Higher-order magnetic anisotropies and the nature of the spin-reorientation transition in face-centered-tetragonal Ni(001)/Cu(001)

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Second- and fourth-order constants K_2, K_4 of the magnetic anisotropy energy in tetragonally distorted Ni(001) ultrathin films on Cu(001) are determined as a function of temperature and film thickness. We measure a positive out-of-plane constant $K_{4\perp} \approx +0.24$ μeV/atom and a negative in-plane constant $K_{4\|}$ near room temperature. This leads to a continuous (second or higher-order) spin-reorientation transition of the easy axis from an in-plane below $\approx$7 ML to a perpendicular orientation for thicker films. $K_{4\|}$ changes sign at $d_{c\|} \approx 6.7$ ML which results in a change of the easy axis from the [100] direction below 6.7 ML to the [110] direction above. These results are quantitatively compared to Ni(111)/W(110) for which no out-of-plane reorientation is observed. [S0163-1829(97)09105-4]

I. INTRODUCTION

In recent years, the spin-reorientation transition (SRT) in ultrathin ferromagnetic layers has attracted much interest. Ultrathin Fe and Co films have been found to exhibit a strongly enhanced magnetic anisotropy energy (MAE) with an easy axis perpendicular to the surface plane.[1–4] At low film thickness this perpendicular anisotropy is sufficient to overcome the demagnetizing field and align the magnetization perpendicular to the plane. The SRT from in-plane to out-of-plane is usually found at a critical number of layers $2 < d_c < 6$ ML, which depends on the temperature.[5,6] Furthermore, a reversible temperature dependent SRT from perpendicular at low temperature to in-plane at higher temperatures has been observed.[2,7,8]

In Ni films, on the other hand, the magnetization switches from an *in-plane state at low thickness* to a perpendicular orientation above $d_c \approx 7$ ML for Ni(001)/Cu(001),[9–13] and at $d_c \approx 5$ ML for Cu/Ni(111)/Cu(111).[1] This reversed thickness dependence is due to the opposite signs of the surface anisotropy K_2^s of Ni and of Fe, Co,[1,14] which favors in-plane magnetization for Ni and out-of-plane magnetization for Fe and Co. The total MAE (Ref. 15) varies as a function of thickness according to $K_2 = K_2^v + 2K_2^s/d$ (K^v is the volume anisotropy). The direction of the magnetization is determined by the balance between K_2^v, K_2^s/d, and the demagnetizing energy $2\pi M^2$. In the monolayer range the surface contribution usually dominates and determines the easy axis of the magnetization. In the cases of Ni(001) and Ni(111) on Cu one has a large volume anisotropy K_2^v which results from the large strain anisotropy due to the $+2.5\%$ elongated nearest-neighbor distance in-plane of Ni on Cu. It favors a perpendicular magnetization and overrides the shape anisotropy $2\pi M^2$.

An interesting question is: Does the transition in Ni/Cu occur continuously or discontinuously, that is to say, is it a first- or second- (or higher-) order phase transition? Theoretically, this transition, as a function of film thickness, has been discussed in terms of phenomenological anisotropy constants only.[1] No microscopic theory, as for example for the temperature driven SRT,[16–20] has been published. For Ni(001)/Cu(001) absolute values for the second-order anisotropy contributions K_2^v and K_2^s have been measured by ferromagnetic resonance (FMR) in UHV.[9] These data agree quantitatively with the anisotropy calculated from a magneto-elasticity model which takes the experimentally determined tetragonal distortion ($+2.5\%$ tensile in-plane and -3.2% compressive strain out-of-plane) into account. Also the critical thickness for SRT, as a function of film thickness, is predicted correctly. The possibility that SRT is driven by a structural or morphological change at d_c has been excluded by quantitative low-energy electron-diffraction[21] [I(E)-LEED] and scanning tunneling microscopy.[22] Based on the available data in the literature,[9] one could argue that the magnetization, as a function of film thickness, switches abruptly in Ni/Cu(001). No stable tilted orientation near $d_c \approx 7$ ML was reported. This is different from results obtained for wedge-shaped Fe and Co layers, where a continuous decrease of the remanent perpendicular magnetization was found when the film thickness increased. This was interpreted as a continuous rotation of the magnetization from out-of-plane to in-plane with stable intermediate angles.[2,8,23]

In the present work we will show that the magnetization in Ni/Cu(001) rotates *continuously* from a perpendicular orientation above 8 ML to the in-plane direction below 7 ML. Absolute values for the magnetic anisotropy up to fourth order are measured by angular-dependent ferromagnetic resonance (FMR) in UHV. These data yield the equilibrium angles (θ_{eq} and φ_{eq}) of the magnetization with respect to the [001] (out-of-plane) and the [100] direction (in-plane) in zero applied magnetic field. A stable intermediate out-of-plane angle θ_{eq} is found. These results are compared to the ones for Ni(111) on W(110) for which the anisotropy, due to strain relaxation as a function of thickness, must be included in the ''surface'' contribution K^s/d.[24,25] The volume part should assume bulk values, which are small. In Ni/Cu, on the other hand, one has a large magnetoelastic contribution, which is *not* thickness dependent (volume-type), and *no* strain relaxation contribution to K^s/d in a large thickness regime. It is worthwhile to note that no perpendicular magnetization in Ni(111) on W(110) was observed although the [111] direction is the easy axis of magnetization in bulk Ni.

Another interesting issue is the correlation between magnetic anisotropy and dimensionality. For both Ni systems a crossover from two- to three-dimensional (3D) behavior at

TABLE I. Equilibrium angles of the magnetization in zero applied magnetic field and corresponding constraints on the anisotropy parameters for fct Ni(001) on Cu(001). Lines (a) to (c) of Fig. 1 correspond to the inequations (a) to (c). For shortness we define $K_{4\perp}+(1/4)(3+\cos 4\varphi_{eq})K_{4\parallel}\equiv X$.

$\theta_{eq}=0°$	(a) $K_2+\frac{1}{2}K_{4\perp}-\frac{1}{8}(3+\cos 4\varphi_{eq})K_{4\parallel}>2\pi M^2$, if $X>0$ (b) $K_2+K_{4\perp}>2\pi M^2$, if $X<0$
$\sin^2\theta_{eq}=\dfrac{K_2+K_{4\perp}-2\pi M^2}{K_{4\perp}+\frac{1}{4}(3+\cos 4\varphi_{eq})K_{4\parallel}}$	(b) $K_2+K_{4\perp}<2\pi M^2$ and (c) $2\pi M^2<K_2-\frac{1}{4}(3+\cos 4\varphi_{eq})K_{4\parallel}$, if $X<0$
$\theta_{eq}=90°$	(a) $K_2+\frac{1}{2}K_{4\perp}-\frac{1}{8}(3+\cos 4\varphi_{eq})K_{4\parallel}<2\pi M^2$, if $X>0$ (c) $K_2-\frac{1}{4}(3+\cos 4\varphi_{eq})K_{4\parallel}<2\pi M^2$, if $X<0$
$\varphi_{eq}=0°$, [100]	$K_{4\parallel}>0$
$\varphi_{eq}=45°$, [110]	$K_{4\parallel}<0$

5–6 ML has been reported.[11,36] However, in these studies the effect on the magnetic anisotropy was not discussed.

