New Model for Magnetism in Ultrathin fcc Fe on Cu(001)

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Using soft-x-ray resonant magnetic scattering in combination with first-principles calculations for noncollinear magnetic configurations we present a new model of the magnetism in ultrathin fcc Fe films on Cu(001). We find the presence of blocks with robust magnetic structure, while the relative directions of the moments of different blocks are sensitive to the detailed atomic structure and temperature. The magnetic noncollinearity is directly demonstrated, which has not been possible so far.

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The investigation of magnetism in low dimensional systems attracts considerable interest. Motivated by the development of new materials in spintronics, a detailed understanding of the organization of magnetic moments at surfaces, interfaces, and in heterostructures is mandatory. When magnetic properties in an embedded ultrathin film are changing at the monolayer level, most of the conventional or advanced methods [1] are limited either by the lack of spatial resolution or by their limited surface sensitivity.

In this context, face centered cubic (fcc) Fe represents an archetype system, since it is notorious for its delicate magnetic properties, both in the bulk and in the ultrathin film limit. Although it has been known since 1967 that fcc Fe can be stabilized when deposited on Cu(001) [2–4] a clear picture of its magnetic structure has not been established yet. Though some consensus exists that fcc Fe films in the thickness range above 4 monolayers (ML) are not ferromagnetic (FM), their detailed magnetic structure is disputed. For instance, theory has favored both, bilayer [5,6] and single layer [7] antiferromagnetic (AFM) structures, the latter suggested by experiment also [4]. Magneto-optic-Kerr-effect (MOKE) data by Qian *et al.* [8] also found a complex structure interpretable as a spin density wave or a noncollinear spin arrangement, however theory has shown that such structures do not correspond to the total energy minimum [9,10]. This calls for a direct analysis of the film's spin structure, which—however—has not been available up to now.

This study provides a clear answer to the long-standing problem of the magnetization profile in fcc Fe by determining the depth-resolved magnetization profile which has not been possible so far by conventional methods such as MOKE and x-ray magnetic circular dichroism experiments. Resonant magnetic scattering experiments in the hard-x-ray regime have been shown to be sensitive to the layer-by-layer magnetic profile [11,12]. In our study, measurements are carried out in the soft-x-ray regime. The components of the magnetization (M) along all directions in space can be determined by collecting reflected intensities up to high scattering angles (85°) as has been shown recently [13].

In combination with first-principles density-functional-theory (DFT) calculations we shine new light on the still unresolved questions regarding the magnetic archetype system fcc-Fe/Cu(001). We find a new physically relevant grouping of the atomic layers reflecting the hierarchy of exchange interactions. The top three layers tend to form a collinear magnetic structure of the form $|\Uparrow\Uparrow\Downarrow|$. The spins of the further layers form strongly AFM coupled pairs $|\Uparrow\Downarrow|$. The exchange coupling between these units is relatively weak that makes the noncollinearity of the spins of different units energetically inexpensive.

The Fe films were grown on Cu(001) by pulsed laser deposition (PLD) in the thickness range between 4 and 8 ML. [14] Based on MOKE experiments carried out at 145 K it was concluded that PLD grown fcc Fe exhibits a spin reorientation transition (SRT) between 4 ML, where M is in-plane and 8 ML, where no remanence in longitudinal (L) geometry was obtained [14]. At 6 ML, MOKE shows a hysteresis loop for both geometries at 145 K.

After depositing the Fe films using a KrF (248 nm) laser as described in Ref. [14], they were capped by a 3 nm thick Au to prevent oxidation. Capping does not affect the magnetic properties in comparison with the uncapped Fe-films as evidenced by *in situ* MOKE experiments.

The XRMS experiments were carried out at the RESOXS end station [12] at the Swiss Light Source in Villigen (Switzerland). The reflectivity was measured in the θ–2θ scan mode. For measurements in L geometry an electromagnet for sample magnetization was used. At each incidence angle θ the reflectivity $I(+)$ and $I(-)$ was obtained by reversing the field ($\mu_0 H = \pm 0.16$ T) while keeping the x-ray helicity unchanged. For measurements in P geometry the sample was magnetized by a permanent magnet ($\mu_0 H = 0.4$ T) followed by data collection in remanence. In this case, $I(+)$ and $I(-)$ were obtained by reversing the x-ray helicity. For all experiments discussed

in the following saturation of the MOKE signal was achieved by applying a field lower than $\mu_0 H = \pm 0.2$ T with a remanence of almost 100%.

