Critical evolution of spin-reorientation transition in magnetic $Co_xNi_{1-x}/Cu(100)$ films upon precise variation of *d*-band filling

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The ultrathin $Co_xNi_{1-x}/Cu(100)$ alloy films with low Co concentration $x \leqslant 10\%$ was prepared for probing the effect of the *d*-band filling on the magnetic behavior. The perpendicular magnetization was observed only for films with $x < 10\%$. The spin-reorientation transition from the in-plane to perpendicular orientation was found to be extremely sensitive to the Co concentration. The critical thickness for the spin reorientation transition was changed drastically from 7.5 to 17.5 ML by varying x from 0 to 8 %. Since no significant change in structure and surface morphology was accompanied, these findings may be attributed to the critical influence of the *d*-band filling on the magnetic anisotropy at variation of alloy composition.

Magnetic ultrathin films reveal unique magnetic properties due to reduced dimensionality and enhanced surface effect. One of the most important properties is the spin-reorientation transition (SRT). The SRT marks the switching behavior of the magnetization orientation at variation of various physical parameters, such as film thickness,[1,2] temperature,[1,4,5] structural transformation,[6,7] and alloy composition.[8] Three different magnetic ultrathin film systems from $3d$-transition elements, such as fcc-like Fe, Co, and Ni films are the most familiar examples revealing the various typical features of the SRT. The SRT for these three systems behaves totally different from each other in spite of only minimum difference in electronic structure or the average *d*-band filling: For Fe/Cu(100) (Refs. 3,4,9–11 or $Fe/Cu_3Au(100)$,[7] the magnetic easy axis is perpendicular to the film plane at low coverages due to the perpendicular magnetocrystalline surface anisotropy, and switches, driven by a fcc-bcc transformation due to the structural instability, to the in-plane orientation above a critical thickness.[6,7] The Co/Cu(100) films show, however, only in-plane magnetization due to the in-plane magnetoelastic anisotropy.[12–15] In contrast to the usual SRT found in Fe/Cu(100),[4,11] Fe/Ag(100),[1] and Co/Cu(111),[16] the Ni/Cu(100) system[16–19,22] reveals an *inverse* SRT at about 7–10 ML, namely, a magnetization switching from the in-plane to perpendicular orientation with increasing thickness. The perpendicular magnetization of fcc-like Ni films can even exist up to 35~70 ML depending on the film preparation. Different from the usual SRT, which is due to an enhanced magnetocrystalline surface anisotropy, the inverse one for the Ni/Cu(100) is attributed to a strain-induced positive volume term, which can overcome the negative shape anisotropy as well as the surface term, and becomes dominate above a critical thickness.[19]

The physical origins of various features of the SRT illustrated above should be traced back to the influence of the average *d*-band filling or $3d$-electron number on the magnetic anisotropy. A direct way to probe this effect is to study the SRT of binary alloy ultrathin films from two neighboring elements such as Fe and Co or Co and Ni, in which a continuous change in *d*-band filling is simulated by varying the alloy composition with the structure invariant. A relatively complete and detailed study on the Fe_xCo_{1-x} alloy films has been reported by A. Dittschar *et al.*[8] Varying the Co composition from 23 to 10 % the critical thickness d_c for the SRT of the Fe_xCo_{1-x} films was shifted from about 2.0 to 3.8 ML. On the other hand, the previous studies on the CoNi/Cu(100) ultrathin alloy film system indicate only in-plane anisotropy without any SRT observed for $x \geqslant 10\%$ and film thickness up to 11 ML.[16] As mentioned above, the behavior of the magnetic easy axis for both Co and Ni films is dominated by the same physical origin, namely, the strain-induced or the magnetoelastic anisotropy. A continuous composition variation of the $Co_xNi_{1-x}/Cu(100)$ alloy films should be an ideal way to monitor the dependence of the strained-induced magnetic anisotropy upon the *d*-band filling. Since the magnetic behavior of the films with the low concentration of Co as well as at the higher coverage remain to be clarified, a systematic and precise investigation of the CoNi films, especially for the lower concentration of Co, is indispensable for this purpose.

In this work, we report a perpendicular anisotropy and critical influence of the Co composition on the SRT for the $Co_xNi_{1-x}/Cu(100)$ alloy films with $x \leqslant 10\%$. In contrast to the Fe_xCo_{1-x} system, an enormous shift of the d_c up to 10 ML for only 8% Co composition difference was observed.

