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2008 J. Phys.: Condens. Matter 20 095217

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Ferromagnetic nanostructures in the generalized Valenta model

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Received 13 October 2007, in final form 18 December 2007

Published 14 February 2008

Online at stacks.iop.org/JPhysCM/20/095217

Abstract

Ferromagnetic nanostructures are an area of great interest for modern physics. A comparison of experimental data and theoretical results shows that the use of the standard molecular field approximation is insufficient for the description of the nanostructure properties. Therefore in the present paper we use the reaction field approach in order to test the Valenta model generalized in this way. The agreement between experiment and theory is then excellent.

1. Introduction

Recently, nanoparticles structures have been intensively discussed in connection with physical phenomena considered at the nanoscale level [1, 2]. In particular, the results obtained for the spin wave resonance in nanoparticles seem to be quite sensitive to the particle geometry and to the surface as well as edge anisotropy. Similarly, we can expect the magnetization to depend on the anisotropy and the particle size.

A comparison of experimental results and theory gives evidence that the standard mean field approach (MFA) is insufficient to explain the properties of ferromagnetic nanostructured samples [3], which are currently of great interest to investigators.

However, it is widely known that the Valenta model [4] used for magnetic thin films leads to their proper description [5] despite the fact that the results of the model are usually considered to be equivalent to the MFA treatment.

Thus, we can see that the experimental data exclude the use of the standard Valenta model for nanostructures, but agreement between the experimental and theoretical results can be obtained when the Valenta model is improved by introducing a modified effective field, e.g. the reaction field approach (RFA) which allows us to apply the model to a description of nanostructured two-dimensional magnetic layers.

Of course, another model can be applied; e.g. the theoretical model based on the Green function method within the random phase approximation [6] has been used to compare experimental data with theoretical expectations [3]. However, the modified version of the Valenta model seems to be methodologically more suitable, taking into account the structural similarity of films and nanoparticles.

Therefore in the present paper we calculate the Curie temperature and magnetization behaviour in terms of the Valenta model [4] generalized by the RFA [7] for different multilayer systems [3]. We then discuss the model in a general context, treating the relationship to the experimental situation [3] as an indication of the improved modelling approach.

Moreover, the example discussed here is an illustration of the thermodynamic behaviour of the system, which can be considered in connection with its general behaviour. The properties of inhomogeneous systems are described in the same way in the Valenta model and in the Green function technique model. The improvement can be considered at different levels of accuracy but is still at the same level as the thermodynamic interpretation. In this context our next goal is to show a comparison between these two theoretical approaches and the convergence of their results. In the light of the above considerations it seems to us that the problem is of a general nature for any thermodynamically inhomogeneous system, not only of magnons but of phonons as well as for objects described by an electronic structure.

2. Experimental data for fcc(100) multilayer systems

Ferromagnetic multilayers separated by a nonmagnetic spacer have shown rich magnetic features, which makes them interesting systems for investigation. In particular, the behaviour of the Curie temperature, one of the most important characteristics of ferromagnets, is worthy of study. Our theoretical considerations and calculations focus particularly on the experiment performed by Scherz and co-workers [3]. They investigated trilayers with two different ferromagnetic films, e.g. Ni film and Co film, having different ordering

temperatures and separated by Cu spacer film. Such a system is a very good basis for illustrating how the Curie temperature is influenced by the thickness of magnetic layers as well as by the thickness of the spacer.

The experimental results show that the Curie temperature of nickel, $T_{C,Ni}$, is lower for Ni embedded in Cu/Ni/Cu(100) system than in the case when Co layers are deposited on Cu, creating a trilayer Co/Cu/Ni/Cu(100) system. In references [3, 5] the authors suggest that the shift of the Curie temperature of nickel ($\Delta T_{C,Ni}$) due to Co deposition is caused by the interlayer exchange coupling through the Cu spacer and it depends on the spacer thickness. The expression for the RKKY coupling between two ferromagnetic layers separated by a nonmagnetic spacer was derived in [8]. The coupling of Ni and Co films was found to be ferromagnetic or antiferromagnetic depending on the spacer thickness. The shift of the Ni ordering temperature as a function of the Cu spacer thickness exhibits oscillations due to the oscillatory character of the coupling. In paper [3] the authors, in order to clarify the notation, labelled the Curie temperature of a Ni film as $T_{C,Ni}$ and the shifted value due to Co deposition as $T_{C,Ni}^*$, to indicate that in a strict thermodynamic sense the latter may not refer to a real phase transition. Nevertheless, it makes sense to call $T_{C,Ni}^*$ a quasi-critical temperature, as it is usually treated in the literature [3], although above this temperature the Ni magnetization does not vanish but is radically decreased (about 100 times) and Ni films can be considered to be in a paramagnetic state. In the present paper we keep this convention as it is practical for making comparisons between the results for different multilayer systems.

Other interesting data have been obtained for Cu/Ni/Cu(100) and Ni/Cu(100) systems [9]. It is then clearly visible that in the case of an uncapped Ni film its Curie temperature is much higher than in the case of Ni capped by Cu.

3. Reaction field approach to the Valenta model

A number of effective field theories which are an improved version of the MFA can be easily found in the literature [10]. The reaction field approximation (correlated effective field approximation) introduced by Onsager [7] and developed by Gusmao and Scherer [11] seems to be one of the most convenient methods for the description of magnetic properties. It consists in the extension of the molecular field appearing in the usual MFA treatment by adding the reaction field of the system (RFA) with respect to the response of the neighbourhood of a given spin to the mutual interaction.

3.1. The background of the Valenta model and its validity

The theory of magnetic thin films known in the literature as the Valenta model [4] consists in two assumptions: (i) about the sample geometry whose discretization reflects the crystallographic lattice and (ii) about the thermodynamics modified for inhomogeneous media using a physical interpretation in terms of Néel sublattices [12]. It is worthwhile noticing that the excellent idea of Néel can be also considered in the context of the Hill approach [13], which has recently been used to describe the properties of nanoparticles [14].

Originally introduced for magnetic films, the Valenta model is now generalized for different systems, taking their inhomogeneity into account. It is extended for diluted alloys [15], multilayers [16] and wires as well as rectangle boxes [17] which can represent the nanoparticle structure. Of course, it is applicable not only for magnetic systems but also for different kinds of objects, e.g. phonons in connection with melting studies [18], and for all cases of a general character in which thermodynamically inhomogeneous samples are involved.

It is important to mention that the background of the Valenta model consists in the division of a sample into homogeneous subsystems. From the quantum mechanics point of view this division implies [4] that the total wavefunction of the electrons of a system practically differs very little from the wavefunction of the state in which the components of magnetic moments in the homogeneous subsystems have well defined values. In these conditions, however, the model corresponds to the situation when the Hamiltonians of the subsystems do not commute with the Hamiltonian of the system. Although the commutative rules are only approached, they are satisfied in a simple case when the interactions between the sublattices are determined by means of the effective fields. Thus, at high temperature, the quantum mechanical construction influences the thermodynamic interpretation. The Hamiltonian of each Néel sublattice corresponds to the integral of motion for each subsystem. This means that the total wavefunction of the state is the product of the wavefunctions with respect to an individual sublattice. From the thermodynamic point of view this fact is equivalent to the factorization of the partition functions. The total partition function is factorized with respect to the partition functions of individual subsystems. The statistical operator of a system is then a product of the statistical operators of subsystems due to the additive character of the effective Hamiltonians. As a consequence, the entropy is a sum of terms describing homogeneous contributions of subsystem entropies that are independent of each other in the calculations.