At first we discuss the 4 ML sample, which according to MOKE shows an in-plane FM order at room temperature (RT). Symbols in Fig. 1(a) represent $I(+)$ and $I(-)$ collected at RT in L geometry at $E = 705.9$ eV. A maximum normal momentum transfer of $q_z = 2k\sin(\theta) = 7.2$ nm^{-1} could be achieved corresponding to $\theta = 85°$. The finite difference between $I(+)$ and $I(-)$ directly proves the presence of magnetic ordering.

In the first step of the analysis, the geometrical film structure was derived by fitting the average signal $\bar{I} = [I(+) + I(-)]/2$ measured at several energies far and near the resonance by allowing thickness, density, and interface roughness of the Au cap layer and the Fe film to vary. The film densities were to within 10% of the bulk values and the thickness of the Au cap layer was derived to 3.0 ± 0.1 nm, very close to the nominal thickness. Similarly, the Fe-film thicknesses were derived to 0.93, 1.39, and 1.90 ± 0.05 nm for the 4, 6, and 8 ML sample, respectively. Roughness is in the range between 0.1 and 0.3 nm, the larger value is found for the Fe/Cu interface, which we attribute to intermixing.

The magnetic profile was then obtained by fitting $I(+)$ and $I(-)$ while keeping the structure parameters constant. Solid lines in Fig. 1(a) represent the calculated reflectivity, $I(+)$ and $I(-)$. In addition, Fig. 1(b) shows the experimental (symbols) and calculated (lines) asymmetries $[A(q_z)]$ given by: $A(q_z) = [I(+) - I(-)]/[I(+) + I(-)]$ for different photon energies around the Fe-L_3 absorption edge. $A(q_z)$ is strongly energy dependent but all curves converge to $A = 0$ at $q_z = 7.2$ nm^{-1}, directly indicating in-plane order since $A(q_z) \propto m_z \sin(\theta)$ [15]. For the calculation of the reflectivity the imaginary parts (f'' and m'') of the energy dependent charge $[f_c = f_0 + f'(E) + \mathrm{i}f''(E)]$ and magnetic $[f_m = m'(E) + \mathrm{i}m''(E)]$ scattering factors were derived from x-ray absorption and XMCD measurements on a reference Fe-film for which sum rules yield a spin moment of $2.1\mu B$. The corresponding real parts (f' and m') are retrieved by applying the Kramers-Kronig relations.

The Fe-film is subdivided into slices, each of which is given a specific orientation expressed by the transverse (m_x), longitudinal (m_y) and polar (m_z) component of M. Their magnitude is linked to the XMCD or m'' (E) amplitude through a weighting factor. In the following a weighting factor of $|m| = 1$ corresponds to a spin moment of $2.1\mu B$. By using a recursive calculation involving the reflection and transmission coefficients at each interface developed by Zak *et al.* [16] for magneto-optics, $I(+)$, $I(-)$ and $A(q_z)$ are calculated for a given magnetization profile.

The measurement of $A(q_z)$ at several energies around the resonance imposes a strong constraint to the analysis if the magnetic profile model is fitted to the whole data set. We benefit from the sharp energy dependence of the scattering factors (change of sign within less than 1 eV) in the vicinity of the absorption edge.

