Magnetization direction reorientation in ultrathin ferromagnetic films: Spin reorientation transition by H adsorption in the Valenta model

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The magnetization of ultrathin ferromagnetic films depends in a very sensitive way on anisotropy fields which favor its particular orientation. We consider the spin reorientation transition in the ferromagnetic thin films within the Valenta model investigating the influence of different anisotropy fields on their magnetization behavior. It is shown that for certain values of the parameters a temperature-driven, a thickness-induced, and a H-induced switching transition from an in-plane to an out-of-plane ordered state may be observed. The use of the Valenta model allows us to present all dependences in the layer resolved mode which leads to the kinetics of the spin reorientation.

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I. INTRODUCTION

The spontaneous magnetization in thin magnetic films is either in plane or perpendicular to the surface plane of atoms. The orientation of the film magnetization is driven by the magnetic anisotropy energy which is determined and, in consequence, can be modified by different contributions, namely, film thickness,[1] temperature,[2] composition,[3] stress,[4] adsorbate coverage,[5] the presence of current,[6] or small external magnetic field.[7] By changing one of these parameters the spontaneous magnetization can switch the direction and a spin reorientation transition (SRT) is observed. The transition can be understood as a result of competing forces that favor different directions of the magnetization such as, e.g., spin-orbit coupling, dipolar interaction, or an external magnetic field. The resulting surface anisotropy fields often favor a perpendicular orientation, while the dipole interaction favors an in-plane magnetization.

With increasing film thickness most frequently a shape anisotropy driven SRT from perpendicular to in plane is observed. An exceptional case is a so-called "reverse SRT," i.e., SRT from in plane to perpendicular. Because of the tetragonal distortion of the film, the most well-known example for this kind of behavior is Ni on Cu(100).[1] Recently, surprising new features for the system Ni/Cu(100) concerning the influence of hydrogen adsorption have been reported.[8] There is a H-induced SRT and it has been shown, by magnetic and crystallographic measurements complemented by gas pressure and surface coverage measurements, that the hydrogen-induced reverse SRT is reversible.[8] The reversibility of the SRT leads to an oscillatory sequence of in-plane and perpendicular magnetizations when the hydrogen coverage is made to oscillate as controlled by the ambient hydrogen pressure. Surprisingly, the orientation switch from in plane to perpendicular to the surface comes with a relaxation, i.e., reduction in the film tetragonal distortion rather than its amplification.

In the present paper we will first consider the magnetic thin films with surface anisotropy. The anisotropy field is assumed to be perpendicular to the film surface. In this case the Curie temperature is lowered when compared with that for films without anisotropy and the effective spontaneous magnetization can simulate the appearance of magnetically dead layers. Next, the spin reorientation phenomena will be analyzed using a Heisenberg model in film geometry with the usual exchange interaction anisotropy terms: the single-site anisotropy and the anisotropy field perpendicular to the film surfaces. For this Hamiltonian model, the H-induced SRT is discussed in detail. Both descriptions are considered within the Valenta model[9] which is based on two assumptions: a discretization of the sample geometry reflects the crystallographic lattice and the thermodynamics is modified for inhomogeneous media. A deep understanding of the SRT phenomena in the ultrathin structures is of a great importance in the context of future spintronic devices that enabled the switching of the individual spin components of the device while avoiding cross-talk at the nanoscale.

II. SPIN REORIENTATION TRANSITION IN VALENTA MODEL

A film can be treated as a sample cut in some crystallographic orientation with respect to the surface of the crystal with a given crystallographic structure characterized by the spectrum of the nearest neighboring atoms. In this case the atoms situated at the surfaces have their neighborhood different than the atoms inside a sample. This geometric situation corresponds then to the different environment in which the atoms at the surface and the atoms inside a sample are embedded. In a natural way, such a film can be divided into monoatomic layers parallel to the surface plane and each layer can be treated as two-dimensional thermodynamically homogenous subsystem[10] interacting with the neighboring subsystems via effective field. The above idea is the basis of the Valenta model concept.

The thermodynamic approach is, in general, based on the free-energy functional construction,

$$F = U - TS, \tag{1}$$

which can be obtained by means of the internal energy U and the entropy S calculations. The entropy is given in the standard form,

$$S = -N^2 \sum_{\nu=1}^{n} \sigma_\nu, \tag{2}$$

where

$$\sigma_\nu = k_B \left[\left(\frac{1}{2} + m_\nu \right) \ln \left(\frac{1}{2} + m_\nu \right) + \left(\frac{1}{2} - m_\nu \right) \ln \left(\frac{1}{2} - m_\nu \right) \right] \tag{3}$$

is the entropy per one lattice site while m_ν denotes the magnetic order parameter.

In order to determine the internal energy we have to introduce the Hamiltonian describing the considered system. In Secs. II A and II B we consider the Hamiltonians with different kinds of anisotropies which allow us to describe different phenomena observed experimentally.

The equilibrium values of the magnetic order parameters m_ν are obtained by minimizing free energy (1) with respect to these parameters. The variational equations are of the form

$$\frac{\partial F}{\partial m_\nu} = 0. \tag{4}$$

The numerical solution of Eq. (4) gives the temperature dependent magnetization m_ν together with its profiles across a film.

A. Model of surface anisotropy perpendicular to thin film

Let us consider a film containing n monoatomic layers with N^2 atoms in each plane. Magnetic properties of an atom can be described by the spin operator $S_{\nu j}$ localized at the lattice site νj, where ν labels monoatomic layers in the direction of the film thickness and j is the position vector in the plane of a layer. We denote the vector of the surface anisotropy field perpendicular to the film surfaces as κ and we assume z as a direction of easy magnetization axis which is determined by the induced average molecular-field direction situated in the plane of the film. The Hamiltonian of the above described system considered in Ising model can be written in the following form:

$$H = -\sum_{\langle \nu j \nu' j' \rangle} J_{\nu\nu'} S^z_{\nu j} S^z_{\nu' j'} - g\mu_B \kappa \sum_{\nu j} S^x_{\nu j} (\delta_{1\nu} - \delta_{n\nu}) - g\mu_B h \sum_{\nu j} S^z_{\nu j}, \tag{5}$$

where the first term of the Hamiltonian determines the exchange energy, the second term expresses the interactions of spins with the surface anisotropy field, and third one is responsible for the interactions between the spins and the external field. We would like to underline that the average field of surface anisotropy with respect to its inhomogeneity across a film is equal to zero even along the direction of its action assumed as axis x. This fact implies that the direction of the anisotropy induced by the exchange interactions in the sense of molecular field is oriented in the plane of a film. In the case of the appearance of perpendicular surface anisotropy the quantization direction will be rotated by an angle ϑ_ν dependent on the parameter κ.