The paper is organized as follows: In Sec. II we present the phenomenological model for the MAE of a tetragonal system like Ni(001) on Cu(001) and the role of the higher-order anisotropy constants is discussed in detail. In Sec. III experimental details for the preparation and structure of Ni(001) on Cu(001) and Ni(111) on W(110) and the FMR experiment are given. In Sec. IV we discuss the role of the second- and fourth-order anisotropy constants for both systems. Section V summarizes this work.

II. SPIN-REORIENTATION TRANSITION AND MAGNETIC ANISOTROPY

The orientation of the magnetization in ultrathin films is determined by the balance between the intrinsic MAE, which arises from spin-orbit coupling, and the shape anisotropy, which is of dipolar origin. The latter always favors an in-plane easy axis for thin films, the former may either favor in-plane or perpendicular orientation. The balance between the two anisotropy contributions changes as a function of film thickness and temperature. In our sign convention a positive anisotropy constant will favor a perpendicular easy axis. A SRT to a perpendicular easy axis occurs when the intrinsic anisotropy becomes positive and dominates over the dipolar shape anisotropy [typically $2\pi M^2(T=0\ \mathrm{K})\approx 12$ μeV/atom for Ni]. It was found experimentally that a perpendicular magnetization can be stabilized in Fe and Co layers *below* a critical number of layers $d_c\approx 3-8$ ML, which depends on the film structure and the temperature.[2,6] Ni(001) and Ni(111) on Cu are the only known systems where this behavior is reversed.

Due to the lack of a microscopic model we will review the phenomenological anisotropy model,[26] which is usually used to describe the SRT as a function of thickness.[27] We start with the anisotropic part of the free-energy density appropriate for a tetragonal ferromagnet like Ni(001) on Cu(001):[9]

$$E=2\pi(N_\perp-N_\parallel)M^2\cos^2\theta-K_2\cos^2\theta-\tfrac{1}{2}K_{4\perp}\cos^4\theta-\tfrac{1}{2}K_{4\parallel}\tfrac{1}{4}(3+\cos 4\varphi)\sin^4\theta, \quad (1)$$

where φ is the angle of the magnetization M with respect to the [100] direction in the (x,y) plane. θ measures the angle from the z axis [001], which is perpendicular to the film plane. $N_\parallel$ and $N_\perp$ are the demagnetization factors in the plane (x,y) and normal to the film plane.[28] K_2, $K_{4\perp}$, and $K_{4\parallel}$ are the second- and fourth-order terms of MAE. For cubic symmetry one has $K_{4\perp}=K_{4\parallel}$ (often denoted as K_1). The easy axis of magnetization is determined by the minimum of the free-energy density [Eq. (1)]. For negative $K_{4\parallel}$ the easy axis is along the [110] direction, for positive $K_{4\parallel}$ along [100]. The out-of-plane orientation is influenced by all anisotropy terms in Eq. (1), also by $K_{4\parallel}$. The equilibrium angles θ_{eq} and φ_{eq} of the magnetization in zero external field are given in Table I. One can identify three stability regions: in-plane, intermediate, and perpendicular. In Fig. 1 we show two planes of the 3D stability diagram of the magnetization: $K_{4\parallel}=0$ [Fig. 1(a)] and $K_{4\perp}=0$ [Fig. 1(b)]. For $K_{4\parallel}=K_{4\perp}=0$ no tilted orientation is possible, and if K_2 changes, a discontinuous flip of the magnetization, that is a first-order phase transition, from in-plane to out-of-plane is expected at $K_2=2\pi M^2$. Interestingly, a tilted orientation out-of-the film plane can be obtained if $K_{4\perp}<0$ [Fig. 1(a)] or $K_{4\parallel}<0$ [Fig. 1(b)]. In real systems K_2 and K_4 change as a function of temperature and film thickness. The system may follow any path within the stability diagram. For example, varying $K_2/2\pi M^2$ from left to right in Fig. 1(b) with $K_{4\parallel}<0$ yields a continuous rotation of the magnetization vector from in-plane to perpendicular. One should note that Figs. 1(a) and 1(b) present only a limited view of the three-dimensional phase diagram which is given by $K_{4\perp}$, $K_{4\parallel}$, and K_2. It turns out that, for example, if $K_{4\parallel}<0$ one can also find a region with tilted orientation for $K_{4\perp}>0$. In Table I the boundary areas between the three regions are listed.

In a thin film two major factors contribute to K_i ($i=2,4,\ldots$) according to $K_i=K_i^v+2K_i^s/d$ namely: (a) a *thickness-independent* contribution usually denoted by K_i^v, and (b) the *thickness-dependent* contribution K_i^s/d. The ''volume'' anisotropy K_i^v includes all *thickness-independent* magnetostrictive contributions and differs in most films very much from K_{bulk}. The ''interface-surface'' anisotropy K_i^s results from the lowered coordination at the substrate/film and film/-vacuum interfaces. Also, any *thickness-dependent* magnetoelastic contribution, which arises from a thickness-dependent relaxation of strain, is included in this term. A special case is the epitaxial system with a medium strain like Ni on Cu(001) ($\eta_{NiCu}=-2.5\%$): The film grows pseudomorphic (coherent) up to a critical thickness d_{cs} in a state of

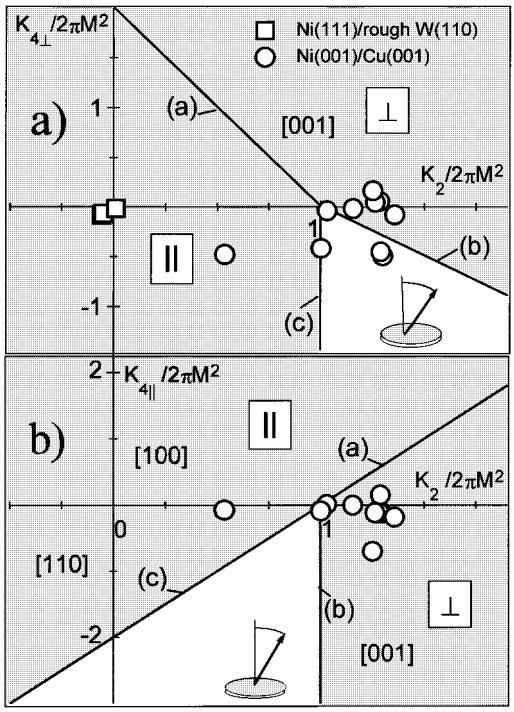

FIG. 1. Stability regions for the easy axis of magnetization. (a) $K_{4\perp}$ versus K_2 normalized to the shape anisotropy $2\pi M^2$. Note that the $K_{4\parallel}=0$ plane is shown only. (b) $K_{4\parallel}$ versus K_2 normalized to $2\pi M^2$ and $K_{4\perp}=0$. A canted $\theta_{\rm eq}$ is found in the unshaded regions only. The phase separation lines are given in Table I. Note that a negative $K_{4\parallel}$ yields an easy in-plane axis along [110]. Experimental K_i values for Ni/Cu(001) and Ni(111)/W(110) projected onto the $K_{4\parallel}=0$ and $K_{4\perp}=0$ plane are shown also.

constant strain. The magnetostrictive contribution to the total anisotropy is then pure volume type. Thickness-dependent relaxation contributions to K_i^s are absent and K_i^s is pure Néel type. For $d>d_{\rm cs}$, a strain relaxation contribution is contained in K_i^s/d. K^v also becomes different.

