# Strongly Enhanced Orbital Moment by Reduced Lattice Symmetry and Varying Composition of $Fe_{1-x}Co_x$ Alloy Films

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We studied tetragonally distorted $Fe_{1-x}Co_x$ alloy films on Rh(001), which show a strong perpendicular anisotropy in a wide thickness and composition range. Analyzing x-ray magnetic circular dichroism spectra at the $L_{3,2}$ edges we found a dependence of the Co magnetic orbital moment on the chemical composition of the $Fe_{1-x}Co_x$ alloy films, with a maximum at $x = 0.6$. For this composition, we observed an out-of-plane easy axis of magnetization at room temperature for film thickness up to 15 monolayers. Since both the magnetic orbital moment and the anisotropy energy show similar composition dependence, it confirms that both quantities are directly related. Our experiments show that the adjustment of the Fermi level by a proper choice of the alloy composition is decisive for the large magnetic orbital moment and for a large magnetic anisotropy in a tetragonally distorted lattice.

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Magnetic anisotropy is related to the magnetic orbital moment ($m_l$) and spin-orbit interaction. Because of symmetry reasons, almost no orbital moment remains in bulk ferromagnets of cubic symmetry. The quenching of the orbital moment can be removed by an appropriate symmetry reduction. The unquenched orbital magnetization is then accompanied by a significant anisotropy of the orbital moment itself, which in turn induces a strong magnetic anisotropy due to spin-orbit coupling [1–3]. In transition metal atoms with a more than half filled $d$-electron shell, the easy magnetization axis is the direction where $m_l$ is largest. Thus, an increasing value of $m_l$ probed along the easy axis is a measure of the increasing anisotropy of $m_l$. The relation between the increasing orbital moment and its anisotropy on one side and the increasing magnetic anisotropy on the other was previously observed for thin Co films on Au(111) [4], Co interfaces in Co/Ni, Co/Pd, and Co/Pt multilayers [5] and for small Co clusters [6]. However, in all these cases the orbital moment and the perpendicular anisotropy were enhanced due to the reduced coordination or due to interface hybridization. Different values of the orbital to spin moment ratio for out-of-plane and in-plane magnetization were also found for Ni in fcc-Co/Ni sandwiches grown on Cu(001) [7].

In this Letter we demonstrate experimentally that the orbital magnetic moment and the magnetic anisotropy can be increased due to a tetragonal distortion in a system with bulklike coordination. A model system is provided by $Fe_{1-x}Co_x$ alloy films, which are distorted due to their pseudomorphic growth on substrates of mismatching lattice constant. No coordination reduction or electronic hybridization at an interface is required in this case. Tetragonal distortion is predicted to reduce the energy separation between the $d_{xy}$ and $d_{x^2-y^2}$ in-plane orbitals [8]. Since in second-order perturbation theory the orbital magnetic moment is inversely proportional to the energy separation between the levels located below and above the Fermi level ($E_F$), the orbital moment could be enhanced due to the distortion. Suitable, tetragonally distorted $Fe_{1-x}Co_x$ alloys were recently realized by growing FeCo/Pt multilayers [9], and by growing $Fe_{1-x}Co_x$ films on a Pd(001) substrate [10]. In both cases, a largely increased perpendicular anisotropy was found experimentally for specific compositions. In the present investigation, we have chosen $Fe_{1-x}Co_x$ alloy films grown on a Rh(001) substrate. This system is a promising candidate for showing a distortion that is near the value of $c/a = 1.22$ with a predicted maximum uniaxial magnetic anisotropy energy of the order of 700–800 $\mu$eV per atom in $Fe_{0.4}Co_{0.6}$ alloys (with $c/a = \sqrt{2}$ for fcc) [8]. The composition of the alloy determines the number of valence electrons, and thus the value of $E_F$ which can be systematically tuned by varying the composition. These alloys are expected to keep a strong perpendicular anisotropy over a wide chemical composition range and to show how the orbital moment behaves with varying Co content. Since the orbital moment is probed by x-ray magnetic circular dichroism (XMCD) spectroscopy, we profit from the element selectivity of the method and measure the orbital moments for both constituent elements. In particular, we show that the orbital moment of Co is strongly dependent on the $Fe_{1-x}Co_x$ alloy composition.

