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LMDAD as a Surface Magnetometry

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ABSTRACT: Linear Magnetic Dichroism in the Angular Dependence (LMDAD) in $l > 0$ core level photoemission can be used as an atom-specific magnetometer. Its large dichroic signal is directly proportional to the surface magnetization and probes in a very efficient way the changes of magnetic moments as a function of the environmental conditions.

1 Introduction

The field of surface magnetism stimulates, for both technological and fundamental reasons, a strong experimental effort. The central problem in understanding electronic and magnetic properties of low dimensional systems is the need of surface-sensitive and atom-specific techniques, able to provide the absolute values and/or the behaviour of either local magnetic moments or magnetic interactions, i.e. need of surface magnetometries. Among these new spectroscopic techniques, fruitful results were obtained from Auger Electron Spectroscopy and Photoemission spectroscopy from core level and valence bands, both in the spin-resolved mode [1, 2], and also from the Spin Polarisation (SP) measurements of the secondary low energy electrons yield [3]. Each of these techniques is a quasi-ideal magnetometry: SP is strongly surface sensitive and can be interpreted semi-quantitatively, but is limited by the intrinsic averaging of the measured magnetisation [4]; on the other hand the application of spin-resolved techniques is so far limited from the low efficiency of spin detectors ($\sim 10^{-3}$). Spectroscopy with circularly and linearly polarised Synchrotron Radiation in the soft X-ray range, on magnetically ordered systems, demonstrated that absorption and photoemission dichroism are large effects and can be developed as powerful techniques to study surface magnetism [5, 6]. Within this scheme, a new approach was recently presented from Roth et al., resulting in a special case of dichroism in photoemission. Experiments performed with linearly polarised light could exploit the angular distribution of photoelectrons from magnetically oriented samples [7], i.e. the LMDAD (Linear Magnetic Dichroism in the Angular Dependence).

Aim of this paper is to show that an atom-specific surface magnetometry could be based on LMDAD experiments, which join the selectivities of the photoemission spectroscopy to a strong magnetic signal directly connected to the surface magnetisation.

2 Principles

Chiralities appear when the experimental geometry is not the mirror image of itself. If the sample is magnetic, mirror experiments

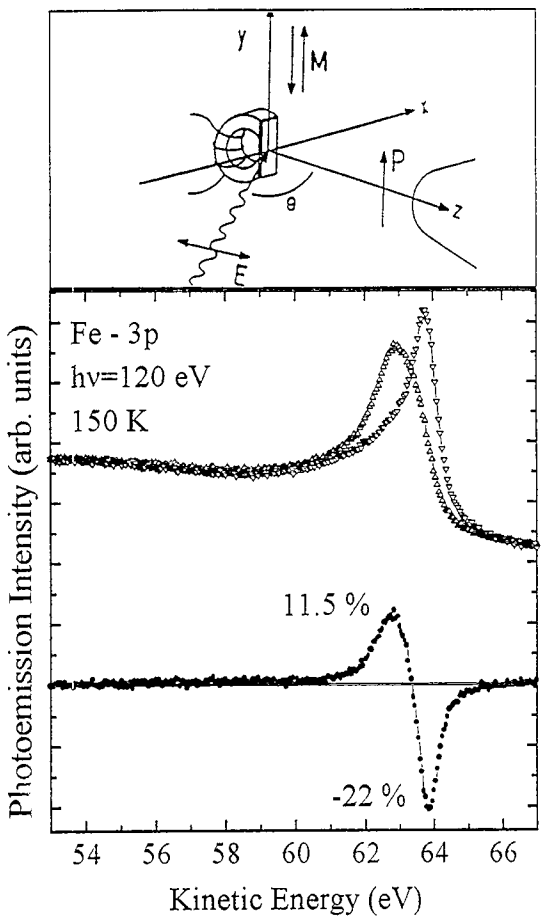

Figure 1. Top: the chiral geometry of the experiment. Bottom: 3p core level spectra as measured for the two magnetisation directions, with the difference curve; the maximum values of asymmetry are indicated.

are obtained, referring to fig.1, by reversing the magnetisation in the $\pm y$ direction with respect to the scattering plane defined by the direction of the Linearly Polarised Synchrotron Radiation (from planar undulator sources at SuperAco) impinging onto the sample and the direction of detection along the z axis (details about mounting system and experiment procedures are given elsewhere [8]). The photoemission intensity difference between two mirror experiments is the LMDAD.