At low temperatures the constructions of a Heisenberg type quantum mechanical theory leads to a solution which can be related to spin wave propagations or, in the magnon representation, to quasi free particles when they are embedded in a heat bath of harmonic oscillators. This level of approximation corresponds to the case when the transverse correlations between spins are neglected, i.e. the Hamiltonian is reduced to an Ising type Hamiltonian whose longitudinal correlations reduce to the MFA results.

Thus, taking into account that the basic nature of the Valenta model is connected with the Heisenberg type interactions, it seems to us to be natural to introduce the reaction field approach instead of the molecular field approximation, which is usually interpreted as the result of the Valenta model while in fact it is only one of various possibilities. Another possibility is to use the reaction field approach, as in the present paper.

3.2. The basic concept of the reaction field approach

Taking into account the original assumptions which are based on the physical background, we can modify the Valenta model by introducing the reaction field instead of the standard molecular field.

The procedure for the reaction field approach consists in adding to the molecular field a correlation dependent term determined by means of the fluctuation–dissipation theorem. The part of the effective field arising from the reaction field does not favour one orientation over another while the molecular field is directed along the spontaneous magnetization axis. The following considerations are based on the pioneering work by Wojtczak and Mrygoń [19], although this kind of approach has also been used in the case of inhomogeneous ferromagnets [20].

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 neighbouring atoms. In this case the atoms situated at the surfaces have a neighbourhood that is different from that of the atoms inside a sample. This geometric situation then corresponds to the different conditions 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 a two-dimensional thermodynamically homogeneous subsystem [21].

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

$$F = U - TS \quad (1)$$

which can be obtained by means of the internal energy U and the entropy S calculations.

To determine the internal energy we introduce the Hamiltonian of the system in the following form:

$$\mathcal{H} = -\frac{1}{2} \sum_{vjv'j',\alpha} J_{vjv'j'}^\alpha S_{vj}^\alpha S_{v'j'}^\alpha - \gamma \sum_{vj\alpha} H_{vj}^\alpha S_{vj}^\alpha \quad (2)$$

where the exchange integral is given by:

$$J_{vjv'j'}^\alpha = J_{vjv'j'} + K_{vj} \delta_{vjv'j'}^{\alpha z} \quad (3)$$

with K_{vj} standing for the anisotropy constant including the volume and the surface anisotropy term and with $J_{vjv'j'}$ representing the exchange integral responsible for the interaction between a given spin and its nearest neighbours in the same magnetic layer ($v = v'$) or in the neighbouring layers ($v' = v \pm 1$). The index v numbers the monoatomic layers of the thin films ($v = 1, \dots, n$) while vector j defines the position of the magnetic atom within a given layer v , z is the direction of spontaneous magnetization, α means direction perpendicular ($\alpha = \perp = x, y$) or parallel ($\alpha = \parallel = z$) to the magnetization, γ stands for $g\mu_B$ factor and H_{vj}^α is an external field.

Hamiltonian (2) can be approximated by an effective Hamiltonian:

$$\mathcal{H}^{\text{eff}} = -\gamma \sum_{vj,\alpha} H_{vj}^{\text{eff},\alpha} S_{vj}^\alpha \quad (4)$$

where $H_{vj}^{\text{eff},\alpha}$ stands for the effective mean field which is given by

$$\gamma H_{vj}^{\text{MFA},\alpha} = \sum_{v'j'} J_{vjv'j'}^\alpha \langle S_{v'j'}^\alpha \rangle + \gamma H_{vj}^\alpha \quad (5)$$

in the standard molecular field approximation (MFA) while in the reaction field approximation (RFA) it takes the following form:

$$\gamma H_{vj}^{\text{RFA},\alpha} = \sum_{v'j'} J_{vjv'j'}^\alpha \langle S_{v'j'}^\alpha \rangle - \lambda \langle S_{vj}^\alpha \rangle + \gamma H_{vj}^\alpha. \quad (6)$$

We introduce here the correlation parameter λ characteristic of the RFA which is in fact independent of (v, j) due to symmetry conditions [11] and is assumed to be homogeneous in the sample.

We can now define the internal energy U as the mean value of the effective Hamiltonian (4), namely

$$U = \langle \mathcal{H}^{\text{eff}} \rangle = - \sum_{vjv'j',\alpha} J_{vjv'j'}^\alpha \langle S_{vj}^\alpha \rangle \langle S_{v'j'}^\alpha \rangle + \sum_{vj,\alpha} (\lambda \langle S_{vj}^\alpha \rangle \langle S_{vj}^\alpha \rangle - \gamma H_{vj}^\alpha \langle S_{vj}^\alpha \rangle). \quad (7)$$

First of all, we calculate the quantity λ considering equation (6). It may be separated into its unperturbed $H_{vj,0}^\alpha$ and perturbed δH_{vj}^α parts. The latter is connected with the response of the system for the existing molecular field and the external field H_{vj}^α . Thus,

$$\gamma H_{vj,0}^\alpha = \sum_{v'j' \in vj} J_{vjv'j'}^\alpha \langle S_{v'j'}^\alpha \rangle_0 - \lambda \langle S_{vj}^\alpha \rangle_0 \quad (8)$$

$$\gamma \delta H_{vj}^\alpha = \gamma H_{vj}^\alpha + \sum_{v'j' \in vj} J_{vjv'j'}^\alpha \delta \langle S_{v'j'}^\alpha \rangle - \lambda \delta \langle S_{vj}^\alpha \rangle. \quad (9)$$

We now introduce the k -space which is usually considered in the case of the thin film geometry [22]. We assume the usual notation, k for 3D systems and h for 2D subsystems so that $k = (\tau, h)$ where τ labels the subsystems. The transformation $T_{vj\tau h}^\alpha$ changes the representation of the Hamiltonian (7) from the configurational space (vj) in which the pair interactions are taken into account to the wavevector (τ, h) -space in which this Hamiltonian is diagonal. Then, the fluctuating term of the effective field δH_{vj}^α (9) can be transformed into (τ, h) -space by multiplying relation (9) by $T_{vj\tau h}^\alpha$ and summing over (vj) , namely

$$\delta \langle S_{\tau h}^\alpha \rangle = \sum_{vj} T_{vj\tau h}^\alpha \delta \langle S_{vj}^\alpha \rangle \quad (10)$$

$$H_{\tau h}^\alpha = \sum_{vj} T_{vj\tau h}^\alpha H_{vj}^\alpha \quad (11)$$

while the equation

$$\sum_{v'j' \in vj} J_{vjv'j'}^\alpha T_{v'j'\tau h}^\alpha = J^\alpha(\tau h) T_{vj\tau h}^\alpha \quad (12)$$

leads to the solution for the transformation coefficient $T_{vj\tau h}^\alpha$. Thus,

$$\gamma \delta H^\alpha(\tau h) = \gamma H^\alpha(\tau h) + (J^\alpha(\tau h) - \lambda) \delta \langle S_{\tau h}^\alpha \rangle. \quad (13)$$

In the (τ, h) -space introduced above we denote h as a wavevector of spin waves propagating in the planes; h is then

perpendicular to j . The mode τ corresponds to the third component of the wavevector oriented perpendicularly to ν . The details of $T_{vj\tau h}^\alpha$ transformation are discussed in [22].

