Local magnetic moment coupling of Gd on Fe(100) studied by magnetic dichroism in angular-dependent photoemission

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We measured the magnetic linear dichroism in the angular distribution (MLDAD) of photoemission of thin Gd layers on Fe(100). At low photon energies large MLDAD asymmetries, up to 40%, in the Gd $4f$ photoemission were observed. The line shape and the photon-energy dependence of the measured MLDAD are in good agreement with theoretical results. Analysis of the Gd $4f$ and Fe $3p$ magnetic signals indicates an antiferromagnetic coupling between Gd and Fe, confirming previous findings. We also demonstrate that the MLDAD plus-minus feature is governed by the orbital magnetic moment of the core hole state. [S0163-1829(98)51234-9]

The microscopic study of the magnetic coupling is of fundamental importance for magnetic thin film technology. Element-specific methods to measure the local magnetic moments have become possible due to recent development of magnetic dichroism techniques in x-ray absorption and photoemission. The latter offers the additional benefit of intrinsic surface sensitivity and it can be employed with linearly polarized x rays. By using a chiral geometry in the photoemission experiment, it is possible to measure with linear polarization[1,2] the magnetic dichroism which is otherwise only accessible by circularly polarized light (MCD): this technique is called magnetic linear dichroism in the angular distribution (MLDAD or LMDAD). Moreover, theoretical analysis of angular-dependent photoemission from Thole and van der Laan clarified the connection between physical properties and linear combination of the measurable fundamental spectra.[3–5] Under specific experimental conditions, several meaningful parameters of the MLDAD can be recognized: (i) the plus-minus $(+/-)$ feature, which can be used to monitor the magnetic coupling between different elements;[6] (ii) the asymmetry, A_{MLDAD}, which is directly proportional to the surface magnetization;[7] (iii) the energy splitting, which in the case of p core levels of Fe and Co is related, through the spin-orbit interaction, to the exchange splitting, i.e., to the local magnetic moment.[8,9] Although the reliability of the MLDAD parameters has been qualitatively demonstrated, a clear confirmation concerning the physical properties which govern the MLDAD effect is still lacking. This fact poses a severe limitation for quantitative analysis of magnetic properties by means of MLDAD.

Since the first experimental evidence of the MLDAD effect,[1] several MLDAD experiments were performed on $2p$ and $3p$ core levels as well as on the valence band (VB) of transition metals (TM) and their interfaces.[10,11] A smaller number of reports dealt with the surface magnetic properties of rare earths (RE) and RE/TM interfaces, where MCD in photoemission and spin-resolved photoemission (SRPES) techniques played a major role. This imbalance is confirmed for the Gd/Fe(100) interface: MCD in photoemission was reported in several papers from Starke *et al.*;[12] SRPES results from Taborelli *et al.*[13] and Carbone and co-workers[14] showed clearly the antiparallel magnetic coupling between Gd and Fe substrate. A previous MLDAD experiment on Gd/Fe(100) was performed by Kinoshita *et al.*,[15] who measured the Gd $4f$ core levels at a fixed photon energy of 90 eV. From this study the authors concluded that the magnitude of the observed MLDAD signal was in disagreement with (i.e., larger than) the theory as deduced from the calculated values for the radial-matrix elements and phase factor of Ce $4f$ by Goldberg *et al.*[16] Here, we report on a detailed comparison between experiment and theory for MLDAD on Gd/Fe(100), concentrating on the Gd $4f$ and Fe $3p$ core levels. The aim of this work is twofold: first, to deepen the understanding of the connection between MLDAD parameters and physical properties, i.e., to assess whether MLDAD is governed by the spin or by the orbital magnetic moment; second, to decide in the interesting debate on whether or not the MLDAD signal is identical to the signal measured by MCD in photoemission.[17] These issues can be suitably investigated by comparing the Fe and Gd magnetic signals, and the Gd/Fe(100) interface is of particular interest, presenting a localized open shell, the $4f$ of Gd, together with a closed core shell, such as the $3p$ of Fe, both producing MLDAD.

