Volume and interface magnetic anisotropy of Fe$_{1-x}$Co$_x$ thin films on GaAs(001)

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Epitaxial Fe$_{1-x}$Co$_x$ alloy films with $x=0.22$ and $x=0.33$ were grown on GaAs(001) by molecular-beam epitaxy in a thickness range of 3 to 80 monolayers (ML). The magnetic properties were investigated by magneto-optic Kerr effect. Ferromagnetic order at room temperature was observed for thicknesses above 4 ML. The in-plane magnetic anisotropy of all films is a superposition of a uniaxial component with the easy axis along [110], which is not discussed here, and a four-fold contribution. The effective uniaxial and four-fold anisotropy constants, $K_{1 \text{eff}}$ and $K_{2 \text{eff}}$, were determined by fits to the hard axis magnetization loops. $K_{1 \text{eff}}$ contains an interface and a volume term which lead to a linear variation of $K_{1 \text{eff}}$ with the inverse film thickness. It turns out that the surface and volume anisotropy constants are of opposite sign for all alloy compositions causing a sign reversal at a critical thickness, $t_{\text{crit}}$. This critical thickness seems to be a universal value, which is caused by a general proportionality between the volume and the interface anisotropy constants with the same negative constant of proportionality for Fe$_{1-x}$Co$_x$/GaAs(001), Fe/Au(001), and Fe/Ag(001). This behavior of the four-fold anisotropy constants is consistently explained within Néel’s pair energy model for a body-centered-cubic ferromagnet. © 2002 American Institute of Physics. [DOI: 10.1063/1.1447207]
A pure interface-induced effect. The uniaxial interface anisotropy constant decreases slightly with increasing Co content, but is not discussed further in this contribution. $K_{\text{eff}}$ depends on both, thickness and alloy composition as can be seen in Fig. 1, where $K_{\text{eff}}$ for different compositions ($x = 0$ to $x = 0.66$) is plotted versus the inverse film thickness. $K_{\text{eff}}$ varies linearly with inverse thickness, which is a consequence of the superposition of a volume and an interface contribution. The data can be fitted by the following linear expression:

$$K_{\text{eff}} = K_{\text{vol}} + K_{\text{int}}/t,$$

where $t =$ film thickness, $K_{\text{vol}}$ is the volume, and $K_{\text{int}}$ is the interface term of the magnetic anisotropy, including contributions of the Fe$_{1-x}$Co$_x$/GaAs interface as well as of the Fe$_{1-x}$Co$_x$/Au interface: $K_{\text{int}} = K_{\text{int,Fe/GaAs}} + K_{\text{int,Au}}$ (see next). For thinner films, $K_{\text{int}}$ becomes more and more dominant and leads to a change of sign of $K_{\text{eff}}$ due to the opposite sign of $K_{\text{vol}}$ and $K_{\text{int}}$. Another possibility to change the sign of $K_{\text{eff}}$ is to vary the composition of the Fe$_{1-x}$Co$_x$ alloys. $K_{\text{vol}}$, the intercept of $K_{\text{eff}}$ for $t \to \infty$, decreases with increasing Co content and becomes negative for alloys with $x > 0.3$, thus being roughly in agreement with measurements for bulk alloys. The difference between positive and negative $K_{\text{eff}}$ shows up in the opposite curvature of the hard axis loops. In agreement with measurements for bulk alloys, the different anisotropy was found with nearly the same critical thickness ($\approx 7$ ML), it seems that the correlation between $K_{\text{vol}}$ and $K_{\text{int}}$ is independent of the particular interface. This also explains why the presence of two different interfaces (Fe$_{1-x}$Co$_x$/GaAs and Fe$_{1-x}$Co$_x$/Au) does not effect the observed $K_{\text{int}}$. However, the universal behavior of ultrathin Fe films on a variety of substrates rules out such a mechanism. Also, magnetoelastic interaction can be ruled out as the dominating contribution, because the value of $K_{\text{int}}$ observed in films under compressive strain (Fe/GaAs) and under tensile strain (Fe/Au) is practically the same.

Since the four-fold anisotropy described here has not been calculated within ab initio theories, a qualitative interpretation within Néel’s pair energy model is attempted. In Néel’s theory, the anisotropy energy is described as the sum of interaction energies of pairs of atoms in the crystal lattice. The interaction energy between two atoms at a distance $r$ is expanded in a series of Legendre polynomials:

$$w = g_2(r) \left( \cos^2 \phi - \frac{1}{3} \right) + g_4(r) \left( \cos^4 \phi - \frac{6}{7} \cos^2 \phi + \frac{3}{35} \right) + \ldots.$$  

Higher terms are neglected, because it is assumed that the series expansion converges rapidly $[g_2(r) \gg g_4(r) \gg g_6(r)]$. $\phi$ is the angle between the unit vector of the magnetization direction $\hat{m}$ and the pair axis unit vector $\hat{p}$ of the two atoms: $\cos(\phi) = p \cdot m$. The interaction energies, only depending on the distance $r$, the pseudo-dipole coefficient $m \cdot \hat{r}$ and the pseudo-quadrupole coefficient $q \cdot \hat{s}$ are the linear interactions. For a distorted lattice, the general expression (3) is simplified in the following way: First, $m \cdot \hat{r}$ and $q \cdot \hat{s}$ can be neglected because the lattice mismatch of 1.4% between Fe$_{1-x}$Co$_x$ and GaAs is rather small and independent of $x$. Second, we are interested only in the four-fold anisotropies within Néel’s model, so we do not consider the isotropic and the two-fold contributions, which are expressed by the $\cos^2 \phi$ terms. Third, we assume the...
magnetization to always lie in the (001)-plane, which means that $m_z = 0$. Then the interaction energy for an atom pair becomes:

$$w_{4} = q(\rho_{x} m_x + \rho_{y} m_y). \quad (4)$$

In order to calculate the anisotropy energy density $\varepsilon_{\text{bulk}}$ inside the crystal, we add up all contributions of the nearest and next nearest neighbor pairs (all other pairs being neglected). With the underlying bcc lattice of the ferromagnet, we get

$$\varepsilon_{\text{bulk}} = \frac{2}{a_0} \left[ \sum_{\text{NN}} w_{4}^{\text{NN}} + \sum_{\text{NNN}} w_{4}^{\text{NNN}} \right]$$

$$= \frac{2}{a_0} \left[ \frac{1}{2} q^{\text{NN}} \sum_{\text{NN}} (\rho_{x} m_x + \rho_{y} m_y)^4 + \frac{1}{2} q^{\text{NNN}} \sum_{\text{NNN}} (\rho_{x} m_x + \rho_{y} m_y)^4 \right]$$

$$= \frac{2}{a_0} \left[ \frac{16}{9} q^{\text{NN}} - 2q^{\text{NNN}} \right] m^2_{x} m^2_{y}, \quad (5)$$

where $a_0$ is the lattice constant, $q^{\text{NN}}$ the interaction energy of nearest neighbors, and $q^{\text{NNN}}$ the interaction energy of next nearest neighbors.

From the comparison of Eq. (5) with $\varepsilon_{\text{bulk}} = K_1^{\text{vol}} a_1^2 a_2^2$, we obtain:

$$K_1^{\text{vol}} = \frac{2}{a_0} \left( \frac{16}{9} q^{\text{NN}} - 2q^{\text{NNN}} \right). \quad (6)$$

The anisotropy energy density of the interface can be calculated in the same way, but now for an atom at the (001) interface, one next nearest and four nearest neighbors are missing. Furthermore, it must be taken into account that the interface anisotropy energy, as introduced, is the difference between the anisotropy energy of an interface and a bulk atom. Hence, we obtain

$$\varepsilon^{\text{int}} = -\frac{1}{a_0} \left[ \sum_{\text{NN}} (w_{4}^{\text{NN}} - w_{4}^{\text{NN}}) + \sum_{\text{NNN}} (w_{4}^{\text{NNN}} - w_{4}^{\text{NNN}}) \right]$$

$$= -\frac{8}{9a_0} q^{\text{NN}} m^2_{x} m^2_{y}; \quad (7)$$

and

$$K_1^{\text{int}} = -\frac{8}{9a_0} q^{\text{NN}}. \quad (8)$$

It can be seen, that the next nearest neighbors do not affect the in-plane anisotropy constant $K_1^{\text{int}}$ at a bcc (001)-interface. For the ratio $K_1^{\text{int}} / K_1^{\text{vol}}$, we get

$$K_1^{\text{int}} / K_1^{\text{vol}} = \frac{-2q^{\text{NN}} a_0}{8q^{\text{NN}} - q^{\text{NNN}}} \quad (9)$$

It is obvious from Eq. (8) that for pure Fe $q^{\text{NN}} = q^{\text{FeFe}}$ is positive, because $K_1^{\text{int}}$ is found to be negative in the experiment. In all other cases, $q^{\text{NN}}$ represents an average value of $q^{\text{FeCo}}, q^{\text{FeCu}}$, and $q^{\text{CoCo}}$. $K_1^{\text{int}}$ becomes positive for $x > 0.3$ according to the data in Fig. 1 and, hence, $q^{\text{NN}}$ more and more negative for increasing Co content. This means that $q^{\text{FeCo}}$ and $q^{\text{CoCo}}$ together are responsible for the negative contribution. In order to separate these two parameters, alloy films with a different degree of chemical short range order should be investigated.

Assuming that we have random alloys, the ratio $q^{\text{NN}} / q^{\text{NNN}}$ can be assumed to be independent of composition because the atomic distances are the same in all samples. Then, it is obvious from Eq. (9) that $K_1^{\text{int}} / K_1^{\text{vol}}$ has the same negative value for all compositions, i.e., the critical thickness of the spin reorientation transition is independent of composition. That exactly is observed experimentally.

Furthermore, we can use our experimental values for $K_1^{\text{int}}$ and $t_{\text{crit}} = -K_1^{\text{int}} / K_1^{\text{vol}}$ to calculate the coupling constants $q^{\text{NN}}$ and $q^{\text{NNN}}$ from Eqs. (8) and (9) for each alloy composition. With $t_{\text{crit}} = 3 a_0$, Eq. (9) yields $q^{\text{NNN}} = 22/27 q^{\text{NN}}$. Numerical values of these parameters for Fe–Fe, Fe–Co, and Co–Co atom pairs could be found from alloys with different chemical order.

In conclusion, it was shown that a universal relation between the four-fold volume and interface anisotropy constants holds for Fe$_{1-x}$Co$_x$(001) films epitaxially grown on Au(110), Ag(001), and GaAs(001). This unexpected behavior can be consistently understood within Néel’s pair energy model of magnetic anisotropy. This agreement also means that the different electronic interface structure does not play a major role for the materials studied here, considering the four-fold anisotropy. It would be interesting to compare the present results with interfaces where strong electronic hybridization is expected like Ni/Cu, etc. However, Néel’s phenomenological theory can not replace high precision ab initio calculations which should provide absolute values of anisotropy constants in the future instead of mere relations between different quantities.

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