In order to find the quantization axis for spins from each layer, we transform the spin components $S^x_{\nu j}$ and $S^z_{\nu j}$ to the system in which the average value $\langle S^{x'}_{\nu j} \rangle$ vanishes. The rotation of the coordinate system is described by the rotation matrix $\mathbf{M}$. Due to the symmetry, the rotation can be confined to the $z-x$ plane without loss of generality. This means that $y' = y$ and that the polar angle ϑ_ν fully characterizes the rotation,

$$\mathbf{M} = \begin{pmatrix} \cos\vartheta_\nu & 0 & -\sin\vartheta_\nu \\ 0 & 1 & 0 \\ \sin\vartheta_\nu & 0 & \cos\vartheta_\nu \end{pmatrix}. \tag{6}$$

The z' axis of a new coordinate system is set to be parallel to the magnetization direction. This gives

$$\langle S^{x'}_{\nu j} \rangle = \langle S^{y'}_{\nu j} \rangle = 0. \tag{7}$$

The magnetization in the primary system can now be read off from Eq. (6), namely,

$$\langle S^x_{\nu j} \rangle = \sin\vartheta_\nu \langle S^{z'}_{\nu j} \rangle,$$

$$\langle S^z_{\nu j} \rangle = \cos\vartheta_\nu \langle S^{z'}_{\nu j} \rangle. \tag{8}$$

$\langle S^x_{\nu j} \rangle$ is the magnetization component normal to the film plane, while $\langle S^z_{\nu j} \rangle$ denotes the component parallel to the film plane. Consequently, $\langle S^{z'}_{\nu j} \rangle$ is the total magnetization. Of course, the angle ϑ_ν is *a priori* unknown.

Taking into account the transformation described above, Hamiltonian (5) can be written in the form equivalent to the Hamiltonian of the effective-field theory, namely,

$$H_{\text{eff}} = \sum_{\nu j} (B^{z'}_\nu S^{z'}_{\nu j} + B^{x'}_\nu S^{x'}_{\nu j}), \tag{9}$$

where

$$B^{z'}_\nu = 2 \sum_{\nu' j' \in \nu j} J_{\nu\nu'} \cos\vartheta_\nu \cos\vartheta_{\nu'} \langle S^{z'}_{\nu' j'} \rangle + g\mu_B h \cos\vartheta_\nu + g\mu_B \kappa (\delta_{1\nu} - \delta_{n\nu}) \sin\vartheta_\nu, \tag{10}$$

$$B^{x'}_\nu = -2 \sum_{\nu' j' \in \nu j} J_{\nu\nu'} \sin\vartheta_\nu \cos\vartheta_{\nu'} \langle S^{z'}_{\nu' j'} \rangle - g\mu_B h \sin\vartheta_\nu + g\mu_B \kappa (\delta_{1\nu} - \delta_{n\nu}) \cos\vartheta_\nu. \tag{11}$$

The condition for $\langle S^{x'}_{\nu j} \rangle$ which has been applied to formulas (10) and (11) implies also the form of Hamiltonian (9) where the dependence on $S^{x'}_{\nu j}$ cannot appear. So, we have to set $B^{x'}_\nu = 0$ and for $h = 0$, we obtain the equation describing the angular distribution of spins, namely,

$$\tan\vartheta_\nu = \frac{g\mu_B \kappa (\delta_{1\nu} - \delta_{n\nu})}{2 \sum_{\nu' j' \in \nu j} J_{\nu\nu'} \langle S^z_{\nu' j'} \rangle}. \tag{12}$$

The internal energy is then expressed in the following form:

$$U = -2z_0\left(\sum_{\nu=2}^{n-1} J_{\nu\nu}m_\nu m_\nu + J_{\nu\nu}m_\nu m_\nu \cos\vartheta_\nu \cos\vartheta_\nu(\delta_{1\nu}+\delta_{n\nu})\right)$$
$$-z_1\left(\sum_{\nu=2}^{n-1} J_{\nu\nu\pm1}m_\nu m_{\nu\pm1} + J_{\nu\nu+1}m_\nu m_{\nu+1}\cos\vartheta_\nu\delta_{1\nu}\right.$$
$$\left.+J_{\nu\nu+1}m_\nu m_{\nu+1}\cos\vartheta_{\nu+1}\delta_{n-1\nu}\right)$$
$$-\kappa\sum_{\nu=1}^{n} m_\nu(\delta_{1\nu}-\delta_{n\nu})\sin\vartheta_\nu, \tag{13}$$

where $m_\nu=\langle S_\nu^{z'}\rangle$ and z_0, z_1 are the numbers of nearest neighbors of a given atom in the same monoatomic layer and in the neighboring layers, respectively.

B. Magnetic anisotropies in thin Heisenberg films

Let us consider a model consisting of n monoatomic layers described by the Hamiltonian,

$$H = -\sum_{\nu j\nu' j'} J_{\nu j\nu' j'}\vec{S}_{\nu j}\vec{S}_{\nu' j'} - \sum_{\nu j} K_{\nu j}S_{\nu j}^x S_{\nu j}^x - \sum_{\nu j}\kappa S_{\nu j}^x, \tag{14}$$

where the first term of the Hamiltonian describes the Heisenberg type of the exchange energy, x is the direction perpendicular to the surface plane, $K_{\nu j}$ is the single-site anisotropy, which can be denoted as K_1 for the surface anisotropy and K_2 for the volume anisotropy constant, and κ is a vector of the anisotropy field perpendicular to the film surfaces. The quantization direction is rotated by the angle ϑ_ν dependent on the parameter κ. The spin components $S_{\nu j}^x$, $S_{\nu j}^y$, and $S_{\nu j}^z$ can be transformed to the new system using Eq. (8) in which the average value of $\langle S_{\nu j}^{x'}\rangle$ vanishes.

