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LETTER TO THE EDITOR

Magnetocrystalline anisotropy of ultrathin Fe films on Ni(110)

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Abstract. We determined the magnetic ground-state moments of ultrathin epitaxial Fe films on Ni(110) and found that the overlayer spins orient at an angle of 55° relative to the substrate magnetization direction. This surprising behaviour is a direct consequence of the contribution of orbital magnetic moment to the magnetocrystalline anisotropy.

The spin alignment in a thin ferromagnetic layer caused by the exchange coupling to an underlying magnetic substrate is a very promising tool for increasing the magnetic homogeneity of the film, which is of importance for further device miniaturization in the magnetic recording industry [1]. This exchange biasing induces magnetic order in the overlayer even for very thin films, which often have Curie transitions below room temperature. Thin overlayer films are desirable for applications since their magnetic moments are generally enhanced due to the reduced symmetry. In a simple model the exchange interaction can be thought of as an effective magnetic field which tries to align spins parallel or antiparallel. One would then expect the overlayer spins to be ordered collinearly with the substrate magnetization direction. This effect, however, can be in competition with the magnetocrystalline anisotropy (MCA) trying to orient the overlayer spins in an easy direction which can be different from that of the substrate. In this letter we show that such a reorientation does indeed occur in ultrathin Fe films grown epitaxially on Ni(110). The strong structural anisotropy of this system makes it an ideal candidate for which to study the interplay between the spin orientation, MCA, exchange interaction, crystal field and spin–orbit coupling. In order to do this we combined circular magnetic x-ray dichroism (CMXD) and first-principles calculations in the local spin-density approximation (LSDA) to characterize and understand the magnetic ground state. We find that the difference between the 3d electron occupation numbers of Ni and Fe introduces a strong enough MCA to overcome the exchange interaction with the substrate. The substrate exchange biasing allowed us also to determine the magnetic moments and MCA of individual Fe islands, which has not been done to our knowledge before.

The ground state of a magnetic material can be characterized by its moments, such as the spin and orbital magnetic moment, the quadrupole moment, and the magnetic dipole moment. Band-structure theory can be used to obtain the expectation values $\boldsymbol{S}, \boldsymbol{L}, \boldsymbol{Q}$, and $\boldsymbol{T}$, respectively, of these moments and of the total energy for the spin-quantization axis along any chosen unit vector $\hat{\boldsymbol{S}}$. The spin moment is determined by the occupation numbers $n^{\uparrow(\downarrow)}$ of majority (minority) bands as $\boldsymbol{S} = \frac{1}{2}(n^{\uparrow} - n^{\downarrow})\hat{\boldsymbol{S}}$ and is constant for all directions $\hat{\boldsymbol{S}}$. The anisotropy of the total energy, E_{MCA}, and the presence of an orbital magnetic moment are

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both due to spin–orbit coupling. As implicitly given by, e.g., Bruno [2] in perturbation theory,

$$\boldsymbol{L} = \mathbf{R}\hat{\boldsymbol{S}} \tag{1}$$

where $\mathbf{R}$ is a (second-rank) tensor. Neglecting any spin–orbit-correlated contributions to $\boldsymbol{T}$ we have

$$\boldsymbol{T} = \tfrac{1}{2}(\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow})\hat{\boldsymbol{S}}. \tag{2}$$

So $\boldsymbol{T}$ is also given by a (second-rank) tensor times $\hat{\boldsymbol{S}}$. The tensor relationships in equations (1) and (2) show that $\boldsymbol{L}$ and $\boldsymbol{T}$ are collinear with $\boldsymbol{S}$ only if the spins are oriented along high-symmetry directions, i.e. where $\boldsymbol{S}$ is an eigenvector of the matrices $\mathbf{R}$ and $\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow}$. At the interface of Fe and Ni this is no longer the case and the moments $\boldsymbol{L}$, $\boldsymbol{T}$, and $\boldsymbol{S}$ are oriented in different directions.

Table 1. A comparison of values for orbital and effective spin moments $\boldsymbol{S}^{eff} \equiv \boldsymbol{S} + \frac{7}{2}\boldsymbol{T}$ $(in\ \mu_B)$ for the Fe/Ni(110) films and bulk Fe. The calculated number of holes $n_h = 3.58$ was used to obtain the absolute values.

