SPIN-FLOP ORDERING FROM FRUSTRATED FERRO- AND ANTIFERROMAGNETIC INTERACTIONS: A COMBINED THEORETICAL AND EXPERIMENTAL STUDY OF A Mn/Fe(100) MONOLAYER

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The magnetic structure of ultrathin antiferromagnetic (AFM) overlayers on ferromagnetic (FM) substrates determines the properties of ferromagnetic and antiferromagnetic multilayers that are key constituents of devices such as exchange-bias or tunneling magnetoresistance recording systems. Complex, non-collinear, magnetic structures are expected at these interfaces, for spin canting minimizes the exchange energy between a ferromagnet and an antiferromagnet that exposes a plane with antiparallel spins. The resulting spin-flop alignment of the moments in the antiferromagnet, perpendicular to the magnetization in the ferromagnet, is the microscopic basis of the large coercive field in exchange-bias devices. In spite of the great interest in these systems, the understanding gained so far on the basis of semiempirical models has neither been validated by accurate first-principles calculations, nor by a direct experimental observation of the non-collinear magnetic order at the interface. In fact, on one hand, accurate, fully unconstrained methods based on density-functional theory (DFT) for studying non-collinear magnetic structures have become available only recently [1]; on the other hand, the simultaneous access to both antiferromagnetic and ferromagnetic ordering, not possible by usual experimental methods, has become possible by magnetic circular and linear dichroism methods using advanced synchrotron sources.

The aim of this work is to determine the magnetic structure of Mn deposited on Fe(100) in the mono-layer regime, using state-of-the-art theoretical and experimental techniques. Mn/Fe(100) has been simulated using fully unconstrained Spinor Density Functional Theory (SDFT) which allows for a proper account of non-collinear magnetic structures [1]. This same system has then been investigated experimentally by means of a combination of X-ray magnetic circular dichroism (XMCD) and X-ray magnetic linear dichroism (XMLD) spectroscopies, which allows for a direct assessment of ferromagnetic and antiferromagnetic ordering, and also for a determination of the moment orientations with chemical sensitivity.

Our SDFT calculations have been performed within the local density approximation and neglecting spin-orbit interactions.

The latter approximation makes the calculated magnetic structures degenerate with respect to an arbitrary overall rotation of the magnetization field. By constraining all the magnetic moments to be collinear a Mn overlayer on Fe can assume one of three distinct configurations: in the *ferromagnetic* (FM) structure all the Mn magnetic moments are parallel to each other and to the magnetization of the ferromagnetic iron substrate; in the second, *anti-ferromagnetic* (AFM), structure the Mn magnetic moments are antiparallel with respect to the substrate magnetization; the third, *ferrimagnetic* (FI), structure is finally characterized by an anti-ferromagnetic arrangement of atomic moments in the overlayer, which result alternatively parallel or anti-parallel to the substrate magnetization. It is interesting to notice that the inequivalence of spin-up and spin-down atoms in the overlayer may determine some buckling in the surface.

When the constraint of spin collinearity is released, the frustration of the magnetic bonds across the interface drives a rotation of the Mn moments resulting in a chessboard arrangement where these moments form angles of ~80° with respect to those of the underlying Fe atoms. This non-collinear (NC) structure is depicted in Figure 1b. We note that, with such arrangement, the magnetic interactions between Mn and Fe are the same for all the Mn atoms, which are therefore all structurally equivalent. The almost perpendicular orientation of the Mn moments corresponds to a quasi-anti-ferromagnetic order in the Mn plane, still avoiding the magnetic frustration experienced in the collinear situations. We find that this NC structure is more stable by about 35 meV/atom than the FI collinear structure (see the top panel of Figure 1a), and should therefore be clearly observable also at room temperature.

