Comparison of magnetic linear dichroism in $4f$ photoemission and $4d-4f$ photoemission from Gd on Y(0001)

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Magnetic linear dichroism (MLD) in $4d-4f$ resonant and $4f$ nonresonant photoemission (PE) is studied from thin epitaxial gadolinium films. In an angle resolved and high-energy resolution mode, experiments were conducted with the electric-field vector of the incident light perpendicular to the sample magnetization. Our results show a significant difference in behavior of MLD in resonant PE as compared to that in nonresonant PE. Off-resonance, the MLD signal is dominated by a negative feature at the low binding energy side of the peak. Near the $4d-4f$ resonance maximum, the MLD displays a plus–minus shape, with a negative signal at the low binding energy side of the $4f$ peak and a positive signal at the high binding energy side. Analysis of MLD in $4d-4f$ resonant PE may provide insight into interactions of the $4d$ core hole with the $4f$ core level in the intermediate state. [S0734-2101(97)03003-0]

I. INTRODUCTION

Magnetic dichroism in core-level photoelectron spectroscopy is a new technique that promises to shed light on the electronic and magnetic properties of bulk and thin-film materials. Synchrotron radiation sources have made it feasible for investigators to study how the relative orientation between the sample magnetization, photoelectron wave vector, and electric field influences the shape of photoemission spectra. We present results that show a change in the line shape of the Gd $4f$ photoemission (PE) peak using linearly polarized light when the direction of sample magnetization is reversed, i.e., magnetic linear dichroism (MLD). Results of MLD in Gd $4f$ photoemission taken off-resonance are compared to those in resonant PE. Our experimental geometry is analogous to the geometry used by Roth *et al.* in their investigations of MLD in the Fe $3p$ core levels.[1] Previous experiments on Gd have focused on magnetic circular dichroism (MCD) in $4f$ and $4d$ resonant[2] and nonresonant photoemission.[3,4]

When photon energies are near the $4d$ threshold a strong Fano resonance is observed in the $4f$ photoionization cross section. The effect is interpreted as a constructive interference between the direct PE channel and the resonant channel. The resonant PE channel is represented by a two-step process. Resonant excitation of a $4d$ electron into an empty $4f$ spin minority state followed by a radiationless Auger decay. Because of the localized nature of the $4f$ state, the resonant channel can be described in an atomic picture. The resonant process is represented schematically as

$$4d^{10}4f^{7}+h\nu\rightarrow 4d^{9}4f^{8}\rightarrow 4d^{10}4f^{6}+\epsilon l,$$

where ϵl represents the continuum state of the photoelectron. Decay from the intermediate state may lead to ${}^{7}F_{J}$ final states as in direct PE or to low spin ${}^{5}X_{J}$ states, where X represents all possible combinations of angular momenta. The low spin states show up as a higher binding energy satellite on the ${}^{7}F_{J}$ peak and are strongly correlated with the fine structure peak on the total yield curve at the onset of the main resonance feature.[5] We note that the $4d$ core hole created in the intermediate states interacts with the $4f$ core levels.

We are, therefore, studying the resonance with MLD in order to understand the contribution of magnetic effects on the resonant PE process. The photoelectron intensity in the $4d-4f$ resonant region leading to the ${}^{7}F_{J}$ final states may be more than an order of magnitude higher than in PE spectra taken off-resonance; therefore, the MLD signal in the spectra is mainly due to the resonant mechanisms. This, in turn, reduces the complication of decoupling the resonant from the nonresonant behavior.

II. EXPERIMENT

The PE experiments were conducted at the Spectromicroscopy Facility (Beamline 7) of the Advanced Light Source in Berkeley at Lawrence Berkeley Laboratory.[6,7] Beamline 7 located at an undulator magnet provided the source of linearly polarized soft x-rays.