Following the procedure described in Sec. II A, we can rewrite Hamiltonian (14) in the coordinate system (x',z') as the effective Hamiltonian expressed by Eq. (9) where $B_\nu^{x'}$ and $B_\nu^{z'}$ are now in the form

$$B_\nu^{x'} = -2\sum_{\nu' j'\in\nu j} J_{\nu\nu'}\sin(\vartheta_{\nu'}-\vartheta_\nu)\langle S_{\nu' j'}^{z'}\rangle$$
$$-2K_{\nu j}\cos\vartheta_\nu\sin\vartheta_\nu\langle S_{\nu' j'}^{z'}\rangle - \kappa\cos\vartheta_\nu, \tag{15}$$

$$B_\nu^{z'} = -2\sum_{\nu' j'\in\nu j} J_{\nu\nu'}\cos(\vartheta_\nu-\vartheta_{\nu'})\langle S_{\nu' j'}^{z'}\rangle - 2K_{\nu j}\sin^2\vartheta_\nu\langle S_{\nu j}^{z'}\rangle$$
$$-\kappa\sin\vartheta_\nu. \tag{16}$$

If we set $B_\nu^{x'}=0$, we obtain the equation describing the angular distribution of spins, namely,

$$\tan\vartheta_\nu = \frac{\sum_{\nu' j'\in\nu j} J_{\nu\nu'}\sin\vartheta_{\nu'}\langle S_{\nu' j'}^{z'}\rangle + K_{\nu j}\sin\vartheta_\nu + \frac{1}{2}\kappa}{\sum_{\nu' j'\in\nu j} J_{\nu\nu'}\cos\vartheta_{\nu'}\langle S_{\nu' j'}^{z'}\rangle}. \tag{17}$$

We can calculate free energy (1) of the system using entropy [Eqs. (2) and (3)] where $m_\nu=\langle S_\nu^{z'}\rangle$ and the internal energy in the form

$$U = -2z_0\sum_\nu J_{\nu\nu}m_\nu m_\nu - z_1\sum_{\nu'\in\nu} J_{\nu\nu'}m_\nu m_{\nu'}\cos(\vartheta_\nu-\vartheta_{\nu'})$$
$$-2\sum_\nu K_\nu m_\nu m_\nu\sin^2\vartheta_\nu - \sum_\nu \kappa m_\nu\sin\vartheta_\nu, \tag{18}$$

where z_0 and z_1 are the numbers of nearest neighbors of a given atom in the same monoatomic layer and in the neighboring layers, respectively.

III. NUMERICAL RESULTS AND DISCUSSION

The minimization of free energy (1) of the system with respect to m_ν gives the temperature dependent magnetization of each layer forming thin film together with the reorientation temperature T_R and the Curie temperature T_C. In Secs. III A and III B, the numerical results of the minimization procedure applied in the case of the systems described by Hamiltonians (5) and (14) are presented and discussed.

A. Magnetically dead layers

Let us consider the thin film of Ni(100) for which $z_0=4$ and $z_1=4$. As it was mentioned in Sec. II A the quantization direction can be rotated by the angle ϑ_ν dependent on the parameter κ. The temperature behavior of the spontaneous magnetization which can switch the direction from in plane to out of plane in the surface layers is shown in Fig. 1. Figure 1(a) presents the temperature dependence of the angle ϑ_ν of the magnetization vector of surface layers for different values of the surface anisotropy field κ for system consisting of five monoatomic layers. It is clearly seen that the larger the value of the parameter κ the smaller the value of the reorientation temperature T_R. For $\kappa=0.1J_{\mathrm{Ni}}$ we have got $T_R=419$ K, for $\kappa=0.5J_{\mathrm{Ni}}$ $T_R=384$ K, while for $\kappa=1J_{\mathrm{Ni}}$ $T_R=371$ K. Starting from $\kappa=1J_{\mathrm{Ni}}$ further increasing of this parameter does not cause a significant decrease in the transition temperature T_R values. It is worth while to notice that the transition temperature T_R is equal to the Curie temperature of the film of given thickness [the vertical line between Figs. 1(a) and 1(b) indicates such behavior in the case of $\kappa=0.5J_{\mathrm{Ni}}$]. In Fig. 1(b) we show the temperature dependence of two magnetic components of the surface layers together with the magnetization curves inside the sample. It is clearly seen that even at low temperatures the directions of the surface magnetization vectors are not entirely in the plane. Above the phase-transition point they are changing their direction to the perpendicular one and one can observe nonzero surface magnetization while the sample inside is in the para-