The vast majority of recent literature discusses the second-order contribution $K_2=K_2^v+2K_2^s/d$ only. As stated above, this restriction does not allow a continuous SRT. For example, in the case of Co (Ref. 30) and Fe (Ref. 31) films it was shown that one can find different types of reorientation transitions (continuous, special, discontinuous) as a function of film thickness if one includes higher-order terms in K^s and K^v. The ratios of $K_{2,4}^v$ and $K_{2,4}^s$ determine an upper d_{c2} and lower critical thickness d_{c1} for a tilted orientation of the magnetization. If $d_{c2}\leqslant d_{c1}$, a first-order thickness-dependent reorientation is possible.[30] $K_{2,4}^s$ are important for the SRT within the film plane[31] too. In the above discussion we have excluded the possibility of domain formation.[18,28] For our FMR experiments this is justified, because one applies a magnetic field of several hundred Gauss, which makes the film single domain. For completeness, we would like to mention that in a cubic or tetragonal film an uniaxial in-plane anisotropy may arise from a vicinal cut of the substrate layer. To account for this contribution one can add a second order in-plane anisotropy $K_{2,2}\cos^2\theta\cos^2\varphi_u$ in Eq. (1), where φ_u measures the angle between the uniaxial symmetry and the [100] direction in the plane.[29]

Another important parameter in the discussion of MAE is the temperature. A strong temperature dependence of all orders of MAE is well documented for bulk samples.[32] For example, the bulk anisotropy $K_{\rm bulk}$ of Ni is known to increase from -3×10^4 erg/cm^3 (-0.2 μeV/atom) at $T/T_c=0.9$ to -6×10^5 erg/cm^3 (-4.1 μeV/atom) at $T/T_c=0.2$.[32] Unfortunately, no general theoretical function for the temperature dependence of MAE is established in the bulk. In ultrathin films the situation is even worse. One has to consider the temperature dependence of surface and volume contributions. It is theoretically unclear if both terms should behave similarly as a function of temperature. Until recently there was only one experimental work[33] which presented few data points for $K_2^s(T)$ and $K_s^v(T)$. Already in the early work[33] it was mentioned that the analysis of $K_2=K_2^v+2K_2^s/d$ should be done at constant $T/T_c(d)$ and not at constant T. This is due to the fact that $T_c(d)$ varies strongly as a function of film thickness.[34,35] Theoretically, it is not clear if K_2 should be analyzed at constant T or T/T_c. Experimentally, however, we[34] and others[33] found that T/T_c is the thermodynamical relevant quantity which yields a $1/d$ linear dependence at many different temperatures. Here, we extend our previous work to the case of Ni(111).[36] Experimentally, this involves the determination of T_c and the determination of the anisotropy at different absolute temperatures for each thickness.

III. EXPERIMENT

All Ni films were deposited in a vacuum better than 1×10^{-10} mbar (base pressure 3×10^{-11} mbar) onto a contamination-free W(110) or Cu(001) substrate, which show a sharp LEED pattern. Film growth was monitored by medium energy electron diffraction in the case of Ni/Cu. Ferromagnetic resonance (FMR) and polar and longitudinal magneto-optic Kerr effect (MOKE) measurements were performed *in situ* immediately after deposition. For FMR, the magnetic field was applied along the Cu[110] direction and along the W[010] direction (direction of largest Ni tensile

TABLE II. FMR conditions used to determine the anisotropy constants for Ni(111) (Ref. 36) and Ni(001). Note, that in Refs. 35 and 36 a different free-energy equation with constants k_2 and k_4 was used. The conversion is $k_2=K_2+K_{4\perp}$, and $k_4=-1/2K_{4\perp}$.

	$H\perp$ film plane	$H\parallel$ film plane
Ni(111)	$\frac{\omega}{\gamma}=H_{R\perp}-4\pi M+\frac{2k_2}{M}$	$\left(\frac{\omega}{\gamma}\right)^2=H_{R\parallel}\left(H_{R\parallel}+4\pi M-\frac{2k_2}{M}-\frac{4k_4}{M}\right)$
fct Ni(001)	$\frac{\omega}{\gamma}=H_{R\perp}-4\pi M+\frac{2(K_2+K_{4\perp})}{M}$	$\left(\frac{\omega}{\gamma}\right)^2=\left(H_{R\parallel}-\frac{2K_{4\parallel}}{M}\right)\left(H_{R\parallel}+4\pi M-\frac{2K_2}{M}+\frac{K_{4\parallel}}{M}\right)$

strain within the plane) which is the easy in-plane axis of Ni(111) films as detected by MOKE. Details have been presented earlier.[9,21,25,35,36]

FMR has been proven to be an excellent tool to determine higher-order magnetic anisotropies quantitatively.[27,37–40] The resonance condition for the Ni/Cu(001) system is derived from Eq. (1) by including the Zeeman energy due to the applied magnetic field:[9,40]

$$\left(\frac{\omega}{\gamma}\right)^2=\left\{H_R\cos(\theta_H-\theta)+\left(-4\pi M+\frac{2K_2}{M}+\frac{K_{4\perp}}{M}-\frac{K_{4\parallel}}{2M}\right)\cos 2\theta+\left(\frac{K_{4\perp}}{M}+\frac{K_{4\parallel}}{2M}\right)\cos 4\theta\right\}$$
$$\times\left\{H_R\cos(\theta_H-\theta)+\left(-4\pi M+\frac{2K_2}{M}+\frac{K_{4\parallel}}{M}\right)\cos^2\theta+\left(\frac{2K_{4\perp}}{M}+\frac{K_{4\parallel}}{M}\right)\cos^4\theta-\frac{2K_{4\parallel}}{M}\right\}. \quad (2)$$

Resonance is observed at the applied magnetic field H_R which depends on the equilibrium angles θ of the magnetization in an applied field and the angle θ_H of the magnetic field with respect to the film normal. The in-plane angle φ_H of the magnetic field was chosen along the [110] direction ($\varphi_H=45°$) in our experiments. The usual Kittel relations for the magnetic field applied either in the plane ($\theta_H=90°$) or perpendicular ($\theta_H=0°$) to the plane are given in Table II. For the Ni(111) system a different uniaxial free-energy model[25,27,35,36] was used. As a result one obtains different resonance conditions if higher-order anisotropies are included (Table II). The quadratic anisotropy constants determined from the perpendicular and parallel resonance conditions for the Ni(111) and Ni(001) can be directly compared. In our analysis we use the field- and temperature-dependent magnetization M of Ni bulk which is proportional to the integrated intensity of the FMR absorption.[34] From the angular-dependent FMR experiment[38] one obtains the anisotropy constants which appear in Eq. (2). The equilibrium angle θ_{eq} of the spontaneous magnetization is calculated according to Table I.