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A Y(0001) single crystal was aligned by Laue diffraction and mechanically polished with a 1 μm diamond solution. The crystal was supported on Ta foil, spot welded to a molybdenum sample puck. The flexibility of the Ta support allowed the height of the crystal face to be easily adjusted to the center of rotation for x-ray photoelectron diffraction (XPD) experiments. The sample was heated by electron bombardment from a filament located behind the substrate. The Gd evaporation cell consisted of a W crucible surrounded by a water-cooled shroud. The crucible was heated by electron bombardment from a nearby filament. A quartz crystal thickness monitor gauged the evaporation rate and film thickness.

The Y(0001) substrate was cleaned by sputtering with a 1.5 keV beam of argon ions and then annealed to 875 K to establish structural order as confirmed by XPD. Films 50 ML thick were grown epitaxially, while the substrate was held at room temperature. The base pressure after bakeout was 5×10^{-10} Torr and during film growth rose up to 3×10^{-9} Torr as determined by an ion gauge. Since the sample could only be cooled to 125 K, a thick film of 50 ML was grown, because the Curie point is expected to be significantly reduced from the bulk value of 293 K for thin Gd films, <15 ML.[8] Long anneal times in combination with elevated temperatures were observed to result in the intermixing of the Y with Gd, which is not surprising since the two materials have similar chemical properties and almost perfect lattice matching.[9] Therefore, we limited ourselves to a short anneal of 45 s at 710 K. This ordered the Gd(0001) film as confirmed by sharp XPD spots, with no intermixing of the substrate with the overlayer as confirmed by x-ray photoelectron spectroscopy (XPS). Moreover, carbon and oxygen XPS peak intensities were below detectable limits. Spin resolved photoemission experiments by McIlroy *et al.* have shown that a Gd(0001) surface exposed to as little as 0.25 L of oxygen forms an oxide with reduced magnetic properties, thus, elimination of oxygen is particularly important.[10] While the film was being cooled to 125 K by convection from a copper braid connected to a liquid nitrogen reservoir, the Gd film was remanently magnetized in plane by pulses from a nearby solenoid. All magnetic measurements were made in remanence at 125 K. From previous studies of Gd(0001)/W(110), the magnetization of the 50 ML film is expected to lie in the plane in a single domain state.[11]

A diagram of the experimental geometry is shown in Fig. 1. The linearly polarized x-rays are incident at an angle of 60° from the x axis with the electric field vector, **E**, in the xy plane. Photoelectrons are collected at normal emission along the x axis with $\pm3°$ angular resolution, with the intensity normalized to the photon flux to account for instabilities in the ring current. The magnetization, **M**, lies in the plane of the sample and is positioned orthogonal to the electric field vector of the exciting radiation. The dichroism is measured by reversing the magnetization by mechanically rotating the sample 180°. The MLD is measured off resonance at 95 eV, where the photoionization cross section is high and these measurements are compared to MLD near the $4d-4f$ resonance maximum.

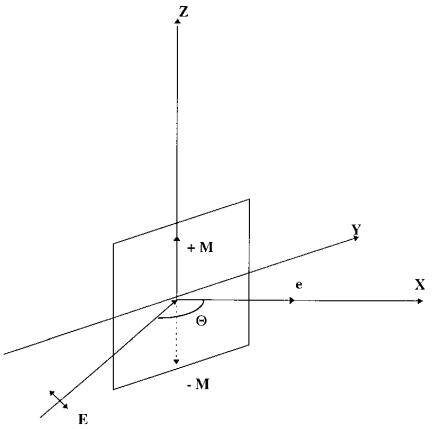

FIG. 1. Experimental geometry: p-polarized light is incident at angle of 60° relative to the x axis. The sample magnetization, **M**, points either parallel or antiparallel along the z axis and is perpendicular to the electric-field vector in the xy plane. The photoelectrons are collected along the sample normal in the x direction.