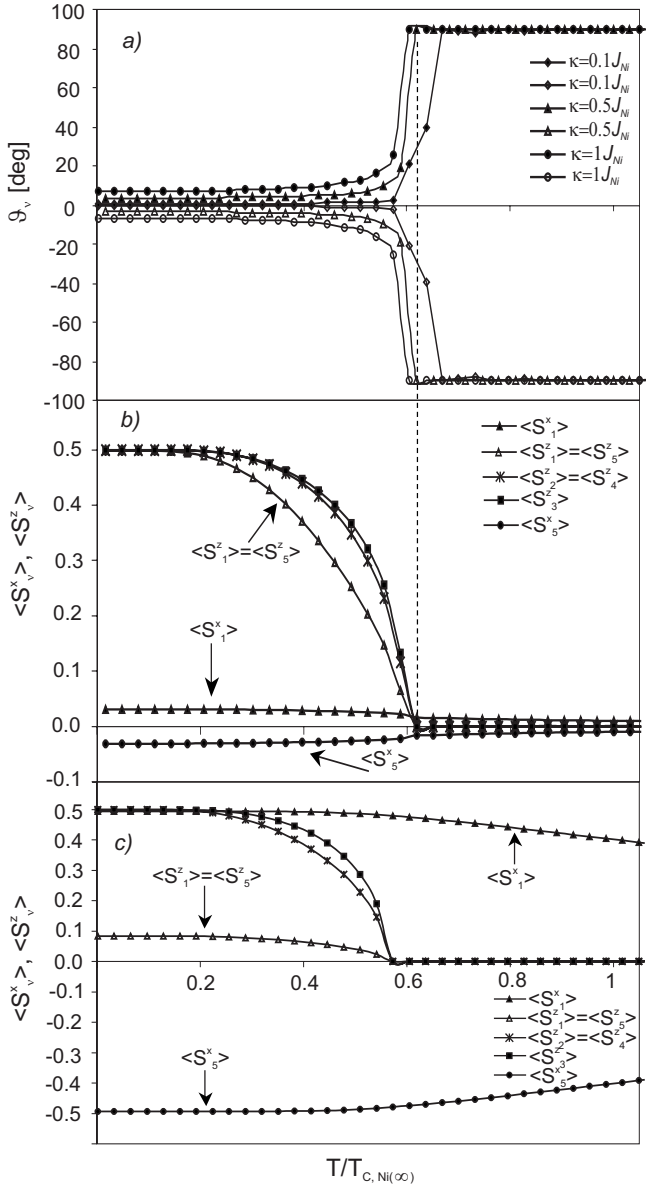

FIG. 1. The temperature dependence of (a) the angle ϑ_ν of the magnetization direction for different values of the surface anisotropy field κ and the average value of two spin components $\langle S_\nu^x \rangle$ and $\langle S_\nu^z \rangle$ (b) for $\kappa=0.5J_{\mathrm{Ni}}$ and (c) for $\kappa=27.5J_{\mathrm{Ni}}$. All dependences were obtained for the system of 5 ML thickness.

magnetic state. As the surface magnetization vectors have the opposite directions, the averaged field is equal to zero even along the direction of its action. In order to obtain the lower values of surface spontaneous magnetization at $T=0$ K then their saturation values, which can be interpreted as a phenomenon of the magnetically dead layers, we have used the anisotropy parameter $\kappa=27.5J_{\mathrm{Ni}}$. The results are presented in Fig. 1(c), where one can see a very low surface magnetization along the easy axis and almost saturation along the perpendicular direction, while the sample inside is ferromagnetic until the Curie temperature.

B. Ni/Cu(100) system

A reversible switching of the easy axis of magnetization for Ni on Cu(100) from in plane to out of plane by changing the partial pressure of hydrogen in the gas phase around the sample, allowing even the oscillations of the magnetization direction, is reported in paper.[8] The hydrogen-induced spin reorientation transition is accompanied by changes in the interlayer spacing between the topmost Ni layer and the rest of the sample upon hydrogen adsorption. Such distortions are directly connected with the changes in magnetic interactions represented by the exchange integrals appearing in the model Hamiltonian used in Sec. II B. Below we present the theoretical analysis of this experimental observations, starting, however, with the discussion of thickness-induced spin reorientation because Ni/Cu(100) system is the most well-known example for the so-called reverse SRT, i.e., SRT from in plane to perpendicular direction and it will complement in the natural way the hydrogen-induced behavior.

1. Thickness-induced spin reorientation

As the SRT is directly connected with the anisotropy parameters, in Fig. 2 we present the behavior of the reorientation temperature T_R normalized to $T_{C,\mathrm{Ni}(\infty)}$ with respect to the anisotropy field κ [Fig. 2(a)], to the surface anisotropy K_1 [Fig. 2(b)], and to the volume anisotropy constant K_2 [Fig. 2(c)] for different thicknesses d of the Ni film in the Ni/Cu(100) system. In Fig. 2(a) we can see that the reorientation temperature T_R increases with the thickness of the Ni film and we can distinguish two regions: first (below T_R) where the magnetization is almost in plane and the second one (above T_R) where the magnetization is directed perpendicularly to the surface plane. Moreover, one can notice that for a given film thickness reorientation temperature T_R decreases with the increase in the parameter κ value. Calculations were performed for $K_1=K_2=0.05J_{\mathrm{Ni}}$. Figure 2(b) exhibits that the reorientation temperature T_R behaves in the same way with the film thickness as in the previous case but in contrast with when the surface anisotropy K_1 increases the T_R also increases for a given thickness of the Ni film. Calculations were performed for $K_2=0.05J_{\mathrm{Ni}}$ and $\kappa=0.02J_{\mathrm{Ni}}$. The dependence of the reorientation temperature T_R on the volume anisotropy K_2 obtained for $K_1=0.05J_{\mathrm{Ni}}$ and $\kappa=0.02J_{\mathrm{Ni}}$ is presented in Fig. 2(c). It seems to be the most interesting behavior because for K_2 values from the interval $(0.002-0.01)J_{\mathrm{Ni}}$ the reorientation temperature T_R decreases with the thickness while starting from $K_2=0.025J_{\mathrm{Ni}}$ this temperature starts increasing with the thickness.

In Fig. 3, we present the thickness dependence of the reorientation temperature T_R normalized to $T_{C,\mathrm{Ni}(\infty)}$ for different values of the anisotropy fields: (a) perpendicular to the film surfaces κ, (b) the surface anisotropy K_1, and (c) the volume anisotropy constant K_2. Figure 3(a) ($K_1=K_2=0.05J_{\mathrm{Ni}}$) shows that T_R increases with the thickness but decreases if the value of parameter κ increases. The first curve from the top represents the case when the Curie temperatures for a given film thickness are equal to their SRT temperatures. Quite similar behavior of T_R as a function of the anisotropy constants K_1 and K_2 can be seen in Figs. 3(b) and 3(c). The reorientation temperature T_R increases if K_1 or K_2 increases for a given Ni film thickness. Additionally, in Fig. 3(b) ($K_2=0.002J_{\mathrm{Ni}}$ and $\kappa=0.02J_{\mathrm{Ni}}$) we can distinguish a set of characteristics $(K_1;d)$ for which the reorientation tem-