The $Fe_{1-x}Co_x$ alloy films were grown in a multichamber ultrahigh vacuum system with a base pressure better than $5 \times 10^{-11}$ mbar and less than $2 \times 10^{-10}$ mbar during deposition. The Rh(001) substrates were prepared using sputter-anneal cycles. The quality of the sample was checked by Auger electron spectroscopy and by scanning tunneling microscopy until a clean surface with nearly equidistant, parallel monatomic steps was obtained. The

 

films were grown at room temperature (RT) by molecular beam epitaxy using two effusion cells as described previously [10]. Low energy electron diffraction (LEED), diffracted intensities vs incident electron energy analysis [$I(V)$-LEED] and Kikuchi patterns of quasielastically backscattered electrons were used to assess quantitatively the amount of tetragonal distortion. Magnetic properties were probed by utilizing the *in situ* magneto-optical Kerr effect (MOKE) and the XMCD at the $L_{2,3}$ absorption edges. XMCD in soft x-ray absorption measurements of the $Fe_{1-x}Co_x$/Rh(001) samples were carried out at the UE56/2-PGM2 beam line at BESSY in Berlin. Absorption spectra were recorded by directly detecting the sample current while scanning the photon energy of the 80% circularly polarized light. The energy resolution was set to 0.25 eV for absorption measurements at both the Fe and Co $L_{3,2}$ edges. XMCD measurements were performed in magnetic remanence after a magnetic field pulse of about 50 mT along the normal of the film plane has been applied (no magnetic field could be applied during the XMCD measurements with our experimental setup).

Both Fe and Co films are subject to lattice misfit when deposited on Rh(001). Namely, while the in-plane lattice constant of Rh(001) amounts to $a_{\mathrm{Rh}} = 2.68$ Å, the corresponding lattice constants of Fe and Co are $a_{\mathrm{Fe}} = 2.54$ Å and $a_{\mathrm{Co}} = 2.51$ Å, respectively, resulting in misfit values of $-5.2\%$ for fcc Fe and $-6.3\%$ for fcc Co [11]. As the Rh(001) in-plane lattice constant is larger, the overlayer lattice has to expand in-plane for pseudomorphic growth. To keep the atomic volume constant, the film contracts in the direction of the $c$ axis, which leads to a $c/a$ ratio of less than $\sqrt{2}$.

The spot positions of the LEED patterns of the films prove a pseudomorphic growth of at least large parts of the $Fe_{1-x}Co_x$ film even at a thickness of 10 monolayer (ML) in the composition range ($0.35 < x < 0.65$). The analysis of the $I(V)$-LEED data and the forward scattering maxima in the Kikuchi patterns at 1 keV [10] gave for the $c/a$ ratio the value of $1.24 \pm 0.04$ for the 10 ML thick $Fe_{0.4}Co_{0.6}$ film [12]. This is similar to the previous measurements for pure Fe and Co films grown on Rh(001), which resulted in $c/a$ ratios of 1.18 for Fe [13] and 1.20 for Co [14] for a little thinner films. Note that the $c/a = 1.24$ is close to 1.22 for which the maximum uniaxial magnetic anisotropy is expected [8]. For the concentration range $0.35 < x < 0.65$ the $c/a$ ratio was found to be only weakly dependent on the Co content. With increasing film thickness, the part of the film that is ordered pseudomorphically to the substrate contributes less to the total film volume and thus does not determine the film properties above a certain thickness.

We found that $Fe_{1-x}Co_x$ alloy films of the composition $x = 0.3$ to $x = 0.6$ show a clear out-of-plane easy axis of magnetization; i.e., rectangular polar MOKE loops were measured up to the thickness $d_c$, which depends on the film composition as shown in Fig. 1 [12]. The easy axis of magnetization changes from perpendicular to in-plane within a thickness of a few monolayers. In this transition thickness range (up to 3–4 ML above $d_c$) the polar remanence decreases (and the polar loops become hard-axis–like), whereas the longitudinal remanence increases (and the longitudinal loops become rectangular). For the alloy films in which the Co content ($x$) is smaller than $x = 0.3$ or larger than $x = 0.6$, an in-plane easy axis of magnetization is detected irrespectively of the film thickness. This is in qualitative agreement with the theoretical values of the uniaxial anisotropy constant $K_u$ calculated for $c/a = 1.24$ [8] and shown in Fig. 1 for comparison. Positive values of $K_u$ exceeding the shape anisotropy could result in the easy magnetization axis being perpendicular to the film plane. This is different from the $Fe_{1-x}Co_x$ films grown on Pd(001) exhibiting the $c/a$ ratio of 1.13, where a perpendicular easy magnetization axis was found only around $x = 0.5$ composition and only at low temperature [10].