The best fit [solid lines in Fig. 1(b)] is obtained by modelling the film with 4 FM ordered slices, each of which

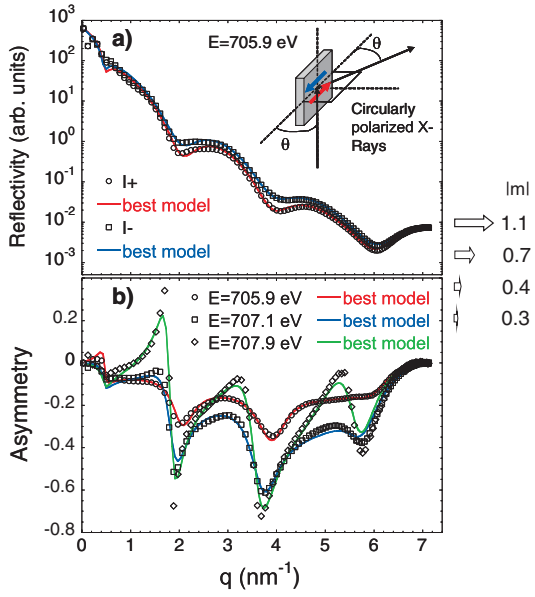

FIG. 1 (color). (Color) (a) Experimental (symbols) and calculated (lines) specular reflectivity at $E = 705.9$ eV for parallel and antiparallel sample magnetization measured at RT for 4 ML Fe/Cu(001). The inset shows the experimental geometry. (b) Corresponding asymmetries for three photon energies around the Fe-L_3-absorption edge. The magnetic structure together with the magnetic weighting factors is shown on the right.

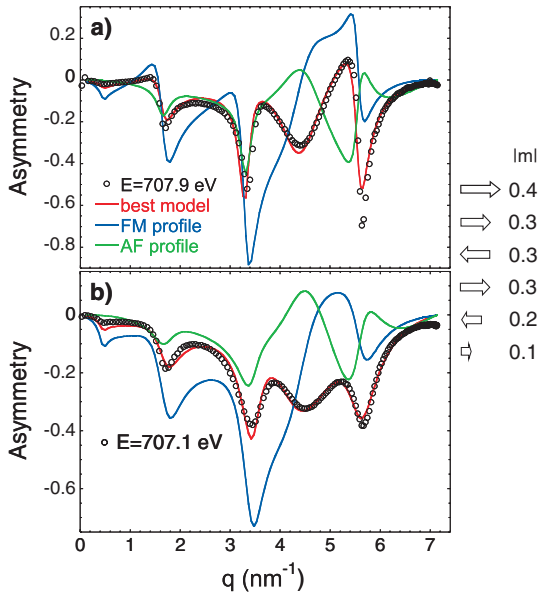

FIG. 2 (color). Experimental (symbols) and fitted (lines) $A(q_z)$ for 6 ML Fe measured at RT in longitudinal geometry for 707.9 (a) and 707.1 eV (b). The magnetization profile for the best fit (red line) is shown on the right. Calculations for FM ordering (blue line) and single layer AFM ordering (green line) are shown for comparison.

characterized by a different magnetic moment and all oriented parallel to the sample plane.

In the next step the 6 ML sample was studied in longitudinal geometry at RT. Symbols in Figs. 2(a) and 2(b) represent the experimental $A(q_z)$ curves for two energies at the L_3-edge together with the best fits shown as red lines. The high quality fits which follow the experimental data in all details correspond to the structure: ⇑⇑⇓⇑⇓⇑ . The upper two layers are FM coupled followed by an AFM coupled third layer. Subsequent layers form units of AFM coupled spins. At this point we emphasize that this grouping of the spins is the basic structure of all samples investigated in the 6 to 8 ML regime, although the relative orientation between the units might be noncollinear.

As compared to the 4 ML sample, the $|m_y(i)|$ values for the individual slices are slightly lower (see Fig. 2 on the right). We tentatively attribute this to the *reduction* of T_C when increasing the coverage from 4 to 6 ML [14]. This involves an enhanced thermal disorder of the spins leading to a reduction of the average magnitude of the spin moments along the L direction.

In order to show the sharp dependence of $A(q_z)$ on the magnetization profile, the blue line is calculated by using FM ordering, while the green line is related to a single layer AFM structure (⇑⇓⇑⇓⇑⇓), in both cases using the same values for $|m_y(i)|$ as for the best fit. Strong deviations from the experimental data are observed clearly ruling out such models.