Using XMCD and XMLD at the Mn-$L_{2,3}$ edges we examined the magnetic structure of sub-monolayer and monolayer (0.1-1.0 ML) Mn films deposited on Fe(100). The samples were grown *in situ* in the 10^{-10} mbar range using a quartz-crystal microbalance to control the thickness. As a first step, an Fe(100) surface was epitaxially grown on a clean and ordered Ag(100) single crystal surface and magnetized in remanence along the
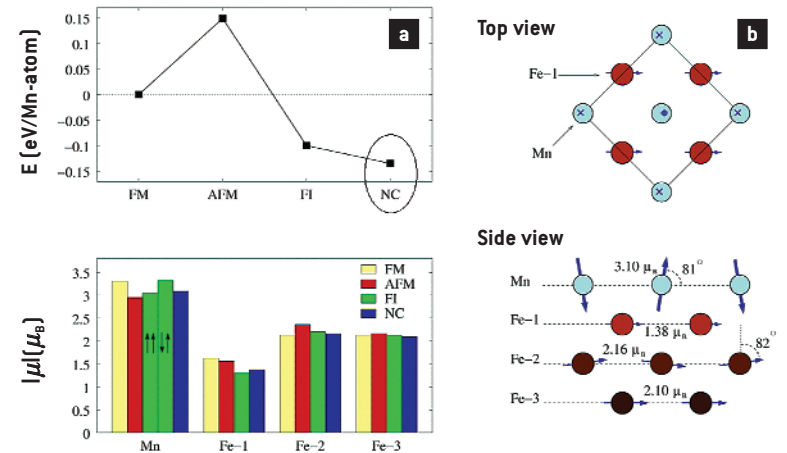
Fe[001]. The thickness of the Fe film was above 50 ML in order to suppress Ag surface segregation. A wedge of Mn in the range of 0-1.1 ML was prepared. The experiments were performed at the 4.2 beamline "*Circular Polarization*".

The magnetization of Mn was investigated by circular dichroism. The XMCD data measurements performed at different Mn thicknesses for the submonolayer regime (not shown here) are in accordance with Ref. 2. For less than 1 ML, we observe an XMCD effect of the Mn adlayer opposite to that of Fe, which is a proof of long-range ferromagnetic order of the adlayer aligned antiparallel to the magnetization in the Fe substrate. The net magnetization of Mn decreases with increasing thickness and approaches zero for 1 ML (Figure 2a), thus pointing out a rather smooth transition from ferro- to anti-ferromagnetic arrangement.

The XMLD was measured rotating the sample in the polar and azimuthal geometries. The spectra recorded for in-plane and for out-of-plane polarization (polar rotation) are shown in Figure 2b. The intensity ratio between the L$_2$ well defined double peak structures (labeled as P_I and P_{II}) turns out to be well suited to detect the spectral changes. The intensity ratios for two geometries, reported in Figure 2a and 2b, show that the polar measurement has a regular dependence as a function of the angle, while the azimuthal one is much less pronounced, which supports the idea that the orientation is perpendicular to the surface.

We have simulated the spectral lineshapes by exact diagonalization of an atomic many-

