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Magnetic anisotropy of Gd(0001)/W(110) monolayers

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The uniaxial magnetic anisotropy of Gd(0001) monolayers and of an 80-Å epitaxial film on W(110) is determined near T_C. Our magnetic resonance technique determines the effective magnetization in absolute magnetic-field units and is sensitive to the crystalline anisotropy below and *above* the Curie temperature. We find a temperature-dependent uniaxial anisotropy in the monolayer which favors alignment of the magnetization normal to the surface and is up to ten times larger than the bulk anisotropy. The dipolar shape anisotropy, however, dominates and the magnetization lies in the basal plane, i.e., the film plane. Furthermore, we demonstrate that the shape anisotropy of a monolayer depends on the lattice type. Summations of the dipolar fields on a square [bcc (100)] and a hexagonal lattice yield a 31% and 6% reduction of the shape anisotropy, respectively, when compared with the calculation for a homogeneous thin film with demagnetization factor $N_\perp = 1$.

Recently, much experimental effort has been put into the determination of the easy axis of magnetization M for epitaxial magnetic monolayer (ML) films prepared under UHV conditions. Results on Fe and Co films show that for layer thicknesses ≥ 5 ML the magnetization lies in the film plane.[1-6] This is the expected behavior due to the dipole interaction, which favors alignment of M in the plane of thin disks. For layer thicknesses below ≈ 5 ML, however, the orientation of M is found to depend on the lattice type, the temperature, and the details of the sample preparation.[2] For a fcc Fe monolayer one finds that the easy axis of M lies normal to the surface,[2,3] while for bcc Fe and hcp Co monolayers[4-6] an in-plane orientation of M is found. This dependence on the lattice structure demonstrates the importance of the spin-orbit interaction[7] on the intrinsic magnetic anisotropy energy. It yields different strengths of the magnetic anisotropy field for different types of lattices.

Existence of a remanent magnetization lying in the surface plane was concluded from spin-polarized low-energy electron diffraction on a 500-Å thick Gd(0001) film[8,9] and from spin-polarized Auger spectroscopy of Gd layers on ferromagnetic Fe(100).[10] These techniques, however, were not sensitive to components of M normal to the surface. For bulk Gd the easy axis of M is known to lie along the hexagonal c axis for temperatures $T \geq 240$ K $(T/T_C \approx 0.8)$,[11] which is normal to the Gd film plane. Thus, depending on the strength of the intrinsic magnetic anisotropy as compared with the dipolar shape anisotropy, one cannot exclude a magnetization inclined at an angle ϕ_{eq} to the film plane (inset Fig. 1). The magnetic resonance appears to be the best-suited experimental technique to determine the orientation of M below and *above* T_C unambiguously, as it is sensitive to both normal and parallel components of M.

In the present work we show results on epitaxial Gd(0001) monolayers on a nonmagnetic W(110) substrate prepared under UHV conditions. Magnetic resonance was performed *in situ* for temperatures $0.8 \leq T/T_C \leq 1.25$. We determine the angle ϕ_{eq} of the magnetization vector by comparing the shape anisotropy of the Gd film with the intrinsic uniaxial anisotropy, which is obtained from our experiment. We find that the intrinsic anisotropy of the Gd monolayers is increased with respect to bulk values up to a factor of 10 and favors alignment of M along the surface normal. But due to the strength of the shape anisotropy the magnetization is forced to lie completely in the film plane for our samples, that is $\phi_{eq} = 0°$.

Films of one to a few atomic layers cannot be treated as a homogeneous medium with a demagnetization factor $N_\perp = 1$ normal to the plane. A single layer of a bcc, fcc,

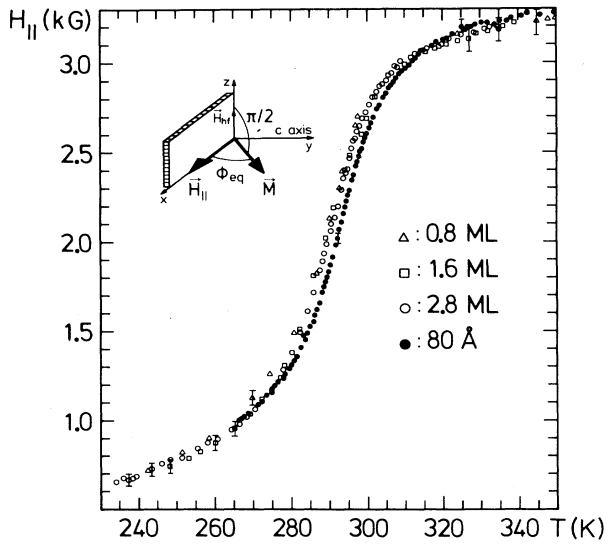

FIG. 1. Magnetic resonance fields at 9 GHz for epitaxial layers Gd(0001)/W(110) as a function of temperature. The dc magnetic field $H_\parallel$ is applied always in the film plane (inset). In uniaxial symmetry the orientation of M is given by ϕ_{eq}.