A. Cu(001)/Ni(001)/vacuum

Recent investigations by quantitative I(E)-LEED analysis[21] and scanning tunneling microscopy[22] have shown that Ni grows pseudomorphic on Cu(001) with an in-plane lattice constant $a_p=2.53$ Å at coverages 3 and 5 ML, and $a_p=2.51$ Å at 11 ML. Compared to the in-plane lattice constant $a_p=2.55$ Å of bulk Cu the Ni a_p is contracted from the first layers on, which indicates that the surface a_p of Cu is already reduced to $a_p=2.53$ Å. This was reported recently.[41] A tetragonally distorted (fct) artificial Ni lattice is stabilized at least up to 11 ML, which in comparison to bulk fcc Ni ($a_p=2.49$ Å) is expanded by 2.5% in the film plane and compressed by -3.2% along the surface normal. There is no characteristic structural or morphological change at $d_c=7$ ML (Refs. 21 and 22) at which the SRT is observed.

B. W(110)/Ni(111)/vacuum

Films were grown on smooth and rough W(110) substrates. Smooth substrates with terrace sizes on the order of >100 Å were produced by high-temperature annealing after sputtering. Rough surfaces were obtained after ''soft'' sputtering at 1 keV (6 μA/cm^2) for 3–5 min and subsequent annealing to 800 K. Such surfaces still showed the bcc(110) LEED pattern with an increased spot diameter ($1.5-2x$) and an intensified background. Such a surface is expected to have small, 1–2 layer deep craters with a mean separation of 20–50 Å. The lattice misfit between fcc Ni(11̲1) and bcc W(110) planes is along the W[001], i.e., Ni[1̲10] direction $f_{[001]}=(a_{\mathrm{Ni}}-a_{\mathrm{W}})/a_{\mathrm{W}}=-2\underline{1}.3\%$ ($a_{\mathrm{Ni}}=2.\underline{4}92$ Å, $a_{\mathrm{W}}=3.165$ Å) and along the W [1̲10], i.e., Ni [11̲2], direction $f_{[1\bar{1}0]}=(\sqrt{3}a_{\mathrm{Ni}}-\sqrt{2}a_{\mathrm{W}})/\sqrt{2}a_{\mathrm{W}}=-3.6\%$. Up to a coverage of 0.75 ML the film grows pseudomorphic with the characteristic (1×1) LEED pattern of W(110). At 300 K growth continues layer-by-layer[42] up to 30 ML. Above 0.75 ML the film relaxes from the bcc(110) type layer to a fcc(111) structure. At 6 ML only the symmetric LEED pattern of a fcc(111) surface with a 3.6% dilated nearest-neighbor distance is observed.[43]

IV. DISCUSSION

A. Second-order surface and volume anisotropy

Before we turn to the discussion of our investigation we will summarize the results reported previously. The reorientation of the magnetization of fct Ni(001) on Cu(001) has been studied by several groups. At room temperature the magnetization is in-plane for film thicknesses below ≈7 ML (Refs. 9, 11, 12, and 45) and above ≈41 ML.[10,44,46] Perpendicular magnetization was reported for the intermediate thickness interval. Near the upper critical thickness (41 ML) evidence for a tilted out-of-plane magnetization was reported.[10] Here, we will focus at thicknesses $d<11$ ML. The results for K_2^v and K_2^s are summarized in Table III. K_2^v of fct Ni(001) (Refs. 9, 37, and 45) is by nearly two orders of magnitude enhanced in comparison to the fourth-order-cubic-bulk anisotropy. The existence of a quadratic ''volume'' anisotropy in Ni(001)/Cu(001) reflects the tetragonal distortion of the fcc Ni lattice on Cu(001). We have shown earlier[9] that the sign and magnitude of K_2^v is in quantitative agreement with the calculated magnetoelastic anisotropy due to the 2.5% lateral expansion of Ni(001) on Cu(001) and the associated tetragonal distortion. The origin of the perpendicular magnetization for $d>7$ ML is K_2^v and *not* K_2^s. The negative surface anisotropy K_2^s favors an in-plane magnetization, and below 7 ML ($-2K_2^s/d+2\pi M^2$) is large enough to compensate $-K_2^v$. In Table III the anisotropies for Ni(001) grown on a Cu buffer layer on Si(001) (Ref. 10) are also listed. K_2^s and K_2^v are different from our values, which is most likely due to the differences in the substrates. For Ni(001) films capped with a Cu layer,[24] one finds that the

TABLE III. Surface and volume anisotropies of fct Ni(001)/Cu(001) and fcc Ni(111)/W(110) films around 300 K [$T/T_c \approx 0.5$ for Ni(111) and $T/T_c \approx 0.7$ for Ni(001)]. Values for films capped with different layers are given also. $K_2^{S,V} < 0$ favors in-plane magnetization. For Ni(001) we use 1 erg/cm^2=334 μeV/atom, 10^6 erg/cm^3=6.8 μeV/atom. For the Ni(111) plane one has 1 erg/cm^2=384 μeV/atom. Note, that the shape anisotropy $2\pi M^2$(300 K)=7.5 μeV (M=420 G) is contained in K_2^V of Ref. 24.

	K_2^S/1 ML (μeV/atom)	K_2^V (μeV/atom)	T (K)	Ref.
Ni(111)/W(110)	−88	10.8	T/T_c=0.54	25
Vac/Ni(111)	−184		300	50
Re/Ni(111)	−73		300	50
Cu/Ni(111)/Cu				
$d<d_{cs}$	−31	+8.8	300	24
$d>d_{cs}$	+104	−6.2	300	24
Cu/Ni(001)/Cu				
$d<d_{cs}$	−134	+26.5	300	24
$d>d_{cs}$	+301	−10.9	300	24
Vac/Ni(001)/Cu				
$d<d_{cs}$	−77	+30	300	9
$d<d_{cs}$	−40	+22	300	10

second Ni/Cu interface roughly doubles the magnitude of K_2^s in the coherent growth regime ($d<d_c$) and modifies the volume contribution by 10% only. If one assumes that the interface of Ni on Cu is the same as that of Cu on Ni, one estimates that the vac/Ni(001) interface anisotropy is negligible.[9,10] Due to the enhanced K_2^s, the lower critical thickness for perpendicular magnetization increases for the Cu/Ni/Cu system.