All of experiments were *in-situ* carried out in a multi-function UHV system with a base pressure of less than 5×10^{-10} mbar. The system is equipped with facilities for low-energy electron diffraction (LEED), auger electron spectroscopy (AES), medium-energy electron diffraction (MEED), film evaporation guns of electron-bombardment type (EFM-3 OMICRON), sputter gun, and magneto-optical Kerr effect (MOKE). The single crystal Cu(100) with miscut $\leqslant 0.5°$ was used as the substrate. The Cu(100) substrate was cleaned after cycles of 2 keV Ar ion sputtering and followed by 5 min. annealing at 800 K for a flat surface. The alloy films were prepared at 300 K by Co-Ni codeposition using two evaporation guns. The film growth was monitored by MEED. In this way, the film thickness and deposition rate can be precisely determined and controlled within 0.05 ML in a layer-by-layer growth. This highly precise controlling of the deposition rate allow us to prepare the desired alloy film

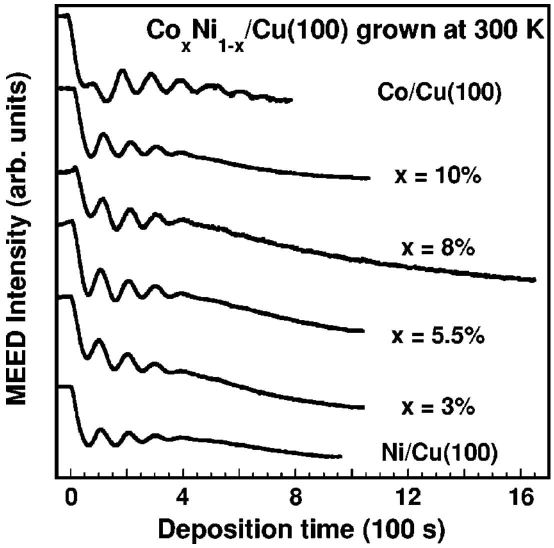

FIG. 1. MEED oscillations for the Co/Cu(100), Ni/Cu(100), and various $Co_xNi_{1-x}/Cu(100)$ films. The growth temperature is 300 K.

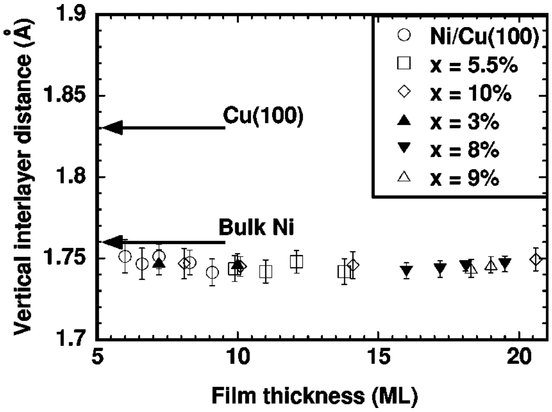

FIG. 2. Average vertical interlayer distance as function of the film thickness for various $Co_xNi_{1-x}/Cu(100)$ films and pure Ni films.

with an accuracy of $\pm 0.5\%$ composition value. A double check of the alloy composition was done by Auger electron spectroscopy.[20] The information on the crystalline structure and the vertical interlayer distance was obtained by LEED and I/V LEED, respectively. The perpendicular and in-plane components of magnetization were monitored by MOKE in polar and longitudinal geometry, respectively.

Figure 1 compiles the results of the MEED oscillations for the CoNi alloy films with different Co composition as well as the pure Co and Ni films grown at room temperature. The pure fcc-like Ni film on Cu(100) grows in a layer-by-layer mode up to 4–5 ML. On the other hand, the pure Co film reveals a bilayer growth at initial stage and, starting from the third layer, keeps on a layer-by-layer growth up to more than 7 ML. Both results of pure Ni and Co films are in good agreement with the previous studies,[16,23] providing a comparative experimental condition. For all of the alloy films with Co composition $x \leqslant 10\%$, the first four peaks of the MEED oscillation can be clearly identified, revealing nearly the same feature as that for the pure Ni film. It is not surprising that in such low Co composition, the Ni dominates the growth behavior and the resulting surface morphology of the alloy films.

