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On the mean-field theory of magnetic multilayers with bilinear and biquadratic Heisenberg exchange

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Abstract

A system consisting of several layers of magnetic ions interacting by both bilinear and biquadratic Heisenberg exchange is studied within the framework of the mean-field approximation. It is shown that for $S = 1$ there exist two types of ordering: ferromagnetic and ferroquadrupolar. The stability of phases as the function of temperature, biquadratic exchange and surface exchange is discussed analytically and numerically and it was shown that similar to bulk samples there appear first- and second-order transitions and a tricritical point may appear depending on system parameters. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

During the last several years the research on thin magnetic films and multilayer structures has been intensified both from the experimental and theoretical point of view. Modern experimental techniques, especially low-energy electron diffraction (LEED) and nuclear magnetic resonance (NMR) enable precise measurements of quantities of local character like magnetization or susceptibility, which gives an additional impetus to the research. There exists also a rising interest for the application of these materials especially in the magnetic recording industry. For example, multilayer magnetic systems with a non-magnetic metallic spacer became the basis for production of various new devices, in particular, ultrasmall memory sensors which make use of the phenomenon of giant magnetoresistance [1].

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In an excellent review article [2] J.C.S. Levy reviews all the experimental techniques suitable for the study of magnetic films, surfaces and interfaces. He also presents some theoretical results for the magnetic films of Heisenberg magnet with bilinear interaction, taking into account the variation of the anisotropy parameter on the surface (with respect to the bulk). There also exist papers analyzing the magnetic characteristics of the films on the basis of the Ising model, taking into account that some physical and chemical conditions at the surface can change the surface exchange parameter $I_S \neq I$ (see, e.g., Ref. [3] and the references cited therein).

The aim of this paper is to study the properties of thin magnetic films where the spins interact not only by bilinear, but also by biquadratic interaction.

The presence of biquadratic interaction in bulk magnetics was established relatively long-time ago ([4–6]). The important role of the biquadratic exchange term in the spin Hamiltonian of some magnetic insulators was stressed by Harris and Owen [7], Joseph [8] and Rodbell et al. [9]. The effects of the biquadratic exchange were analyzed in numerous papers using various theoretical approaches (MF, GF, etc), to mention just some; Refs. [10–15] etc.

On the other hand, biquadratic exchange between the spins of ultrathin ferromagnetic films (in multi-layered magnetics and sandwich structures) was observed only recently. First, in the sandwich structure Fe|Cr|Fe with a wedge spacer, it was discovered that the sign and magnitude of the exchange interaction changes oscillatory with the spacer width by using scanning electron microscopy (SEM) [16] and Brillouin-light scattering (BLS) [17]. Furthermore, in the intermediate ranges between ferromagnetic and antiferro-magnetic bilinear interaction, rather unusual domain samples were detected by magneto-optical Kerr effect (MOKE) [18,19]. These unusual characteristics were ascribed to the existence of canting between the directions of the magnetization in two Fe slabs, which could not be explained in terms of the conventional ferro- or antiferromagnetic bilinear interaction between layers. The introduction of biquadratic interaction between layers [18] can explain the magnetization dependence on the field at $T = 0\,\mathrm{K}$ and it was shown that the canting angle depends on the ratio of bilinear and biquadratic interactions. More recently, using surface MOKE and ferromagnetic resonance (FMR) measurements in the three-layer sample Fe|Cu|Fe [20] the simultaneous existence of both bilinear and biquadratic interactions between magnetic layers was established.

Quite recently, MOKE and BLS were used to demonstrate the existence of both bilinear and biquadratic interactions in the epitaxial Fe|Cr|Fe|Ag|GaAs(1 0 0) structures [21], then biquadratic interaction in Fe|Zn superlattices ([22] by FMR and vibrating-sample magnetometer VSM), while in Fe|FeSi multilayers [23] the transition from antiferromagnetic biquadratic interaction was shown with temperature decrease.

The possible physical origins of the biquadratic interaction in general are discussed thoroughly in now-a-days classical work of Nagaev [24]. Its origin in multilayer magnetic systems in particular, was first discussed by Slonczewski [25], who suggested an essentially macroscopic model, in which the biquadratic interaction arises due to spatial fluctuation of the bilinear interaction caused by "terraced" width fluctuations (at the monolayer scale) of the nonmagnetic spacer. After that, Barnás and Grünberg [26,27] elaborated two possible microscopic mechanisms which lead to biquadratic interaction even in the case of ideally flat interfaces. According to the first mechanism, the biquadratic term appears as a result of the competition between the inter- and intra-layer exchange interaction, while the second mechanism is based on the fact that the electron wave functions responsible for the interaction, depend on the relative orientation of the magnetization in the ferromagnetic films.

One should necessarily mention the work of Erickson et al. [28] in which the exchange interaction between the ferromagnetic films and the films of transition metals separated by a paramagnetic spacer was treated within the framework of the free-electron model. The model was applied to three-layer films, such as Fe|Cr|Fe or Co|Ru|Co, where the minority-spin (↓) energy bands are matched with the ones in the paramagnetic spacer, while the majority-spin (↑) electrons experience the repulsive potential (barrier with the height proportional to the exchange energy gap), caused due to insufficiency of the corresponding states in the spacer. It was shown within the framework of this model that the expansion of the coupling leads to an infinite series of terms corresponding to bilinear, biquadratic and higher-order couplings between magnetic moments in the ferromagnet. The coefficients corresponding to bilinear (A_{12}) and biquadratic (B_{12}) exchange oscillate with spacer width, so that in some ranges of spacer width it is possible that B_{12} becomes larger than A_{12}.

The above-mentioned experimental and theoretical results inspired us to analyse some characteristic properties of thin magnetic films which depend on the relation between bilinear and biquadratic exchange interaction. We shall study a simplified model with simple cubic structure in the nearest-neighbours approximation where both types of interactions between spins exist both between the layers and within the layers.

The structure of the paper is as follows: The model Hamiltonian of the system is defined in Section 2, together with some thermodynamic properties of the film in the mean-field (MF) approximation. The results of theoretical analysis are presented in Section 3, while numerical calculations are discussed in Section 4 together with final remarks.