		L_M	S_M^{eff}	L_M/S_M^{eff}
Bulk Fe	LSDA	0.110	1.10	0.100
	Stearns [8]	0.14	1.12	0.12
	Clemens [7]	0.12	1.02	0.12
	Chen *et al* [6]	0.085	0.99	0.086
Fe/Ni(110)	0.4 ML	0.22 ± 0.04	0.93 ± 0.07	0.23 ± 0.04
	2.0 ML	0.22 ± 0.03	0.97 ± 0.07	0.22 ± 0.03
	3.0 ML	0.18 ± 0.03	0.82 ± 0.07	0.22 ± 0.04

Table 2. Calculated LSDA results for the expectation values of the quadrupole moment for the majority/minority bands, the tensors $\frac{1}{2}(\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow})$ and $\mathbf{R}$, and the spin magnetic moment (in μ_B).

	$x = [1\bar{1}0]$	$y = [001]$	$z = [110]$
$\mathbf{Q}^{\uparrow}$	0.024	−0.008	−0.016
$\mathbf{Q}^{\downarrow}$	0.238	−0.071	−0.176
$\frac{1}{2}(\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow})$	0.106	−0.031	−0.080
$\mathbf{R}$	0.211	0.288	0.242
$\boldsymbol{S}$	1.427	1.427	1.428

For a quantitative evaluation of the moments in equations (1) and (2) and analysis of our experimental results we performed fully relativistic first-principles LSDA calculations using the linear muffin-tin orbital method with the atomic-sphere approximation [3]. Following Skriver *et al* [4] we included the electric dipole Madelung potential which has been found to be important for surfaces. It is well known that LSDA calculations give values for $\boldsymbol{L}$ that are too small compared to experimental values because of an incorrect implementation of Hund's second rule. To overcome this problem, we used an additional orbital polarization term $-\frac{1}{2}BL_M^2$ to maximize $\boldsymbol{L}$ within the mean-field LSDA approach [5]. The Racah coefficient B was determined self-consistently, making our method parameter-free. We obtain good agreement with the magnetic moments of bulk Fe, which have been measured with CMXD

[6, 7] and neutron scattering [8], as shown in table 1. Moments for thin Fe films were calculated using an fcc (110) slab consisting of seven Ni layers covered on both surfaces with one monolayer (ML) of Fe. The Fe–Ni interlayer distance was chosen to be identical to the Ni–Ni layer separations of 1.24 Å. Varying the interlayer spacings did not change the results significantly. The results are summarized in table 2. With the calculated 3d occupation number of 4.64 electrons the majority band is almost completely filled, leading to the small values of $\mathbf{Q}^{\uparrow}$. The minority band contains 1.78 electrons which mainly occupy the $d_{x^2-y^2}$ and d_{xy} orbitals with one-electron quadrupole moments $q_{zz} = -\frac{4}{7}$, $q_{xx} = q_{yy} = \frac{2}{7}$, and the d_{xz} orbital with $q_{yy} = \frac{4}{7}$, $q_{xx} = q_{zz} = \frac{2}{7}$. This results in the strong anisotropy of $\mathbf{Q}^{\downarrow}$.

The calculated total energy of the slab has a minimum for the spins pointing along the [001] crystallographic axis, which happens to be the easy-magnetization axis of bulk bcc Fe. Due to the relationship between E_{MCA} and $\boldsymbol{L}$ [2] the maximum component of $\boldsymbol{L}$ along $\hat{\boldsymbol{S}}$ corresponds to the minimum value of the total energy. This suggests that a thin epitaxial Fe film grown on a Ni(110) surface prefers a magnetization direction different from the one in the Ni substrate which has a $[1\bar{1}1]$ easy-magnetization axis.

For an experimental corroboration we performed CMXD measurements on beamline 1.1 of the Synchrotron Radiation Source at Daresbury with 80% circularly polarized x-rays. The Ni(110) substrate was a picture-frame crystal magnetized along its in-plane $[1\bar{1}1]$ easy axis by a current pulse through a coil. The surface was cleaned in the usual way via Ar-ion sputtering and annealing. Residual C contamination was removed by adsorbing O and subsequent annealing. The surface cleanliness was checked by x-ray photoelectron spectroscopy (XPS) and the surface order with low-energy electron diffraction (LEED). The Fe coverage was determined with XPS. No change in the 1×1 LEED pattern of the clean surface was observed in the coverage range below 3 ML studied here. This and the fact that the XPS Fe uptake curve can be described by straight-line segments with a break point around 2 ML indicate that the Fe film is well ordered and justify the assumption in the LSDA calculations of an fcc-like thin film.