and hcp structure (that is to say a square lattice with next-nearest-neighbor distance a_0, $a_0\sqrt{2}$, and a triangular net, respectively) represents a lattice of discrete magnetic moments. A summation of the dipolar fields over discrete lattice sites is necessary in order to obtain the true demagnetizing field. This will be described in a later part of this paper.

Gd(0001) films of 0.8 ML, 1.6 ML, 2.8 ML, and 80-Å thickness were prepared on W(110) and characterized in UHV as described earlier.[12] Magnetic resonance at 9 GHz was performed *in situ* on freshly evaporated films.[13] The Curie temperatures T_C of these films were determined to be 271 ± 2, 278 ± 2, 282 ± 2, and 287 ± 2 K, respectively.[12,13]

In the present analysis we use the magnetic free-energy density given for axial symmetry (inset Fig. 1) by[14]

$$E=-H_{\parallel}M\cos\phi+2\pi M^2(N_{\perp}\sin^2\phi+N_{\parallel}\cos^2\phi)-K_2\sin^2\phi\,. \tag{1}$$

The first term is the Zeeman energy for the external field $H_{\parallel}$ applied in the surface plane. The angle ϕ measures the inclination of M with respect to the film plane. The second term represents the demagnetization energy caused by the dipole-dipole interaction of the magnetic moments. The last term is the first-order uniaxial magnetic anisotropy appropriate to an hexagonal lattice. Higher-order terms like $K_4\sin^4\phi$ can be neglected in the case of Gd for $T\geq240$ K.[11] The equilibrium position ϕ_{eq} of the magnetization is found by minimizing the free energy [Eq. (1)]

$$-MH_{\parallel}=[4\pi(N_{\perp}-N_{\parallel})M^2-2K_2]\cos\phi_{eq}\,. \tag{2}$$

From Eq. (1) it is seen that E is lowest for $\phi=90°$ in zero external field, if $K_2\geq2\pi(N_{\perp}-N_{\parallel})M^2$, i.e., the magnetization pointing along the film normal. M lies in the plane for two limits: (i) $K_2<0$, (ii) $0\leq K_2<2\pi(N_{\perp}-N_{\parallel})M^2$.

The condition for magnetic resonance in our experimental geometry (Fig. 1 inset) is given by [14,15]

$$\left(\frac{h\nu}{g\mu_B}\right)^2=H_{\parallel}[H_{\parallel}+4\pi(N_{\perp}-N_{\parallel})M(T,H_{\parallel})-2K_2(T)/M(T,H_{\parallel})]\,, \tag{2a}$$

where ν is the microwave frequency and $g=1.97$ the g factor for Gd metal. The last two terms in the brackets can be combined to give the effective magnetization M_{eff}

$$4\pi(N_{\perp}-N_{\parallel})M_{eff}=4\pi(N_{\perp}-N_{\parallel})M(T,H_{\parallel})-2K_2(T)/M(T,H_{\parallel})\,. \tag{2b}$$

M_{eff} is determined directly from the shift of the resonance field $H_{\parallel}$ with respect to the $g=1.97$ resonance position (high-temperature limit). To extract K_2 both values $M(T,H_{\parallel})$ and $H_{\parallel}(T)$ have to be obtained independently. The demagnetization factors $N_{\perp}=1$ and $N_{\parallel}=0$ are usually used for thin films of several nanometers thickness in the literature. These values are correct for a homogeneous medium. For films of one to a few atomic layers, however, they are inappropriate as discussed above. Recently, it has been shown[16] that the N tensor can be obtained from a summation over a finite number of n point dipoles on a lattice. The asymptotic values N_{ij}^{∞} (for $n\rightarrow\infty$) for a given spin direction are extrapolated by fitting $N_{ij}(n)$ to

$$N_{ij}=N_{ij}^{\infty}+a_1n^{-1/3}+a_2n^{-2/3}+a_3n^{-1}\,. \tag{3}$$

The last three terms reflect the contributions from moments at the surfaces, edges, and corners of the sample.