III. RESULTS

Figure 2 illustrates MLD in Gd $4f$ PE at 95 eV off-resonance, left upper panel, and MLD in resonant $4d-4f$ PE at 150 eV, right upper panel. The open squares correspond to magnetization up, and the black filled squares correspond to magnetization down. The lower panels show a plot of the peak normalized difference (PND) for a quantitative comparison. The PND is obtained by subtracting a suitable integral background from both spectra, magnetization up, and magnetization down. The difference in intensity at each binding energy is then calculated and normalized to the peak maximum intensity of the sum of the two spectra after background subtraction. Representing the dichroism in this fashion eliminates the problems that occur when calculating the asymmetry by dividing the difference of small numbers by the sum of small numbers. A similar procedure for quantitative comparison of MLD in the Fe $2p$ levels was applied by Hillebrecht *et al.*[12] The spectra at 95 eV show a negative dichroism in the low binding energy side of the peak with MLD of 6%. However, the MLD signal near the resonance maximum shows a significant change in sign and shape with a plus–minus feature of about 10%. The spectrum taken at 150 eV with the magnetization up has a maximum peak intensity that is greater than the spectrum obtained with the magnetization reversed, while the maximum peak height intensity is larger for the magnetization down for the spectrum taken at 95 eV. As illustrated in Fig. 2, the ^7F_J MLD in the PE spectra near the resonance maximum is markedly different from the spectra taken off-resonance.

IV. DISCUSSION

An atomic model of MLD in nonresonant PE, as described by Thole and van der Laan, has proven successful in interpreting MLD in Fe $3p$ core-level photoemission.[13,14]