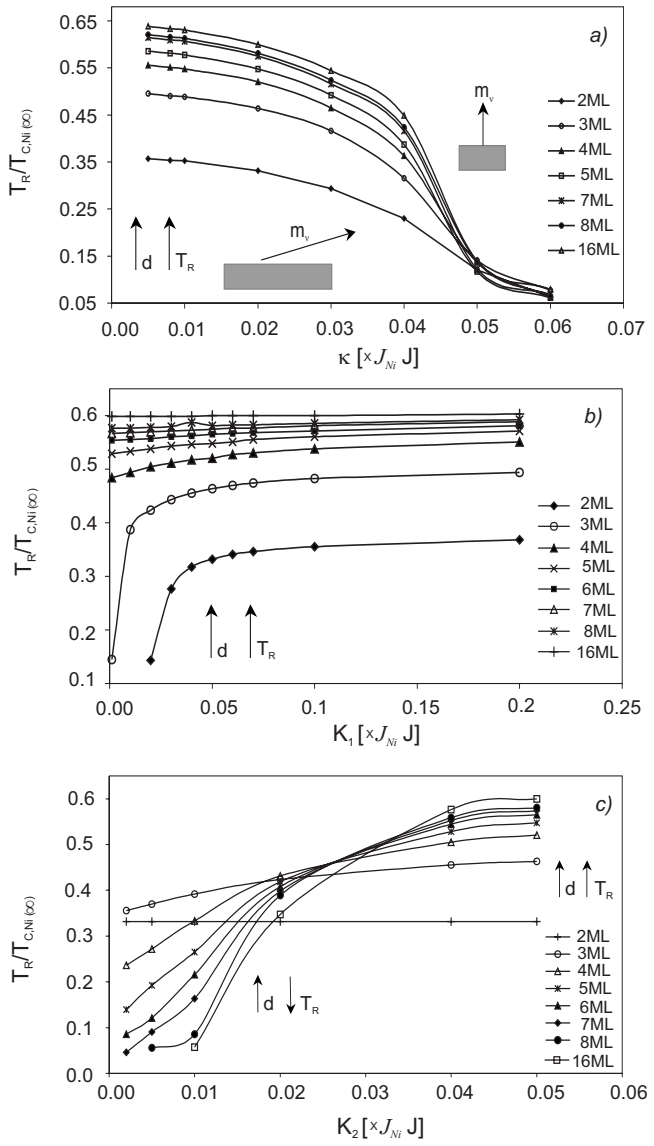

FIG. 2. The reorientation temperature T_R normalized to $T_{C,\mathrm{Ni}(\infty)}$ as a function of (a) the anisotropy field perpendicular to the film surfaces κ, (b) the surface anisotropy K_1, and (c) the volume anisotropy constant K_2 for different Ni film thicknesses.

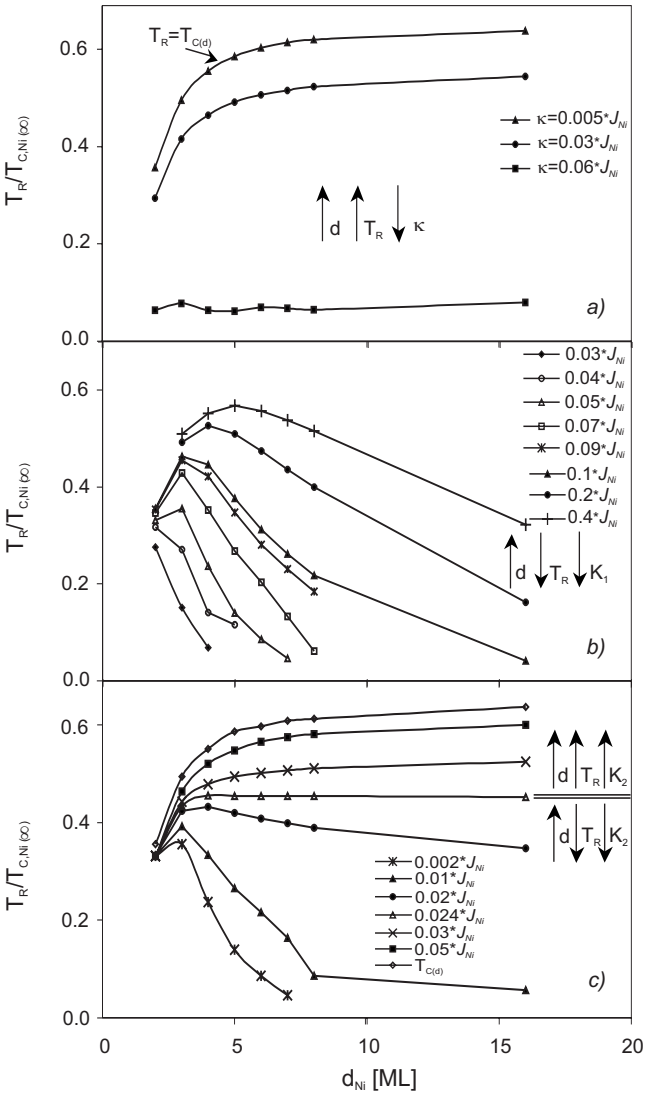

FIG. 3. The thickness dependence of reorientation temperature T_R normalized to $T_{C,\mathrm{Ni}(\infty)}$ for different values of (a) the anisotropy field perpendicular to the film surfaces κ, (b) the surface anisotropy K_1, and (c) the volume anisotropy constant K_2.

perature T_R increases and another one for which this temperature decreases. As for parameter $K_2=0.024J_{\mathrm{Ni}}$ we could find its critical value K_2 when the reorientation temperature does not depend on the Ni film thickness. For higher values of K_2 temperature T_R increases, while for lower values it decreases with the thickness [see Fig. 3(c); $K_1=0.05J_{\mathrm{Ni}}$ and $\kappa=0.02J_{\mathrm{Ni}}$].