Angle resolved experiments measure the differences in the angular dependent emission of photoelectrons because of the nonvanishing contributions in the interference channels between the $l+1$, $l-1$ final state wavefunctions. If spin-orbit is present in the initial state ($l > 0$) [9, 10] the differences in the angular distribution give rise to strong dichroism, as shown in fig.1 for the 3p core level of Fe(100). The magnetic asymmetry is measured as $A = \frac{I_{up}-I_{down}}{I_{up}+I_{down}}$, where $I_{up(down)}$ are the photoemission intensities for the $up(down)$ward direction of the imposed magnetization. In the conditions of present experiment its value is the 22% of the total photocurrent, for the clean Fe(100) surface, and we measure the relative variations of this value. Photoelectron Diffraction effects modify the asymmetry when forward scattering conditions are met [11].

3 LMDAD as a diagnostic

LMDAD provide a direct diagnostic of the magnetic ordering and coupling of interfaces, by means of the tunable sensitivity to different elements which is intrinsic of the photoemission technique. Among the obtained results, we cite: a) the analysis of the in plane magnetisation direction of epitaxial fcc-Fe thin films on CuAl(100) substrate[12]; b) the control of magnetic ordering at different stages of the Cr/Fe(100) and Fe/Cr/Fe(100) interfaces formation, in which the top Fe layer grown epitaxially on a 5 ML Cr interlayer shows an antiferromagnetical coupling to the Fe(100) substrate across the Cr [8].

Moreover, LMDAD magnetic hysteresis on selected core level can be measured [13]; the 3p core level hysteresis for a polycrystalline Fe layers grown on a Vitrovac (amorphous

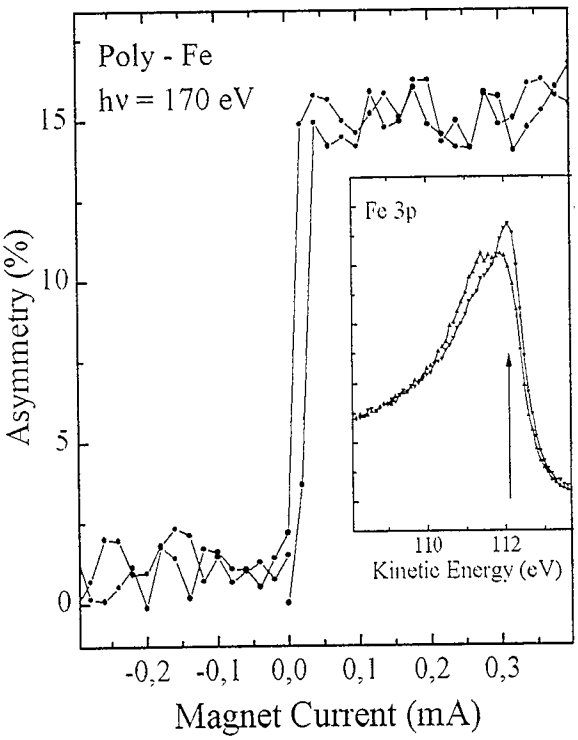

Figure 2. LMDAD hysteresis, measured at the Fe-3p core level on 10 Å of poly-Fe deposited on Vitrovac substrate.

Co-based soft magnetic ribbon) substrate is shown in fig.3. The hysteresis is obtained, as a function of the applied magnetic field, in constant final state mode by setting the analyser at the kinetic energy corresponding to the maximum LMDAD asymmetry (as shown in the inset of Fig.2). This application is confined to soft magnetic systems and their overlayers, because of a limitation is set by the requirement of minimal external fields which perturb the photoemission measurements.

LMDAD is then a Kerr-like probe for magnetic surfaces and interfaces, but with in addition surface and chemical selectivity: we want to stress that this diagnostic costs only two times more the effort of the standard photoemission spectroscopy.