In addition to the growth behavior, the structural properties of the alloy films with $x \leqslant 10\%$ are also almost the same as that of the pure Ni film. There is no significant difference in the LEED patterns between the alloy and pure Ni films. The spacing of the LEED spots keeps almost the same for all the films investigated, indicating a pseudomorphic growth of the films. In application of I/V LEED measurement, the average vertical interlayer distance can be extracted within the kinematic approximation by using the Bragg condition[7,24,25]

$$a_{\perp}(n) = \frac{n\pi\hbar}{\sqrt{2m(E_p + V_0)}\sin\theta}. \tag{1}$$

Here, the integer n is the order of the corresponding interference, E_p the primary energy of the electron, V_0 the additional energy shift due to the average innerpotential in the crystal, m electron mass, and θ the incident angle with respect the sample surface. The vertical interlayer distance can be thus determined by a linear regression of the E_p versus n^2.[25] The results are depicted in Fig. 2 for different alloy films. The values of the vertical interlayer distance in Fig. 2 are smaller than those for the Cu(100) and bulk Ni due to the in-plane tensile strain (-2.5%), revealing tetragonal distortion. One can also see that the vertical interlayer distances for all of the alloy films investigated are around 1.745 Å and almost the same as that of the pure Ni films. This indicates that not only the growth mode or the surface morphology as shown above, but also the crystalline structure for the alloy films with $x \leqslant 10\%$ is nearly the same as that for the pure Ni films. The effects of the 10% Co composition of the CoNi/Cu(100) alloy film on the structural and morphological properties are negligible. The composition variation can be thus interpreted as the d-band filling evolution with d-electron number.

The structural behavior found above is consistent with that of the previous study by dynamical LEED for the Ni/Cu(100) films,[9] which indicated also that both the in-plane and vertical spacing of the Ni layers are largely independent of both depth and film thickness. The values of the vertical spacing we obtained are, however, about 0.04 Å ($\sim 2\%$ of the lattice spacing) larger than those in Ref. 9, giving also about 2% difference at determining the magnetoelastic or strained-induced anisotropy. The effect of this uncertainty on the SRT will be shown to be minimal later.

An interesting phenomenological $(1/d)^{2/3}$ power dependence of both in-plane and out-of-plane strain for Cu/Ni/Cu/Si(100) films was found by Ha *et al.*, however, in a higher Ni thickness range of 30 Å $\leqslant d \leqslant$ 150 Å.[10] As mentioned above, the film growth in this work is pseudomorphic and tetragonal distorted throughout the thickness range investigated ($\leqslant 35$ Å). This behavior is largely the same as the finding of Ref. 9. The relaxation of the strain with the thickness seems to appear at higher coverages in this system, and both the in-plane and out-of-plane strain can be considered as constant throughout the thickness range investigated in this work.

Figure 3 presents the longitudinal and polar MOKE hysteresis loops for the pure Ni films and the

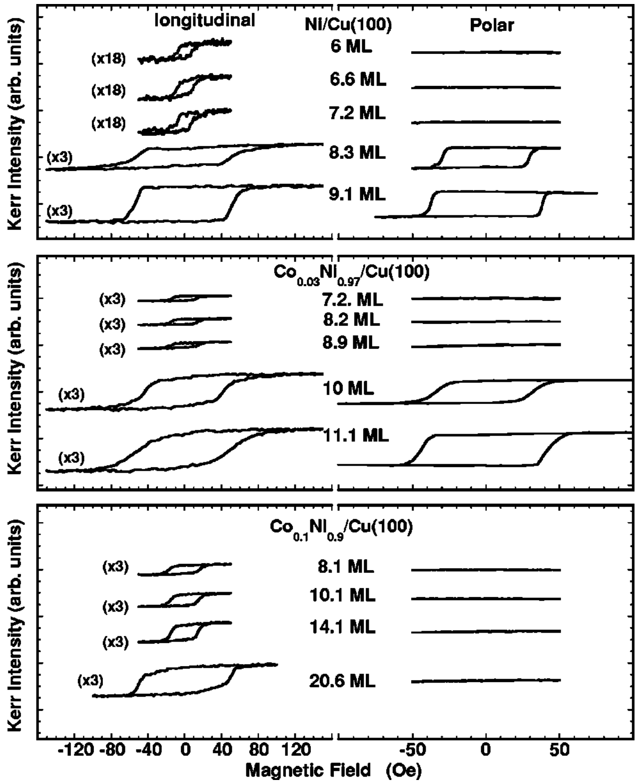

FIG. 3. Compliation of longitudinal and polar Kerr hysteresis loops for the Ni/Cu(100), $Co_{0.03}Ni_{0.97}$/Cu(100), and $Co_{0.1}Ni_{0.9}$/Cu(100) films. The measurement temperature is 110 K.