Now, we present the detailed calculations leading to the expressions for λ and other quantities which are necessary to describe our system.

First of all, following the procedure reported in [11], we obtain the expressions for the susceptibilities, parallel or perpendicular with respect to the spontaneous magnetization oriented along the z -axis. In the space (τ, h) they are of the forms:

$$\chi^\parallel(\tau h) = \chi^{0\parallel} \frac{1}{1 - \frac{\chi^{0\parallel}}{\gamma^2} (J^\parallel(\tau h) - \lambda)} \quad (14)$$

and

$$\chi^{0\perp} = \frac{\gamma^2 \langle S_{vj}^z \rangle_0}{\gamma H_{vj0}^z} \quad (15)$$

which can be transformed by means of (8) and (10) together with relation (12) to the form:

$$\frac{\chi^{0\perp}}{\gamma^2} = \frac{1}{(J^z(1, 0) - \lambda)}. \quad (16)$$

The effective susceptibility $\chi^{0\parallel}$ of the system consists of the non-interacting localized spins in the presence of an external field H_{vj}^α and it satisfies the summation rule:

$$\chi^{0\parallel} = \frac{1}{nN^2} \sum_{\tau h} \chi^\parallel(\tau h) \quad (17)$$

where nN^2 denotes the number of spins in the system consisting of n layers while N stands for the number of spins in the linear dimension of the plane.

We assume that our system is isotropic in the sense that spins which are embedded in molecular and external fields create as response to these fields their own isotropic field which does not favour one orientation over another. This yields the equality

$$\chi^{0\perp} = \chi^{0\parallel} \quad (18)$$

which can be considered as the equation with respect to λ and which allows us to derive it using the summation rule (17).

By substituting (14) into (17) and using (16) we have

$$1 = \frac{J^z(1, 0) - \lambda}{J(1, 0)} \frac{1}{nN^2} \sum_{\tau h} \frac{1}{\frac{J^z(1, 0)}{J(1, 0)} - \frac{J(\tau, h)}{J(1, 0)}} \quad (19)$$

where $J^z(1, 0) = J(1, 0) + K$ and $J(\tau, h) = J^\perp(\tau, h)$ and K is the anisotropy of each layer.

On the basis of the above equation we can introduce the quantities defined in the following way:

$$s = \frac{J^z(1, 0)}{J(1, 0)} \quad (20)$$

and

$$G(s) = \frac{J(1, 0)}{nN^2} \sum_{\tau h} \frac{1}{J(1, 0) + K - J(\tau, h)}. \quad (21)$$

The last quantity is known as ‘lattice Green function’ and it depends on the number of layers forming the thin film. We can see from (19) that the parameter λ can be written in the form:

$$\lambda = J(1, 0) \left(s - \frac{1}{G(s)} \right). \quad (22)$$

Now, we have to calculate $J(\tau, h)$ which is dependent on the crystallographic structure of the system. We use the relation:

$$J(\tau, h) = \frac{1}{N} \sum_{vj} \sum_{v'j'} T_{v\tau} T_{v'\tau} e^{ih(j-j')} J_{vjv'j'}. \quad (23)$$

For the coefficients $T_{v\tau}$ we have the well-known equations [22]

$$T_{\nu+1, \tau} + T_{\nu-1, \tau} = (2 \cos \vartheta_\tau) T_{\nu\tau} \quad \text{for } \nu = 2, 3, \dots, n-1 \quad (24)$$

together with the orthonormality condition

$$\sum_{\nu} T_{\nu\tau} T_{\nu\tau'} = \delta_{\tau\tau'} \quad (25)$$

and the boundary conditions

$$\begin{aligned} (\kappa_S - 2 \cos \vartheta_\tau) T_{1\tau} + T_{2\tau} &= 0, \\ (\kappa_S - 2 \cos \vartheta_\tau) T_{n\tau} + T_{n-1, \tau} &= 0 \end{aligned} \quad (26)$$

where ϑ_τ is connected with the perpendicular wave amplitudes $T_{v\tau}$ and can be interpreted as the third component of the propagation wavevector. In the case of thin films, parameter ϑ_τ runs over a discrete spectrum of values which can be determined by the difference equations (24)–(26) [23, 24] and which depends on constant κ_S , being the perpendicular anisotropy, which describes only the properties of the boundary surfaces.

Taking into account the relations (24) and (25), we can rewrite equation (23) in the form

$$\begin{aligned} J(\tau, h) &= \frac{1}{N} \sum_{j'_v \in j_v} J_{vjv'j'} e^{ih(j_v - j'_v)} \\ &+ \frac{1}{N} 2 \cos \alpha_\tau \sum_{j'_\mu \in j_\nu} J_{vj\mu j'} e^{ih(j_\nu - j'_\mu)} \end{aligned} \quad (27)$$

where $\mu = \nu \pm 1$.

Next, we can define the following coefficients which determine the structure of the considered system:

$$f_0 = \sum_{j'_v \in j_v} e^{ih(j_v - j'_v)} \quad (28)$$

where the sum runs over spins j' belonging to the same layer ν and

$$f_{|\mu|} = \sum_{j'_\mu \in j_\nu} e^{ih(j_\nu - j'_\mu)} \quad (29)$$

where the sum runs over spins j' belonging to the neighbouring layers $\mu = \nu \pm 1$.

In this way the ‘lattice Green function’ given by (21) is determined and we can write the final expression for the internal energy U as follows:

$$\begin{aligned} U = \langle \mathcal{H}^{\text{eff}} \rangle &= -z_0 \sum_{\nu} J_{\nu\nu} m_\nu m_\nu - z_1 \sum_{\nu \in \nu'} \frac{J_{\nu\nu'}}{2} m_\nu m_{\nu \pm 1} \\ &- \sum_{\nu} (K_\nu - \lambda) m_\nu m_\nu - \gamma \sum_{\nu} H_\nu^z m_\nu \end{aligned} \quad (30)$$

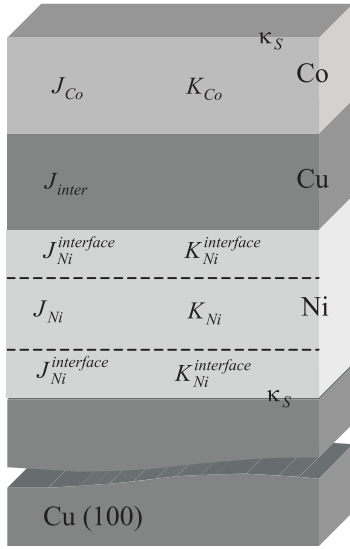


Figure 1. The trilayer system Co/Cu/Ni/Cu(100) with the exchange couplings and anisotropies used in the theoretical model considered here.

for internal layers $\nu = 2, 3, \dots, n - 1$, with boundary conditions for $\nu = 1$, and for $\nu = n$, given by

$$U_1 = -z_0 \sum_{\nu} J_{\nu\nu} m_{\nu} m_{\nu} - z_1 \sum_{\nu' \in \nu} \frac{J_{\nu\nu'}}{2} m_{\nu} m_{\nu+1} - \sum_{\nu} (K_{\nu} - \lambda) m_{\nu} m_{\nu} - \gamma \sum_{\nu} H_{\nu}^z m_{\nu} \quad (31)$$

$$U_n = -z_0 \sum_{\nu} J_{\nu\nu} m_{\nu} m_{\nu} - z_1 \sum_{\nu' \in \nu} \frac{J_{\nu\nu'}}{2} m_{\nu} m_{\nu-1} - \sum_{\nu} (K_{\nu} - \lambda) m_{\nu} m_{\nu} - \gamma \sum_{\nu} H_{\nu}^z m_{\nu} \quad (32)$$

where

$$m_{\nu} \equiv \langle S_{\nu}^z \rangle \quad (33)$$

$K_{\nu} \equiv K$ while z_0, z_1 are the numbers of nearest neighbours of a given atom in the same monoatomic layer and in the neighbouring layers, respectively.