The main goal of our estimate is the calculation of N_{ij} for one layer to analyze our data for 0.8 and 1.6 ML. The demagnetizing field of a multilayer has to be calculated slightly differently by averaging over the fields of each individual layer.[17] This goes beyond the present communication.

We calculate N_{ij} for a hexagonal (0001) lattice with lattice constants $a_0=3.46$ Å, $c/2=2.89$ Å, i.e., Gd(0001)/W(110) [Fig. 2:(Δ)], as well as for $a_0=3.6$ Å, $c/2=2.89$ Å [Gd-bulk, Fig. 2:(+)]. Point dipoles in a thick disk of 1–11 layers with a disk diameter of up to 2000 lattice sites are included in the summation. To check the reliability of the extrapolated N_{ij}^{∞} we also determine N_{ij} for a bcc (100) lattice with $a_0=2.87$ Å [Fe, Fig. 2:(○)]. For the monolayer, i.e., a square lattice, we obtain $N_{\perp}=0.692(1)$. This is in agreement with Ref. 1 where $N_{\perp}-N_{\parallel}=0.692-0.154=0.538$ (see Fig. 2). Yafet and Gyorgy obtain $N_{\perp}=0.719$.[18] $N_{\perp}^{\infty}$ for spins pointing normal to the film is plotted in Fig. 2 as a function of the number of layers. $N_{\parallel}^{\infty}$ for spins lying in the plane is also calculated and the known relationship $2N_{\parallel}=1-N_{\perp}$ is confirmed.

We find that $N_{\perp}$ for a monolayer depends on the lattice type. For a hexagonal monolayer $N_{\perp}=0.94$ and for a square lattice, bcc (100) monolayer, with nearest-neighbor distance a_0 we calculate $N_{\perp}=0.692$. Interestingly, $N_{\perp}$ of the densely packed hexagonal lattice deviates only by about 6% from the homogeneous value, while the more open square monolayer yields a reduction of 31%. Differences between an average $N_{\perp}^{\infty}$ for a multilayer and the $N_{\perp}^{\infty}$ of only the central plane of this multilayer become irrelevant for the hexagonal structure.

A plot of the experimental resonance fields as a function

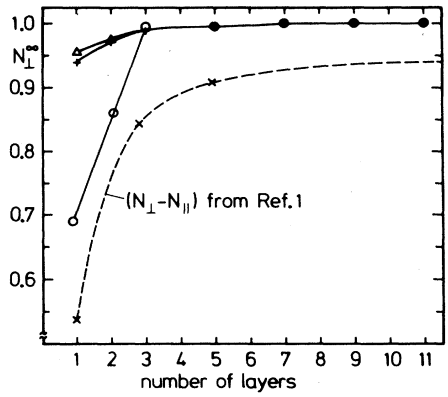

FIG. 2. Demagnetization factor $N_{\perp}^{\infty}$ of the central layer as a function of atomic layers for the hcp(0001) and bcc(100) lattices as described in the text. Values for $N_{\perp}-N_{\parallel}$ [Eq. (2a)] from Ref. 1 are shown also. They were calculated for a spherical distribution of dipole density around each lattice site.

of temperature is shown in Fig. 1. For all layer thicknesses one observes a shift to lower magnetic fields with decreasing temperature. Using Eqs. (2a) and (2b) one sees that this shift to lower resonance fields determines the effective magnetization M_{eff}, which increases when the temperature is lowered through T_C. Thus, we observe an in-plane magnetization for the monolayers and the 80-Å film. The small changes in T_C and $H_\parallel$ for the different films are difficult to see in Fig. 1. It is better observed in a log-log plot of $M/H_\parallel$ over reduced temperature and has been discussed earlier.[13]