Furthermore, the direction of the magnetization vector is a very sensitive function of temperature. In Figs. 4(a) and 4(b) we present the thickness dependence of the angle ϑ_ν of the magnetization vector in different temperatures for freestanding Ni(100) film and for Ni film in the Ni/Cu(100) system ($K_1=0.1J_{\mathrm{Ni}}$, $K_2=0.002J_{\mathrm{Ni}}$, and $\kappa=0.02J_{\mathrm{Ni}}$), respectively. It is clearly seen that the role of Cu substrate is crucial for the direction of the magnetization vector. The rotation of the magnetization by the angle of 90° appears for the Ni film of 5 ML thickness at $T=300$ K, while its simple deposition on Cu substrate causes such rotation at the same temperature in the thinner Ni films. The reorientation temperature for 5 ML Ni film on Cu(100) is lower than that for the freestanding 5 ML Ni film (Fig. 4). Moreover, Fig. 4 shows how the magnetization vector rotates in a given temperature when the film thickness increases. Particularly interesting behavior can be noticed for 2 ML Ni films when the sudden jump of the angle ϑ_ν value is observed for certain temperatures so that the 2 ML thickness can be considered as the critical one in these temperatures.

2. H-induced spin reorientation

A very interesting situation appears when we consider a hydrogen adsorption on the top of Ni/Cu(100) system. For the hydrogen covered 8 ML Ni/Cu system the low-energy electron-diffraction (LEED) measurements and their quantitative analysis[8] indicate the layer relaxation induced in the Ni

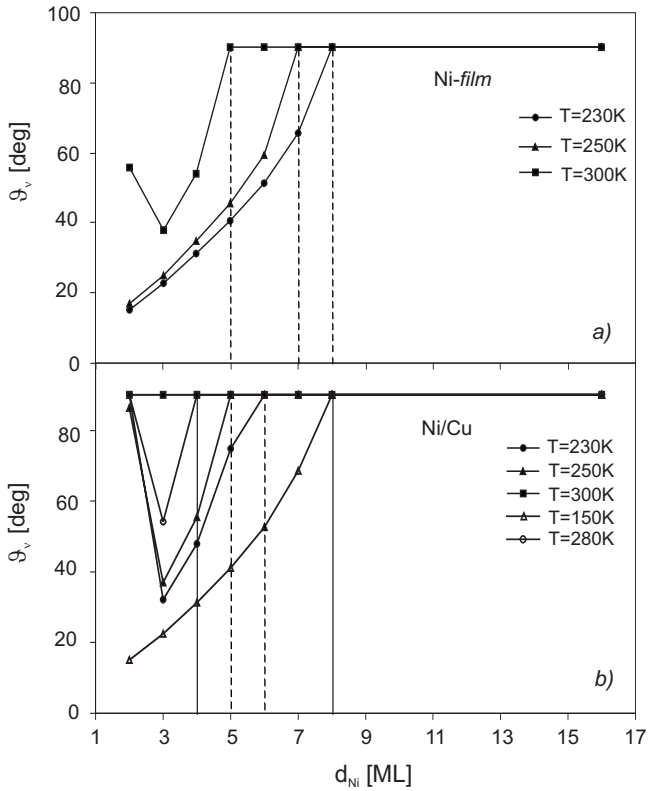

FIG. 4. The thickness dependence of angle ϑ_ν of the magnetization direction obtained in different temperatures for two systems: (a) freestanding Ni film and (b) Ni/Cu. The vertical lines indicate the thickness of the film in which the magnetization is directed perpendicularly to the surface.

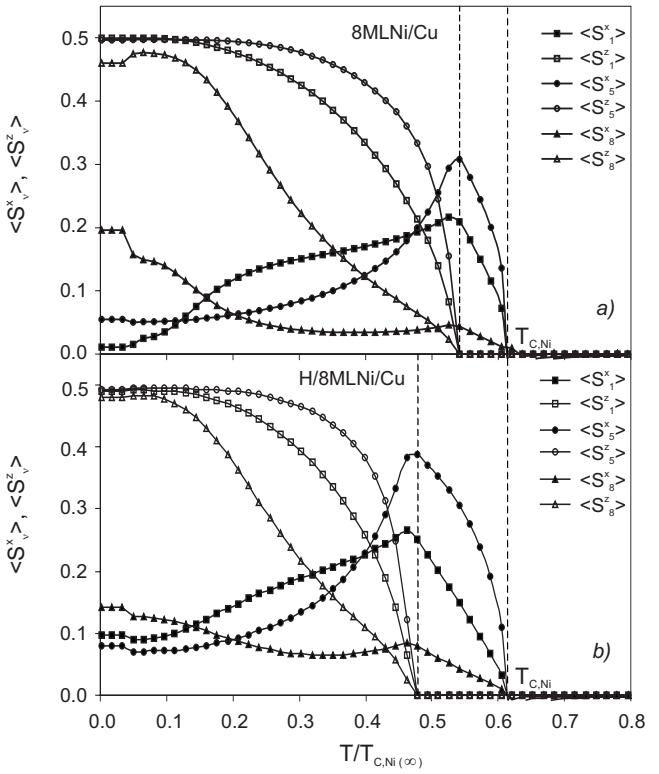

FIG. 5. The average value of two spin components $\langle S_\nu^x \rangle$ and $\langle S_\nu^z \rangle$ as a function of temperature normalized to $T_{C,\mathrm{Ni}(\infty)}$ for (a) 8 ML Ni/Cu system and (b) H/8 ML Ni/Cu system. The Curie temperature $T_{C,\mathrm{Ni}}$ for both systems is equal to 391 K. The vertical lines indicate the reorientation temperatures T_R=341 K and T_R=301 K for the system without and with hydrogen, respectively.

film. The most important feature is that the topmost layer spacing d_{12} is relaxed from 1.675 to 1.770 Å. This is remarkable result. The SRT of the clean Ni film is driven by the tetragonal distortion and one could speculate that the H-induced compression of the layer spacing is responsible for the H-induced SRT. Instead, the H-induced relaxation reduces the tetragonal surface distortion, but the average tetragonal distortion is only little modified[8] so the SRT driven by H-induced enhanced magnetoelastic anisotropy is excluded. The *ab initio* calculations[11,12] indicate that an outword relaxation of the topmost Ni-layer spacing is caused by the strong hybridization between H and Ni leading to the magnetic-moment reduction in the surface Ni layer and in consequence to a reduction in the magnetic surface anisotropy. Thus, the existence of the hydrogen in the H/Ni/Cu(100) system is included in our calculations first by means of proper value of the exchange integral $J_{12} \neq J_{\mathrm{Ni}}$ which describes the interactions between the surface monoatomic layer and the second layer of the system and second by the change in magnetic surface anisotropy parameter which follows in a natural way due to assumption that $K_1 = 0.05 J_{12}$.