The entropy is given in the standard form:

$$S = -N^2 \sum_{\nu=1}^n \sigma_{\nu} \quad (34)$$

where

$$\sigma_{\nu} = k_B \left[\left(\frac{1}{2} + m_{\nu} \right) \ln \left(\frac{1}{2} + m_{\nu} \right) + \left(\frac{1}{2} - m_{\nu} \right) \ln \left(\frac{1}{2} - m_{\nu} \right) \right] \quad (35)$$

is the entropy per single lattice site.

3.3. Application of the Valenta model modified by RFA to multilayer systems

The present paper is especially focused on the trilayer system which consists of Ni film placed on fcc-Cu(100) substrate and covered with Co film separated by a nonmagnetic Cu spacer (figure 1). We study the influence of Co film as well as Cu spacer thickness on the Curie temperature for Ni film in the system in question.

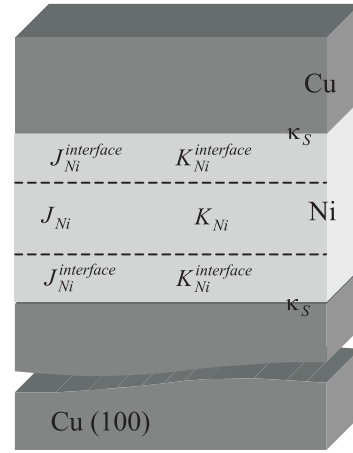


Figure 2. The bilayer system Cu/Ni/Cu(100) with the exchange couplings and anisotropies used in the theoretical model considered here.

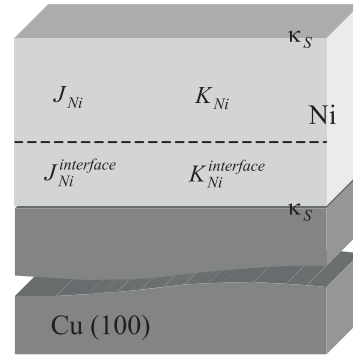


Figure 3. The single Ni/Cu(100) system with the exchange couplings and anisotropies.

Taking into account the theoretical model introduced above, we can divide our system with thickness $d = d_{\text{Ni}} + d_{\text{Co}}$ into several parts with respect to different exchange couplings $J_{\nu j \nu' j'}$ namely, for interior Ni and Co layers we have J_{Ni} and J_{Co} while for nickel layers which are directly connected with copper we put an interface exchange coupling $J_{\text{Ni}}^{\text{interface}}$. The Co/Cu interface is not differentiated because it is approximately compensated by the enhancement of the magnetic moment in the topmost layer facing the vacuum [25] and we take the same values of the exchange integral for all monoatomic layers forming the Co film. The trilayer system shown in figure 1 can be modified for simpler systems like a Ni layer embedded in the fcc-Cu host (figure 2), a Ni overlayer on the fcc-Cu(100) substrate (figure 3) or a freestanding Ni layer (figure 4).

In the Ni/Cu(100) system we do not distinguish the values for the exchange integral at the surface from those inside the Ni film, with the exception of the interface layer. This assumption is based on the theoretical calculations of magnetic moments in the Ni/Cu system performed by means of the Korringa–Kohn–Rostoker Green function method [26]. It has been shown [26] that the enhancement of the surface magnetic moment on Ni film compared to the value for the interior Ni atoms is rather

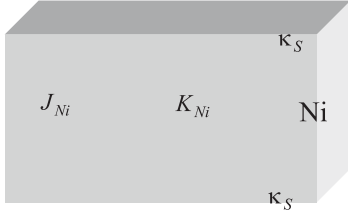


Figure 4. The freestanding Ni layer with the exchange couplings and anisotropies.

small (6%), while the reduction of the interface magnetic moment of Ni is much more pronounced in comparison to its bulk value (38%). That is why we distinguish only the interface exchange integral $J_{\text{Ni}}^{\text{interface}}$, treating the surface interactions as bulk-like coupling.

The interaction between two different magnetic films Ni and Co takes place via the interlayer exchange coupling (IEC), which depends on the thickness of the nonmagnetic spacer, and for this coupling we use J_{inter} (IEC).

Apart from the $J_{vjv'j'}$ a crucial role is played by the anisotropy K_v . In each magnetic layer we distinguish two kinds of anisotropies—in-plane K_{Ni} and K_{Co} —while in the interface Ni/Cu layer we have $K_{\text{Ni}}^{\text{interface}}$. In order to take into account the boundary conditions connected with the lack of nearest neighbours on the surface we also consider a perpendicular anisotropy κ_S introduced by equations (24)–(26).

In order to perform the calculations for each of the above mentioned systems and to compare the Curie temperature of Ni layers we have to determine the equation (27) for the case of the fcc(100) thin film system. It is easy to see that it can be written in the form

$$J(\tau, h) = J_1 f_0 + J_2 2 \cos \vartheta_\tau f_{|1|} \quad (36)$$

with coefficients expressed as follows:

$$f_0 = 2 \cos\left(h_x \frac{a}{2}\right) \cdot 2 \cos\left(h_y \frac{a}{2}\right) \quad (37)$$

$$f_{|1|} = 2 \left(\cos\left(h_x \frac{a}{2}\right) + \cos\left(h_y \frac{a}{2}\right) \right) \quad (38)$$

and determining the structure of the considered system.

Transforming the coordinates to spherical system:

$$h_x = h \cos \varphi, \quad h_y = h \sin \varphi \quad (39)$$

we get $J(1, 0)$ appearing in equations (18)–(21) equal to:

$$J(1, 0) = z_0 J_1 + z_1 J_2 2 \cos \vartheta_\tau \quad (40)$$

where $z_0 = 4$ and $z_1 = 4$ are the numbers of nearest neighbours of a given atom in the same monoatomic layer and in the neighbouring layers, respectively.

We have determined all quantities to perform the calculations giving us the temperature dependent magnetization of each layer forming our system. For this purpose we have to find the equilibrium values of m_v by minimizing the free energy of the system (1) with respect to m_v :

$$\frac{\partial F}{\partial m_v} = 0, \quad v = 1, 2, \dots, n \quad (41)$$

where n represents the number of layers in the whole system.

Thus, the theoretical results for the Curie temperature are obtained in the present paper where the method of calculation corresponds to the procedure of the Valenta model [4] applied now to the molecular field modified by the reaction field term. The procedure consists in the minimization of the thermodynamic free energy taken for the internal energy (30) with respect to the magnetization profiles. Close to the Curie temperature the problem is equivalent to the set of linear equations (41) whose solution leads to the Curie temperature and the temperature dependent magnetization of each monoatomic layer.