2. Model and the mean-field approximation

We have mentioned above that we shall study the ferromagnetic film with N layers, with translational symmetry in XY-plane and z-axis perpendicular to the film. The interaction between magnetic moments is both bilinear Heisenberg exchange (I_{nm}) and biquadratic exchange ($K_{nm} = aI_{nm}$). We shall consider the simplest case where we consider only the nearest neighbours' interaction which will be set equal to I in the bulk and between the surface layers and the bulk. The influence of surfaces will be expressed by the variation of the surface interaction with respect to the bulk ($\varepsilon_1 = I_1/I$ and $\varepsilon_N = I_N/I$), which is definitely a simplification of the real situation, but our own aim is to study the influence of the biquadratic interaction on the system properties.

The Hamiltonian of the system can be written in the form [10,29]

$$H = -\frac{1}{2}\sum_{ij} I_{ij}\boldsymbol{S}_i\boldsymbol{S}_j - \frac{a}{2}\sum_{ij} I_{ij}(\boldsymbol{S}_i\boldsymbol{S}_j)^2 - g\mu_B\mathscr{H}\sum_i S_i^z \tag{1}$$

where we have introduced also the Zeeman term of the interaction with the external field along quantization axis (z-axis), which, in fact, introduces "Ising-type" symmetry into the Hamiltonian. We shall introduce, besides the operators of the dipole magnetic moments $(\hat{S}_x, \hat{S}_y, \hat{S}_z)$ also the operators of the quadrupole moments [10]:

$$\hat{Q}_{\boldsymbol{n}}^0 = 3(\hat{S}_{\boldsymbol{n}}^z)^2 - S(S+1), \quad \hat{Q}_{\boldsymbol{n}}^2 = (\hat{S}_{\boldsymbol{n}}^x)^2 - (\hat{S}_{\boldsymbol{n}}^y)^2, \quad \hat{Q}_{\boldsymbol{n}}^{\alpha,\beta} = \hat{S}_{\boldsymbol{n}}^\alpha\hat{S}_{\boldsymbol{n}}^\beta + \hat{S}_{\boldsymbol{n}}^\beta\hat{S}_{\boldsymbol{n}}^\alpha$$
$$(\alpha \neq \beta = x, y, z) \tag{2}$$

and in this way the Hamiltonian takes the form convenient for the analysis in the mean-field approximation:

$$\hat{H} = -\frac{1}{2}\sum_{ij} I_{ij}\left(1-\frac{a}{2}\right)\boldsymbol{S}_i\boldsymbol{S}_j - \frac{a}{2}\sum_{ij} I_{ij}\left(\frac{1}{6}\hat{Q}_i^0\hat{Q}_j^0 + \frac{1}{2}\hat{Q}_i^2\hat{Q}_j^2 + \frac{1}{2}\sum_{\alpha\neq\beta}\hat{Q}_i^{\alpha\beta}\hat{Q}_j^{\alpha\beta}\right)$$
$$-g\mu_B\mathscr{H}\sum_i \hat{S}_i^z\,. \tag{3}$$

The mean-field Hamiltonian will be introduced in the standard manner ($\hat{A} = \langle\hat{A}\rangle_{MF} + (\hat{A} - \langle\hat{A}\rangle_{MF}) = \langle\hat{A}\rangle_{MF} + \delta\hat{A}$ and neglecting the terms of the form $(\delta\hat{A})^2$), having in mind that due to the Ising symmetry and translational symmetry in the XY-plane ($\boldsymbol{i} = (\boldsymbol{n}, z = na_o)$) : $\langle S_i^x\rangle = \langle S_i^y\rangle = 0$, $\langle Q_i^z\rangle = \langle Q_i^{\alpha,\beta}\rangle = 0$, $\langle S_i^z\rangle = \sigma_n, \langle Q_i^0\rangle = q_n$ ($n = 1, 2, \ldots, N$)

$$\hat{H}_{MF} = \hat{H}_0 - \tfrac{1}{2}\langle\hat{H}_0\rangle\,, \tag{4}$$

where

$$\hat{H}_0 = -N_2\sum_{n=1}^{N}(\bar{\mathscr{H}}_n\hat{S}_n^z + \bar{\mathscr{K}}_n\hat{Q}_n^0)\,. \tag{5}$$

N_2 represents the number of the sites in XY-plane and the average fields $\bar{\mathscr{H}}_n$ and $\bar{\mathscr{K}}_n$, for free-boundary conditions, have the form

$$n = 1:$$

$$\bar{\mathscr{H}}_1 = \left(1-\frac{a}{2}\right)(4I_1\sigma_1 + I\sigma_2) + g\mu_B\mathscr{H}\,,$$

$$\bar{\mathscr{K}}_1 = \frac{a}{6}(4I_1q_1 + Iq_2)\,.$$

$$n = N:$$

$$\bar{\mathscr{H}}_N = \left(1-\frac{a}{2}\right)(4I_N\sigma_N + I\sigma_{N-1}) + g\mu_B\mathscr{H}\,,$$

$$\bar{\mathscr{K}}_N = \frac{a}{6}(4I_Nq_N + Iq_{N-1})\,.$$

$2 \leqslant n \leqslant N-1$:

$$\bar{\mathscr{H}}_n = \left(1-\frac{a}{2}\right) I(4\sigma_n + \sigma_{n+1} + \sigma_{n-1}) + g\mu_B \mathscr{H} ,$$

$$\bar{\mathscr{K}}_n = \frac{a}{6} I(4q_n + q_{n+1} + q_{n-1}) . \tag{6}$$

The constant term in Eq. (4) does not influence the eigenvalues of the Hamiltonian, of course, but it is the part of the free energy (F) and in this way influences the self-consistent magnetization of the system, which depends both on the average dipole (σ_n) and average quadrupole (q_n) moments.

We shall discuss further on the particular case of the spin $S=1$ since, in this case, in the bulk system (with Ising symmetry) there arise two types of ordering [10]:

(a) ferromagnetic one, for $0 < a < 1$, characterized by the parameters $\sigma_n \neq 0$ and $q_n \neq 0$ for $0 \leqslant T \leqslant T_c$ and

(b) ferroquadrupolar, for $a > 1$, characterized by the parameters $\sigma_n = 0$ and $q_n \neq 0$ for $0 \leqslant T \leqslant T_c$.

We shall see later that even in thin films with Ising symmetry, there also appear two types of ordering.