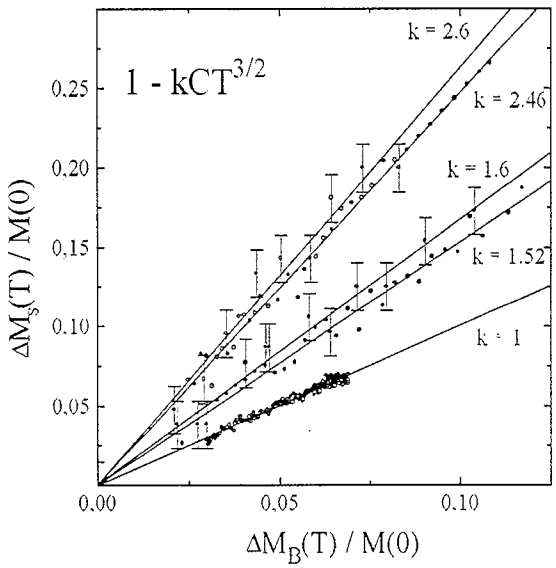

Figure 3. Thermal decrease of the relative surface magnetization as measured by Fe 3p LMDAD asymmetry (*circles with error bars*) and by the SP of the secondary electron yield (*circles*) versus the thermal decrease of the relative bulk magnetization measured in situ by the magneto-optic Kerr rotation. The data are measured for the clean Fe(100) surface (*open symbols*) and for the c(2x2)S/Fe(100) superstructure (*filled symbols*). The solid lines are the theoretical curves fitted to the data.

4 LMDAD vs Temperature

After annealing a Fe(100) single crystal up to 700 C, a monolayer of sulfur segregates on the surface with a clear c(2x2) surface reconstruction [14]. The temperature dependence, in the range $T \leq 0.4\ T_C$, of the Fe-3p maximum LMDAD asymmetry for the Fe(100) clean surface and of the sulfur segregated one is shown in fig.3. The data are compared with SP values measured with a 100 KV Mott scattering experiment and with bulk-sensitive Kerr rotation data obtained "in situ" for the same sample mounting. SP and LMDAD results are in good agreement,

resulting in a thermal decrease of the relative surface magnetisation $\Delta M_S(T)/M_S(0)$ that follows the law $M(T)/M(0) = 1 - kCT^{3/2}$. According with spin waves theory in this range of temperature, C is a constant describing the decrease of the bulk magnetisation due to spin waves and k depends on $J_\perp/J$, i.e. on the ratio of interatomic exchange interaction between the surface and the bulk [15].

The same law is expected for both the bulk (M_B) and the surface (M_S) magnetisation, but with different values of k, which represents the coupling factor between surface and bulk; $k = 1$ corresponds to the bulk behaviour, as measured from the Kerr-rotation. A surface enhancement factor of $k \simeq 2.5$ is obtained for the clean surface, representing the reduced exchange interaction of surface atoms in a direction perpendicular to the surface [16]. The value $k \simeq 1.6$ obtained for the sulfur segregated surface indicates a reduced surface anomaly with respect to the clean Fe(100) surface, which is consistent with the reduced magnetic moment of the iron surface atoms in presence of sulfur segregation. In Fig.4 are reported the data near T_C, which follows the law $M_{B(S)} \propto (1 - T/T_C)^{\beta_{B(S)}}$ for the vanishing of bulk and surface magnetization within the mean field theory. The critical exponents obtained are: for the surface $\beta_S = 0.81 \pm 0.01$ and for the bulk $\beta_B = 0.38 \pm 0.01$ respectively from the LMDAD and the Kerr measurements [17]. The LMDAD data are well in agreement with the results from different surface sensitive techniques [18]. Moreover, others LMDAD experiments on the temperature dependence of the magnetisation, in two-dimensional fcc-Fe films, also show good agreement with the prediction of mean field theory [19].

These are strong evidences that the LMDAD asymmetry is directly proportional to the magnetic order parameter $< M_S >$ of the surface atoms and shows the relative changes of the exchange interaction at the surface.

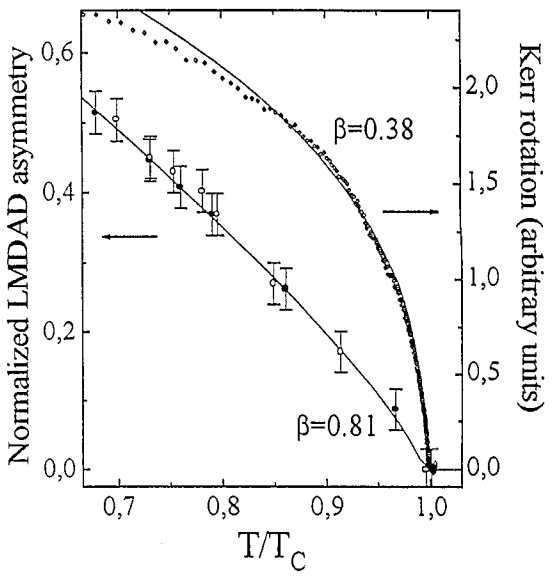

Figure 4. LMDAD (*circles*) and Kerr rotation (*diamonds*) as a function of the reduced temperature in the near critical region. The curves were normalised to 1 at T=0.. The critical exponents were obtained by maximizing the linearity of *log* [LMDAD] vs. *log* [1-T/T_c]. The solid lines are the theoretical curves fitted to the data.