The result obtained in this satisfies the size effect property, i.e. the dependence of the Curie temperature on the film thickness.

4. Numerical results and discussion

For all systems analysed here we use the following values of exchange integrals and anisotropy parameters: $J_{\text{Ni}} = 1.7 \times 10^{-21}$ J, $J_{\text{Ni}}^{\text{interface}} = 4.97 \times 10^{-22}$ J, $J_{\text{Co}} = 3 \times 10^{-21}$ J, $J_{\text{inter}} = 4.97 \times 10^{-23}$ J, $K_{\text{Ni}} = 0.001 J_{\text{Ni}}$, $K_{\text{Ni}}^{\text{interface}} = 0.001 J_{\text{Ni}}^{\text{interface}}$, $K_{\text{Co}} = 0.001 J_{\text{Co}}$, $\kappa_S = -1$ for the systems with Co film and $\kappa_S = -0.1$ for the systems without Co film. The values for the exchange integrals are slightly changed from those proposed in [3] by Scherz *et al*, in connection with the fitting procedure, while the values for other parameters come directly from the procedure of best fitting to the experimental data.

It is worthwhile noticing that the real anisotropy results from various physical sources. One of them is the spin-orbit coupling whose influence on the anisotropic behaviour of the magnetic system can be described by the effective term in the Hamiltonian (2). The evaluation of the effective constants is then dependent on the particular sample, i.e. the anisotropy constant can be treated as phenomenological and its value should be related to the experiments by the best fitting procedure. It is also well known [26] that the values of the exchange integrals depend on the layer-resolved magnetic moments which are by themselves dependent on the film thickness. However, for the purpose of the present considerations we keep them constant for the reasons mentioned in section 3.3 and, on the other hand, for clear comparison with the results presented in paper [3]. The weak point is connected with the value of $J_{\text{Ni}}^{\text{interface}}$ which is much smaller than the other values of the exchange integrals in the system and can be considered as an unrealistic modification. However, some experimental measurements reported in [27–29] show that the magnetic moments are reduced by more than a factor of 2 in comparison to the Ni bulk. Such a large reduction in the total magnetic moment cannot be due to the existence of some magnetic dead layers since a clear ferromagnetic response has been found down to 1.6 ML Ni on Cu(001) [27] but it can be attributed to the interlayer mixing on the interface going in the real systems deeper than one monoatomic layer.

Table 1. Calculated critical temperatures of Ni films in freestanding (fs) Ni layer, single Ni/Cu film, bilayer Cu/Ni/Cu system and trilayer 3 ML Co/Cu/Ni/Cu system. The last column represents the shift of the Ni temperature caused by the exchange coupling in the presence of Co film.

Ni (ML)	$T_{C,Ni}$ (K)									$\Delta T_{C,Ni}$ (K)
	fs-Ni	Ni/Cu(100)	Cu/Ni/Cu(100)			Co/Cu/Ni/Cu(100)				
			RFA	MFA	Exp [3]	RFA	MFA	Exp [3]		
1	91	27	14	73		45	79		31	
2	103	71	31	109	30	101	113	100	70	
3	198	173	131	261	147	199	263	214	68	
4	287	271	246	375	237	319	376	314	73	
5	366	339	334	423		392	423		58	
6	432	422	407	447		445	447		38	

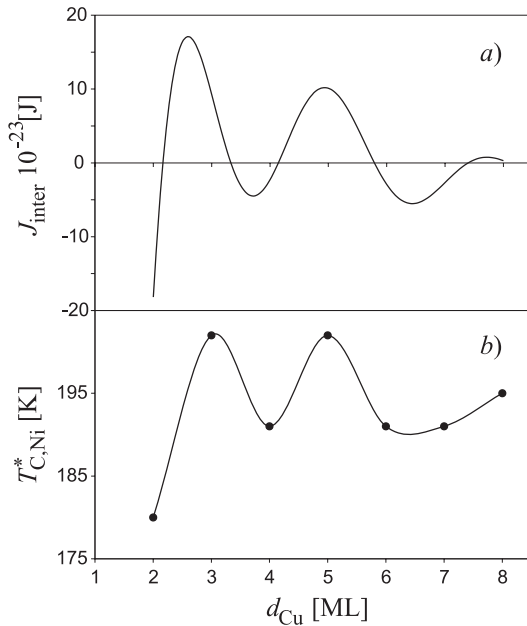


Figure 5. The interlayer exchange coupling (a) and the Curie temperature (b) as a function of the Cu spacer thickness for the 3 MLCo/Cu/3 MLNi/Cu(100) system.

The values of J_{inter} are directly connected with the thickness d of the Cu spacer by the well-known Bruno expression [8], namely

$$J_{inter}(d) = \frac{1}{d^2} \left\{ A_1 \sin\left(\frac{2\pi d}{\Lambda_1} + \Phi_1\right) + A_2 \sin\left(\frac{2\pi d}{\Lambda_2} + \Phi_2\right) \right\} \quad (42)$$

where $\Lambda_1 = 2.56$ ML with $\Phi_1 = \frac{\pi}{2}$ and $\Lambda_2 = 5.88$ ML with $\Phi_2 = \pi$ while the ratio of the amplitudes, obtained from measurements, is $\frac{A_1}{A_2} = 1.3(5)$. The above dependence obtained for the system with 3 ML of Ni capped with 3 ML of Co is presented in figure 5. We can notice the oscillatory character of interlayer exchange coupling (figure 5(a)) and consequently the same behaviour of the Curie temperature (figure 5(b)). For the positive and negative values of the IEC two magnetic layers are coupled ferromagnetically or antiferromagnetically, respectively.

The calculations consists in resolving the set of equations (41) for the free energy of the system (1) described

in the frame of the reaction field approximation modifying the Valenta model of thin films. They allow us to obtain the values of the Curie temperature and to compare their behaviour as well as to present the temperature dependent magnetization of the ferromagnetic Ni film in different systems. The experimental data for the Curie temperature are taken from [3] where the method of their evaluation is given. Starting from the simplest freestanding Ni film we extend this system by depositing it on the Cu substrate and then putting on the top of that system a nonmagnetic Cu film and finally covering it with a magnetic film of Co. We then analyse the influence of the Ni/Cu interface and of the IEC between Ni and Co films on the Curie temperature of the Ni film. The results for different thicknesses of the Ni film are presented in table 1.

Analysing the data in table 1 for a given Ni film thickness we can see that the Curie temperature for a freestanding film is higher than in the case of Ni/Cu and Cu/Ni/Cu systems. The values of temperature obtained for the single Ni film and the bilayer system are in a good agreement with the results reported in [9] where it is shown experimentally that an uncapped Ni film has a higher temperature than a film embedded in the Cu/Ni/Cu(100) system. Moreover, a similar behaviour of transition temperature has been shown theoretically in [30] for Co film, while for Fe film the inversed behaviour has been reported. This last case can be easily described within our approach by using a different interface exchange integral, namely $J_{Fe}^{interface} > J_{Fe}$.