The free energy of the system in MFA can be found from the partition sum Z of the Hamiltonian $\hat{H}_0(\Theta = k_B T)$:

$$F = -\Theta \ln Z_0 - \tfrac{1}{2}\langle H_0 \rangle , \tag{7}$$

where

$$Z_0 = \left(\prod_{i=1}^{N} \mathscr{Z}_i\right)^{N_2} . \tag{8}$$

Simple calculation leads to the free energy per site in the layer:

$$\mathscr{F} \equiv \frac{F}{N_2} = -\Theta \sum_{n=1}^{N} \left\{ \frac{\bar{\mathscr{K}}_n}{\Theta} + \ln[\mathrm{e}^{-3\bar{\mathscr{K}}_n/\Theta} + 2\cosh(\bar{\mathscr{H}}_n/\Theta)] \right\} + \frac{1}{2}\sum_{n=1}^{N} (\sigma_n \bar{\mathscr{H}}_n + q_n \bar{\mathscr{K}}_n) \tag{9}$$

and the ground-state energy ($T = 0$ K):

$$\mathscr{E}_0 \equiv \frac{E_0}{N_2} = -\frac{1}{2}\sum_{n=1}^{N} [\sigma_n(0)\bar{\mathscr{H}}_n(0) + q_n(0)\bar{\mathscr{K}}_n(0)] . \tag{10}$$

In the case of the ferromagnetic (FM) ground state ($S=1$), $\sigma_n(0) = 1$ and $q_n(0) = 1$, it follows from Eq. (10) (for $\mathscr{H} = 0$)

$$\mathscr{E}_0^{FM} = -\frac{1}{2}\left(1-\frac{a}{3}\right) [4I_1 + 4I_N + I(6N-10)] . \tag{11}$$

For ferroquadrupolar (FQ) ground state $\sigma_n = \langle S_n^z \rangle = 0$; $\langle (S_n^z)^2 \rangle = 0$ and $q_n = -2$, we obtain

$$\mathscr{E}_0^{FQ} = -\frac{a}{3}[4I_1 + 4I_N + I(6N-10)] . \tag{12}$$

From Eqs. (11) and (12), it follows that at $T = 0$ for $0 \leqslant a < 1$, the system is in the ferromagnetic state, while for $a > 1$ it is in the ferroquadrupolar state. For $a = 1$ there occurs a degeneration, since both states possess the same energy, which agrees with the results for bulk [10].

At temperatures $T \neq 0$, the average dipole $\sigma_n(T)$ and quadrupole $q_n(T)$ moment per layer is obtained from the condition of minimal free energy:

$$\frac{\partial F}{\partial \sigma_n} = 0 \quad \frac{\partial F}{\partial q_n} = 0 ,$$

giving

$$\sigma_n = \frac{2\sinh(\bar{\mathscr{H}}_n/\Theta)}{e^{-3\bar{\mathscr{K}}_n/\Theta} + 2\cosh(\bar{\mathscr{H}}_n/\Theta)} , \tag{13}$$

$$q_n = \frac{6\cosh(\bar{\mathscr{H}}_n/\Theta)}{e^{-3\bar{\mathscr{K}}_n/\Theta} + 2\cosh(\bar{\mathscr{H}}_n/\Theta)} - 2 = 3\sigma_n \coth(\bar{\mathscr{H}}_n/\Theta) - 2 \tag{14}$$

for $n = 1, 2, \ldots, N$.

The above expressions turn into the bulk relations for σ and q [10], if we set the following values for the average fields

$$\mathscr{H}_n = \bar{\mathscr{H}}_n \left(1 - \frac{a}{2}\right) J(0)\sigma; \quad \bar{\mathscr{K}}_n = \frac{a}{6} J(0) q; \quad J(0) = 6I .$$

The complex form of Eqs. (13) and (14) clearly indicates that they can be solved only numerically in the whole range of temperatures $(0 \leqslant T \leqslant T_c)$, which shall be done in the next section. An analytical solution can be obtained only by expansion in the vicinity of a phase transition. In this way, we shall be able to deduce for which values of a there appear various types of ordering and a tricritical point in the film. An alternative procedure would be to diagonalize first the quadratic part of the free energy in the vicinity of a phase transition, yet due to the existence of biquadratic interaction, this seem to be a rather formidable task. We shall now study the phase transitions in the system, first analytically, then numerically.

3. Phase transitions in the system

Let us now return to the case of N-layered film. First we linearize the Eqs. (13) and (14) in terms of σ_n, which enables us to find the possible values of critical temperatures.

The linearization of Eq. (13) in terms of σ_n ($q_n \simeq \sigma_n^2$) in the vicinity of the phase-transition temperature ($\sigma_n \simeq 0$, $q_n \simeq 0$, $\Theta \leqslant \Theta_C$) leads to the following system (for $\mathscr{H} = 0$):

$$\hat{D}_N \boldsymbol{\sigma} = 0\,, \tag{15}$$

where $\hat{D}_N = \beta \hat{I}_N - \hat{A}_N$,

$$\hat{A}_N = \begin{pmatrix} 4\varepsilon_1 & 1 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 \\ 1 & 4 & 1 & 0 & \cdots & 0 & 0 & 0 & 0 \\ 0 & 1 & 4 & 1 & \cdots & 0 & 0 & 0 & 0 \\ . & . & . & . & \ddots & . & . & . & . \\ 0 & 0 & 0 & 0 & \cdots & 1 & 4 & 1 & 0 \\ 0 & 0 & 0 & 0 & \cdots & 0 & 1 & 4 & 1 \\ 0 & 0 & 0 & 0 & \cdots & 0 & 0 & 1 & 4\varepsilon_N \end{pmatrix}, \quad \boldsymbol{\sigma} = \begin{pmatrix} \sigma_1 \\ \sigma_2 \\ . \\ . \\ . \\ \sigma_N \end{pmatrix}. \tag{16}$$

$\hat{I}_N$ is a unit $(N \times N)$ matrix and $\beta = 3\Theta/I(2-a)$.

