Magnetoelastic mechanism of spin-reorientation transitions at step-edges.

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Abstract

The symmetry-induced magnetic anisotropy due to monoatomic steps at strained Ni films is determined using results of first-principles relativistic full-potential linearized augmented plane wave (FLAPW) calculations and an analogy with the Néel model. We show that there is a magnetoelastic anisotropy contribution to the uniaxial magnetic anisotropy energy in the vicinal plane of a stepped surface. In addition to the known spin-direction reorientation transition at a flat Ni/Cu(001) surface, we propose a spin-direction reorientation transition in the vicinal plane for a stepped Ni/Cu surface due to the magnetoelastic anisotropy. We show that with an increase of Ni film thickness, the magnetization in the vicinal plane turns perpendicular to the step edge at a critical thickness calculated to be in the range of 16-24 Ni layers for the Ni/Cu(1,1,13) stepped surface.

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Ni/Cu(001) is known as a unique system to show an in-plane to out-of-plane spin-reorientation transition with an increase of Ni-overlayer thickness [1,2]. This is likely due to the positive magnetoelastic energy contribution to the magnetic anisotropy energy (MAE), caused by tetragonal strain in Ni-films due to the film/substrate lattice mismatch [3].

We have investigated the uniaxial MAE for flat and stepped Ni films, and use the results of first-principles MAE calculations for the strained Ni-films to estimate the in-plane to out-of-plane spin reorientation transition thickness. Further, we also use an analogy with the Néel model with parameters chosen from our first-principles calculations to estimate the step-induced contribution to the MAE and to discuss the possibility of a spin-reorientation transition at the Ni/Cu(001) monoatomic step.

The relativistic self-consistent version [5] of the full-potential linearized augmented plane-wave (FLAPW) method [6] is employed to perform self-consistent calculations for strained Ni films and bulk Ni with an (001) magnetization direction. The local force theorem for magnetization rotation is then used for the MAE calculations [7].

For strained bulk Ni, we choose the bct (001) plane lattice constant as 4.781 a.u. (equal to the Cu substrate) and the (001) interlayer distance as 3.222 a.u. in accordance with the experimental strained structure of a Ni magnetic overlayer on Cu(001) [8]. Here, 343 k-points in 1/8th of the tetragonal 3D BZ were used for the self-consistent calculations. Then 1728 k-points in 1/8th of the 3D BZ (13824 in the full BZ) are used for the MAE calculations. The MAE is shown in Table I. There is a very good agreement between our results and results of previous perturbative state-tracking FLAPW calculations [3] and relativistic full-potential linear muffin-tin orbital (FLMTO) calculations [4].

For Ni-films with a thickness (d) of 3, 5, 7, 9, 11 layers we choose a slab geometry with assumed experimental strained structure of Ni magnetic overlayer on Cu(001) which is the same as in the strained bulk Ni calculations. Here, 100 k-points in 1/4th of the 2D BZ (400 k-points in the full 2D BZ) were used for the self-consistent calculations and 1600 k-points (6400 k-points in the FBZ) in 1/4 of the 2D BZ were used for the MAE calculations.

The calculated MAE as a function of Ni-film thickness is shown in Fig.1. There is
a very good linear dependence of the MAE on film thickness. The uniaxial MAE of d
magnetic bct(001) layers in the [(100),(010),(001)] coordinate frame (denoted by V) is then
approximated by

\[ E = -d \cdot K_v \cdot V_z^2 - 2 \cdot K_s \cdot V_z^2 \]  

(1)

where, \( K_v \) is a “volume” MAE per atom, \( K_s \) is a “surface” MAE per atom, and \( V_z \) is
the magnetization direction cosine with respect to the z(V)-axis. By fitting the calculated
MAE (cf., Fig. 1) by Eq.(1), we obtain the “volume” \( K_v = 83.5 \mu eV \) and “surface”
\( K_s = -446.5 \mu eV \) contributions. The calculated “volume” contribution is in very good
quantitative agreement with the calculated uniaxial MAE for strained bulk-Ni (cf., Table I).
This is clear numerical evidence that the “volume” contribution to the MAE in strained Ni
films is the same as the MAE of strained bulk Ni, which is qualitatively consistent with Néel
model arguments [15,16] and supports quantitatively an analysis performed in Refs. [1,3].