This kind of behaviour of the Curie temperature as well as the decrease of the magnetization caused by the capping Cu layer is attributed to hybridization between the bonding d-states of the ferromagnet and the Cu in the interface layer [9]. The hybridization leads to reduction of the interacting magnetic moments related to each crystalline lattice site. Spins connected with magnetic moments, interacting with their nearest neighbours and with the molecular field, generate their own reaction field. Therefore, a given spin is affected by both the molecular field as well as the reaction field. We can conclude that the interface in the Ni/Cu system and the additional interface in the Cu/Ni/Cu system modify the magnetic moments in different ways, which influences the reaction field and consequently the $T_{C,Ni}$ of the magnetic film.

An interesting temperature behaviour of the magnetization appears if we put on the top of the bilayer Cu/Ni/Cu system another kind of magnetic film, like Co film, building a trilayer

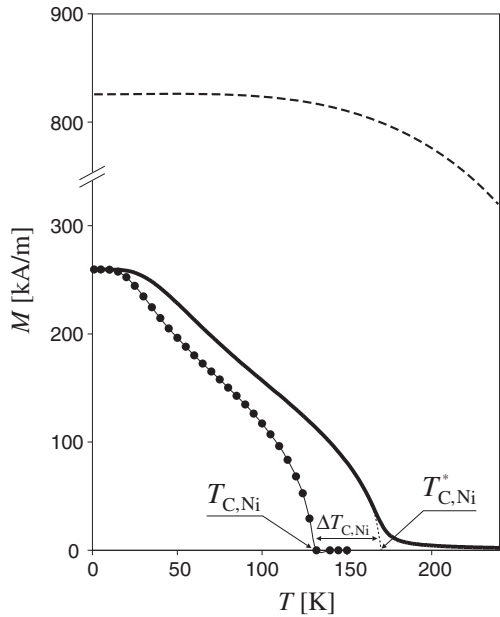


Figure 6. Calculated Ni magnetization of a bilayer Ni film (dotted line) and of Ni film coupled to Co film (solid line) as a function of temperature. The dashed line refers to the Co magnetization. By applying $J_{\text{inter}} = 0$ we identify $T_{\text{C,Ni}}^*$.

Co/Cu/Ni/Cu system. Now, we examine and compare the temperature dependent magnetization for a bilayer composed of 3 ML of Ni embedded between Cu films—Cu/Ni/Cu(100)—and for trilayer when 2 ML of Co are deposited on the top of the above system—Co/Cu/Ni/Cu(100). The magnetization curves obtained as an average over all Ni and Co layers, respectively for Ni and Co films, are shown in figure 6. We can notice two different phase transition temperatures for these different magnetic films: $T_{\text{C,Ni}}^* = 168$ K and $T_{\text{C,Co}} = 386$ K (to estimate $T_{\text{C,Ni}}^*$ we fit the magnetization curve m_{Ni} when $J_{\text{inter}} = 0$). For the whole range of temperatures the magnetization of Ni film is much smaller than for the Co one, which is caused by the difference in the values of magnetic moments reported for these two materials [3]. The third curve shown in figure 6 (dotted line) is calculated for the bilayer system Cu/Ni/Cu which exhibits a phase transition at $T_{\text{C,Ni}} = 131$ K, lower than $T_{\text{C,Ni}}^* = 168$ K, which is characteristic of the system with 2 ML of Co deposited on the top. The influence of the capped Co layers on Ni phase transition temperature means the influence of the IEC on magnetization and its behaviour in the vicinity of $T_{\text{C,Ni}}$ is clearly seen. These results are in good agreement with experimental data and with theoretical calculations obtained from the Green function approach [3]. It is worthwhile noticing that the change of the critical temperature of Ni film $T_{\text{C,Ni}}^*$ in the Co/Cu/Ni/Cu system with respect to its value $T_{\text{C,Ni}}$ in the Cu/Ni/Cu system is always towards higher temperatures independently of the sign of the IEC, while we observe the oscillatory character of the shift of phase transition temperature $\Delta T_{\text{C,Ni}} = T_{\text{C,Ni}}^* - T_{\text{C,Ni}}$ (figure 5).

In addition, for a better comparison of these two systems we analyse the influence of Ni film thickness on its Curie temperature in the case of the Co/Cu/Ni/Cu(100) system with 3 ML of Co and the Cu/Ni/Cu(100) system. The results are

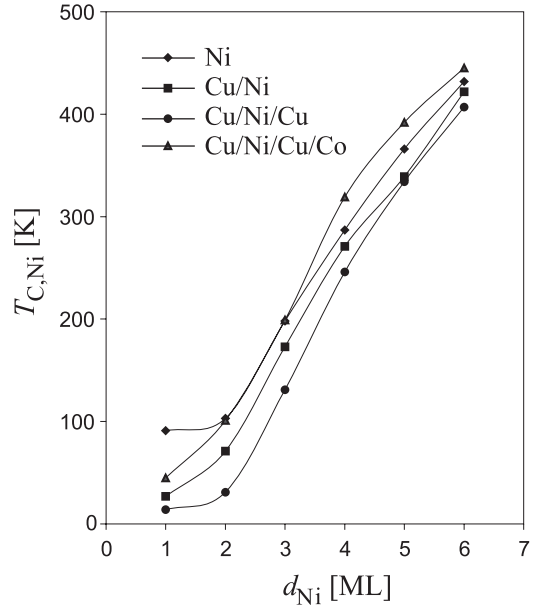


Figure 7. The thickness dependence of Curie temperature $T_{\text{C,Ni}}$ of Ni film for the following systems: Co/Ni/Cu/Cu(100) with 3 ML of Co, Cu/Ni/Cu, Ni/Cu and the freestanding Ni film.

gathered in table 1 where we also present the results from MFA calculations. We can see that the values of Curie temperatures calculated within MFA are quite different from those obtained in RFA. In each system an increase of transition temperature with Ni film thickness is observed. The comparison with experimental data shows quite good agreement (table 1). Taking into account the fact that in the case of experimental data [3] the systems are not perfectly built, as complete monolayers, we can consider this agreement as excellent.

The Curie temperatures obtained for the Co/Cu/Ni/Cu system indicate that the Co film causes an increase of the Curie temperature of the Ni film, which is also reported in [3, 5, 9, 31, 32]. It is obvious that the reaction field in Ni layers is now changed due to the presence of another magnetic material.

The results from table 1 are presented in figure 7. It is worthwhile noticing that the curves for Cu/Ni/Cu and Co/Cu/Ni/Cu systems are in good agreement with those obtained within the microscopic many-body Green function theory (e.g. [3]).

The shift of Ni Curie temperature with a change of the Ni film thickness observed in the Co/Cu/Ni/Cu system (see the last column in table 1) is presented in figure 8 for two values of J_{inter} , namely $J_{\text{inter}} = 4.97 \times 10^{-23}$ J and $J_{\text{inter}} = 10.8 \times 10^{-23}$ J. The value of $\Delta T_{\text{C,Ni}}(d_{\text{Ni}})$ exhibits a maximum for $d_{\text{Ni}} = 4$ ML, which is in agreement with the results reported in [32].