If the system is to possess spontaneous magnetization ($\sigma_n \neq 0$ even for vanishing external field for $\Theta \leqslant \Theta_C$), the determinant $D_N \equiv \det|\hat{D}_N|$ must vanish, the condition which provides us with the possible values of phase-transition temperatures:

$$\Theta_C^{\nu} = \beta_\nu \frac{(2-a)I}{3} \quad \nu = 1,2,\ldots,N\,, \tag{17}$$

where β_ν are roots of the equation $D_N = 0$:

$$D_N(\rho) = T_N(\rho) + (\gamma_1 + \gamma_N)T_{N-1}(\rho) + \gamma_1\gamma_N T_{N-2}(\rho)\,. \tag{18}$$

The above polynomial of Nth order can be expressed in terms of either β or $\rho = \beta - 4$; $\gamma_1 = 4(1-\varepsilon_1)$, $\gamma_N = 4(1-\varepsilon_N)$ where $T_N(\rho)$ is Chebyshev's polynomial of the second kind:

$$T_N(\rho) = \begin{vmatrix} \rho & -1 & 0 & 0 & \cdots & 0 & 0 & 0 & 0 \\ -1 & \rho & -1 & 0 & \cdots & 0 & 0 & 0 & 0 \\ 0 & -1 & \rho & -1 & \cdots & 0 & 0 & 0 & 0 \\ . & . & . & . & \ddots & . & . & . & . \\ 0 & 0 & 0 & 0 & \cdots & -1 & \rho & -1 & 0 \\ 0 & 0 & 0 & 0 & \cdots & 0 & -1 & \rho & -1 \\ 0 & 0 & 0 & 0 & \cdots & 0 & 0 & -1 & \rho \end{vmatrix}. \tag{19}$$

It is important to stress that according to the definition of D_N, β_ν are the eigenvalues of the matrix $\hat{A}_N$ so they depend solely on the parameters ε_1 and ε_N since they are the only system parameters involved in the definition of the matrix $\hat{A}_N$. Exact analytical solution can be found only for $N = 2$ and $N = 3$, and some particular sets of parameters for higher N, but here we shall present some general properties of the solutions.

According to Gershgorin's theorem [30], the number of positive eigenvalues depends on the matrix elements and in our particular case, for $\varepsilon_1 > 0.25$ and $\varepsilon_N > 0.25$, all eigenvalues β_ν are positive. For smaller values of ε_1 and ε_N, there can arise for $N \geqslant 3$

at most two negative roots, while for $N = 2$, one root is always positive and the other is negative.

To each root β_ν there corresponds one ordered phase $\Phi^{(\nu)}$ with its set of $2N$ parameters $(\sigma_1^\nu, \sigma_2^\nu, \ldots, \sigma_N^\nu)$ and $(q_1^\nu, q_2^\nu, \ldots, q_N^\nu)$. The most stable phase (in fact, the single stable phase for $0 \leqslant \Theta \leqslant \Theta_C$) is the phase corresponding to the maximal temperature Θ_C which we denote by β_1 (or Θ_C^1). We are going to demonstrate this at the end of this section after presenting the simplified expression for the free energy. However, it is important to stress that according to Perron's theorem [30] which deals with positive matrices (all matrix elements positive) which is our case, there exists always one positive root larger than the modulus of all others, to which there corresponds positive eigenvector (all components positive). This means that all σ_n corresponding to this phase are positive, so it corresponds to ferromagnetic ordering in each of the layers. Due to hermicity of the matrix and the orthogonality of eigenvectors, it follows that phases corresponding to other eigenvectors do not manifest ferromagnetic ordering in all layers.

One can easily deduce the following relations from Eq. (15) (in the vicinity of each temperature $\Theta \simeq \Theta_C^\nu$):

$$S_n = \beta\sigma_n \,, \tag{20}$$

where we have introduced the following notation:

$$\begin{aligned} &S_n = 4\sigma_n + \sigma_{n-1} + \sigma_{n-1}, \quad Q_n = 4q_n + q_{n-1} + q_{n-1}, \quad n \neq 1, N \,, \\ &S_1 = 4\varepsilon_1\sigma_1 + \sigma_2, \quad Q_1 = 4\varepsilon_1 q_1 + q_2 \,, \\ &S_N = 4\varepsilon_N\sigma_N + \sigma_{N-1}, \quad Q_N = 4\varepsilon_N q_N + q_{N-1} \,. \end{aligned} \tag{21}$$

In order to determine the possible types of ordering (ferromagnetic or ferroquadrupolar) and possible types of transitions (first or second order), we must expand Eqs. (13) and (14) up to terms of order σ^2 using the Eq. (20). The first step is to express Q_n in terms of σ_n^2. It follows from Eq. (14) that

$$q_n \approx \frac{1}{\alpha\beta} Q_n + \frac{3}{4\beta^2} S_n^2 \,, \tag{22}$$

where $\alpha = \dfrac{2-a}{a}$.

Using the relations between Q_n and q_n and Eq. (22), we shall obtain the systems of equations which express Q_n in terms of S_n^2 (i.e. $(\beta\sigma_n)^2$) and which can be put into the matrix form

$$\hat{\Delta}_N(x)\boldsymbol{Q} = \frac{3x}{4}\hat{A}_N\boldsymbol{\sigma}^2 \,, \tag{23}$$

where

$$\hat{\Delta}_N(x) = x\hat{I}_N - \hat{A}_N \, ; \tag{24}$$

$$\boldsymbol{\sigma}^2 = \begin{pmatrix} \sigma_1^2 \\ \sigma_2^2 \\ \vdots \\ \sigma_N^2 \end{pmatrix}; \qquad \boldsymbol{Q} = \begin{pmatrix} Q_1 \\ Q_2 \\ \vdots \\ Q_N \end{pmatrix}; \qquad x \equiv \alpha\beta \, . \tag{25}$$

An important comment is necessary here. The matrices $\Delta_N(x)$ and $D_N(\beta)$ have the same form, so that their determinants have similar form:

$$D_N = \prod_\nu (\beta - \beta_\nu); \qquad \Delta_N = \prod_\nu (x - \beta_\nu) \, ,$$

where β_ν are the solutions of $D_N = 0$.

The relation (23) enables us to conclude when the system transits to the ferro-quadrupolar ordering. Obviously, when Δ_N vanishes, σ must vanish too, otherwise, Q would diverge.

