Lattice Relaxation Driven Reorientation Transition in $Ni_n/Cu(100)$

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The magnetic anisotropy energy of $Ni_n/Cu(100)$ is calculated in terms of the spin-polarized fully relativistic Korringa-Kohn-Rostoker method including surface relaxation by using 2D structure constants originally described for low-energy electron diffraction calculations. Investigating different relaxations, an explanation for the reorientation transition from in-plane to perpendicular can be given. For a relaxation of -5.5% ($c/a = 0.945$) this reorientation occurs at about seven layers of Ni and yields second order terms to the magnetic anisotropy energy that are in excellent agreement with experiment. [S0031-9007(98)08322-7]

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Thin films of Ni on Cu(100) show an unexpected behavior of magnetic phase transitions [1–12]. In contrast to quite a few other magnetic thin films on noble metal surfaces, where with increasing film thickness the moments first line up perpendicular to the surface and then reorientate to an in-plane direction, for $n < 7$ monolayers of Ni on Cu(100) an in-plane direction of the magnetization M is observed [5,6,10] that reorientates [5,6,10,11] at about $n = 7$ to perpendicular to plane, switching eventually back to in plane for $n > 37$ [5]. The anisotropic part of the free energy for magnetic multilayer systems (see, e.g., Ref. [2] and references therein) can phenomenologically be described by

$$E = 2\pi M^2 \cos^2\theta - K_2 \cos^2\theta\,, \tag{1}$$

$$K_2 = K_2^v + K_2^s/d\,, \tag{2}$$

where K_2 refers to the second order term of the magnetic anisotropy energy (MAE) and θ denotes the angles of M with respect to the surface normal. As indicated in Eq. (2), where d refers to the film thickness, K_2 is thought to consist of two parts, namely, a thickness-independent "volume"-like contribution K_2^v and a thickness-dependent "interface-surface"-like contribution K_2^s. It is more or less the interplay of these two constants that together with the shape anisotropy (magnetic dipole-dipole interaction) determines the unique features of the reorientation properties of thin films of Ni on Cu(100).

In the present paper the fully relativistic spin-polarized screened Korringa-Kohn-Rostoker (KKR) method [13,14] was applied using the spin-polarized local density functional as given by Vosko *et al.* [15]. In order to be able to treat layer relaxation, the occurring screened structure constants [16,17] have been derived for a system of layers which have only the same in-plane translational symmetry [18–20], but otherwise can differ in the respective interlayer distance. Self-consistency for the effective potentials and effective exchange fields (with a uniform orientation perpendicular to plane) was obtained using 45 $k_\parallel$ points in the irreducible part of the surface Brillouin zone (ISBZ). In all cases three layers of Cu served as "buffer" at the Cu/Ni interface and three "empty" layers as buffer to the Ni/vacuum interface; i.e., for a given number n of Ni monolayers the total number L of atomic layers investigated self-consistently is $L = n + 6$. It should be noted that the present calculations refer to free surfaces of Ni on Cu(100), which implies that in here actually relaxation effects for a semi-infinite system are included and not bulklike relaxations as in the paper by Hjortstam *et al.* [8].

The magnetic anisotropy energy ΔE_a,

$$\Delta E_a = E(\parallel) - E(\perp)\,, \tag{3}$$

defined as the energy difference between a uniform in-plane (perpendicular to the surface normal in all planes of atoms) and a uniform perpendicular (along the surface normal in all planes of atoms) orientation of the magnetization of the system was obtained [13,14] by making use of the force theorem approximation, namely, as a sum of the respective band energy difference ΔE_b and the magnetic dipole-dipole energy contribution ΔE_{dd},

$$\Delta E_a = \Delta E_b + \Delta E_{dd}\,. \tag{4}$$

In order to evaluate ΔE_b 990 $k_\parallel$ points in the ISBZ were used, guaranteeing well converged quantities.

In Fig. 1 the magnetic anisotropy energy ΔE_a is shown together with the corresponding band energy difference ΔE_b and magnetic dipole-dipole energy difference ΔE_{dd} versus the number of Ni layers. Three cases are displayed, namely: (a) The values corresponding to an fcc "parent lattice" [21] with respect to the experimental lattice spacing of fcc Cu ($a_0 = 6.8309$ a.u.); (b) those corresponding to a (uniform) relaxation of the interlayer distance by -2.5%; and (c) for an interlayer relaxation of -5.5%. It should be noted that case (b) refers to a c/a ratio of 0.975 and (c) to a c/a ratio of 0.945 (experimental value [4,8]), since the (two-dimensional) lattice spacing within the planes of atoms remains unchanged. As can be seen from Fig. 1, the shape of the ΔE_a curve arises mainly from the band energy contribution ΔE_b, since ΔE_{dd} scales linearly with the volume and is always negative (ΔE_{dd} tends to turn the magnetization in plane). Relaxation of the interlayer distances leaves ΔE_{dd} nearly unchanged, however, as can be seen from Fig. 1, it is of crucial importance for ΔE_b.