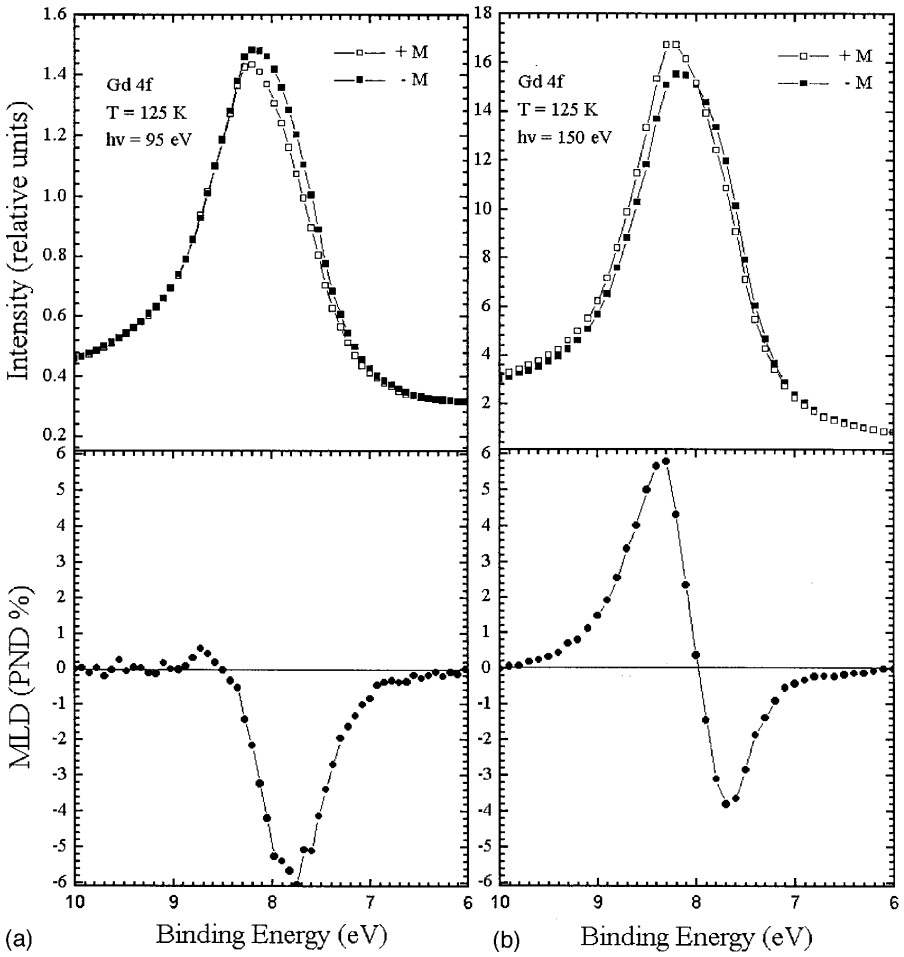

FIG. 2. (a) Upper panel: Gd $4f$ core-level spectra taken off-resonance at 95 eV. Open squares represent magnetization up, $+z$ direction; whereas the filled squares represent magnetization down, $-z$ direction. Lower panel: plot of peak normalized difference (%), see the text. (b) Upper panel: Gd core-level spectra taken near $4d-4f$ resonance maximum at 150 eV. Lower panel: plot of peak normalized difference (%).

The localized nature of the $4f$ states suggests that an atomic model would be appropriate in describing MLD in the Gd $4f$ PE spectra. In the method by Thole and van der Laan, the dipole matrix elements are written in terms of the creation and annihilation operators of an l shell electron with magnetic component m. By using this approach, terms pertaining to the geometry of the experiment are separated from terms describing the interactions of the core hole left in the final state leading to multiplet splitting. For PE from an f shell, the MLD would be described as a linear combination of the fundamental spectra, I^x, where $x=1,3,5$. Thole and van der Laan have calculated the line shape of the fundamental spectra for Gd^{3+} $4f$ photoemission.[14] Moreover, both MLD and MCD measure the same set of fundamental spectra; thus, similar results might be expected from both experiments. However, we measure a MLD signal that shows a predominate contribution on the low binding energy side of the $4f$ peak as shown in Fig. 2, which is very different from the strong MCD effect observed in Gd $4f$ PE by Starke *et al.*[3]

On resonance, our results indicate that there is a strong interaction between the Gd $4d$ core hole and the $4f$ electrons. This is shown by flipping the magnetization, i.e., the majority spin direction, with respect to the electric-field vector. Thus, the matrix elements for excitation out of the $4d_{5/2}$ and $4d_{3/2}$ manifolds into the unpopulated $4f$ minority state changes. This leaves a different intermediate state for the $4d$ core hole to interact with the magnetic $4f$ electrons. Resonances in rare-earth materials are well known, and combining MLD with resonant PE in a single experiment may yield new information. For example, recent magnetic dichroism studies in resonant PE have been used to interpret the $2p$ core-level absorption structure in Ni.[15]

Because the PE intensity in the resonant region leading to ${}^7F_J$ states is substantially enhanced, as in Fig. 2, the interference of the resonant channel with the direct channel is small, thus, contributions to the spectra are mainly from the resonant process. Although not presented here, MLD spectra at several photon energies in the range of 138–154 eV were recorded in the $4d-4f$ resonant region and show results similar to those in Fig. 2. Analysis of data demonstrating MLD as a function of photon energy in the resonant region would provide a novel approach in understanding $4d-4f$ resonant PE. However, quantitative analysis would be complicated by several factors. In the case of $4d-4f$ resonant

PE, the spin–orbit splitting of the $4d$ levels is the same size as the exchange splitting in the intermediate state between the $4d$ hole and $4f$ electrons, so that separation of resonant behavior due to transitions from the $d_{5/2}$ and $d_{3/2}$ channels is not straightforward, which is in contrast to the case of Gd $3d-4f$ resonant PE where distinct multiplets are observed in the absorption spectrum.[16,17] Moreover, interpretation of MLD in the resonant PE may be further complicated by a Faraday rotation of the incident radiation's electric-field vector as the light penetrates the sample. These effects have been found to be significant near core-level absorption edges[18] and, thus, may contribute in resonant PE.

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