Expanding Eq. (13) up to order σ_n^3 and using Eqs. (17) and (22), one gets

$$\frac{9}{8}\sigma_n^2 - \frac{3}{2x}Q_n = 3\left(\frac{\beta_\nu}{\beta} - 1\right) , \tag{26}$$

or in the matrix form

$$\frac{3}{4}\hat{I}_N\boldsymbol{\sigma}^2 - \frac{1}{x}I_N\boldsymbol{Q} = 2\left(\frac{\beta_\nu}{\beta} - 1\right)\mathbf{1}_N \, , \tag{27}$$

where we distinguish the unit matrix $\hat{I}_N$ and the column matrix $\mathbf{1}_N$ with all elements equal to 1. From (23) it follows that

$$\boldsymbol{Q} = \frac{3x}{4}\hat{\Delta}_N^{-1}\hat{A}_N\boldsymbol{\sigma}^2 \tag{28}$$

Combining (27) and (28), we obtain

$$\hat{\Delta}_0^{(N)}\boldsymbol{\sigma}^2 = \frac{8}{3\beta}(\beta_\nu - \beta)\hat{\Delta}_N\mathbf{1}_N \, , \tag{29}$$

where we have introduced

$$\hat{\Delta}_0^{(N)} = \Delta_N - \hat{A}_N = x\hat{I}_N - 2A_N \, . \tag{30}$$

The condition for the appearance of the tricritical point is

$$\det|\hat{\Delta}_0^{(N)}| \equiv \Delta_0^{(N)} = 0 \, , \tag{31}$$

since then one cannot determine $\sigma_n^2 = f(\Theta_C - \Theta)$.

In the most general case, since the equation $\hat{\Delta}_N(x) = 0$ has the solutions $x_\nu = \beta_\nu$ $(\nu = 1, 2, \ldots, N)$ i.e. if

$$\Delta_N(\beta_\nu) = |\beta_\nu \hat{I}_N - \hat{A}_N| = 0\,, \tag{32}$$

then also $\Delta_0^{(N)}(x = 2\beta_\nu) = 0$. It can be easily proved, since

$$\Delta_0^{(N)}(2\beta_\nu) = |2\beta_\nu \hat{I}_N - 2\hat{A}_N| = 2^N |\beta_\nu \hat{I}_N - \hat{A}_N| = 2^N \Delta_N(\beta_\nu) = 0\,. \tag{33}$$

This means that the relation

$$\Delta_N(x) = \prod_{\nu=1}^{N}(x - \beta_\nu)$$

implies the relation

$$\Delta_0^{(N)}(x) = \prod_{\nu=1}^{N}(x - 2\beta_\nu)\,.$$

Formally, one can conclude that for any phase Φ^μ there appear N tricritical points in the (β, α) plane: $(\beta_\mu, \alpha_\mu^\nu \equiv 2\beta_\nu/\beta_\mu)$. If we introduce the notation $\beta_1 > \beta_2 > \cdots$ where $\beta_1 = (3\Theta_C^1/(2-a)I)$ corresponds to the transition point of the stable phase (defined in the range $0 \leqslant \Theta < \Theta_C^1$), then there appears maximal value of the parameter α : $\alpha_\mu^1 = 2\beta_1/\beta_\mu$ leading to the highest value for the tricritical point of the given phase. This means, that during the decrease of α, the change of the character of the transition occurs at this point, so all lower values of α are irrelevant. Since we are mostly interested in the stable phase corresponding to β_1, we see that tricritical point occurs for $\alpha = 2$.

Similar reasoning can be repeated for the transition to ferroquadrupolar ordering $(\Delta_N(x) = 0)$, leading to the conclusion that it occurs for $\alpha = 1$ in the stable phase.

Let us finally analyze the free energy for any phase Φ_ν near the critical temperature β_ν. Expanding the free energy in the vicinity of Θ_C^ν in terms of $\{\sigma_n\}$ up to order σ^4,

$$\mathscr{F}_N = \mathscr{F}_N(0) + \sum_{n=1}^{N}\left[\left(\frac{\sigma_n}{2} - \frac{(2-a)I}{6\Theta}S_n\right)\frac{(2-a)I}{2}S_n + \left(\frac{q_n}{2} - \frac{aI}{6\Theta}Q_n\right)\frac{aI}{6}Q_n \right.$$
$$\left. + \frac{(2-a)^4 I^4}{36\cdot 2^4\Theta^3}S_n^4 - \frac{1}{3\Theta^2}\frac{Ia}{6}Q_n S_n^2 + \mathrm{O}(\sigma^6)\right] \tag{34}$$

and using

$$q_n - \frac{Ia}{3\Theta}Q_n \approx \frac{(2-a)^2 I^2}{12\Theta^2}S_n^2 = \frac{3}{4\beta^2}S_n^2, \quad S_n \simeq \beta_\nu \sigma_N,$$

we have

$$\mathscr{F}_N = \mathscr{F}_N(0) + \sum_{n=1}^{N}\left[\frac{(2-a)^2 I^2}{12\Theta}(\beta - \beta_\nu)\beta_\nu\sigma_n^2 - \frac{aI}{4^2}Q_n\sigma_n^2 + \frac{3(2-a)I\beta}{4\cdot 4^2}\sigma_n^4\right]$$
$$= \mathscr{F}_N(0) + \sum_{n=1}^{N}\left[\frac{(2-a)I}{4}(\beta - \beta_\nu)\sigma_n^2 + \frac{aI}{4^2}\sigma_n^2\left(\frac{3\alpha\beta}{4}\sigma_n^2 - Q_n\right)\right]\,. \tag{35}$$

Finally, using Eq. (26), the total free energy in the vicinity of $\Theta \simeq \Theta_C^{\nu}$, for the phase Φ^{ν} is

$$\mathscr{F}_N = \mathscr{F}_N(0) - \frac{(2-a)I}{8}(\beta_\nu - \beta)\sum_{n=1}^{N}\sigma_n^2 < \mathscr{F}_N(0). \tag{36}$$

Let us now discuss this expression in more detail. It is obvious that the free energy corresponding to the phase with highest transition point (Θ_C^1) is the lowest till temperatures $\Theta \leqslant \Theta_C^1$. It is rather difficult to compare this free energy with the one corresponding to some phase with lower transition point ($\Theta_C^{\nu} < \Theta_C^1$), yet all numerical studies indicate that it is lower, so this is the reason why we assume this phase to be the only stable one.

Let us now look at some numerical examples.

4. Numerical study of the phase transitions

The numerical study was based on the numerical solving of the coupled system of nonlinear equations, Eqs. (13) and (14) under the condition of minimal free energy. The results agree with our analytical study in the sense that main conclusions concerning the character of ordered phases and transition points are confirmed.