The Valenta model of thin films improved by the reaction field approach (RFA) allows us, in particular, to calculate the temperature dependence of magnetization for each monoatomic layer which builds the trilayer system. We show in figures 9(a) and (b) the layer-dependent spontaneous magnetization as a function of temperature determined for two systems: the first one consists of 3 ML of Ni and 3 ML of Co, the second one is composed of 5 ML of Ni and 3 ML of

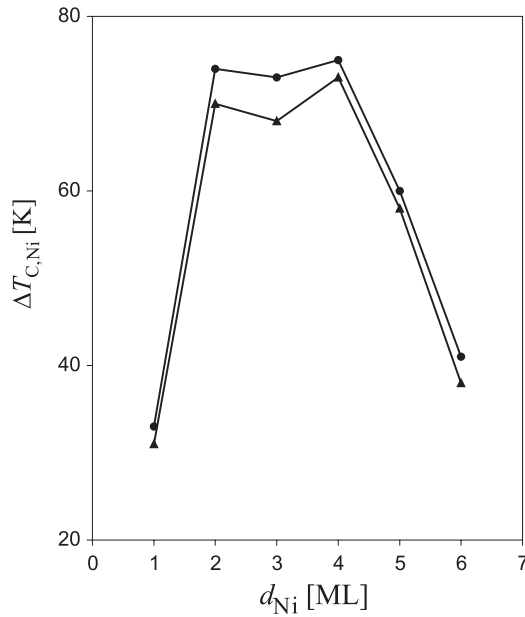


Figure 8. The shift $\Delta T_{C,Ni}$ (K) of Ni Curie temperature as a function of the Ni film thickness observed in the Co/Cu/Ni/Cu system for different interlayer couplings $J_{inter} = 4.97 \times 10^{-23}$ J (▲) and $J_{inter} = 10.8 \times 10^{-23}$ J (●).

Co while in both cases the Ni film is separated from the Co film by a Cu spacer stacked along the (100) direction. We can see that the distribution of magnetization along the direction perpendicular to the surface has the same character for both systems. First of all, the magnetization is lower at surfaces of Ni and Co films than in the middle layers and its distribution is asymmetric due to different boundary conditions. Moreover, the difference between the magnetization of the surface and middle layers is much more pronounced in the case of the thicker system. On the other hand, the existence of an exchange coupling integral J_{inter} causes small tails in the Ni magnetization curves, which are similar to those observed in the Curie point region for phase transition in the system under an external magnetic field.

In figure 10 we present the average magnetization normalized to $m_{Ni}(T=0)$ as a function of temperature normalized to $T_{C,Ni}$ (phase transition temperature of Ni in the Cu/Ni/Cu system) for different Ni film thicknesses. Each curve shows a tail, but the thicker the Ni film is the weaker the observed influence of IEC on magnetization and the shorter the tail in the magnetization curve. The first curve in figure 10 (looking from the left) represents the case of a bilayer system with 4 ML of Ni placed between Cu films. Unfortunately, quite a strong influence of IEC on magnetization in the systems with one and two Ni monolayers causes difficulties in determining their $T_{C,Ni}^*$.

Our theoretical calculations show the difference between the magnetic properties of Ni film considered as a freestanding Ni layer and placed in a single Ni/Cu film, a bilayer Cu/Ni/Cu and a trilayer Co/Cu/Ni/Cu system. These different properties reflected by different Curie temperatures can be explained as being caused by the interface with a nonmagnetic Cu layer and by the interlayer exchange coupling with a Co magnetic

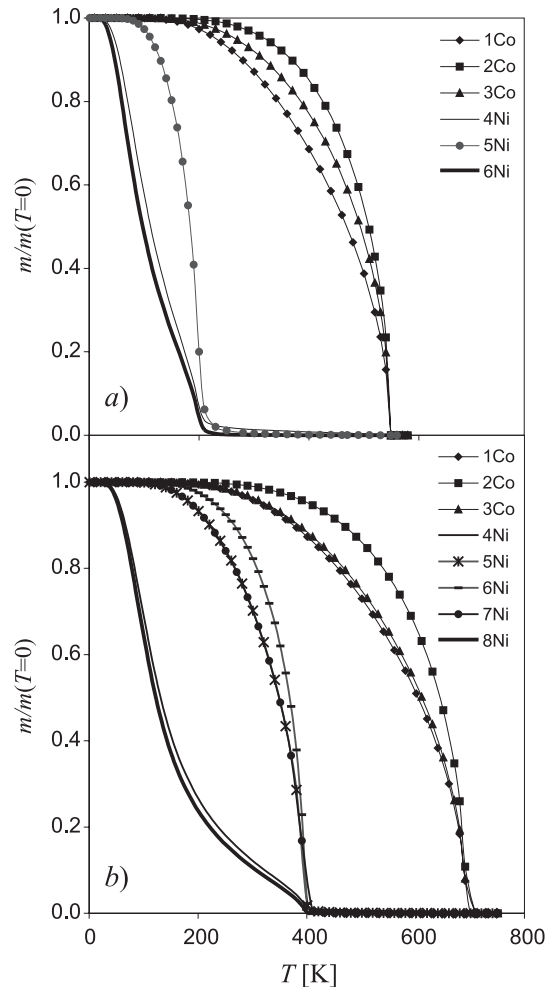


Figure 9. The layer-dependent spontaneous magnetization as a function of temperature determined for 3 MLCo/Cu/3 MLNi/Cu(100) system (a) and for 3 ML Co/Cu/5 ML Ni/Cu(100) system (b).

film. The explanation for such behaviour is related to the existence of the field in which the spins are embedded in different environments and to which they react.

5. Final remarks

The Valenta model modified by means of the reaction field approach seems to be an excellent illustrative example which shows that the origin of the Valenta model usually related to the molecular field approximation can be in fact of wider applicability.

The Valenta model of thin films improved by the reaction field approximation seems to be a very good approach and a useful method to calculate some magnetic properties of different multilayer systems, giving results similar to those obtained within the Green function theory [3].

It is worthwhile noticing that within the theoretical model for thin films used here the Curie temperature in the case of isotropic interactions is tending to zero. This corresponds to the anisotropy constant $K_v = 0$ ($K_{Ni} = 0, K_{Co} = 0$, respectively). In the light of the present approach the stability of magnetic order in thin films can be considered as resulting

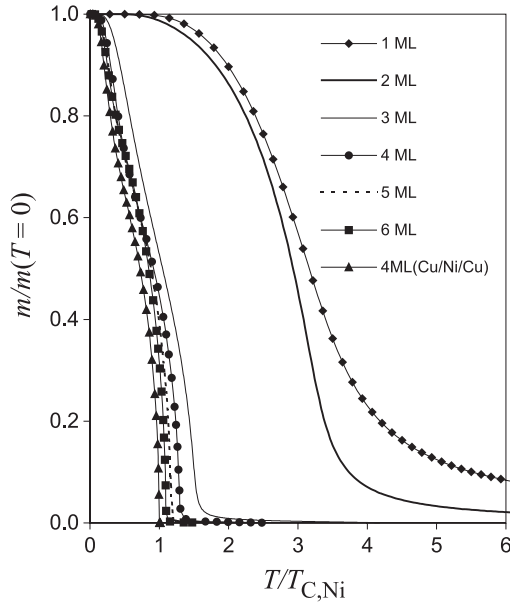


Figure 10. The average magnetization normalized to $m_{\text{Ni}}(T = 0)$ as a function of temperature normalized to $T_{\text{C,Ni}}$ for different Ni film thicknesses in the trilayer system with 3 ML of Co. The first curve from the left represents the case of a bilayer system with 4 ML of Ni placed between Cu films.

from the anisotropic character of the fluctuating part of the reaction field. This anisotropy is caused by the shape of correlation functions which are connected with the geometry of a sample.