Figure 1 shows a typical Fe $L_{2,3}$ x-ray absorption spectrum (XAS) monitored by detecting the sample drain current. The spectrum was taken with the helicity vector at 45° to the $[1\bar{1}1]$ surface direction (see the inset in figure 1). In order to take only transitions into empty 3d states into account a background due to transitions into 4s orbitals has to be subtracted. Since the detailed shape of this background is unknown we used the step function indicated in figure 1 by the dotted line. The CMXD (XAS) spectrum is the difference (sum) in absorption with the photon helicity and the sample magnetization aligned parallel and antiparallel with areas ΔA (A). For the ground-state expectation values of the moments along the direction of the incident light $\hat{\boldsymbol{P}}$ the sum rules [9, 10] give

$$\hat{\boldsymbol{P}} \cdot \boldsymbol{L} = -\frac{4}{3}\frac{\Delta A_3 + \Delta A_2}{A_3 + A_2} n_h \tag{3}$$

$$\hat{\boldsymbol{P}} \cdot \boldsymbol{S}^{eff} \equiv \hat{\boldsymbol{P}} \cdot \boldsymbol{S} + \tfrac{7}{2}\hat{\boldsymbol{P}} \cdot \boldsymbol{T} = \frac{\Delta A_3 - 2\Delta A_2}{A_3 + A_2} n_h \tag{4}$$

where we can obtain the components $\hat{\boldsymbol{P}} \cdot \boldsymbol{L}$ etc by using the calculated value $n_h = 3.58$ for the number of 3d holes. As we will show below, $\boldsymbol{S}$, and hence $\boldsymbol{L}$ and $\boldsymbol{T}$, are in the surface plane, so we can simplify the discussion using

$$\hat{\boldsymbol{P}} \cdot \boldsymbol{L} = \hat{\boldsymbol{P}}_{\parallel} \cdot \boldsymbol{L} = \hat{\boldsymbol{M}} \cdot \boldsymbol{L} \cos 45^{\circ} \equiv \boldsymbol{L}_M \cos 45^{\circ} \tag{5}$$

where $\hat{\boldsymbol{P}}_{\parallel}$ is the projection of $\hat{\boldsymbol{P}}$ on the surface plane and $\hat{\boldsymbol{M}}$ is a unit vector along $\hat{\boldsymbol{P}}_{\parallel}$. The values of $\boldsymbol{L}_M$ and $\boldsymbol{S}_M^{eff}$ are listed in table 1.

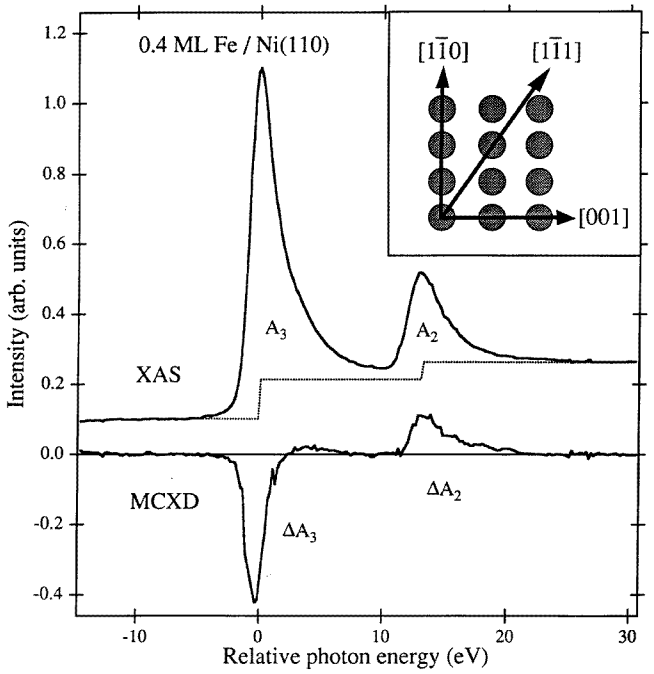

Figure 1. The sum (XAS) of and difference (CMXD) between electron yield spectra with photon helicity parallel and antiparallel to the magnetization direction. The CMXD spectrum is corrected for the 45° light incidence angle. The inset shows a top view of the Ni(110) surface.