Our main interest was to study numerically the influence of the boundaries and the change of order parameters σ_n and q_n along the layers, since these are difficult to obtain analytically.

To be more specific, the values of order parameters σ_n and q_n depend on the layer index ($n = 1, 2, \ldots, N$), the boundary conditions ($\varepsilon_1, \varepsilon_N$), the parameter of the biquadratic exchange (a) and the temperature, here expressed in terms of the dimensionless temperature $t = k_B T/I$. We are going to present some of the plots obtained for selected parameters.

Fig. 1 shows the values of the magnetization and quadrupole moment over the layers of the symmetric film ($N = 10$), in different phases and for different values of the surface parameters at the temperature $t = 2.5$. Fig. 1a describes the ferromagnetic phase ($a = \frac{1}{4}$) when $\sigma_n > q_n > 0$, and the Fig. 1b treats the ferroquadrupolar phase ($a = \frac{5}{4}$) when $\sigma_n = 0$; $q_n < 0$. In both phases, the values of the order parameters are smaller at the film surfaces if $\varepsilon < \frac{5}{4}$. These differences manifest themselves in the first 3–4 surface layers. One should note that for $\varepsilon = \frac{5}{4}$ order parameters obtain the same value at all film layers. This is simply the consequence of the fact that in this case the total exchange interaction in surface layers $4\varepsilon I + I$ has the same value $6I$ as in the bulk. Finally, for $\varepsilon > \frac{5}{4}$, order parameters have larger values at the surfaces than in the bulk.

A more detailed analysis of the influence of the surface parameters (Fig. 2) shows that their decrease can cause a drastic reduction of the order parameter value at the film surfaces. For example, for $\varepsilon = 0.1$, the magnetization in the surface layer is about 10% of the bulk value, while for the quadrupole moment it is 30%. In inner layers

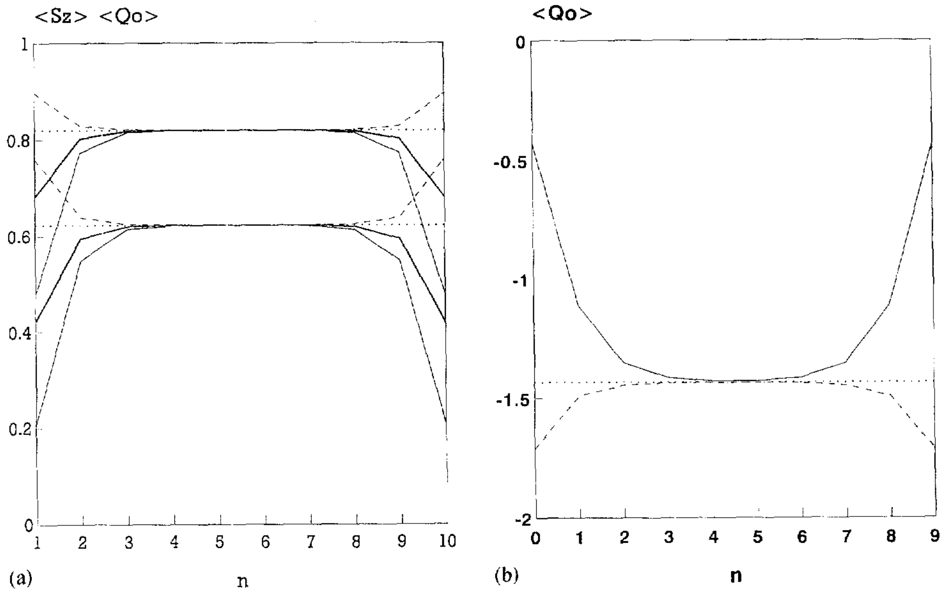


Fig. 1. The magnetization $\langle S^z \rangle$ (upper plots) and average quadrupole moment $\langle Q^0 \rangle$ (lower plots) in various layers of symmetric film at the temperature $k_B T/I = 2.5$, for several values of $\varepsilon : \varepsilon = \frac{3}{4}$ thin solid line; $\varepsilon = 1$ solid line; $\varepsilon = \frac{5}{4}$ dotted line and $\varepsilon = \frac{3}{2}$ dashed line. (a) Ferromagnetic phase $a = 0.25$; (b) ferroquadrupolar phase, $a = 1.25$.

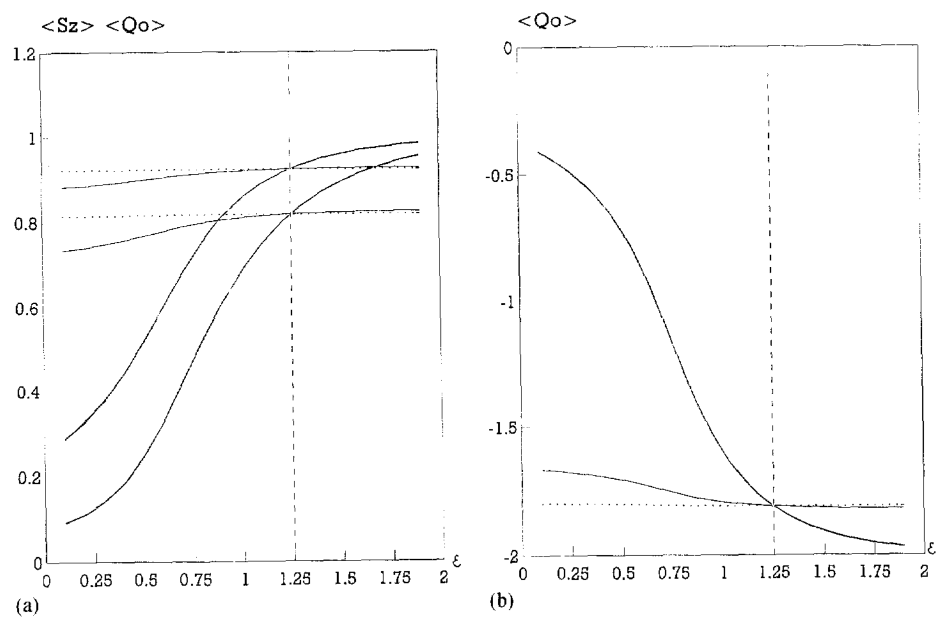


Fig. 2. The magnetization $\langle S^z \rangle$ (upper plots) and average quadrupole moment $\langle Q^0 \rangle$ (lower plots) in various layers of symmetric film versus surface parameter ε for $k_B T/I = 2 : n = 1$-solid line; $n = 2$-thin solid line and $n = 3$-dotted line. (a) Ferromagnetic phase, $a = 0.25$; (b) ferroquadrupolar phase, $a = 1.25$.