Hjortstram et al. [4] have calculated the uniaxial MAE and magnetostriction in strained
bulk Ni and found good agreement between the linear magnetoelastic theory and the results
of first-principles calculations for strain-induced MAE in the limit of small strains. They
also found that for assumed experimental strained structure of Ni films the MAE value for
strained bulk Ni agrees well with measured “volume” contribution to the MAE for Ni films
on Cu(001) [1]. The present calculations for both strained bulk Ni and Ni films (cf., Table
I) agree well with the results of [4]. The fact, that “volume” MAE (85 \( \mu eV \)) is somewhat
bigger than the MAE for strained bulk Ni (57 \( \mu eV \)) can be due to computational differences
and neglect of the higher order terms in MAE expression Eq.(1) (which are small but not
necessarily negligible). Taking into account relatively small calculated MAE values we argue
that both the “volume” MAE for Ni films and MAE for strained Ni bulk agree surprisingly
well. This good numerical agreement makes it possible to conclude that “volume” MAE
in Ni films is due to the strain induced by film/substrate lattice mismatch. This confirms
quantitatively conclusions of Hjortstram et al. [4] based on comparison between experimental
data for flat Ni films and the results of strained bulk Ni calculations.
Finally, using Eq. (1) and the calculated MAE data (cf., Table III), we calculate the in-plane to out-of-plane spin-reorientation transition thickness as $d_c = 11$ ML, which agrees qualitatively with the experimentally observed spin-reorientation transition thickness of 7 ML for Ni/Cu(001). (The demagnetization energy is omitted here because it is very small for Ni [5].) It is necessary to mention that the use of free-standing Ni-films to analyze the MAE for Ni/Cu(001) is rather qualitative since we do not take into account the interface contribution to the MAE and the possible oscillations of the MAE with Ni-film thickness at the Ni/Cu(001) interface [3]. Moreover, the chosen geometry is an “average” experimental geometry and deviations due to relaxation are likely to occur in the real interface.

To proceed further, let us now assume the Néel pair-interaction form of the magnetic anisotropy energy ($E_{AB}$) [15] for the pair of atoms A and B connected by $R_{AB} = \vert R_{AB} \vert (\beta_x, \beta_y, \beta_z)$, with magnetic moment direction $\mathbf{m} = (V_x, V_y, V_z)$, where $\beta, V$ are the direction cosines; $L_{AB}$ is a Néel coefficient (the isotropic term is omitted). Hence,

$$E_{AB} = -L_{AB}\left(\sum_{i=x,y,z} \beta_i V_i\right)^2$$  \hspace{1cm} (2)

The “volume” and “surface” MAE contributions in Eq. (1) are then expressed as:

$$K_v = +L(6\beta_z^2 - 3)$$  \hspace{1cm} (3)

$$K_s = -L(3\beta_z^2 - 1)$$  \hspace{1cm} (4)

where, following Victora et al. [11], we consider only nearest-neighbor interactions and do not take into account the change of the Néel parameter due to the strain (which seems to be reasonable assumption in the limit of small strains, considered here).