Results on the magnetism of Ni(111)/W(110) (Table III) have been reported previously.[25,35,36] At about 5 ML a crossover from three- to two-dimensional behavior is found. The analysis of the ferromagnetic resonance data for Ni(111) was performed with a free-energy model appropriate for a hexagonal system[35] with negligible in-plane anisotropy. Based on the LEED finding this seems appropriate for $d>5$ ML, where isotropic strain is observed. The results obtained in our earlier publications are summarized below. (1) No dimensionality effect in the anisotropy is observed as a function of film thickness. (2) K_2^s is negative and K_2^v is positive, and the total anisotropy K_2 is for all thicknesses smaller than $2\pi M^2$. The magnetization lies in the plane for all thicknesses. (3) The large K_2^v at T/T_c=0.54 (Table III) corresponds to an additional magnetoelastic anisotropy $K_{\rm bulk}+K_{\rm me}$ with $K_{\rm me}=1.5\lambda\varepsilon$. Using the bulk magnetostriction constant $\lambda=-2.5\times10^{-5}$ one calculates a tensile strain ε=4.2%,[25] which is in reasonable agreement with the measured 3.6%. (4) The volume anisotropy of Ni(111) films grown on rough W(110) is smaller than the one for films on a smooth substrate.[36] This indicates the effect of the increased number of defects in the rough films, which cause a faster relaxation to the unstrained bulk lattice. The surface anisotropy, however, remains the same (Table III). This is surprising since one would expect that the strain relaxation contribution to K_2^s is different for the rough and smooth surface. Consequently, the Néel-type interface contribution

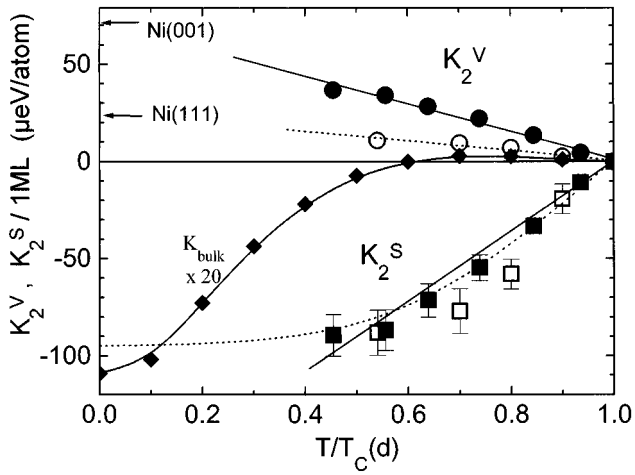

FIG. 2. Temperature dependence K_2^s (squares) and K_2^v (circles) for Ni(111)/smooth W(110) (open symbols) and Ni/Cu(001) (solid symbols). The anisotropy of bulk Ni (Ref. 32) is given also (solid diamonds). Dashed and solid lines are guides to the eyes only.

at the W(110) interface must be different. In the following, we discuss K_2^v, K_2^s of Ni(001) and Ni(111) as a function of reduced temperature $T/T_c(d)$ (Fig. 2).

K_2^v varies almost linearly as a function of T/T_c in both systems. Extrapolation of the linear behavior to T=0 K yields $K_2^v \approx 72$ μeV/atom for Ni(001) and $K_2^v \approx 25$ μeV/atom for Ni(111). A total-energy calculation for bulk fct Ni(001) with the lattice parameters of Ni/Cu(001) (Ref. 21) yields $K_2^v \approx 140$ μeV/atom at T=0 K.[49] The agreement is not satisfactory and becomes even worse if one considers an extrapolation based on experimental bulk Ni magnetostriction constants.[13] One finds that the strain anisotropy $K_{\rm me}=3/2\,\lambda_{100}\,(\varepsilon_\perp-\varepsilon_\parallel)\,(c_{11}-c_{12})$ (Ref. 40) calculated with $\varepsilon_\parallel$=2.5%, $\epsilon_\perp=-3.2\%$ and, taking the temperature dependences of λ_{100}, c_{11}, and c_{12} of bulk Ni (Ref. 48) into account, agrees within the error bars with K_2^v. It is also worthwhile to mention that there may be errors in the calculation since the same theory yields a wrong MAE for bulk Ni.[49] K_2^v of Ni(111) is always smaller than that of fct Ni(001). At first this seems surprising since the average in-plane strain ε=3.6% for Ni(111) (as detected by LEED) is larger than the one for fct Ni(001). However, the magnetostrictive constant λ_{111} is by a factor of 2 smaller than λ_{100}. This explains why for a larger strain a smaller anisotropy results.

Interestingly, the surface anisotropy of Ni(001) ($d<d_{cs}$) and Ni(111) is nearly the same for all temperatures. Considering that for Ni(111) there is a magnetoelastic and a Néel-type contribution to K_2^s and in Ni(001) only a Néel-type contribution, this agreement seems to be accidental. The simple explanation that the Néel-type contribution in both structures is the same and dominates over any small magnetoelastic contribution seems unlikely to us. No straightforward explanation can be given, since there is no possibility to analyze the temperature dependence of the magnetoelastic and the Néel contributions separately. Also, no microscopic theoretical model for the temperature dependence of K_2^s exists to our knowledge. For lack of better models we have added a linear fit to K_2^s of Ni(001) which should be seen as a guide to the eye only. If one follows the linear trend to T/T_c=0, one finds K_2^s (T=0 K)≈−180 μeV/atom which