Since pure Fe and Co films grown on Rh(001) are spontaneously magnetized in-plane, it is reasonable to assume that the surface and Fe-Co/Rh interface anisotropies do not contribute significantly to the perpendicular anisotropy of the $Fe_{1-x}Co_x$/Rh(001) system. Also, the shape anisotropy is only weakly dependent on the composition due to slightly decreasing saturation magnetization with increasing Co content. Then we can take $d_c$ as directly

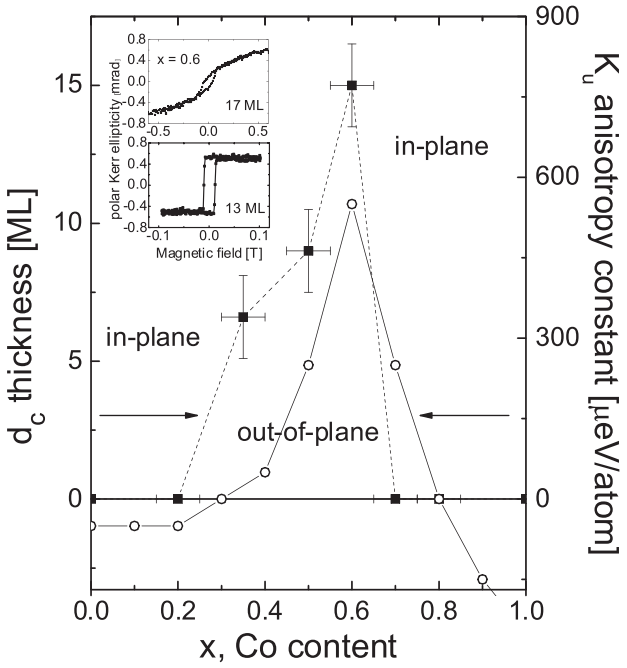


FIG. 1. Maximum thickness up to which the magnetization of the $Fe_{1-x}Co_x$ alloy films grown on Rh(001) is oriented out-of-plane at room temperature ($d_c$) versus the film composition—full points; the dotted line is a guide for the eyes. The inset shows typical polar MOKE loops for the $x = 0.6$ film measured below and just above $d_c$. Note that the field scale is different for 13 and 17 ML thick films. Theoretical values of the uniaxial anisotropy constant $K_u$ calculated for $c/a$ ratio of 1.24 (open circles) are taken from Fig. 1 in [8].

related to the volume magnetocrystalline anisotropy: the larger the anisotropy is, the larger is the thickness $d_c$. We stress that only if the film thickness is kept constant, the anisotropy can be considered as depending on the chemical composition only. With changing film thickness, the distortion will also change and influence the magnetocrystalline anisotropy independently of the composition.

The magnetic orbital moments of Co and Fe in $Fe_{1-x}Co_x$/Rh(001) films were investigated by using element-selective XMCD spectroscopy. Figure 2 shows the representative XMCD spectra at the Co and Fe $L_{3,2}$ edges, respectively, recorded at RT for 6 ML thick films of different composition. Absorption spectra were recorded at the incident photon direction parallel to the film normal and also at other incidence angles to verify the direction of remanent magnetization. A reduction of the XMCD signal by the direction cosine was found, ensuring that the easy magnetization axis is normal to the film plane. There is a clear composition dependence of the $L_3$ and $L_2$ XMCD for both Co and Fe.

A more quantitative analysis of the data is achieved by means of the sum rule, which links the integrated intensity differences at the $L_3$ and $L_2$ edges to the atomic orbital and spin moments. This intensity was assumed to correspond to 2.55 unoccupied $d$ states for Co and 3.34 for Fe [15]. The values of the spin moment $m_s$, orbital moment $m_l$, and the ratio between $m_l$ and $m_s$ for Co and Fe atoms obtained at RT for the $Fe_{1-x}Co_x$/Rh(001) films of varying composition are plotted in Fig. 3. The error bars shown there do not include uncertainty in the degree of circular polarization and in the number of unoccupied $3d$ states.