In Fig. 2 the layer resolved band energy differences ΔE_b^p for six, nine, and twelve Ni layers are shown for the three cases mentioned above. One can see that the surface and interface contributions are negative, the surface giving the larger contribution, and that relaxation predominantly increases the contribution from the interior of the Ni films. Defining therefore the following quantities:

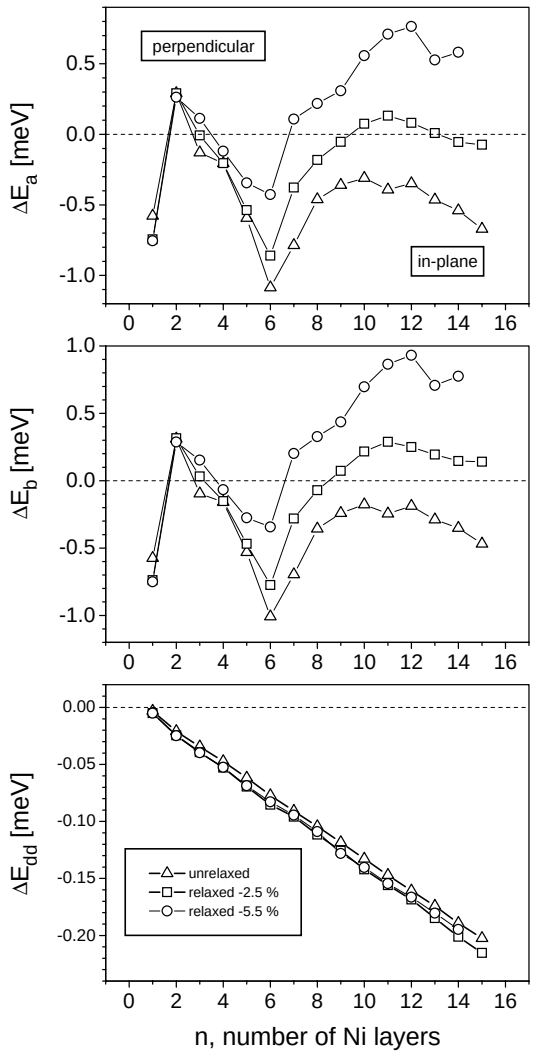

FIG. 1. Magnetic anisotropy energy ΔE_a (top), band energy difference ΔE_b (middle), and magnetic dipole-dipole energy difference ΔE_{dd} (bottom) versus the number of Ni layers on Cu(100). Triangles, squares, and circles refer in turn to a uniform relaxation by 0%, -2.5%, and -5.5%, i.e., to a c/a ratio of 1, 0.975, and 0.945.

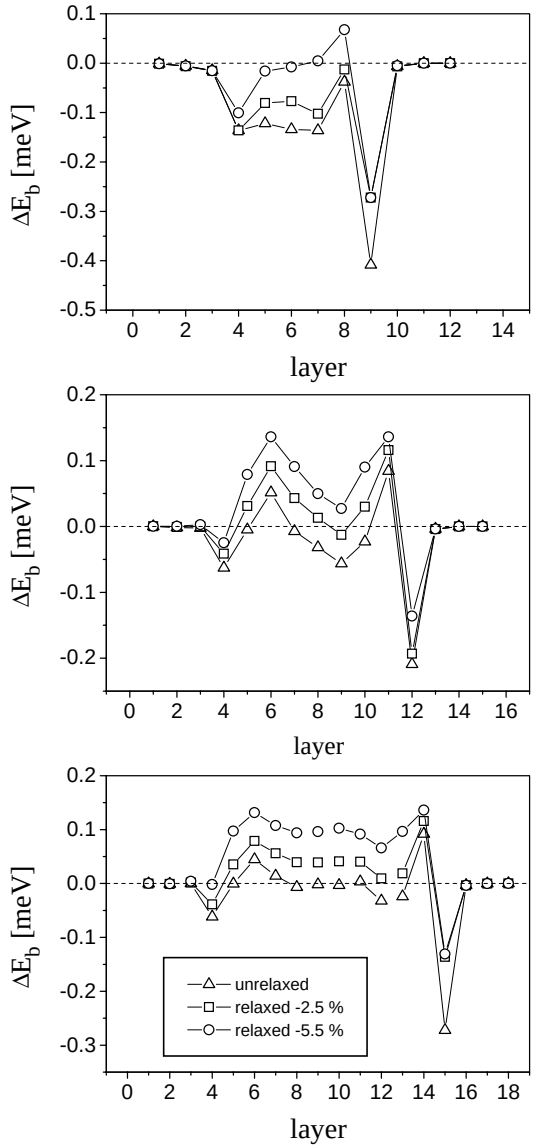

FIG. 2. Layer resolved band energy differences ΔE_b^n for six (top), nine (middle), and twelve (bottom) Ni layers on Cu(100). Triangles, squares, and circles refer in turn to a uniform relaxation by 0%, -2.5%, and -5.5%, i.e., to a c/a ratio of 1, 0.975, and 0.945.