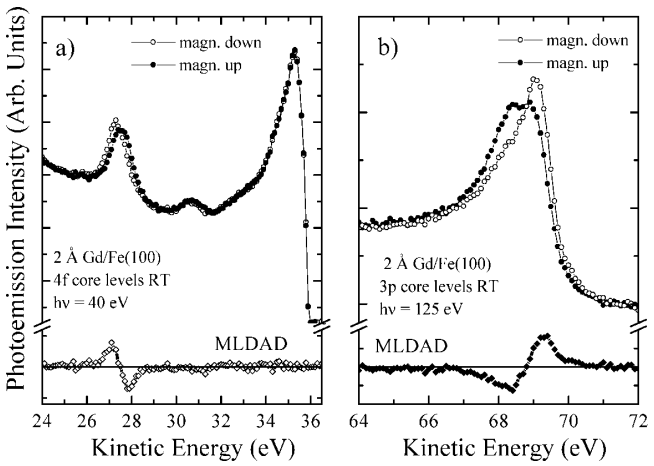

FIG. 1. (a) Top: Photoemission spectra in the region of Gd $4f$ core level and valence band for 2 Å Gd on Fe(100), measured at $h\nu=40$ eV with linearly polarized x rays, for magnetization up (filled circles) and down (open circles). Bottom: the corresponding MLDAD (open diamonds). (b) Top: magnetization-dependent photoemission spectra of the Fe $3p$ core level (open and filled circles), at $h\nu=125$ eV, for the same Gd/Fe(100) sample as (a). Bottom: the corresponding Fe $3p$ MLDAD (filled diamonds).

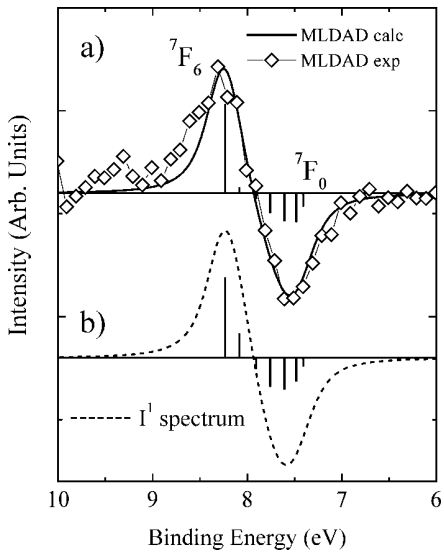

FIG. 2. (a) Comparison between the theoretical (drawn line) and the measured [open diamonds, $h\nu=40$ eV as Fig. 1(a)] Gd $4f$ MLDAD, normalized to the same height. (b) The theoretical angle-integrated MCD spectrum, I^1 (dashed line). The sticks give the $^7F_{0,\ldots,6}$ multiplet lines which have been convoluted with a Gaussian of $\sigma=0.085$ eV and a Lorentzian of $\Gamma=0.18$ eV (Ref. 4).

Measurements were performed at the SU3 Swiss-French undulator beamline of the Super-ACO storage ring at Orsay, in the same experimental setup and geometry as in Refs. 7, 8. All photoemission spectra were measured in remanence and at room temperature (RT). Ultrathin Gd films were evaporated from a tungsten basket onto an atomically clean, unannealed, Fe(100) surface of a Fe3%Si single crystal. Base pressure was 6×10^{-11} mbar, rising to 1×10^{-10} mbar during evaporation.[18] After deposition of 2 Å of Gd the low-energy electron diffraction (LEED) showed the characteristic faint pattern of a disordered growth of Gd onto Fe(100), confirming previous findings.[14,15] Figure 1(a) shows the magnetization-dependent spectra for the Gd $4f$ core levels and the VB taken at $h\nu=40$ eV for a Gd thickness θ of 2 Å, together with the MLDAD spectrum. A rather intense Gd $4f$ MLDAD is found around 27 eV kinetic energy (KE), despite the presence of a bump around 30 eV KE, which is due to oxygen $2p$ derived states and which is indicative of surface contamination. From the relative cross sections at $h\nu=40$ eV, we infer that the oxygen content in the Gd film is less than 3%. In order to perform energy-dependent measurements on the same sample we checked periodically the MLDAD reduction at $h\nu=40$ eV; we noticed a decrease of the magnetic signal over time. We considered the magnetic film ''alive'' below an acceptable reduction of 25% of the MLDAD signal as in Fig. 1(a): this corresponded to 3 h under given vacuum conditions. Ultrathin Gd films prepared with various θ values showed a sharp decrease of the $4f$ MLDAD signal with increasing θ, to be connected with the change of the Gd magnetization $\mathbf{M}_{\mathrm{Gd}}(T,\theta)$ as a function of temperature and thickness, and to the value of the surface Curie temperature. Figure 1(b) shows the Fe $3p$ photoemission spectra of the same Gd/Fe(100), at $h\nu=125$ eV: it is seen that the $(+/-)$ structure of the Fe $3p$ MLDAD is opposite in sign compared to the Gd $4f$ one.