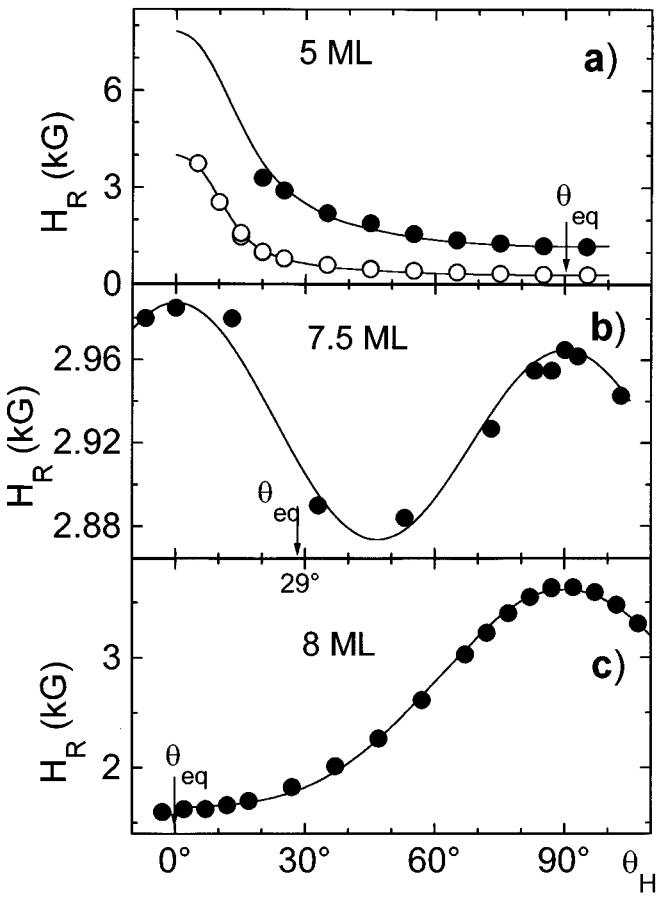

FIG. 3. Angular dependence of ferromagnetic resonance field at $T/T_c \approx 0.7$ for three thicknesses of Ni/Cu(001) (solid circle) 9 GHz; (open circle) 4 GHz. Fits according to Eq. (2) (solid lines) with $M \approx 420$ G, $g = 2.18$ yield: (a) $K_2 = 1.6$, $K_{4\perp} = -0.48$, $K_{4\parallel} = -0.55$; (b) $K_2 = 6.59$, $K_{4\perp} = -0.15$, $K_{4\parallel} = -0.14$; and (c) $K_2 = 9.5$, $K_{4\perp} = 0.1$, $K_{4\parallel} = -1.7$ μeV/atom.

yields $K_2^{\mathrm{ML}} = K_2^v + 2K_2^s = -270$ μeV/atom at $T = 0$ K for the Ni(001) monolayer. This value does not agree with $K_2^{\mathrm{ML}} = -94$ μeV/atom calculated for a Ni(001) monolayer with the Cu lattice constant.[47] On the other hand, if one chooses an extrapolation as indicated by the dotted line, one obtains K_2^{ML} ($T = 0$ K) $= -98$ μeV/atom in much better agreement with the calculation, but with a large error bar. We like to point out, however, that this extrapolation is ambiguous and was added only to reflect the fact that there is a curvature in the $K_2^s(T/T_c)$ data. All anisotropies in Ni vanish at the Curie temperature. This is not generally true in ferromagnets. For Gd(0001) films on W(110) it was found that K_2^v remains finite at T_c,[34] as it is also known for the bulk.

B. $K_{4\perp}$, $K_{4\parallel}$, and the spin-reorientation transition

As pointed out in Sec. II, an analysis of the anisotropy in quadratic order only is a severe restriction in the investigation of SRT. In the following we will discuss the reorientation transition in Ni/Cu(001). The fourth-order contributions in Eq. (1) can be determined by angular-dependent FMR according to Eq. (2). The experimental resonance field H_R as a function of magnetic field angle θ_H is shown in Fig. 3 for three different thicknesses. The temperature for all films is about $T/T_c = 0.7$. Already, the reversed angular dependence [Figs. 3(a) and 3(c)] for the 5 and 8 ML film unambiguously shows that the easy axis of magnetization is parallel and perpendicular, respectively. This is confirmed by the fit (solid line) according to Eq. (2), which yields the parameters K_2, $K_{4\perp}$, and $K_{4\parallel}$. The demagnetization energy $2\pi M^2 = 7.5$ μeV/atom of bulk Ni is used. Figure 3(b) shows only a small, unusual variation of H_R (note the different y scale). The fit determines anisotropy constants K_i, which yield an equilibrium angle $\theta_{\mathrm{eq}} = 29°$ of the magnetization in zero magnetic field (Table I). From this we conclude that the SRT as a function of film thickness is of second or higher order. This was reproduced for several other thicknesses and a narrow thickness interval from 7 to 8 ML is found where stable canted orientations of the magnetization exist.

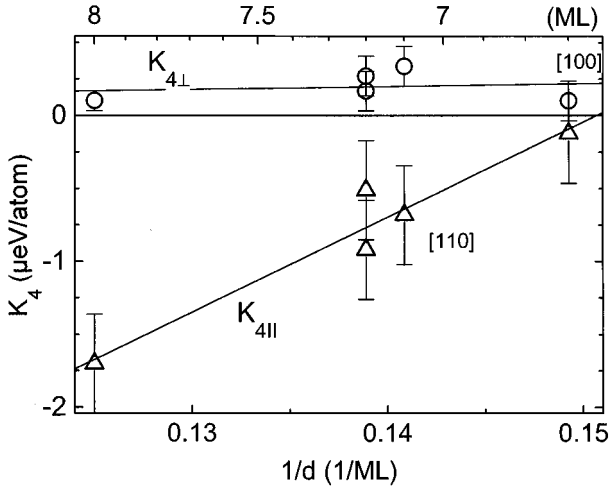

FIG. 4. Fourth-order anisotropy constants $K_{4\parallel}$ and $K_{4\perp}$ as a function of reciprocal film thickness for Ni(001) near $T/T_c \approx 0.8$. $K_{4\parallel}$ changes sign at $d \approx 6.7$ ML.

Results for $K_{4\perp}$ and $K_{4\parallel}$ as a function of film thickness at $T/T_c \approx 0.8$ are given in Fig. 4. One finds that $K_{4\perp} \approx 0.24$ μeV/atom is nearly thickness independent for 6 ML $< d <$ 8 ML,[40] and much smaller than $K_2 \approx 10$ μeV/atom. Interestingly, due to the negligible thickness dependence $K_{4\perp}$ is not expected to be the dominant mechanism for the thickness-dependent out-of-plane SRT. $K_{4\parallel}$, on the other hand, shows a stronger thickness dependence and follows a $1/d$ law within the error bars. One may conclude that the fourth-order contribution $K_{4\parallel}$ is the intrinsic origin for the continuous out-of-plane reorientation. $K_{4\parallel}$ changes sign at approximately 6.7 ML (Fig. 4). This indicates that an in-plane SRT occurs. The easy axis of magnetization rotates from [100] in the film plane to [110] for $d > 6.7$ ML at this temperature. However, a clear correlation between in-plane and out-of-plane SRT in the sense that they dependent on each other cannot be concluded from our data.

Linear regression of the data in Fig. 4 yields the fourth-order surface ($K_{4\parallel}^s$,$K_{4\perp}^s$) and volume ($K_{4\parallel}^v$,$K_{4\perp}^v$) anisotropies given in Table IV. The second- and fourth-order contributions are different in sign, that is to say for an infinitely thick fct Ni K_2^v favors the [001] and $K_{4\parallel}^v$ the [110] direction. $K_{4\parallel}^v$ and $K_{4\perp}^v$ differ by two orders of magnitude and have negative signs. This is different to the cubic anisotropy of bulk Ni $K_{\mathrm{bulk}} \approx +10^{-3}$ μeV/atom [$0.75 < T/T_c < 0.87$ (Ref.