The big influence of Co film on the Curie temperature of the Ni film in the case of very thin Ni films (1 ML, 2 ML) can be explained by taking into account the size effect when a 2D system should be considered instead of a 3D system and the enhanced spin fluctuations have to be introduced. The IEC between the two ferromagnetic films suppresses these spin fluctuations and induces a sizeable magnetization in the Ni film. It can be easily related to the idea of a generated reaction field discussed here.

The improvement obtained by means of the generalized Valenta model has a great advantage due to the thermodynamic structure of the theory. The entropy is then still factorized while the internal energy is calculated by means of the spin correlations. This fact confirms the use of the present method and its priority among others by the interpretation from the physical point of view. Moreover, in this context the approach modifying the Valenta model leads to an excellent agreement between experimental and theoretical results collected for the Curie temperature.

From the point of view of the physical interpretation we can see that the molecular field MFA corresponds to the spin correlation functions for which the short range determined by the parameter λ is not taken into account ($\lambda = 0$). On the other hand the RFA approach corresponds to the case when the short range correlations appear ($\lambda \neq 0$). This correlation behaviour can also be observed for the model based on the spin pair entropy [21].

Thus we can see that the entropy factorization leads in both cases considered to different correlation ranges in the

superficial plane and in the direction perpendicular to the surface.

The simplest case is for $\lambda \neq 0$ in the plane which corresponds to a thermodynamically homogeneous subsystem and $\lambda = 0$ in the direction perpendicular to the surface which corresponds to interaction between two homogeneous subsystems. The considered situation describes the interaction between the homogeneous subsystems by means of the molecular field terms, which is the main assumption of the Valenta model based on the Néel sublattice thermodynamics.

The situation discussed in the present paper, $\lambda \neq 0$ for all directions, satisfies the Valenta model construction in the generalized sense when the interaction between monoatomic layers considered as homogeneous subsystems is still of a molecular field nature but now in RFA terms.

Appendix. A Green function approach

In [3] the experimental results are explained by means of the Green function approach which was found to be a suitable tool for this purpose. The Green function method is of general character and it is used in many different problems. In particular, in the case of thin film geometry the method was introduced by Brodhorb and Haubenreisser [33] and developed by many authors [34]. It is well known that the formula which determines the magnetization distribution $\langle S_{\nu j}^z \rangle$ across a film with layer structure numbered by ν , applied in a variety of papers, can be written for $S = \frac{1}{2}$ as (e.g. [35]):

$$\langle S_{\nu j}^z \rangle = \frac{1}{2} + \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\text{Im} \langle \langle S_{\nu j}^+ | S_{\nu j}^- \rangle \rangle_{\omega}}{\exp(\omega/k_B T) - 1} d\omega \quad (\text{A.1})$$

where $\langle \langle S_{\nu j}^+ | S_{\nu j}^- \rangle \rangle_{\omega}$ is taken as the Fourier representation of the Green function defined by:

$$\langle \langle S_{\nu j}^+(t) | S_{\nu j}^-(t') \rangle \rangle = -i\theta(t - t') \langle [S_{\nu j}^+(t), S_{\nu j}^-(t')] \rangle \quad (\text{A.2})$$

in the configuration space νj and satisfying the equation:

$$\frac{d \langle \langle S_{\nu j}^+(t) | S_{\nu j}^-(t') \rangle \rangle}{dt} = -i\delta(t - t') 2 \langle S_{\nu j}^z \rangle \delta_{\nu j \nu' j'} + \theta(t - t') \langle [[S_{\nu j}^+, H], S_{\nu j}^-] \rangle \quad (\text{A.3})$$

in which the considered system is described by the Hamiltonian H .

Let us consider the Hamiltonian H in the form of a set of magnetic moments represented by the spin operators embedded into the molecular $H_{\nu j}^m$ and external H magnetic fields. Thus, the Hamiltonian takes the form

$$H = -g\mu_B \sum_{\mu\rho} (H + H_{\mu\rho}^m) S_{\mu\rho}^z \quad (\text{A.4})$$

which allows us to find the imaginary parts of the proper Green functions:

$$\langle \langle S_{\mu\rho}^+ | S_{\mu'\rho'}^- \rangle \rangle = \frac{2 \langle S_{\mu\rho}^z \rangle \delta_{\rho\rho'} \delta_{\mu\mu'}}{\omega - g\mu_B (H + H_{\mu\rho}^m) + i\varepsilon} \quad (\text{A.5})$$

which are of the form

$$\text{Im} \left\langle \left\langle S_{\mu\rho}^+ \left| S_{\mu'\rho'}^- \right. \right\rangle \right\rangle = -2\pi (\varpi - g\mu_B (H + H_{\mu\rho}^m)) \times \langle S_{\mu\rho}^z \rangle \delta_{\rho\rho'} \delta_{\mu\mu'} \quad (\text{A.6})$$

We can see that only diagonal functions ($\rho' = \rho, \mu' = \mu$) are different from zero. Moreover, the average values of $\langle S_{\mu\rho}^z \rangle$ do not depend on ρ because of the thermodynamic homogeneity of the layers treated as homogeneous subsystems, i.e.

$$\langle S_{\mu\rho}^z \rangle = \langle S_{\mu}^z \rangle \quad (\text{A.7})$$

and consequently

$$H_{\mu\rho}^m = H_{\mu}^m.$$

Taking into account the relation (A.6) for condition (A.7) we obtain the formula (A.1) in the form

$$\langle S_v^z \rangle = \frac{1}{2} - 2 \langle S_v^z \rangle \int_{-\infty}^{+\infty} \frac{\delta(\varpi - g\mu_B (H + H_v^m))}{e^{\varpi/k_B T} - 1} d\omega \quad (\text{A.8})$$

hence

$$\langle S_v^z \rangle = \frac{1}{2} th \left[\frac{g\mu_B (H + H_v^m)}{2k_B T} \right] \quad (\text{A.9})$$

which corresponds exactly to the solution within the Valenta model for

$$g\mu_B H_v^m = J \sum_{v' \in v} z(v, v') \langle S_{v'}^z \rangle. \quad (\text{A.10})$$

Thus, the problems are equivalent with respect to the final result for $\langle S_v^z \rangle$ when we assume the Ising model and the random phase approximation (RPA) decoupling for the proper Green functions.

Let us consider the Hamiltonian in the case of the molecular field given by (A.4) but for the $H_{\mu\rho}^m$ given by the RFA calculations (6). Then the Green function approach allows us to obtain the average value of spin component z , i.e. $\langle S_v^z \rangle$, given by (A.9) where H_v^m takes now its value (6) with λ given by (22) where s and $G(s)$ are given by (20) and (21), respectively.

We can see that the equivalence between the results of the Valenta model and the Green function approach in the random phase approximation for the Ising model is evident in their property of general character. The extension of the Valenta model applied in the present paper by the introduction of the reaction field correction corresponds to the situation in which the spin correlations are taken into account. Although this idea is common for considerations within the Valenta model and the Green function approach, as far as the physical interpretation goes the results are now not identical due to the different descriptions of the magnetization profiles.

The particular results considered in the present paper provide an illustrative example whose generalization seems to us to have a general meaning for the discussion of methods and their comparison at different levels of accuracy.

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