A detailed analysis of the line shape and energy dependence for the Fe $3p$ has been already given in Refs. 2, 19–21, so that it is sufficient here to discuss the Gd $4f$ photoemission. A detailed theoretical analysis[22,23] shows that the MLDAD spectrum strongly resembles the MCD spectrum. However, there are small differences, as displayed in Fig. 2. In terms of fundamental spectra I^x (as shown in Fig. 3 of Ref. 5), the angle-integrated Gd $4f$ MCD photoemission is given by the I^1 spectrum [Fig. 2(b)]. The MLDAD is primarily composed of the I^1 spectrum, but also contains a small contribution due to I^3 and I^5 [Fig. 2(a)]. The agreement between experimental and calculated MLDAD spectrum is excellent. The signature, i.e., the $(+/-)$ or $(-/+)$ of the MLDAD spectra can be used to determine whether the coupling between Gd and Fe atoms is ferromagnetic or antiferromagnetic. The Gd ^7F final state is split by spin-orbit interaction into a septet $J=0,1,\ldots,6$ manifold. Since the dominant contribution in the Gd $4f$ MLDAD originates from the I^1 spectrum, its magnitude is related to the expectation value of the core-hole orbital moment. If we define as positive the sign of the orbital moment when it is parallel to the ground-state spin, then the $^7F_6$ level corresponds to positive orbital moment and the $^7F_0$ corresponds to a negative orbital moment [cf. Fig. 2(a)]. Moreover, because the spin and orbit prefer to be coupled antiparallel for a less than half-filled shell, the $^7F_6$ level is located at the high-binding energy (BE) side, while the $^7F_0$ level is at the low BE side of the spectrum. The Gd $4f$ I^1 spectrum displays therefore a $(+,-)$ signature when viewed with decreasing BE (increasing KE; Fig. 2). The Fe $3p$ I^1 spectrum has a $(-,+)$, which is thus opposite to the Gd $4f$ signature. In the case of the Fe $3p$ final state, the exchange interaction and the spin-orbit coupling are of equal importance. With only a single hole in the $3p$ shell the spin and the orbit prefer to couple parallel, so that the level with the lowest BE corresponds to a positive orbital moment, then to a parallel alignment between Fe and Gd *orbital* magnetic moments. For MLDAD we further have to consider the signs of the radial-matrix elements and the

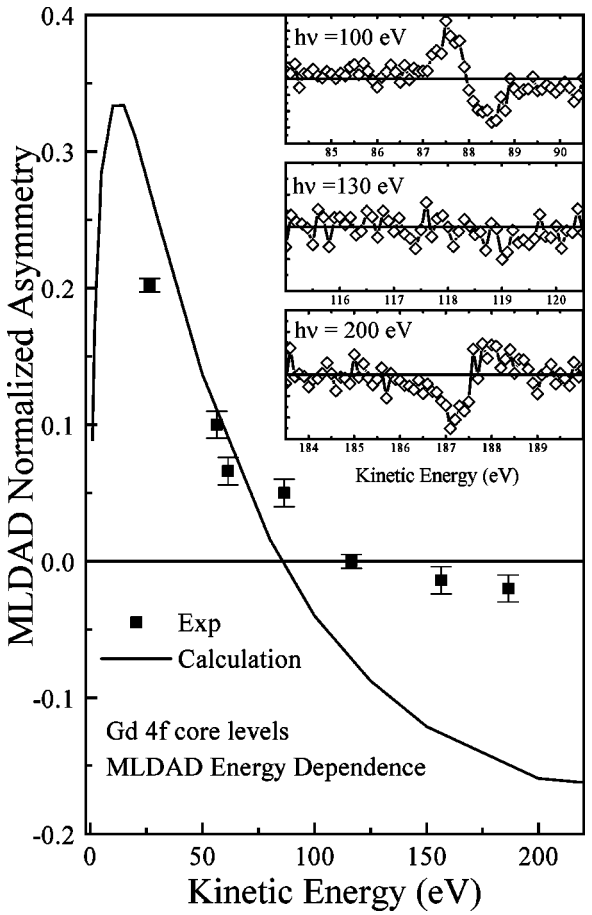

FIG. 3. Theoretical (drawn line) and experimental (filled squares with error bars) KE dependence of the A_{MLDAD} for the Gd $^7F_6$ final state level at 8.2 eV BE (as in Fig. 2). Inset: the corresponding Gd $4f$ MLDAD differences measured at $h\nu=100$ eV, $h\nu=130$ eV, and $h\nu=200$ eV.

phase factor. If we compare the Gd $4f$ photoemission at 30 eV KE with the Fe $3p$ photoemission at 100 eV KE, the radial-matrix elements have the same signs, but the phase factors have opposite signs.[20] From this, we can conclude that when the MLDAD signatures are the same for the Fe $3p$ and Gd $4f$ levels, their *total* magnetic moments are coupled ferromagnetic. The experimental results show that the MLDAD signatures have opposite signs, so that the coupling must be antiferromagnetic, which is known from spin-resolved Auger spectroscopy[13] and SRPES experiments.[14] Therefore, we can consider this result as *a clear proof that the MLDAD* $(+/-)$ *signature is governed by the orbital moment*.