For the 6 ML thick $Fe_{1-x}Co_x$/Rh(001) films with $0.35 < x < 0.6$, 100% remanent magnetization was found at RT, indicating the preservation of the spin alignment along the perpendicular field applied before. In the case of $x = 0.65$ film, however, the easy axis of magnetization is not normal to the film plane anymore. Thus, this film was not magnetically saturated in remanence, and only the projections of the Co and Fe moments to the film normal were probed. The saturation values of $m_s$, which is, in fact, the effective spin moment including also the dipolar term, show a rather weak composition dependence both in the case of Co and Fe. The application of the sum rule gives a magnetic spin moment of $(1.60 \pm 0.09)\mu_B$ per Co atom and $(2.13 \pm 0.13)\mu_B$ per Fe atom. It is worth noting that the average magnetic spin moment is of the order of $1.89\mu_B$ for $x = 0.6$, which is less than the theoretical value expected for the $Fe_{1-x}Co_x$ alloys of this composition. However, it must be taken into account that our XMCD spectra were measured at RT for films of only 6 ML thickness, and thus the values of the magnetic moment could be underestimated due to the magnetic finite-size effect. The thickness of 6 ML was chosen to cover a wide composition range in which the remanent magnetization points out of the film plane.

In contrast to the cubic structure where the orbital moment is nearly completely quenched, a strong enhancement

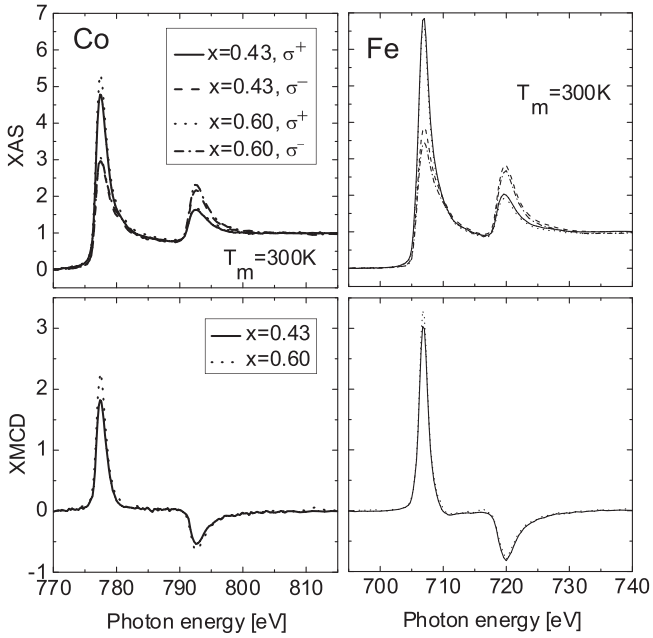


FIG. 2. X-ray absorption spectra (XAS) and corresponding XMCD spectra at the $L_{3,2}$ absorption edge of Co and Fe. The absorption spectra were recorded at $T_m = 300$ K with circularly polarized light of opposite helicity, $\sigma^+$ and $\sigma^-$, at normal incidence. The spectra are shown for 6 ML thick $Fe_{1-x}Co_x$ films of two different $x$.

of the Co orbital moment, $m_l$, occurs in the tetragonally distorted $Fe_{1-x}Co_x$ films. Since the XMCD experiments were performed for films of constant thickness, an increase of the magnetic orbital moment with increasing Co content is related to the adjustment of the Fermi level with respect to the energy levels of the relevant orbitals. The value of the orbital moment of roughly $0.32\mu_B$ at $x = 0.6$ is larger than the largest interface value measured for Co/Pd multilayers with perpendicular anisotropy [5]. The increasing orbital moment probed along the film normal, which is the distortion axis and the easy axis of magnetization, immediately implicates that also the anisotropy itself must increase because the symmetry and thus the orbital moment is not changed in the film plane. In the case of the $x = 0.65$ film, the measured values of $m_l$ are reduced in comparison to the value obtained for $x = 0.6$ both for Co and Fe since only the projections of the moments on the film normal were probed. In order to extract a possible change of $m_l$ itself, the $m_l/m_s$ ratio was calculated and plotted vs film composition in Fig. 3(c). For $x = 0.65 \pm 0.05$, the $m_l/m_s$ is found to be reduced in comparison to the $x = 0.6$ film of the same thickness. Since there is no reason for $m_s$ to differ remarkably between $x = 0.65$ and $x = 0.6$, a reduced value of $m_l$ at $x = 0.65$ can be concluded.