TABLE IV. Second- and fourth-order surface and volume anisotropy constants for Ni(001) ($T/T_c \approx 0.8$) and Ni(111) on rough W(110). For easier comparison the original Ni(111) data (Refs. 27 and 36) have been recalculated (Table II) in terms of the free energy given in Eq. (1).

		K_i^V μeV/atom	K_i^S/1 ML (μeV/atom)
Ni(001)	$K_{4\parallel}$	−9.8	+33
	$K_{4\perp}$	−0.08	+1.0
	K_2	+29	−77
Ni(111)	K_2	+2.3	−18
	$K_{4\perp}$	+2	−20

48)], and shows that the tetragonal distortion not only introduces a second-order term K_2 [Eq. (1)] but also changes fourth-order contributions. $K_{4\perp}^s$ and $K_{4\parallel}^s$ are both positive in difference to a negative K_2^s. For completeness we would like to mention that this evaluation holds only at the given temperature and has been obtained in a narrow thickness interval only. Preliminary results which will be discussed elsewhere indicate that K_{4i} also strongly depends on temperature and sample history.

According to Fritsche *et al.*[30] three different types of SRT as a function of d can be distinguish if one considers second- and fourth-order contributions. Following their arguments we calculate the critical thicknesses[40] for full in-plane and out-of-plane orientation at $T/T_c \approx 0.8$:

$$d_{c2} = \frac{2(K_2^s + K_{4\perp}^s)}{2\pi M^2 - (K_2^v + K_{4\perp}^v)} = 7.1 \pm 1 \text{ ML}, \tag{3a}$$

$$d_{c1} = \frac{2K_2^s - K_{4\parallel}^s}{2\pi M^2 - K_2^v + 0.5 \times K_{4\parallel}^v} = 7.1 \pm 1 \text{ ML}. \tag{3b}$$

Within the error bar these values are equal and lie within the thickness interval for a canted magnetization observed by angular FMR. From $d_{c1} \approx d_{c2}$ one might have concluded that the thickness-dependent reorientation is of first-order or a *special* transition. Note that according to Fritzsche *et al.*[30] a *special* transition for $d_{c1} = d_{c2}$ is the only one at which the total anisotropy really disappears. Figure 3, however, unambiguously demonstrates that a stable canted magnetization exists and that the SRT is of second- or higher order. For Ni(111)/W(110) an analysis of fourth-order anisotropy at different film thicknesses was only possible for the rough substrate.[36] For shortness we will not repeat the free-energy model used in the analysis of Ni(111),[36] but present the result only (Fig. 5). To ease the comparison between the different free-energy models we recalculated the original Ni(111) data in terms of Eq. (1). The recalculated Ni(111) data are shown in Fig. 5 as a function of film thickness and are also plotted in Fig. 1(a) normalized by $2\pi M^2$. Unfortunately, the FMR measurements were not taken at the same reduced temperature, resulting in the scatter of the data points in Fig. 5. Nevertheless, one can estimate the higher-order surface and volume contributions given in Table IV (last two lines) with a large error bar. Interestingly, in Ni(111) the magnitude of $K_{4\perp}^v$ is comparable to K_2^v, which may indicate that uniaxial distortion plays a negligible role in this thickness regime. This is different to the Ni(001) fct case, where the strongly enhanced second-order term reflects the large tetragonal distortion. The magnitude of $K_{4\perp}^s$ relative to K_2^s has no special implications, since there is no thickness dependent SRT at any thickness.

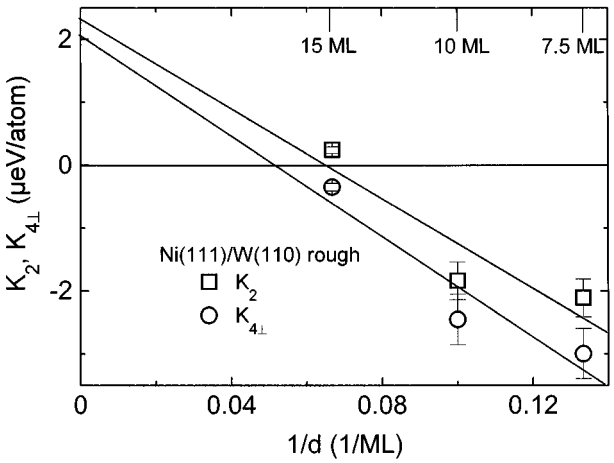

FIG. 5. Second- and fourth-order anisotropy for Ni(111)/rough W(110) as a function of reciprocal film thickness (1 ML = 2.035 Å). Values for 15, 10, and 7.5 ML were obtained at 550 K ($T/T_c = 0.9$), 300 K ($T/T_c = 0.5$), and 300 K ($T/T_c = 0.6$).

V. SUMMARY

The out-of-plane reorientation of the magnetization in fct Ni/Cu(001) has been identified as a second- or higher-order phase transition. Between 7 and 8 ML stable intermediate angles of the magnetization have been measured by angular dependent FMR. Near 6.7 ML a reorientation within the film plane from [100] to [110] is also found. The origin for the continuous transition is tentatively ascribed to the fourth-order in-plane MAE $K_{4\parallel}$, which is experimentally determined. A consequence of $K_{4\parallel}$ may also be the occurrence of the in-plane reorientation at about the same thickness. Theoretically, however, both reorientations are not expected to be related. Furthermore, we have studied the temperature dependence of the surface and volume magnetic anisotropy in Ni(001)/Cu(001) and in Ni(111)/W(110). A linear extrapolation of K_2^v as a function of T/T_c yields experimental values for the anisotropy at 0 K of tetragonally distorted Ni(001) bulk and isotropically strained Ni(111) bulk, which can be compared to theoretical calculation. For fct Ni(001) we extrapolate $K_2^v \approx +72$ μeV/atom. An extrapolation of K_2^s ($T \to 0$ K) yields a monolayer magnetic anisotropy of -98 μeV/atom which agrees very well with a theoretical value $K_2^{\mathrm{ML}} = -94$ μeV/atom obtained for the Ni monolayer on Cu(001).[47]

Note added in proof. After submission of this manuscript an SRT calculation by Millev *et al.*[51] was published, which is in good agreement with our analysis.