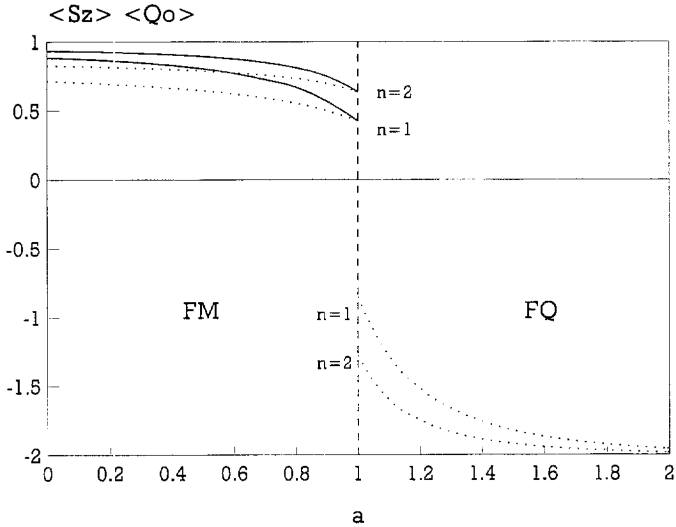
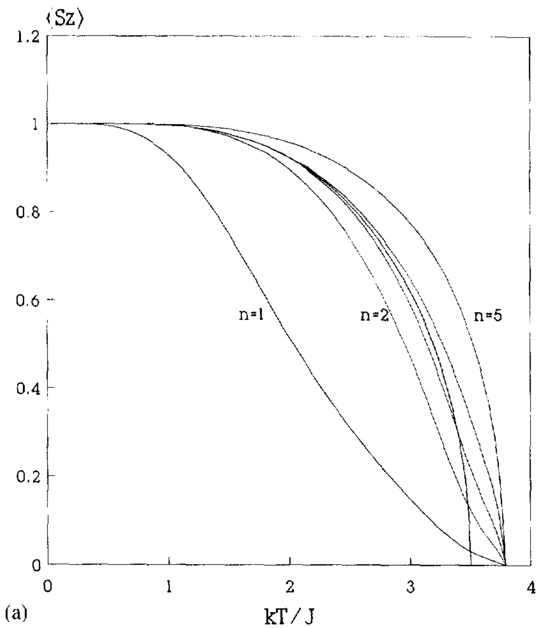


Fig. 3. The magnetization $\langle S^z \rangle$ (solid line) and average quadrupole moment $\langle Q^0 \rangle$ (dotted line) for the first two layers ($n = 1,2$) versus biquadratic exchange parameter a, for $N = 10$ layer film with free surfaces ($\varepsilon = 1$) at the temperature $k_B T/I = 2$.

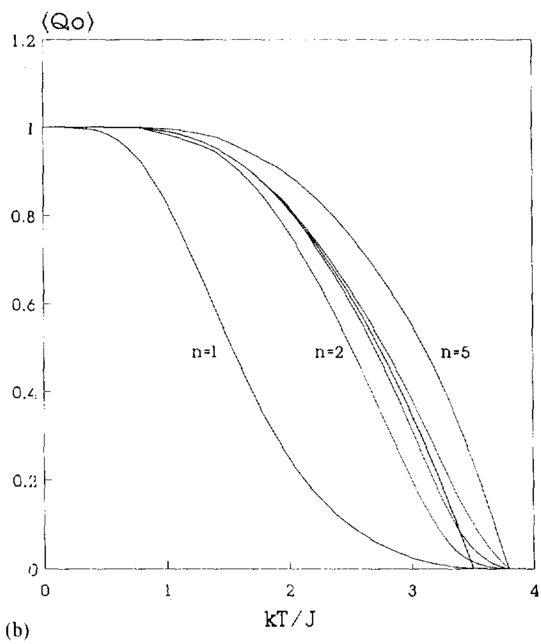
this influence is decreased and becomes negligeable after the third layer. The plots all intersect for $\varepsilon = 1.25$, since in this case the order parameters are the same in all layers. Further increase of the surface parameter ($\varepsilon > \frac{5}{4}$) has not so pronounced influence on order parameters.

Using the mean-field Eqs. (13) and (14), we have also studied the influence of the biquadratic exchange parameter. The results of this analysis for ten-layer film ($N = 10$) with free surfaces ($\varepsilon_1 = \varepsilon_N = 1$) at the temperature $t = 2$ are shown at the Fig. 3. One can see from the plot that the increase of the biquadratic interaction (a) in the ferromagnetic phase leads to the decrease of the order parameters. There arise two degenerate sets of solutions for $a = 1$ just as the case of bulk ferromagnetic with biquadratic interaction. For one set, order parameters have the same values at particular film layers ($\sigma_n = q_n$), while for the other one $\sigma_n = 0$, $q_n < 0$ at each layer. Both sets lead to the same free energy and coexist at all temperatures below the critical one. For $a > 1$, the system transits to the ferroquadrupolar phase where the increase of biquadratic exchange leads to a sudden decrease and then a saturation of the average quadrupolar moments.

Using Eqs. (13) and (14) we have finally studied the temperature dependence of the order parameters for various film layers. This dependence is for the sake of simplicity plotted for the case of five-layered film with asymmetric boundary conditions ($\varepsilon_1 = 0.5$, $\varepsilon_N = 1.5$) in the ferromagnetic phase ($a = 0.25$). It can be seen from the Fig. 4 that the order parameters in various layers might have the values smaller or higher than the bulk ones, depending exclusively on the boundary conditions. Another important



(a)



(b)

Fig. 4. The temperature dependence of the magnetization (a) and quadrupole moment (b) in the ferromagnetic phase ($a = 0.25$, $N = 5$) of the asymmetric film ($\varepsilon_1 = 0.5$, $\varepsilon_N = 1.5$). Solid line represents bulk quantities.