The composition dependencies of the orbital moments and of the $m_l/m_s$ ratios of Co and Fe (Fig. 3) coincide well with the composition dependence of $d_c$, which is related to the strength of the uniaxial magnetocrystalline anisotropy (Fig. 1). Both $m_l/m_s$ and $d_c$ increase up to $x = 0.6$ and

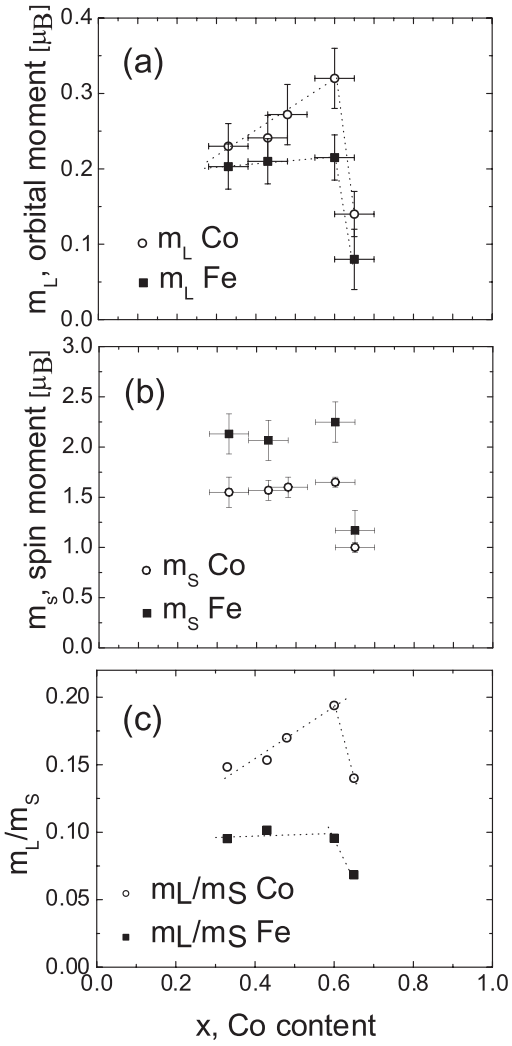


FIG. 3. Results of the sum rule application for the magnetic orbital (a) and spin moments (b) (in units of $\mu_B$), and the ratio between the orbital and spin moments ($m_l/m_s$) (c) of Fe and Co, measured in remanence for 6 ML thick films at RT, plotted vs film composition $x$. The dotted lines are guides for the eyes.

start to decrease above this composition. For $x = 0.65$, the 6 ML thick film does not exhibit perpendicular magnetization ($d_c < 6$ ML), and $m_l/m_s$ is reduced below its maximum value that is reached at $x = 0.6$. A larger orbital magnetic moment forces the spin moment stronger to a perpendicular alignment, and consequently the magnetic anisotropy energy is larger.

The values of the orbital moment $m_l$ show a strong composition dependence in the case of Co, whereas the orbital moment of Fe shows a weaker dependence. The orbital moment is expected to change drastically when $E_F$ is moving by varying the number of valence electrons [3]. Our experimental result reflects a tendency known from the calculations for fcc(001) monolayers of Fe, Co, and Ni [3]. The calculated orbital magnetic moment depends on the number of valence electrons and shows a clear maximum for Co. Also, in the case of multilayers and clusters, a strong magnetic anisotropy of Co rather than of Fe is observed. Moreover, by using Bruno's formula [3] we can estimate the spin-orbit anisotropy energies of the Co atoms, which coincide with the values expected for the $Fe_{1-x}Co_x$ films of this composition and this distortion range [8]. One could say that the $Fe_{1-x}Co_x$ alloys combine a large saturation magnetization determined mostly by Fe and a large uniaxial magnetocrystalline anisotropy energy determined mostly by Co.

To summarize our results, we show that a strong perpendicular anisotropy, outbalancing the shape anisotropy, can be achieved for $Fe_{1-x}Co_x$ alloy films on Rh(001) at room temperature in a wide composition and thickness range. This allowed us to measure the magnetic orbital moment along the easy magnetization axis for relatively thick films (6 ML) in remanence over a wide composition range. At constant thickness, the magnetic orbital moment of Co depends significantly on the film composition. The composition dependence of the orbital magnetic moment coincides with a similar dependence of the calculated [8] and experimentally estimated (by the thickness $d_c$ up to which the magnetization is oriented out-of-plane) uniaxial anisotropy energy both showing a maximum at $x = 0.6$.

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