To extract the easy direction of the magnetization in the limit of zero applied field, we determine the first-order uniaxial magnetic anisotropy constant K_2 from the effective magnetization [Eq. (2b)]. The demagnetization factors for the different films are taken from our calculation (Fig. 2). In our previous analysis of the susceptibility of the 80-Å film[12] we found a bulklike behavior. For this film, we use the bulk magnetization which has been experimentally determined as a function of the applied magnetic field and of the temperature.[19] For the monolayer films, on the other hand, the magnetic field and temperature dependence of M is not known experimentally. Our measurement of the susceptibility above T_C revealed two-dimensional (2D) Ising-like magnetic behavior of these films.[12] Thus, we use the theoretical temperature dependence of the spontaneous magnetization for a 2D Ising system near T_C, i.e.,

$$M_s(t)=M_s t^\beta, \quad t=1-T/T_C, \quad 0<t\leq 0.1 \tag{4}$$

with $\beta=\frac{1}{8}$ and $M_s(T=0\text{ K})=214$ G.[11] For the data above T_C we obtain M according to

$$M(T,H_\parallel)=C(T-T_C)^{-\gamma}H_\parallel, \quad \gamma=\tfrac{7}{4} \tag{5}$$

with C being the Curie constant of bulk Gd. For $T\leq T_C$ we neglect the effect of $H_\parallel$ on M. Putting these magnetization values into Eqs. (2a) and (2b) together with the correct demagnetization factors $N_\perp$ for the different films we calculate $K_2(T)$. The result is shown in Fig. 3 as a function of the reduced temperature T/T_C. One finds a positive anisotropy constant K_2 for all film thicknesses. The temperature dependence of the anisotropy for the monolayer films is qualitatively the same: no intrinsic anisotropy above T_C and a sharp increase of K_2 just below T_C with a maximum around $T/T_C\approx 0.96$. The anisotropy of the 80-Å film is much smaller and differs from zero even above the ordering temperature. This behavior is known from bulk Gd measurements[11] and may be ascribed to a field-induced anisotropy. As stated earlier this strong *positive* anisotropy favors alignment parallel to the film normal, i.e., the c axis. Comparison with the demagnetization energy (Fig. 3, solid lines), however, shows that even for the highest anisotropy values the demagnetization energy overcomes the intrinsic spin-orbit anisotropy. Thus, we find that for a monolayer and an 80-Å thick film of Gd(0001) on W(110) *the magnetization lies completely in the film plane* ($\phi_{eq}=0°$, Fig. 1 inset). Also for films of several hundred Å thickness M was found to lie in the basal plane,[8,9] i.e., the film plane, opposite to the known behavior of bulk Gd. From the previous discussion the

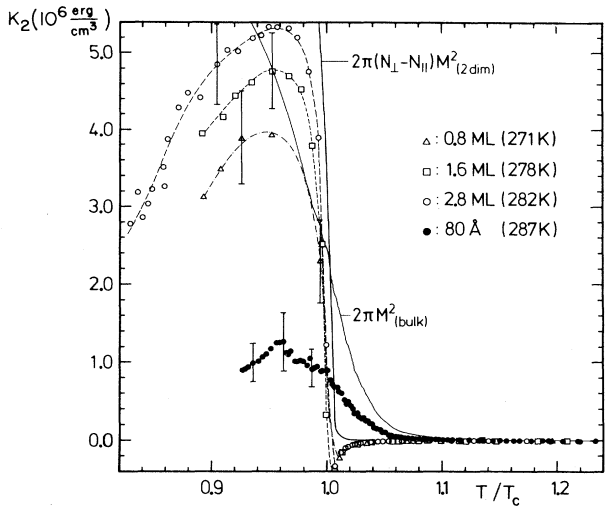

FIG. 3. Experimental values of the anisotropy constant K_2 vs relative temperature T/T_C. Curie temperatures of each film are given in brackets. The shape anisotropy energy as a function of T/T_C for the theoretical $M(T)$ of a 2D-Ising system (thick solid line) and for the experimental $M(T,H_\parallel)$ of bulk Gd (thin solid line) is also shown.

reason is obvious: The dipolar shape anisotropy of thin films overcomes all other contributions, but is missing in spherical bulk samples.

The magnitude of K_2 is approximately one order of magnitude larger than the bulk anisotropy, a behavior which has also been found for Fe and Ni monolayers.[7] An analysis of our anisotropy data in terms of a surface anisotropy[20] seems not useful to us and somewhat arbitrary. For the symmetry of the hcp surface layer one would expect a Néel-type surface anisotropy equal to zero according to Ref. 21. And for the monolayer film a surface layer is not defined. The strong increase in K_2 measured in erg/cm^3 is due to strain in the lattice which is caused by the lattice mismatch of Gd(0001) and W(110). The Gd monolayer lattice constant is known to be compressed by about 5% compared to the bulk value.[22] This compression can be related in a simple picture to an increase in the uniaxial anisotropy energy[23] for Gd. A dilatation of the monolayer lattice would yield a reduced uniaxial anisotropy. In this model it becomes also understandable that the anisotropy of the 80-Å thick sample is lower than the one of the monolayer. An adjustment of the lattice constant with increasing thickness takes place and reduces the strain in the film. Residual stresses in this film may cause our experimentally determined higher volume anisotropy.