Co_xNi_{1-x}/Cu(100) films with $x=3$ and 10 %. For the pure Ni film, the critical thickness for the SRT from the in-plane to perpendicular orientation is found to be around 7.5 ML. This value agrees well with those of previous studies.[16,19,22] After the SRT, as early reported by Huang *et al.*,[16] the in-plane component coexists with the perpendicular one. The detailed analysis of this behavior, which will appear in our forthcoming paper,[21] indicates that the coexistence of the longitudinal and polar Kerr signals is related to a canted magnetization after the SRT due to high order term of the magnetic anisotropy.[5] Here, we will focus on the finding of the significant shift of the d_c for the SRT at variation of the alloy composition. As shown in Fig. 3, the $Co_{0.03}Ni_{0.97}$/Cu(100) film reveals the in-plane magnetization below ~9 ML, and an evident perpendicular Kerr component is found at coverages above 10 ML. The d_c is estimated to be around 9.5 ML, which is deviated clearly from the 7.5 ML for the pure Ni film. Only few percentages change in Co concentration can already delay the SRT up to 2 ML. This effect is much larger than that in the FeCo/Cu(100) system, where 2 ML d_c shift can be obtained only by Co composition variation larger than 15%.[8] Increasing the Co composition to 10%, the CoNi alloy film reveals only the in-plane magnetization up to ~21 ML. As compared to the growth and structural properties of the alloy film, the magnetic properties, or more precisely saying, the d_c for the SRT is much more sensitive to the Co composition.

Figure 4 shows a phase diagram of the magnetization orientation for the CoNi alloy films at variation of Co concentration and film thickness. It is clear to see that the critical thickness of the SRT increases drastically with increasing Co

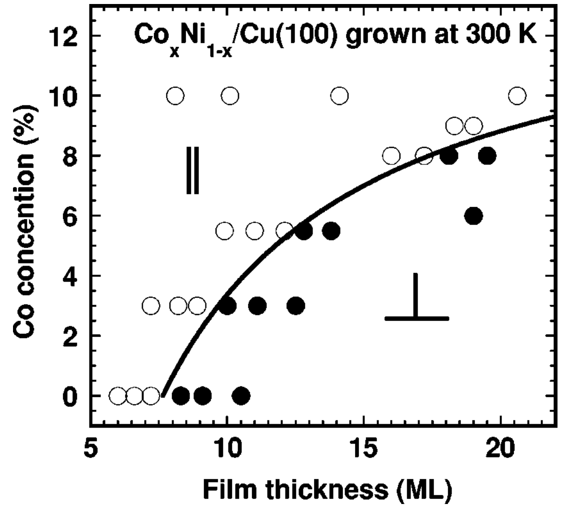

FIG. 4. The magnetic phase diagram of the magnetic easy axis at variation of Co concentration and film thickness. The in-plane and perpendicular orientations are denoted by empty and solid circles, respectively. The solid curve presents the calculated value using Eq. (3) and parameters described in text.

concentration. With the Co concentration only less than 8%, the d_c of the SRT changes from the 7.5 to ~17 ML. As indicated above, this d_c shifting is much larger than that observed in the FeCo alloy film. This implies that the magnetic anisotropy for the CoNi films is more sensitive to the change of *d*-band filling as compared to the FeCo system. This highly sensitive effect may explain the absence of the observation of the perpendicular CoNi/Cu(100) film in the previous study, for which a precise controlling of the Co concentration down to 10% is required.

At variation of the Co concentration, two possible modifications in crystalline structure and electronic structure are expected. First, the strain of the CoNi films must alter with the Co concentration because of different lattice mismatches for Co/Cu(100) (−1.9%) and Ni/Cu(100) (−2.5%). This effect is, however, negligible because the maximum value of the Co concentration in our alloy films is only 10% and the induced change in the lattice mismatch should be less than 0.06%. This can also be verified by our structural and morphological studies, as indicated above by MEED and I/V LEED data, where the crystalline structure and surface morphology of the CoNi films investigated are nearly invariant. Secondly, the composition variation changes the *d*-band filling or the electronic structure of the film and in turn may alter the strain-induced magnetoelastic anisotropy constant and magnetic moment. This evolution could make a crossover from the positive magnetic anisotropy to the negative one with the increasing Co concentration. A simple phenomenological analysis for our data is made as follows.