If we first assume $\boldsymbol{L}$ and $\boldsymbol{S}^{eff}$ to be along $\hat{\boldsymbol{M}} = [1\bar{1}1]$ we find a 50 to 100% increase in magnitude of the orbital moment $\boldsymbol{L}$ for the thin Fe films on Ni(110) compared to bulk measurements with CMXD [6, 7] and neutron scattering [8]. This dramatic enhancement is in agreement with the calculation and indicates the different electronic structure of the fcc-like film compared to that of the bcc bulk. On the other hand we find $\boldsymbol{S}^{eff}$ close to the bulk value (where $\boldsymbol{T}$ may be neglected and so $\boldsymbol{S}^{eff} = \boldsymbol{S}$), whereas it is well established that the spin moment is enhanced in thin Fe films [11] and also our calculation predicts a 30% increase. Finally we find a slight increase with decreasing coverage of the moments listed in table 1. However, due to the large systematic errors introduced by the n_h-weighting, the variation remains within the experimental uncertainty.

The low magnitude of $\boldsymbol{S}^{eff}$ relative to that of $\boldsymbol{L}$ indicates that the increase in the spin moment of our thin film compared to that of Fe bulk is compensated by $\boldsymbol{T}$, keeping $\boldsymbol{S}^{eff}$ low in magnitude. It is not trivial to take $\boldsymbol{T}$ into account because $\boldsymbol{L}$, $\boldsymbol{S}$, and $\boldsymbol{T}$ are in general not collinear, except when they are along a symmetry axis. Although our calculation gives the [001] symmetry axis as the easy direction, we have to consider the possibility that the actual direction of $\boldsymbol{S}$ in the surface layer is different due to the interaction with the underlying Ni magnetized in the $[1\bar{1}1]$ direction. $\boldsymbol{S}$ may then not be along a main axis and due to equations (1) and (2) $\boldsymbol{L}$, and especially $\boldsymbol{T}$, can enclose a large angle with $\boldsymbol{S}$.

An important consequence of the anisotropy of $\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow}$ and $\mathbf{R}$ is that $\boldsymbol{L}_M/\boldsymbol{S}_M^{eff}$ is not a constant but can have any value. In figure 2 we show the values of $\boldsymbol{S}_M^{eff}$, $\boldsymbol{L}_M$, and $\boldsymbol{L}_M/\boldsymbol{S}_M^{eff}$ calculated using equations (1) and (2) with the components $\mathbf{Q}^{\downarrow} - \mathbf{Q}^{\uparrow}$ of and $\mathbf{R}$ in table 2 for all directions of $\boldsymbol{S}$ in the surface plane. Indeed $\boldsymbol{S}_M^{eff}$ and $\boldsymbol{L}_M$ have their

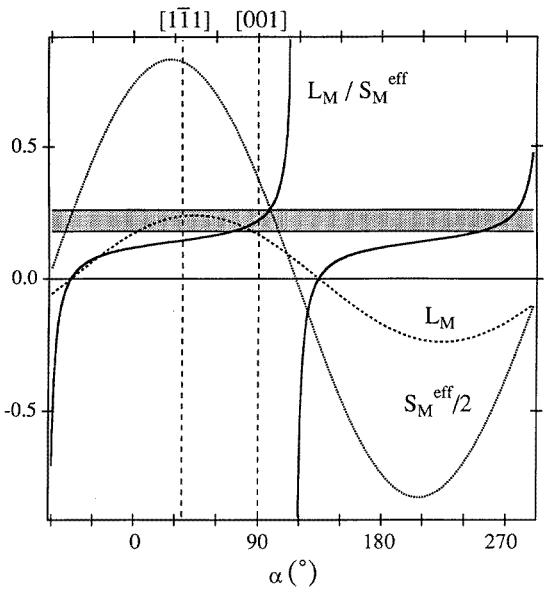

Figure 2. The orbital and effective spin magnetic moment (in μ_B) and their ratio projected onto the $\boldsymbol{M} = [1\bar{1}1]$ direction versus the in-plane magnetization direction. The calculated ratios are compared with the experimental value (the shaded horizontal band).

extremes and zeros at different angles and so $\boldsymbol{L}_M/\boldsymbol{S}_M^{eff}$ is not a constant. The experimental $\boldsymbol{L}_M/\boldsymbol{S}_M^{eff}$ is indicated by the shaded band. We use this ratio because it does not suffer from uncertainties connected to n_h and allows a more accurate comparison of experiment and LSDA calculations. We see that the theoretical and experimental ratios agree at around [001], which was also the easy direction obtained in the calculation. A [1$\bar{1}$1] spin direction, on the other hand, is clearly outside the experimental error. Magnetization normal to the surface can also be excluded since the spins would then be perpendicular to the substrate magnetization direction and the film would split up into magnetic domains with both up and down directions resulting in a net zero overlayer magnetization.