In Figs. 5(a) and 5(b) the temperature dependences of the average values of two spin components $\langle S_\nu^x \rangle$ and $\langle S_\nu^z \rangle$ for 8 ML Ni/Cu and H/8 ML Ni/Cu systems are presented, respectively. When the magnetization component $\langle S_\nu^z \rangle$ (open marks) tends to zero the component $\langle S_\nu^x \rangle$ (full marks) achieves the largest value at the reorientation point in both cases. However, the presence of the hydrogen causes the lowering of the reorientation temperature in comparison to its value in the 8 ML Ni/Cu system while the Curie temperature is almost unchanged. It is worth while to notice that in both cases the direction of magnetization is not completely in plane even in the low temperatures exhibiting a small value of x component. The interesting point is that the hydrogen suppresses x component in the interface layers while they are clearly enhanced in the middle and surface layers.

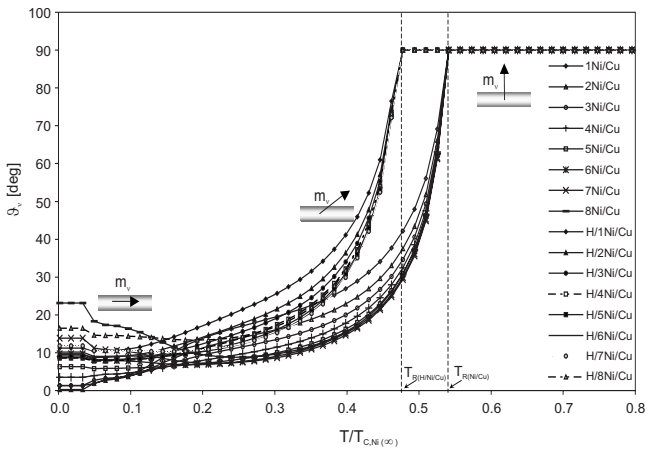

FIG. 6. Kinetics of the spin reorientation in two systems: 8 ML Ni/Cu and H/8 ML Ni/Cu.

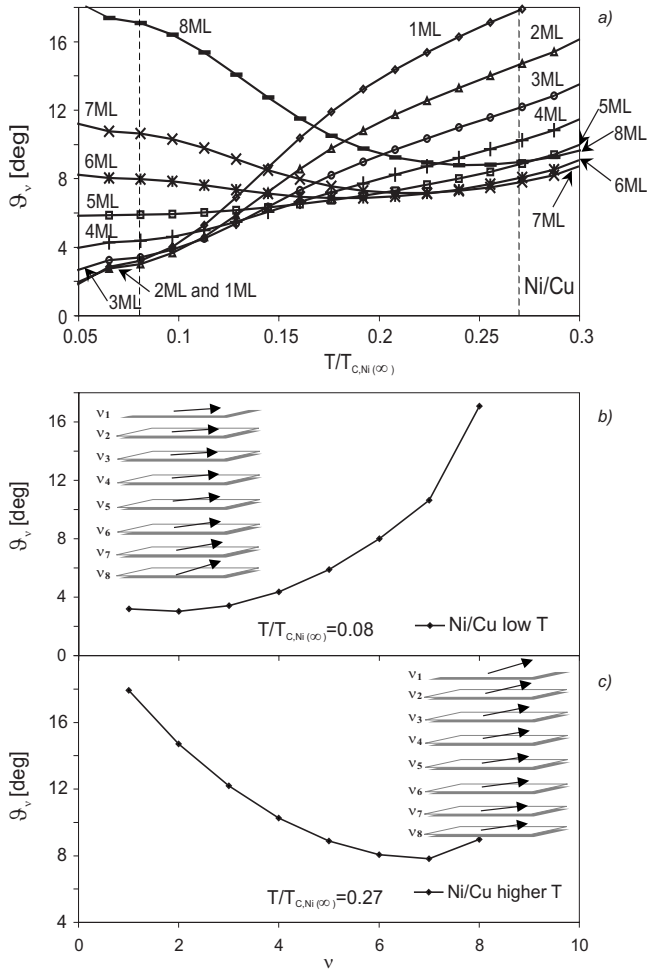

FIG. 7. The behavior of the layer resolved magnetization direction in the low-temperature region for 8 ML Ni/Cu system: (a) the temperature dependence of the angle ϑ_ν of the magnetization direction for each monoatomic layer and the angular profile of magnetization direction across the film thickness obtained (b) for $T_{C,\mathrm{Ni}(\infty)}=0.065$ and (c) for $T_{C,\mathrm{Ni}(\infty)}=0.29$, respectively.