In conclusion, our magnetic resonance study of a monolayer Gd(0001) on W(110) reveals that the magnetization lies completely in the surface plane in zero external field in contrast to results reported for Fe monolayers. As an advantage in comparison to most other techniques applied to magnetic monolayers we like to point out that our measurement yields the effective magnetization, Eq. (2b), in absolute field units. A quantitative comparison to theoretical calculations is possible. This, and the possibility of measuring at T_C^- *and* T_C^+, makes the UHV ESR a good

method to study critical phenomena of ultrathin films. It is also demonstrated that the demagnetization energy of a magnetic monolayer depends on the lattice structure and is significantly different from the one calculated for an homogeneous medium.

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[1]B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, Phys. Rev. Lett. **59**, 1756 (1987).

[2]C. Liu, E. R. Moog, and S. D. Bader, Phys. Rev. Lett. **60**, 2422 (1988).

[3]D. Pescia, M. Stampononi, G. L. Bona, A. Vaterlaus, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 2126 (1987).

[4]M. Stampanoni, A. Vaterlaus, M. Aeschlimann, and F. Meier Phys. Rev. Lett. **59**, 2483 (1987).

[5]C. M. Schneider, J. J. de Miguel, J. Garbe, S. Ferrer, and J. Kirschner (unpublished).

[6]D. Pescia, G. Zampieri, M. Stampanoni, G. L. Bona, R. F. Willis, and F. Meier, Phys. Rev. Lett. **58**, 933 (1987).

[7]J. G. Gay and R. Richter, Phys. Rev. Lett. **56**, 2728 (1986).

[8]D. Weller, S. F. Alvarado, W. Gudat, K. Schroeder, and M. Campagna, Phys. Rev. Lett. **54**, 1555 (1985); D. Weller and S. F. Alvarado, Z. Phys. B **58**, 261 (1985).

[9]D. Weller, Ph.D. thesis, University of Cologne, 1985 (unpublished).

[10]M. Taborelli, R. Allenspach, G. Boffa, and M. Landolt, Phys. Rev. Lett. **56**, 2869 (1986).

[11]W. D. Corner and B. K. Tanner, J. Phys. C **9**, 627 (1976); B. Coqblin, *The Electronic Structure of Rare-Earth Metals and Alloys: The Magnetic Heavy Rare-Earths* (Academic, London, 1977).

[12]M. Farle and K. Baberschke, Phys. Rev. Lett. **58**, 511 (1987); K. Baberschke, M. Farle, and M. Zomack, Appl. Phys. A **44**, 13 (1987).

[13]K. Baberschke, M. Zomack, and M. Farle, in *Magnetic Properties of Low-Dimensional Systems,* edited by L. M. Falicov and J. L. Moran-Lopez, Springer Proceedings in Physics Vol. 14 (Springer-Verlag, Berlin, 1986), p. 84.

[14]C. Chappert, K. Le Dang, P. Beauvillain, H. Hurdequint, and D. Renard, Phys. Rev. B **34**, 3192 (1986).

[15]S. V. Vonsovskii, *Ferromagnetic Resonance* (Pergamon, Oxford, 1966).

[16]C. D. H. Williams, D. Evans, and J. S. Thorp, J. Magn. Magn. Mater. **73**, 123 (1988).

[17]A. S. Arrott (private communication).

[18]Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).

[19]H. E. Nigh, S. Legvold, and F. H. Spedding, Phys. Rev. **132**, 1092 (1963); M. N. Deschizeaux and G. Develey, J. Phys. (Paris) **32**, 319 (1971).

[20]R. Bergholz and U. Gradmann, J. Magn. Magn. Mater. **45**, 389 (1984).

[21]I. S. Jacobs and C. P. Bean, in *Magnetism III,* edited by G. T. Rado and H. Suhl (Academic, London, 1963), p. 314.

[22]J. Kolaczkiewicz and E. Bauer, Surf. Sci. **175**, 487 (1986).

[23]A. H. Eschenfelder, *Magnetic Bubble Technology* (Springer-Verlag, Berlin, 1981), p. 23.