The 6 ML thick sample magnetized in L geometry represents a special case in which all units are perfectly collinear aligned, however as will be shown next noncollinearity appears by rotation of the spin units relative to each other. As it is known from MOKE experiments, the 6 ML sample shows a SRT to perpendicular magnetization with decreasing temperature. Thus we decided to characterize the magnetization profile at low temperature (40 K) in remanence after the field was applied perpendicular to the sample plane.

Experimental and calculated $A(q_z)$ curves for 6 and 8 ML are shown in Figs. 3(a) and 3(b), respectively. The corresponding magnetization profiles are shown on the right. Here the magnetic profiles involve a noncollinearity of the spins, but the orientation within the units remains fixed (AFM in deeper layers and ⇑⇑⇓ for the upper three layers). The magnetization of the upper layers is oriented out of plane.

The net perpendicular magnetization in remanence is very similar for both thicknesses and the difference originates mainly from the Fe atomic layers at the Fe/Cu interface, which is more in-plane in the case of the 6 ML sample. The value of the net magnetization in perpendicular remanence corresponds to the magnetization of about 2 ML of Fe which is in a good agreement with the MOKE results showing the saturation Kerr signal decreasing after the maximum around 4 ML of Fe [14]. In correspondence with the low sample temperature (40 K) we find large magnitudes of the magnetic moment $|m|$, which equal to 0.9 ± 0.1 $(1.9\mu_B)$ in the interior of the film and $1.3 \pm 0.1(2.7\mu_B)$ and $0.6 \pm 0.1(1.3\mu_B)$ at the top and at the bottom interface, respectively.

The experimental results are supported by DFT calculations taking into account the complete film structure including the Cu-substrate. Modern DFT computer packages allow to study noncollinear magnetic configurations [17]. The calculations were started with various noncollinear magnetic configurations. The magnetic structure varies from iteration to iteration until numerical self-consistency is reached. We found that numerically self-consistent structures obtained at the end of the iteration process depend on the initial configuration. This feature is rather unusual for DFT calculations and reflects the property of fcc Fe to have many quasistable magnetic states.

In agreement with previous studies [9,10] none of the noncollinear magnetic states was lower in energy than the energy of the lowest-energy collinear state. The energy difference between noncollinear and collinear states was, however, typically rather small in the range of 1–2 meV per atom. Most importantly, the analysis of various quasistable states obtained in the calculations shows that they possess a number of common features that reveal a certain hierarchy of the exchange interactions in the fcc-Fe/Cu(001) films: (i) In all cases the magnetic configuration of the three surface layers is very close to collinear. The moments of the upper two layers are parallel to each other and antiparallel to the moment of the third layer. (ii) Deeper layers form pairs with almost antiparallel orientation between the moments. (iii) Surprisingly, the relative orientation of the moments of different groups varies strongly from one

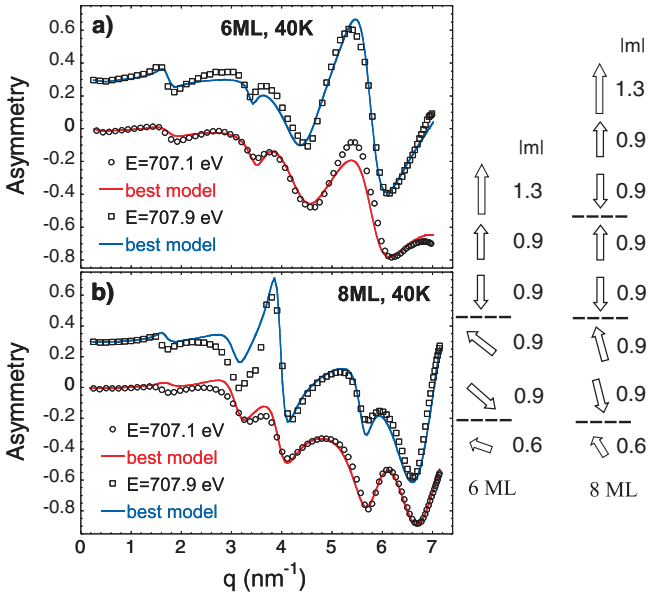

FIG. 3 (color). Experimental (symbols) and calculated (lines) $A(q_z)$ for 6 (a) and 8 ML (b) at 40 K measured in remanence after applying a perpendicular field of $\mu_0 H = 0.4$ T. Magnetization profiles are shown on the right. Dashed lines separate strongly coupled spin units.