$$K_2^{S_1} = \sum_{p=1,4} \Delta E_b^p, \qquad K_2^{S_2} = \sum_{p=L-3,L} \Delta E_b^p, \quad (5)$$

$$K_2^I = \frac{1}{(L-8)} \sum_{p=5,L-4} \Delta E_b^p, \quad (6)$$

$$\Delta E_b = K_2^{S_1} + K_2^{S_2} + (L-8)K_2^I, \quad (7)$$

namely, a contribution to ΔE_b from the Cu/Ni interface ($K_2^{S_1}$), from the Ni/vacuum interface ($K_2^{S_2}$) and an averaged contribution from the interior (K_2^I), then one easily can identify these quantities with the second order constants in Eq. (2),

$$K_2^v = K_2^I, \qquad K_2^s = K_2^{S_1} + K_2^{S_2}. \quad (8)$$

Quite clearly, the only relevant quantity in Eqs. (5)–(8) is ΔE_b; the grouping of terms in the sum over layer dependent contributions to ΔE_b serves merely purposes of interpretation. For this very reason a comparison to the approach used by Cinal *et al.* [22] in calculating the magnetocrystalline anisotropy in ferromagnetic films does not seem to be very useful.

In Table I the present values for a relaxation of -5.5% ($c/a = 0.945$) are compared to the corresponding constants obtained experimentally. It should be noted that the "error bars" in this table for the present values arise from the fact that the theoretical data for both nine and twelve layers of Ni, shown in Fig. 2, were used. It is evident from this table that the present values agree very well with the experimental data. Furthermore, it is interesting to note that in comparison to Co and Fe on Cu(100), the sign of $K_2^{S_1}$ and $K_2^{S_2}$ is reverse, which in turn explains the in-plane orientation in the regime of $3 \leq n \leq 7$ Ni layers.

From a theoretical standpoint of view it is tempting to ask whether a kind of frozen potential approximation can be applied to this particular problem of relaxation, i.e., whether from the self-consistent potentials for the unrelaxed geometry one can calculate ΔE_b for a relaxed geometry. In Fig. 3 the band energy differences for nine layers of Ni on Cu(100) as calculated with this frozen potential approximation are compared to those corresponding to self-consistently determined potentials. As one can see from this figure only up to rather small relaxations ($\sim 2\%$) this kind of approximation seems to be useful. It is worthwhile to mention that up to -5.5% ΔE_b, as obtained from self-consistent potentials, varies almost linearly with the relaxation.

In conclusion, we have investigated the reorientation transition of the magnetization for free surfaces of Ni on Cu(100) by calculating the magnetic anisotropy energy within the fully relativistic spin-polarized KKR method including relaxation of the Ni layers. We found a reorientation transition from in-plane to perpendicular at ten and seven layers of Ni for -2.5% and -5.5% relaxation, respectively. This compares very well to the experimental results. Furthermore, not only the experimental observation of a negative surface and interface anisotropy constant could be confirmed, but also the actual values for the second order anisotropy constants are in excellent agreement with experiment. We can state therefore unambiguously that at $T = 0$ the reorientation transition of free surfaces of Ni on Cu(100) is driven by tetragonal distortion and that the actual number of layers at which this transition occurs is caused by a delicate balance between the interface ($K_2^{S_1}$), the surface ($K_2^{S_2}$), the interior (K_2^I) contributions to K_2, and, of course, the magnetic dipole-dipole contribution ΔE_{dd} to the magnetic anisotrop y energy ΔE_a.

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TABLE I. Volume and surface anisotropy constants (μeV) for free surfaces of Ni on Cu(100) at $T = 0$ K.

	K_2^v	K_2^s	c/a
Experiment [6,11]	70 ± 20	-100	~ 0.945
Ref. [4]	140^{+36}_{-90}		0.945
Present calculation	80 ± 20	-100 ± 20	0.945

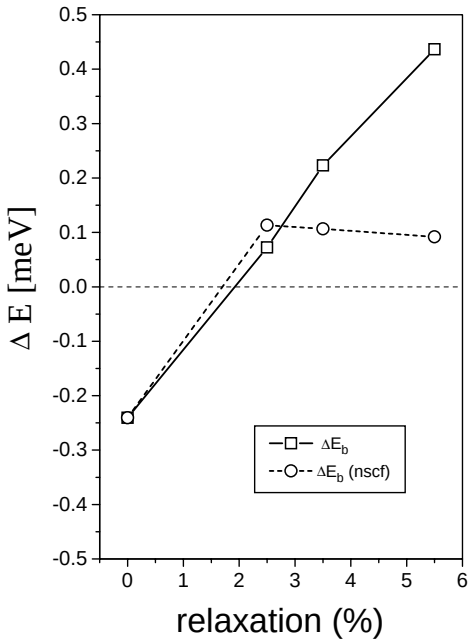

FIG. 3. Band energy difference (ΔE_b) for nine layers of Ni on Cu(100) versus relaxation. Circles denote a calculation using unrelaxed potentials, while squares refer to self-consistently relaxed potentials.

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