The value of the Néel parameter, $L = -396 \mu eV$, is then calculated using Eq. (3), the calculated value of the “volume” anisotropy for strained bulk-Ni (cf., Table I) and $\beta_z^2 = 0.476$. The value of the “surface” anisotropy is then calculated using Eq. (4) and is 169.5 \mu eV. A comparison with the result of first-principle calculations (cf., Table III) clearly shows that this “isotropic” variant of the Néel model does not even reproduce the sign calculated from the first-principles “surface” MAE.
It is known [13] that there is a significant change in the electronic structure at the Ni-
surface compared to the bulk. This change is especially pronounced for the “surface” layer
and leads to significant modifications of the electronic density of states near the Fermi level
for the spin-minority band. This should lead to a significant difference in “in-plane” and
“inter-plane” Néel parameters for the surface layer. Therefore, it is quite natural to introduce
an additional in-plane Néel parameter $M$ to describe the pair-interaction, Eq.(2), within the
surface layer. This gives the following parametrization for the “surface” anisotropy:

$$K_s = -L(3\beta_z^2 - 1) + (L - M)$$

(5)

Then, we approximate the MAE of a strained Ni-film by choosing the Néel parameter
$L$ to fit the “volume” contribution to the MAE, Eq.(3), and then determine the in-plane
parameter $M$ to fit the “surface” contribution, Eq.(5). Using the calculated values of the
“volume” and “surface” anisotropies for the strained Ni-film (cf. Table I), we obtain the
following values for the Néel parameters, $L = -580 \mu eV$ and $M = 115 \mu eV$.

As shown in Refs. [16,14], the anisotropy due to the step along (110) on the fct(001)
surface can be written in terms of the contributions from “step - corner” and “step - edge”
atoms (we use the bct-system and a fct (110) step is a bct (100) step):

$$E_{st} = -K_{se}V_x^2 - K_{sc}^1V_z^2 - K_{sc}^2V_xV_z$$

(6)

where, $K_{se}$ is a “step-edge” atom’s MAE, and $K_{sc}^1$ and $K_{sc}^2$ are a “step-corner” atom’s MAE.

Within the assumption of only nearest-neighbor interactions one can express $K_{se}, K_{sc}^1$
and $K_{sc}^2$ in terms of the Néel parameters $L$ and $M$:

$$K_{sc}^1 = 0.5L(3\beta_z^2 - 1)$$

$$K_{sc}^2 = -L\beta_x\beta_z$$

$$K_{se} = -0.5M$$

(7)

The values of $K_{se}, K_{sc}^1$ and $K_{sc}^2$ are then calculated for the Ni- step using Eq.(7) and are
shown in Table II together with values of the “volume” and “surface” anisotropies.
The symmetry induced uniaxial one-step MAE (per one atom in the xy-plane) is then given \([17]\) by:

\[
E = -D \star dK_vV_z^2 - 2 \star D \star K_sV_x^2 - K_{se}V_x^2 \\
\quad -K_{sc}^1 V_z^2 - K_{sc}^2 V_x^2
\]  

(8)

where \(D\) is the number of one-step atoms (along \(x\)). We consider only the “surface” step and neglect the “interface” step. As follows from Eq.(8), the direction of the magnetization in the xy-plane is determined by the sign of \(K_{se}\). In the case of a Ni stepped surface, the magnetization is directed along the step-edge (perpendicular to the \(x\)-axis) due to the negative sign of \(K_{se}\) (cf. Table [1]). This is in qualitative agreement with experimental data \([12]\) for thin Ni-films on stepped Cu(001).

Next, we transform Eq.(8) to a vicinal coordinate frame (\(U\)) using the relation between the old (\(V\)) and new (\(U\)) magnetization direction cosines in terms of the vicinal angle \(\alpha\). Keeping terms up to \(\alpha^2\), and with

\[
V_x = (1 - 0.5\alpha^2)U_x + \alpha U_z
\]

\[
V_z = (1 - 0.5\alpha^2)U_z - \alpha U_x
\]

(9)

we come to the following expression for the MAE in the xy-vicinal plane

\[
E^U_{xy} = (-D \star dK_v\alpha^2 - 2D \star K_s\alpha^2 - K_{se}(1 - \alpha^2) \\
\quad -K_{sc}^1\alpha^2 + K_{sc}^2\alpha) U_x^2
\]  

(10)