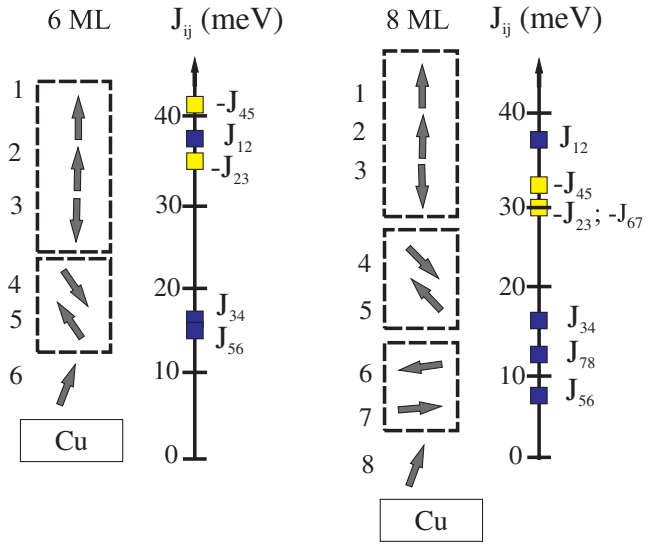

FIG. 4 (color online). Typical quasistable magnetic structures for 6 and 8 ML films obtained from self-consistent DFT calculations. Corresponding exchange interactions parameters (J_{ij}) between nearest neighbor layers are shown on the right. Negative J_{ij} (yellow symbols) indicate AFM coupling. Boxes indicate strongly coupled spin units.

quasistable state to another. Typical quasistable states for 6 and 8 ML films are shown in Fig. 4.

This theoretically derived magnetic structure is in perfect agreement with the experimental results as far as the basic unit structure is concerned. With regard to the relative orientation between the spin units, the calculations cannot account for them since they do not take into consideration the spin orbit coupling and therefore no information about the magnetic anisotropy is provided. The latter is known to be strongly influenced by experimental details such as atomic structure, temperature, etc., and thus might depend on the experimental conditions and the preparation procedure.

To get further insight into the physical origin of these results the interlayer exchange interactions were calculated. Starting with the collinear ground state the moments of layers i and j were, first, both rotated clockwise by angle θ and, second, rotated in opposite directions by the same angle. The energy difference between the two states $\Delta E(\theta)$ is used to estimate the Heisenberg-type parameters of the interlayer exchange interactions $J_{ij}(\theta) = \Delta E(\theta)/[1 - \cos(2\theta)]$. In Fig. 4 we show the nearest neighbor (nn) exchange parameters J_{ij} obtained for films of 6 and 8 ML thickness using the value of angle $\theta = 15°$ [18]. Interactions between more distant layers are weaker by about an order of magnitude and are not shown.

With regard to their magnitude there is a clear separation of the nn exchange parameters into two groups: stronger interactions between layers 1–2, 2–3, 4–5, and 6–7 and weaker interactions between layers 3–4, 5–6, and 7–8. This indicates much stronger exchange interactions within suggested groups of layers compared with the interactions between layers belonging to different groups.

In summary, we have applied a novel experimental technique, XRMS, which allows the direct analysis of the layer resolved spin structure in the fcc Fe films on Cu(001). Our results provide an answer to the long standing problem of the magnetic structure in the coverage regime above 4 ML, which is commonly referred to as ''antiferromagnetic'', but whose detailed structure was never solved. We find a new model, which is characterized by units of strongly coupled spins, whose relative orientation can vary easily involving noncollinearity of the spins. The experimental results are in perfect agreement with DFT calculations. Our study is not only important for fcc Fe films on Cu(001) but also has wide applications for complex magnetic systems in general.

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