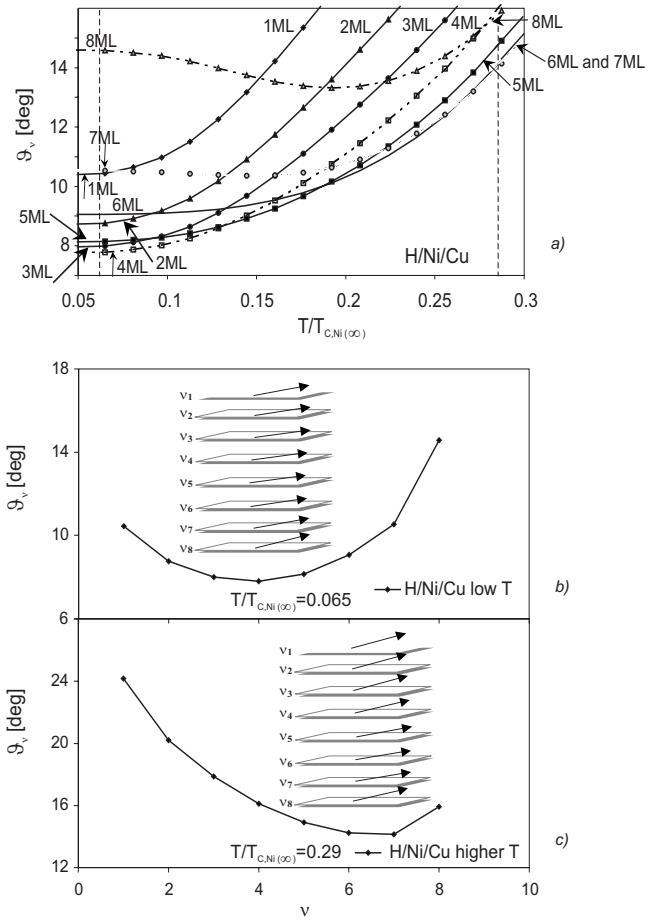

FIG. 8. The behavior of the layer resolved magnetization direction in the low-temperature region for H/8 ML Ni/Cu system: (a) the temperature dependence of the angle ϑ_ν of the magnetization direction for each monoatomic layer and the angular profile of magnetization direction across the film thickness obtained (b) for $T_{C,\mathrm{Ni}(\infty)}=0.065$ and (c) for $T_{C,\mathrm{Ni}(\infty)}=0.29$, respectively.

The time dependence of the SRT in response to a change in hydrogen partial pressure is reported in paper[8] as determined by the dynamics of the H coverage on the Ni surface. At the adsorption temperature of 322 K, the H coverage causes the relaxation of Ni layer mentioned above and the switch of easy axis of magnetization from in plane to out of plane is observed.[8] Our calculations indicate slightly different transition temperature. However, the calculated shift of the transition temperature from $T_R=341$ K for 8 ML Ni/Cu system to $T_R=301$ K for H/8 ML Ni/Cu system determines the interval which includes this experimental value. A particular interest of H/8 ML Ni/Cu system is connected with the fact that at this temperature interval the desorption rate for hydrogen is sizable and it seems that H is an ideal adsorbate for reversible SRT. The switching of the easy axis of magnetization from in plane to out of plane and back due to the hydrogen partial pressure has been observed experimentally.[8]

The Valenta model allows us to present the dependences described above in the layer resolved mode which leads to the kinetics of the spin reorientation. We show it in Fig. 6 for both systems. One can easily follow how the angle ϑ_ν of the magnetization vector changes from one monoatomic plane to another with increasing temperature. Unfortunately it is not easy because of many changes in the low-temperature region. In order to analyze the angular distribution of magnetization in this region, we provide, in Figs. 7 and 8, the kinetic process from the most interesting temperature interval in which the most remarkable changes in the magnetization direction are observed, for 8 ML Ni/Cu and H/8 ML Ni/Cu systems, respectively. We can divide this interval in two parts. In the right part (higher temperatures) the sequences of angles are 1, 2, 3, 4, 5, 8, 6, 7 and 1, 2, 3, 4, 8, 5, 6, 7 for Ni/Cu(100) and H/Ni/Cu(100) systems, respectively. In the left part (lower temperatures) the sequences of angles are 8, 7, 6, 5, 4, 3, 2, 1 and 8, 7, 1, 6, 2, 5, 3, 4 for Ni/Cu(100) and H/Ni/Cu(100) systems, respectively. In all cases sequence means from bigger angles toward the smaller ones. For more precise viewing we present [Figs. 7(b), 7(c), 8(b), and 8(c)] these angular distributions as layer dependent profiles of ϑ_ν in a given temperatures marked in both Figs. 7 and 8 by the dashed lines. In between these temperatures we

can observe the most impressive changes in the magnetization direction in each particular monoatomic layer. In the higher-temperature region [Figs. 7(a) and 8(a)] the reorientation from in-plane to out-of-plane direction is going smoothly starting at the surfaces of both systems. It is evident that the hydrogen adsorption has influence not only on the reorientation temperature but also, in quite spectacular way, on the distribution of magnetization direction between the monoatomic layers.

IV. FINAL REMARKS

Theoretical description of the ferromagnetic thin-film systems based on the Valenta model seems to be a very good approach and useful method to calculate some magnetic properties of different multilayer systems.[13] Originally, the Valenta model was constructed for the homogeneous angular distribution of magnetic moments oriented in agreement with the quantization axis. In the present considerations we have extended the Valenta model to the discussion concerning the angular distribution of magnetization in terms of the variational procedure derived in the discrete space. We have obtained then not only two orientations of homogeneous distribution but also their inhomogeneous distribution among the sublattices.

The evaluation of topical phenomena predicted for new nanotechnology shows that their description can be related to the fundamental model for thin film in two aspects, geometry and thermodynamics, in classical formulation. Various modifications of the model make it more adequate for actual thin films whose technology is constantly developing and today it approaches better the ideal picture of theoretical films. The actual thin films, also other nanostructures, can be easily described within the model slightly extended in the expected way but still based on the fundamental predictions originally used for its construction.[9] In the methodological context, the model of magnetic thin films introduced by Valenta seems to be one of the most important methods with respect to the thermodynamics of inhomogeneous systems formulated in the Néel representation. The present example of its applicability confirms that the methodological background is sufficiently strong to assume that the Valenta model construction is not only a weak approximation but it is also able to make predictions for the methodological formulation of thermodynamics of thin films or, even more generally, of inhomogeneous small systems. The method can be considered as a reference theory with respect to phenomena of fine structure and their interpretation. It is convenient and successful for a characterization of phase transitions, for analysis of boundary conditions, and, in particular, for the multilayered structure description. The important advantage of the present approach among others (Monte Carlo simulations, Green's-function techniques, etc.) is its conceptual and mathematical simplicity leading to the correct results and their quantitative agreement with the experimental data.

Our results show that we can observe the spin reorientation transition phenomenon as well as we can easily obtain proper Curie temperature of the Ni film considering not only different thicknesses of the film but also the Ni film with the same thickness but embedded in different environment [Ni/Cu(100), H/Ni/Cu (100)]. The most interesting result is connected with a complicated behavior of the magnetization vector in the sample with the hydrogen-induced layer relaxation when the distorted interlayer exchange interaction is included, and this drives the adsorbate-induced SRT. Very likely these features bear the potential to construct a hydrogen-sensitive layer resolved magnetic switch for spintronic devices.

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