Phenomenologically, considering only the low order terms, the critical thickness d_c is determined by the magnetic volume (K_v) and surface (K_s) anisotropies as

$$d_c=\frac{-2K_s}{K_v-2\pi M^2}, \tag{2}$$

where $-2\pi M^2$ stands for the shape anisotropy and M denotes the magnetic moment density. The K_v in this system is mainly contributed by the strain-induced magnetoelastic anisotropy.[14,19,26] Assuming that the magnetic moment density M, volume anisotropy K_v, and surface anisotropy K_s vary linearly with the Co concentration x, d_c can thus be expressed as

$$d_c = \frac{-2[xK_s^{\mathrm{Co}} + (1-x)K_s^{\mathrm{Ni}}]}{[xK_v^{\mathrm{Co}} + (1-x)K_v^{\mathrm{Ni}}] - 2\pi[xM^{\mathrm{Co}} + (1-x)M^{\mathrm{Ni}}]^2}, \tag{3}$$

where x is the composition. The solid curve depicted in Fig. 4 presents the calculated d_c, by substituting the values of M, K_v, and K_s from the previous studies into Eq. (3). [$M^{\mathrm{Ni}} = 0.57\ \mu_{\mathrm{B}}$,[27] $M^{\mathrm{Co}} = 1.8\ \mu_{\mathrm{B}}$,[27] $K_v^{\mathrm{Ni}} = 29\ \mu$eV/atom,[19] $K_v^{\mathrm{Co}} = -73.8\ \mu$eV/atom,[14] $K_s^{\mathrm{Ni}} = -77\ \mu$eV/atom,[19] $K_s^{\mathrm{Co}} = -55.8\ \mu$eV/atom (Ref. 14)]. The solid curve agrees surprisingly well with the crossover boundary or the experimental values of the d_c. This clearly indicates that the drastic effect of the Co composition on the SRT can be mainly explained by the evolution of the magnetic anisotropy and magnetic moment. Since the crystalline structure of all the films investigated did not show any significant difference, the drastic influence of the Co concentration on the SRT in the CoNi alloy films should be traced back to the change in electronic structure.

The influence of the fractional $3d$-band filling on the magnetic moment and anisotropy has been studied in detail with a first-principles calculation.[29,28] For a bulk system of the $3d$ transition binary alloy, the magnetic moment at variation of the alloy composition was calculated as a function of the hole number of the $3d$-band.[29] A linear dependence of the average magnetic moment on the effective Z alloy number was found for the the Ni alloyed with Co and Fe (but not Cu). The Ni magnetic moment in such alloys is saturated (constant) by the magnetic surrounding Fe or Co, which has a larger magnetic moment than the Ni. This behavior in the bulk seems also true in a thin film system, as indicated by the surprising matching between the calculated d_c from Eq. (3) and the measured ones shown in Fig. 4.

Concerning the variation of the magnetic anisotropy in a binary alloy system with two neighboring elements, James *et al.* has reported that in particular for the *d*-electron number close to that of Ni, the orbital moment anisotropy of the binary alloy CoNi varies drastically with the *d*-band filling and even change their sign.[28] Moreover, as indicated in Ref. 28, in particular for the CoNi alloy, there is a linear relationship between the calculated orbital moment anisotropy and magnetostriction. Since there is also a linear relationship between the orbital moment anisotropy and strained-induced magnetic anisotropy, the results in Ref. 28 suggests thus that, in addition to the linear variation of the magnetic moment mentioned above, the variation of strain-induced magnetoelastic anisotropy due to the modulation of the magnetostriction coefficients with the *d*-band filling should be responsible for the dramatic shift of the d_c in the binary CoNi alloy films. The agreement of the analysis by the phenomenological model [Eq. (3)] with the experimental SRT results indicates thus that the alloying effect on the SRT is attributed to the variation of the magnetoelastic anisotropy and magnetic moments as a function of *d*-band filling.

Finally, it should be pointed out that, as mentioned above, the magnetoelastic anisotropy may have about 2% uncertainty because the vertical interlayer distance obtained in this work is ~2% larger than the one in Ref. 9. However, substituting this value into Eqs. (2) or (3), this effect may only lead to a minimal change of ~2% in the d_c (about 0.2 ML for 10 ML thickness) of the SRT.