This is a surprising result and shows clearly that for Fe/Ni(110) the MCA is strong enough to overcome the exchange interaction with the Ni(110) substrate. Since the clean Ni(110) surface is known to have an easy [1$\bar{1}$1] axis the change to [001] upon Fe deposition can be mainly attributed to the difference in 3d-band filling. Such a behaviour is evident from the relationship of E_{MCA} and $\boldsymbol{L}$ [2]. An increased 3d occupation number reduces the size of $\boldsymbol{L}$ and consequently E_{MCA}, thus enabling the exchange biasing to orient the surface spins along the bulk easy axis. On a non-magnetic substrate long-range ferromagnetic order sets in only after a continuous film is formed by island coalescence [12]. It is, therefore, interesting that the observed spin reorientation not only happens for the films that are several ML thick but also for Fe islands on the surface. The exact size of these islands formed at submonolayer coverage of course depends on the deposition parameters and the growth mode. But our lowest measured coverage of 0.4 ML is small enough to ensure that island coalescence does not occur [12]. Our results show that Fe atoms in the islands have the same orbital and spin moment as in the continuous film and do not adopt random spin orientations but are still parallel or antiparallel to the easy [001] magnetization axis.

In conclusion we have studied thin Fe films epitaxially grown on the fcc Ni(110) surface

with circular magnetic x-ray dichroism and first-principles calculations, and found strongly enhanced values for the magnetic ground-state moments compared to those for bcc bulk Fe. We showed that the magnetic dipole moment is directly related to the quadrupole moment. The crystal structure of the Fe/Ni(110) system imposes a strong anisotropy onto the quadrupole moment and consequently onto the magnetic dipole term which is comparable in size to the spin moment. This enabled us to separate the spin and dipole moment and to establish a different spin orientation in the Fe overlayer compared to that of the Ni substrate. We used the exchange biasing of the substrate to determine the magnetic properties of the Fe film at submonolayer coverage and found the spin reorientation to persist to this regime.

References

[1] Speriosu V S, Herman D A, Sanders I L and Yogi T 1990 *IBM J. Res. Dev.* **34** 884
[2] Bruno P 1989 *Phys. Rev.* B **39** 865
[3] Andersen O K 1975 *Phys. Rev.* B **12** 3060
Temmermann W M, Sterne P A, Guo G Y and Szotek Z 1989 *Mol. Simul.* **4** 153
[4] Skriver H L and Rosengard N M 1991 *Phys. Rev.* B **43** 9538; 1992 *Phys. Rev.* B **46** 7157
[5] Eriksson O, Johansson B, Albers R C, Boring A M and Brooks M S S 1990 *Phys. Rev.* B **42** 2707
[6] Chen C T, Idzerda Y U, Lin H-J, Smith N V, Meigs G, Chaban E, Ho G H, Pellegrin E and Sette F 1995 *Phys. Rev. Lett.* **75** 152
[7] Clemens W *Thesis* University of Cologne
[8] Stearns M B 1986 *Magnetic Properties of 3d, 4d and 5d Elements, Alloys and Compounds, Landolt–Börnstein, New Series* Group III, vol 19a, ed K-H Hellwege and O Madelung (Berlin: Springer) and references therein
[9] Thole B T, Carra P, Sette F and van der Laan G 1992 *Phys. Rev. Lett.* **68** 1943
[10] Carra P, Thole B T, Altarelli M and Wang X 1993 *Phys. Rev. Lett.* **69** 2307
[11] Bland J A C, Daboo C, Heinrich B, Celinski Z and Bateson R D 1995 *Phys. Rev.* B **51** 258
[12] Elmers H J, Hauschild J, Höchle H, Gradmann U, Bethge H, Heuer D and Köhler U 1994 *Phys. Rev. Lett.* **73** 898