We now turn to the energy dependence of the MLDAD signal, which is proportional to the transition matrix elements for the $l-1$ and $l+1$ continua times a phase factor.[5] We calculated the radial-matrix elements and the phase factors using Cowan's code.[24] The energy dependence of the Fe $3p$ was already given in Fig. 1 of Ref. 21. The calculated asymmetry, $A_{MLDAD}=I_{DIFF}/I_{SUM}$, for the Gd $^7F_6$ final state level at 8.2 eV BE (Fig. 2) is given by the drawn line in Fig. 3. The MLDAD will be maximum when the phase difference between the two continua goes to $\delta\rightarrow\pi/2$, which occurs around 15 eV and 300 eV KE. At low energies the matrix element for the g channel is three times larger than that of the d channel, which yields a maximum value for the MLDAD $4f$ asymmetry of 0.34, around 15 eV KE. This is thus significantly smaller than for the Fe $3p$ MLDAD, where the two photoemission channels are of comparable strength. At higher photon energy of few hundred eV the g channel is an order of magnitude stronger than the d channel, resulting in a reduced asymmetry. The MLDAD vanishes when the phase shift goes to zero, which occurs around 80 eV KE. In the calculation we have not included the resonant photoemission due to the $4d\rightarrow4f$ absorption at 140–150 eV, which has a strong influence on the cross section, phase factor, and angular dependence.[25] The $4d$ absorption removes the chirality in the experimental geometry, thereby strongly reducing the MLDAD asymmetry. However, the details depend strongly on the coherence in the resonant photoemission process, as discussed in Ref. 25. Figure 3 also shows the experimental values with relative error bars of the normalized $A_{MLDAD}=I_{DIFF}/I_{SUM}$ corresponding to the Gd $^7F_6$ final state level (Fig. 2), where I_{DIFF} and I_{SUM} are, respectively, the difference and the sum of the magnetization-dependent photoemission spectra. The peak to peak A_{MLDAD} measured at $h\nu=40$ eV, corresponding to Fig. 1(a), results in $\sim$40%. Normalization of sum spectra was obtained using an integral background subtraction of Shirley type, without data smoothing. The inset shows the MLDAD differences before ($h\nu=100$ eV), near ($h\nu=130$ eV), and far above ($h\nu=200$ eV) the zero-line crossing, where the sign of MLDAD changes. The overall agreement between experiment and theory is good: the quantitative differences can be ascribed to the $M_{\mathrm{Gd}}(T,\theta)$ value of Gd/Fe(100) at RT and perhaps to surface contamination. Moreover, at energies around and above 100 eV the influence of the giant resonance and the strongly reduced absolute value of the photoemission cross-section intensity can lead to large deviations.[25,26]

For angular-dependent photoemission it is also important to consider the photoelectron diffraction (PED) effects.[27] These can lead to intensity modulations with respect to the atomic dependence as a function of photon energy and emission angle. They can strongly modify the line shapes of the total photoemission spectra and the magnitude of MLDAD asymmetry,[27,28] and also mix the different fundamental spectra, as described in Sec. IV of Ref. 5. In our case, PED in the Gd energy dependence is expected to be small due to the low Gd coverage (although backscattering is still possible); moreover, experiments were performed on the unannealed Fe(100) surface, and the faint LEED pattern testifies that experimental conditions were not favorable for PED effects. At all measured photon energies we found that (i) the magnetization averaged photoemission spectra did not change in line shape, and (ii) the Gd $4f$ MLDAD spectrum always resembled the calculated spectrum given in Fig. 2(a).

In summary, we demonstrated that for the Gd $4f$ and Fe $3p$ MLDAD there is good agreement between experiment and theory. This makes this technique attractive to use for quantitative studies of magnetic materials. We showed that the shape of the MLDAD is very similar to that of the MCD photoemission spectrum, not only for a p shell but also for an f shell. From the comparison of the MLDAD $(+/-)$ signatures for the Gd $4f$ and Fe $3p$ we were able to confirm the antiferromagnetic coupling between the magnetic moments of the two materials. These results can also be turned around and regarded as an experimental proof that the signature

(+/−) or (−/+) of the MLDAD is due to the orientation of the core hole orbital moment with respect to the magnetization. To deduce this we have made direct use of the fact that for the less than half-filled Gd $4f^6$ final state, the spin and orbit prefer to couple antiparallel, giving rise to a parallel alignment of the orbital moments of substrate and adsorbate, hence an antiparallel alignment of their total magnetic moment, i.e., the known antiferromagnetic coupling of Gd and Fe(100). The important conclusions from the comparison between experiment and theory are that (i) even differences up to one order of magnitude in the radial-matrix elements for the $l+1$ and $l-1$ photoemission channels can still lead to a relatively strong MLDAD effect, which can be turned to practical use; (ii) in order to determine the sign of the magnetic coupling with MLDAD, extreme care should be exercised, and a calculation providing the signs of the matrix elements and phase factor is required to support the analysis.

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