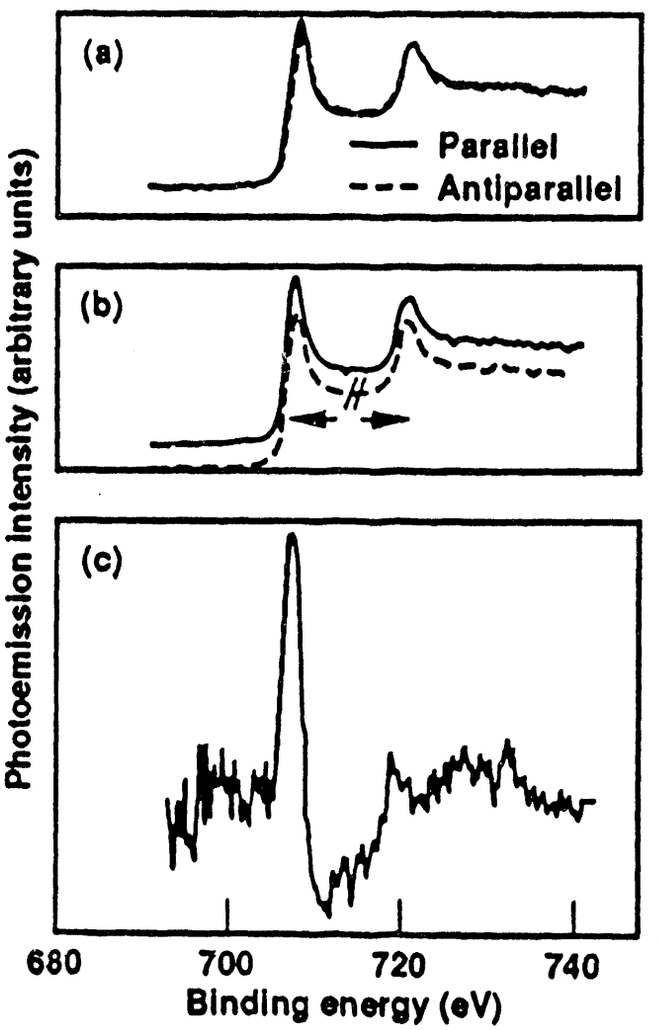


Figure 3 Spin-specific photoelectron diffraction using magnetic circular dichroism. (a) Upper panel shows the unshifted iron 2p spectra for scattering along the [111] (crystalline) direction and the (b) middle panel demonstrates how the peaks are shifted to remove exchange effects from the comparison of intensities. (c) Intensity asymmetry along the [111] direction summed over 30 pairs of spectra with parallel and anti-parallel photon and minority electron-spin alignment. The 2p3/2 peak asymmetry at a binding energy of 707 eV is about 3%.

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