5 LMDAD as a Local Magnetometer

The exchange interaction between the d-electrons and the core hole, together with the spin-orbit interaction, is responsible of the splitting into "m_J" levels of the core level. But, the cited J value and the splitting in m_J levels is no longer an exact picture: Zeeman effect retains until $J = L + S$ is still a good quantum number, i.e. when the magnetic field is treatable as a perturbation respect to the spin-orbit splitting of the J levels. With

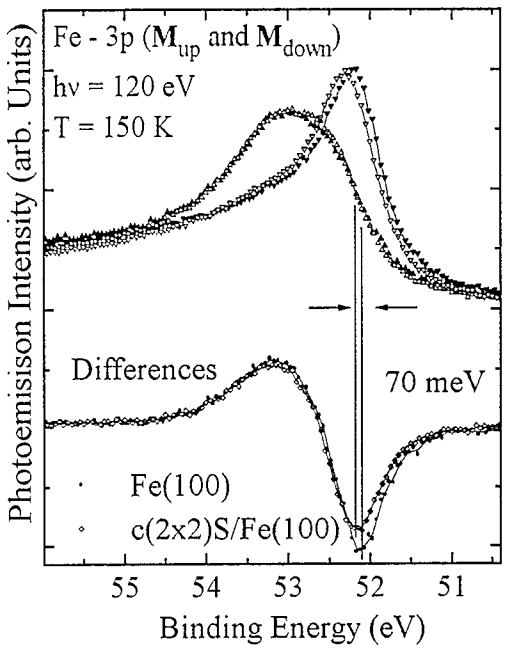

Figure 5. Fe 3p LMDAD spectra for the clean Fe(100) surface (*filled symbols*) and the sulfur segregated c(2x2)S/Fe(100) surface (*open symbols*). All the spectra were measured at T=150K. The reduced width of the dichroism spectrum reflects the reduced splitting of the J=3/2, $m_j = +3/2$ and $m_j = -3/2$ sublevels.

this in mind, the m_J levels are simply the components of the splitting caused from a *particular* magnetic field, the exchange field, acting only on spin [10].

Within an atomic picture the "width" of the dichroism is related to the magnitude of the magnetic interaction and is a direct measure of the atomic exchange interaction for the 3p core hole [8, 9, 10, 20]. The resulting splitting is of 1.06±0.01 eV for the clean Fe(100) surface. In figure 5, the Fe 3p spectra for the clean Fe(100) surface are compared with the ones of the sulfur segregated c(2x2)Fe(100) surface, with their relative LMDAD curves. Besides the 10 % reduction in the dichroism intensity, other effects of the presence of Sulfur at the surface are: a) the clear shift of the peak dominated from the minority spin, that can be due to a preferential hybridization with the S bands [21]; b) the reduced dichroism width of the sulfurated surface to 0.99±0.01 eV and the appearance of dichroism on the Sulfur atoms at the surface [17].

These results demonstrate that LMDAD could provide atom-specific *quantitative* informations of changes in magnetic moments.

6 Outlook

An atom specific surface magnetometry can be based on the LMDAD effect of core level. We have shown that this allows to measure relative changes of the local exchange interaction, and of magnetic moment, as well as extended properties, i.e. the coupling between surface and substrate. High resolution photoemission experiments with unpolarized (which means partially linear polarised) ultraviolet or X-ray laboratory sources showed that the detection of magnetic asymmetries is possible [22, 23]. Thus, the LMDAD technique extends to the domain of surface and interface magnetism the powerful photoemission probe with an increase of only a factor two more measurements, without need of spin-resolution or circular polarisation.

These facts imply that no compromises are imposed by LMDAD magnetometry on energy resolution, lateral resolution or time resolution of the state of art photoemission experiments. The stage for developing such applications of magnetometry is therefore set.

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