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[1] U. Gradmann, *Handbook of Magnetic Materials* (Elsevier, Amsterdam, 1993), Vol. 7, p. 1.
[2] R. Allenspach, J. Magn. Magn. Mater. **129**, 160 (1994), and references therein.
[3] J. Thomassen, F. May, B. Feldmann, M. Wuttig, and H. Ibach, Phys. Rev. Lett. **69**, 3831 (1992).
[4] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **70**, 1006 (1993).
[5] D. P. Pappas, K. P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).
[6] Dongqi Li, M. Freitag, J. Pearson, Z. Q. Qiu, and S. D. Bader, Phys. Rev. Lett. **72**, 3112 (1994).
[7] A. Berger and H. Hopster, Phys. Rev. Lett. **76**, 519 (1996).
[8] G. Garreau, E. Beaurepaire, K. Ounadjela, and M. Farle, Phys. Rev. B **53**, 1083 (1996).
[9] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13 467 (1994).
[10] G. Bochi, C. A. Ballentine, H. E. Inglefield, C. V. Thompson, R. C. O'Handley, Hans J. Hug, B. Stiefel, A. Moser, and H.-J. Güntherodt, Phys. Rev. B **52**, 7311 (1995).
[11] F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, Phys. Rev. B **49**, 3962 (1994).
[12] W. L. O'Brien and B. P. Tonner, Phys. Rev. B **50**, 12 672 (1994).
[13] M. Farle, W. Platow, A. N. Anisimov, B. Schulz, and K. Baberschke, J. Magn. Magn. Mater. (to be published).
[14] P. Bruno and J. P. Renard, Appl. Phys. A **49**, 499 (1989).
[15] We chose the nomenclature which distinguishes spin-orbit (K_i) and dipolar ($2\pi M^2$) contributions. In other literature a mixed term $K_{2\text{eff}}=K_2+2\pi M^2$ is sometimes used.
[16] A. Moschel and K. D. Usadel, Phys. Rev. B **51**, 16 111 (1995).
[17] A. Hucht and K. D. Usadel, J. Magn. Magn. Mater. **156**, 423 (1996), and references therein.
[18] P. J. Jensen and K. H. Bennemann, Phys. Rev. B **52**, 16 012 (1995); **42**, 849 (1990).
[19] D. L. Mills, J. Magn. Magn. Mater. **100**, 515 (1991), and references therein.
[20] S. T. Chui, Phys. Rev. B **50**, 12 559 (1994); Phys. Rev. Lett. **74**, 3896 (1995).
[21] S. Müller, B. Schulz, G. Kostka, M. Farle, K. Heinz, and K. Baberschke, Surf. Sci. **364**, 235 (1996).
[22] J. Shen, J. Giergiel, and J. Kirschner, Phys. Rev. **52**, 8454 (1995).
[23] R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385 (1992).
[24] R. Jungblut, M. T. Johnson, J. de Steege, A. Reinders, and F. J. A. den Broeder, J. Appl. Phys. **75**, 6424 (1994).
[25] Yi Li, M. Farle, and K. Baberschke, J. Magn. Magn. Mater. **93**, 345 (1991).
[26] R. F. Pearson, in *Experimental Magnetism*, edited by G. M. Kalvius and R. S. Tebble (Wiley, New York, 1979), Vol. 1, p. 138.
[27] A. Berghaus, M. Farle, Yi Li, and K. Baberschke, *Magnetic Properties of Low-Dimensional Systems II: New Developments*, Springer Proceedings in Physics Vol. 50 (Springer-Verlag, Berlin, 1990), p. 61.
[28] A. Aspelmeier, M. Tischer, M. Farle, M. Russo, K. Baberschke, and D. Arvanitis, J. Magn. Magn. Mater. **146**, 256 (1995).
[29] W. A. Lewis, M. Farle, B. M. Clemens, and R. L. White, J. Appl. Phys. **75**, 5644 (1994).
[30] H. Fritzsche, J. Kohlhepp, H. J. Elmers, and U. Gradmann, Phys. Rev. B **49**, 15 665 (1994).
[31] H. Fritzsche, H. J. Elmers, and U. Gradmann, J. Magn. Magn. Mater. **135**, 343 (1994).
[32] S. Chikazumi, *Physics of Magnetism* (Krieger, Malabar, 1964).
[33] R. Bergholz and U. Gradmann, J. Magn. Magn. Mater. **45**, 389 (1984).
[34] G. André, A. Aspelmeier, B. Schulz, M. Farle and K. Baberschke, Surf. Sci. **326**, 275 (1995).
[35] M. Farle, A. Berghaus, Yi Li, and K. Baberschke, Phys. Rev. B **42**, 4873 (1990).
[36] Yi Li and K. Baberschke, Phys. Rev. Lett. **68**, 1208 (1992); in *Magnetic Surfaces, Thin Films, and Multilayers*, edited by S. S. P. Parkin *et al.*, MRS Symposia Proceedings No. 231 (Materials Research Society, Pittsburgh, 1992), p. 289.
[37] B. Schulz, A. Aspelmeier, and K. Baberschke, Vacuum **46**, 1189 (1995).
[38] Full angular scans of H_R yield more detailed information than techniques which measure only one in-plane and one perpendicular orientation with respect to the film plane. In Ref. 27, for example, it is shown that the later measurement can give ambiguous results. A full angular scan, yields a unique set of higher-order anisotropy parameters.
[39] B. Heinrich, J. F. Cochran, M. Kowalewski, J. Kirschner, Z. Celinski, A. S. Arrott, and K. Myrtle, Phys. Rev. B **44**, 9348 (1991).
[40] B. Schulz, Ph.D. thesis, Freie Universität, Berlin, 1995.
[41] S. Müller, A. Kinne, M. Kottcke, R. Metzler, P. Bayer, L. Hammer, and K. Heinz, Phys. Rev. Lett. **75**, 2859 (1995).
[42] G. Lilienkamp, C. Koziol, and E. Bauer, in *Reflection High Energy Electron Diffraction*, edited by P. K. Larsen and P. J. Dobson (Plenum, New York, 1988).
[43] K. P. Kämper, W. Schmitt, and G. Güntherodt, Phys. Rev. B **38**, 9451 (1988).
[44] G. Bochi, C. A. Ballentine, H. E. Inglefield, S. S. Bogomolov, C. V. Thompson, and R. C. O'Handley, in *Magnetic Ultrathin Films, Multilayers and Surfaces/Interfaces and Characterization*, edited by B. T. Jonker *et al.*, MRS Symposia Proceedings No. 313 (Materials Research Society, Pittsburgh, 1993), p. 309; R. Naik, C. Kota, J. S. Payson, and G. L. Dunifer, Phys. Rev. B **48**, 1008 (1993).
[45] K. Baberschke, Appl. Phys. A **62**, 417 (1996).
[46] G. Bochi, C. A. Ballentine, H. E. Inglefield, C. V. Thompson, and R. C. O'Handley, Phys. Rev. B **53**, 1729 (1996).
[47] T. H. Moos, W. Hübner, and K. H. Bennemann, Solid State Commun. **98**, 639 (1996).
[48] M. B. Stearns, in *Magnetic Properties of Metals, 3d, 4d and 5d Elements, Alloys, and Compounds*, Landolt-Börnstein, Vol. III/19a, edited by H. P. J. Wijn (Springer, Berlin, 1986).
[49] J. Trygg, B. Johansson, O. Erickson, and J. M. Wills, Phys. Rev. Lett. **75**, 2871 (1996); O. Eriksson (private communication).
[50] U. Gradmann, J. Magn. Magn. Mater. **54-57**, 733 (1986).
[51] Y. Millev *et al.*, Phys. Rev. B **54**, 4137 (1996).