In conclusion, a perpendicular anisotropy was found in the CoNi binary alloy films for Co concentration less than 10%. In contrast to the FeCo system, the critical thickness for the *inverse* SRT alters drastically with Co concentration. This effect can be traced back to the critical change of the magnetic anisotropy as well as the dependence of average magnetic moment upon *d*-band filling variation near the Ni. Our result provides a direct insight into the connection between the magnetic anisotropy and the *d*-band filling or stoichiometry.

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[1] D. P. Pappas, C. R. Brundle, and H. Hopster, Phys. Rev. B **45**, 8169 (1992).

[2] J. A. C. Bland, G. A. Gehring, B. Kaplan, and C. Daboo, J. Magn. Magn. Mater. **107**, 173 (1992).

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

[4] D. P. Pappas, K.-P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).

[5] K. Baberschke and M. Farle, J. Appl. Phys. **81**, 5038 (1997).

[6] S. Müller, P. Bayer, C. Reischl, K. Heinz, B. Feldmann, H. Zillgen, and M. Wuttig, Phys. Rev. Lett. **74**, 765 (1995).

[7] M.-T. Lin, J. Shen, W. Kuch, H. Jenniches, M. Klaua, C. M. Schneider, and J. Kirschner, Phys. Rev. B **55**, 5886 (1997).

[8] A. Dittschar, M. Zharnikov, W. Kuch, M.-T. Lin, C. M. Schneider, and J. Kirschner, Phys. Rev. B **57**, R3209 (1998).

[9] W. Platow, U. Bovensiepen, P. Poulopoulos, M. Farle, K. Baberschke, L. Hammer, S. Walter, S. Müller, and K. Heinz, Phys. Rev. B **59**, 12 641 (1999).

[10] K. Ha, M. Ciria, R. C. O'Handley, P. W. Stephens, and P. Pagola, Phys. Rev. B **60**, 13 780 (1999).

[11] J. Thomassen, F. May, B. Feldmann, M. Wuttig, and H. Ibach, Phys. Rev. Lett. **69**, 3831 (1992).

[12] C. M. Schneider, P. Bressler, P. Schuster, J. Kirschner, J. J. d. Miguel, and R. Miranda, Phys. Rev. Lett. **64**, 1059 (1990).

[13] C. C. Kuo, W. C. Lin, and M.-T. Lin (unpublished).

[14] M. Kowalewski, C. M. Schneider, and B. Heinrich, Phys. Rev. B **47**, 8748 (1993).

[15] P. Krams, F. Lauks, R. L. Stamps, B. Hillebrands, and G. Güntherodt, Phys. Rev. Lett. **69**, 3674 (1992).

[16] F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, Phys. Rev. B **49**, 3962 (1994).

[17] W. L. O'Brien and B. P. Tonner, Phys. Rev. B **49**, 15 370 (1994).

[18] G. Bochi, C. A. Ballentine, H. E. Inglefield, C. V. Thompson, R. C. O'Handley, H. J. Hug, B. Stiefel, A. Moser, and H. J. Gunderodt, Phys. Rev. B **52**, 7311 (1995).

[19] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13 467 (1994).
[20] W. C. Lin, C. C. Kuo, and M.-T. Lin (unpublished).
[21] C. C. Kuo, W. C. Lin, and M.-T. Lin (unpublished).
[22] S. Z. Wu, G. J. Mankey, F. Huang, and R. F. Willis, J. Appl. Phys. **76**, 6434 (1994).
[23] J. Shen, J. Giergiel, and J. Kirschner, Phys. Rev. B **52**, 8454 (1995).
[24] M. Zharnikov, A. Dittschar, W. Kuch, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **76**, 4620 (1996).
[25] M.-T. Lin, J. Shen, W. Kuch, J. Jenniches, M. Klaua, C. M. Schneider, and J. Kirschner, Surf. Sci. **410**, 290 (1998).
[26] R. Jungblut, M. T. Johnson, J. a. d. Stegge, A. Reinders, and F. J. A. d. Broeder, J. Appl. Phys. **75**, 6424 (1994).
[27] R. F. Willis, J. A. C. Bland, and W. Schwarzacher, J. Appl. Phys. **63**, 4051 (1988).
[28] P. James, O. Eriksson, O. Hjortstam, B. Johansson, and L. Nordström, Appl. Phys. Lett. **76**, 915 (2000).
[29] P. James, O. Eriksson, B. Johansson, and I. A. Abrikosov, Phys. Rev. B **59**, 419 (1999).