Figure 1. (a) Top panel: total energies of the FM, AFM, FI, and NC structures relative to the FM structure. Bottom panel: magnitude of the atomic magnetic moments in the various structures. For the FI structure ↑↑ and ↑↓ indicate the Mn atom with magnetic moment parallel and antiparallel to the underlying Fe atoms, respectively. **(b)** Schematic view of the Mn/Fe magnetic interface.
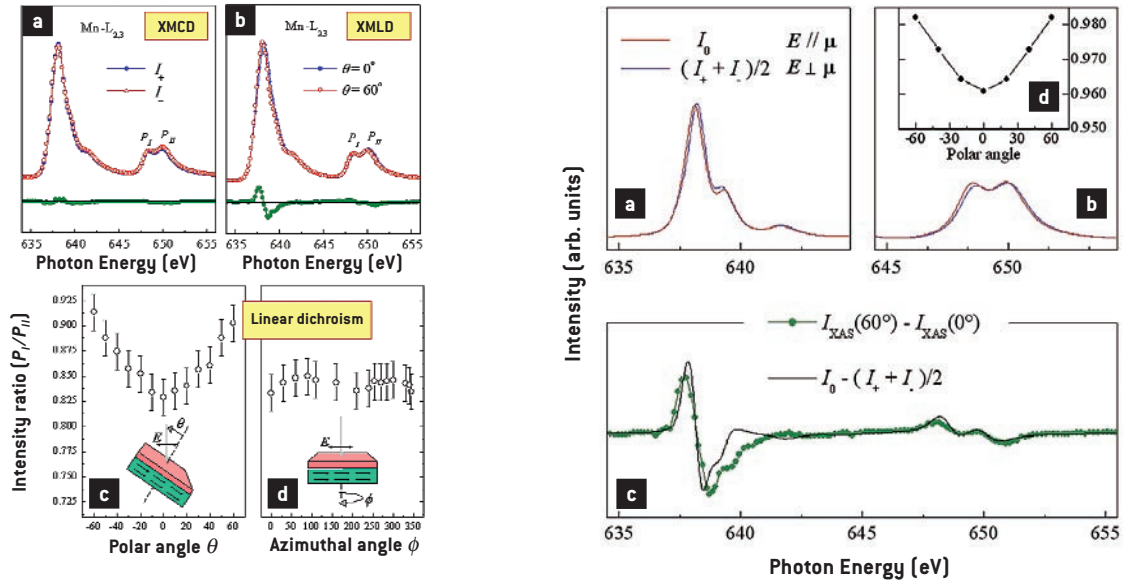
Figure 2. Top: **(a)** XMCD data at the $L_{2,3}$ edges for 1 ML of Mn; **(b)** Mn $L_{2,3}$ X-ray-absorption spectroscopy for 1 ML of Mn as a function of the angle between E and the surface normal. Bottom: XMLD effect for the polar **(c)** and for the azimuthal **(d)** rotations; the intensity ratio of the Mn-L_2 double peak structures is used to measure the intensity of the effect.

Figure 3. (a) and **(b)** Simulation of the absorption spectra for the two polarization directions after suitable broadening. **(c)** Comparison between the simulated and the experimental XMLD. **(d)** Simulated P_I/P_{II} intensity ratio.

body Hamiltonian, based on a fully coherent spectral function given by:

$$I_q(\omega) = \sum_n \left| \sum_{v,c} \left\langle \Psi_f^n(N) \left| a_v^+ a_c \right| \Psi_G(N) \right\rangle \left\langle v \left| rC_q^{(1)} \right| c \right\rangle \right| \times \delta \left(\hbar\omega - \left[E_f^n(N) - E_g(N) \right] \right)$$

The irreducible components of the dipole operator are defined as: $T_q^{(1)} = rC_q^{(1)}$, where $q = 0, +1, -1$ correspond to z-linearly polarized, right circularly polarized and left circularly polarized light, respectively, and $C_q^{(1)}$ q are normalized spherical harmonics [3].

In Figure 3a,b we show the results of the spectral simulations. By using different combinations of the irreducible components of the dipole operator $T_q^{(1)}$ we are able to simulate the Mn-$L_{2,3}$ spectra as a function of the orientation between the magnetic moment on Mn and $\vec{E}$: I_0 for $\vec{E} // \vec{\mu}$ and $(I_+ + I_-)$ for $\vec{E} \perp \vec{\mu}$. In terms of the lineshape, the agreement with the experimental spectra of Figure 2b is very good. As shown in Figure 5c, the XMLD spectrum, which carries all information about the lineshape changes, is well reproduced by the simulated XMLD lineshape calculated as $I_0 - (I_+ + I_-)/2$. The calculated P_I and P_{II} intensity ratio, assuming perpendicular orientation of the Mn moments with respect to the surface plane, also simulates the angular dependence of the

experimental results. We can conclude that the magnetic moments of Mn are aligned out of the surface plane, perpendicular to the Fe magnetization.

The non-collinear magnetism of the system intrinsically derives from the competing exchange interactions between the monolayer and the substrate moments. This mechanism may also lead to the development of similar spin-flop structures in other monolayer systems, where the exchange interaction within the antiferromagnetic monolayers prevails, but does not overwhelm, the coupling with the ferromagnetic substrate.

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