Using the relation between \(\alpha\) and \(D\) (\(\alpha \approx l_{op}/(a_{up}D) \equiv x/D\)) and neglecting \(1/D^2\)-order terms we transform Eq.(10) into:

\[
E^U_{xy} = (-d \times^2/D K_v - 2x^2/D K_s - K_{se} \\
\quad +x/DK_{sc}^2) U_x^2
\]  

(11)

We next apply Eq. (11), with the parameters determined within the framework of the modified Néel model (cf., Table [1]), to calculate the MAE at the (1,1,13) stepped surface.
Using the strain parameter $x = 0.674$ and step length $D = 6$, we obtain the following contributions to the MAE:

(1) The contribution due to the “volume” magnetoelastic anisotropy is $-6.322 \times d \, \mu eV$; it turns the magnetization perpendicular to the step and increases with an increase of the film thickness;

(2) The contribution due to the “surface” MAE is $68 \, \mu eV$; it turns the magnetization parallel to the step and does not depend on the film thickness;

(3) The contribution due to the “step-edge” MAE is $57.5 \, \mu eV$; it turns magnetization parallel to the step and does not depend on the film thickness;

(4) The contribution due to “step-corner” MAE is $+22 \, \mu eV$, and it turns the magnetization parallel to the step and does not depend on the film thickness;

Finally, for thicknesses below 24 ML, the contributions (2),(3) and (4) will keep the magnetization parallel to the step in the vicinal (xy)-plane and the critical thickness for the magnetization to turn perpendicular to the step edge is then estimated to be 24 ML.

In conclusion, Eq.(11) shows that there is a “volume” uniaxial magnetoelastic anisotropy contribution to the symmetry-induced magnetic anisotropy in the vicinal plane of the stepped surface, and it is caused by tetragonal strain in the magnetic film due to film/substrate lattice mismatch. For monoatomic steps on Ni-films, this magnetoelastic contribution leads to an additional spin-reorientation transition in which the magnetization turns from parallel to perpendicular to the step alignment with increase of the Ni-film thickness. Using the results of first-principles calculations for the strained Ni film and a Néel model-type analysis, we estimate the critical Ni-film thickness for this transition to be 24 ML for the (1,1,13) stepped surface. One has to take into account the difference between the surface anisotropy in Ni/Cu(001) films and in the calculated free-standing Ni-films: if we use the experimentally derived (at zero temperature) “surface” MAE $[1]$ (i.e. $-250 \, \mu eV$), we expect a critical thickness of 16 ML for this transition. This result shows the uncertainty and range of values possible at this level of theory.

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The expression Eq. (10) differs from Eq. (3) of Ref. [14] due to an additional uniaxial volume magnetoelastic term.
TABLES

TABLE I. Uniaxial MAE of tetragonal strained bulk Ni ($K_v$), “volume” ($K_v$) and “surface” ($K_s$) contributions to the MAE for strained Ni-films. All data are given in $\mu eV$.

|          | This work | Exp. $^a$ | FLAPW $^b$ | FLMT$^c$ |
|----------|-----------|-----------|------------|-----------|
| Bulk-Ni  | $K_v$     | 57        | 65         | 60        |
| Ni-film  | $K_v$     | 83.5      | 30 (60-70) |           |
|          | $K_s$     |           | -446.5     |           |

$^a$ Ref. [1] (the data obtained for 300 K, in parentheses, are the result of a linear extrapolation of the temperature dependent MAE to T=0 K)

$^b$ Ref. [3], $^c$ Ref. [4].

TABLE II. Values of $K_v$, $K_s$, $K_{se}$, $K_{sc}^1$ and $K_{sc}^2$ (in $\mu eV$) for the Ni-step.

| $K_v$  | $K_s$  | $K_{se}$ | $K_{sc}^1$ | $K_{sc}^2$ |
|--------|--------|----------|------------|------------|
| +83.5  | -446.5 | -57.5    | -124       | 195        |
Figures

Fig. 1 The MAE (in $\mu$ eV) for the strained Ni-films as a function of the film thickness d (in ML).