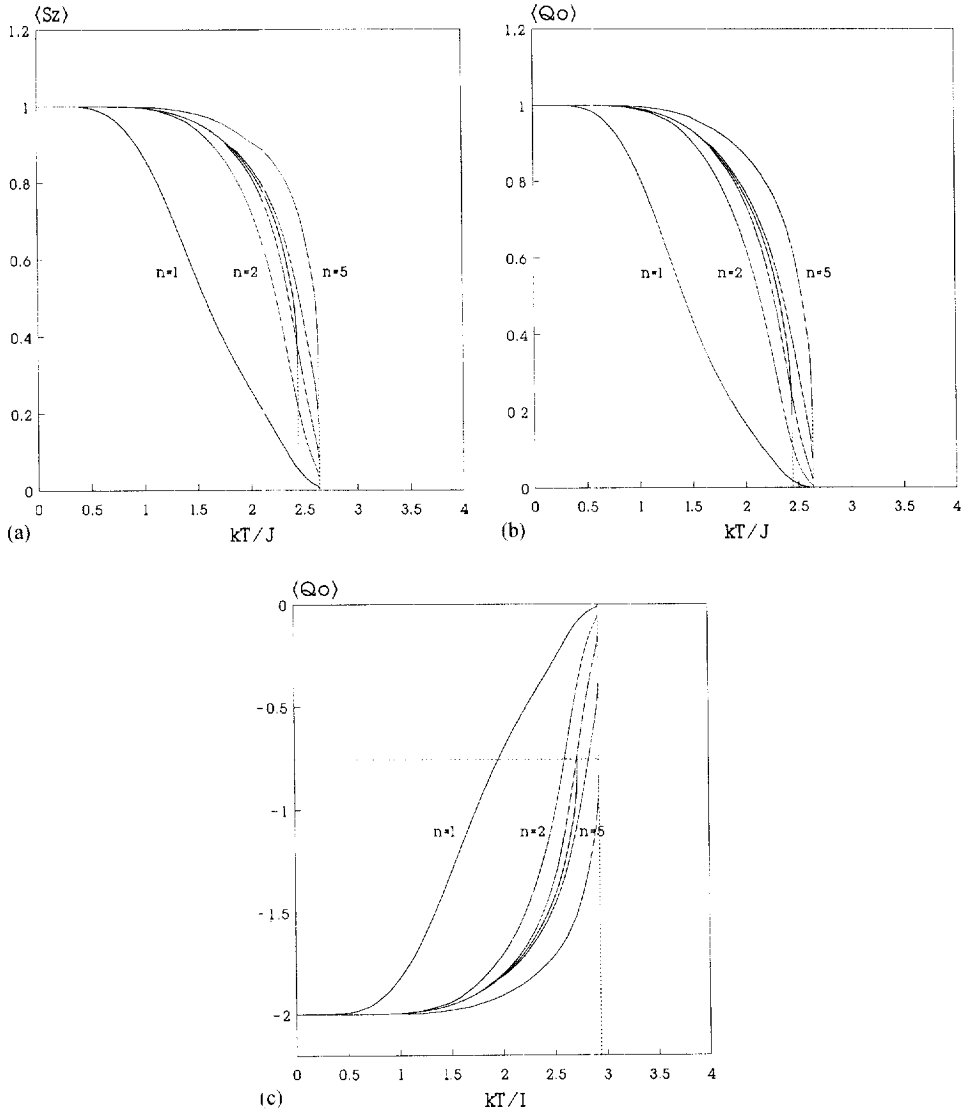


Fig. 5. The temperature dependence of the magnetization (a) and quadrupole moment (b) in the ferromagnetic phase ($a = 0.8$, $N = 5$) of the asymmetric film ($\varepsilon_1 = 0.5$, $\varepsilon_N = 1.5$), (c) ferroquadrupolar phase ($a = 1.25$).

conclusion is that opposite to the case of semi-infinite structure, there occurs a unique phase-transition temperature in the thin films. The values of transition temperatures agree with the analytical estimates (Eq. (17)).

Fig. 5 represents the temperature dependence of the magnetization and quadrupole moment under the same conditions except for $a = 0.8$. The influence of the surface parameters is similar as in the previous example. However, in this case the order parameters vanish abruptly in the vicinity of the transition temperature, indicating to

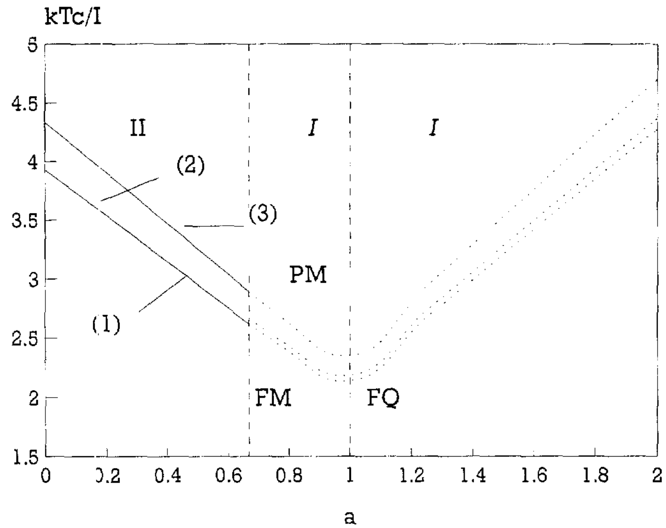


Fig. 6. Phase diagram of the $N = 10$ layer symmetric system, $1 - \varepsilon = 0.25$, $2 -$ bulk, $3 - \varepsilon = 1.5$.

the change of the character of phase transitions for $a > \frac{2}{3}$. This phenomenon was already discussed analytically in the previous section.

First-order transitions also occur in the ferroquadrupolar phase, as can be seen from Fig. 5c, where the temperature dependence of the quadrupolar moment over the film layers is presented for $a = 1.25$. All other conditions are the same as for the previous two figures.

All conclusions are summarised by the phase diagram (Fig. 6) where kT_C/I is plotted versus biquadratic parameter a, so that the tricritical point corresponds to $a = \frac{2}{3}$ and transition to ferroquadrupolar ordering corresponds to $a = 1$.

Finally, let us note that the expansion of Eq. (36) is meaningfull only for $\alpha > 2$, i.e. when second-order transition occurs. For the analytical study of first-order transition, an expansion of the free energy to higher orders of σ, would be necessary. We have avoided this by studying this region only numerically.

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