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# Spatial distribution of Gaussian fluctuations of the molecular field and magnetization in the pyramid-like Ising nanoscopic system interacting with the substrate

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## ABSTRACT

We study thermodynamic properties of an Ising model of a ferromagnetic nanoscopic pyramid deposited onto a ferromagnetic bulk substrate. The influence of the interaction between the pyramid and the substrate is calculated in terms of the equilibrium reduced-state (density) operator used for description of thermodynamic properties of nanoscopic systems. The spatial distribution of the fluctuations of molecular field and magnetization in the nanoscopic pyramid is obtained in the Gaussian fluctuations approximation. Experimental consequences for the magnetic force measurements are briefly discussed.

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## 1. Introduction

During the past two decades we have witnessed significant advances in the ability to synthesize nanoscale structures as well as development of novel experimental method allowing exploration of their physical properties [1]. This is exciting for two reasons. Firstly, new forms of matter with no counterpart in nature and revealing unique physical properties have been fabricated. Secondly, we have now realized nanostructures that open new avenues for development of very small devices.

Nanosopic magnetic systems [2] have always been very attractive from the theoretical point of view. On the other hand, studies of nanoscopic systems are characterized by a close coupling between theory and experiment because of rapidly increasing number of experimental works on real nanoscopic materials [1]. These materials often correspond remarkably close to certain idealized spin models and are great challenges not only for physicists, but also for chemists and engineers.

There is a great interest in producing two-dimensional arrays of magnetic nanodots which may serve e.g. as magnetic recording media. Due to small dimensions of these particles quantum-mechanical effects cannot be ignored. Effects observed in quantum dots are relevant also in quantum computing and spin electronics. It was shown recently that nanoscopic structures in the form of

pyramids or similar shapes can be fabricated on a bulk substrate [3–5].

In experiments aiming at the investigation of nanoscopic systems we deal frequently with such systems deposited on a bulk substrate. However, most often in the theoretical description of such experiments the interaction between the nanosystem and the substrate is neglected. The main purpose of this work is to find a way to include this and calculate its effect on the thermodynamic properties of the Ising model of a ferromagnetic nanoscopic system. To achieve this we apply the equilibrium reduced-state (density) operator (ERSO) [6] and carry out the calculations within the Gaussian fluctuations approximation (GFA).

ERSO is the most general equilibrium state operator. It has been derived from the generalized Schrödinger variational principle and not on the quantum statistical mechanics, i.e. without statistical hypotheses. When the term describing the interaction between the system and its environment is neglected ERSO takes the form of the statistical operator of the Gibbs canonical distribution. ERSO applied to an exactly solvable microscopic model leads to exact results.

GFA is a modified version of the high density expansion method as has been proposed in Refs. [7,8]. GFA is an improvement over the molecular field approximation (MFA) due to the self-consistent inclusion of Gaussian fluctuations of this field. The essential new element of GFA is the summing up of the partial sums of Feynman diagrams of the same structure of recurrent formulae at each stage of the calculations. GFA is based on a classification of the Feynman diagrams in terms of  $1/z$ , where  $z$  is the effective number of spins

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interacting with any given spin. Owing to this procedure the theory becomes internally consistent and does not lead to unphysical results such as, for example, a complex Curie temperature [9].

## 2. The model

In this paper we consider magnetic properties of a nanoscopic pyramid (nanopyramid). They are well described by the simple model of localized and ordered spins with the following spin-1/2 Hamiltonian

$$H = -\frac{1}{2}I \sum S_{fl}^z S_{fl'}^z, \quad (1)$$

where  $I$  is the coupling parameter and  $\sum$  stands for summation over pairs of different simple cubic (sc) lattice points. Our considerations are restricted to nearest-neighbor interaction only.

In expression (1)  $\mathbf{f}$  denotes the two-dimensional position vectors of a spin belonging to a given monoatomic layer  $l = 1, 2, 3, 4$  of the pyramid. There are four spins in the  $l = 1$  layer, 16 spins in the  $l = 2$  layer, 36 spins in the  $l = 3$  layer and 64 spins in the  $l = 4$  layer. The total number of spins in the pyramid is 120 (see Fig. 1).

We assume that the nanoscopic pyramid is deposited on a bulk ferromagnetic substrate sufficiently well described by the spin-1/2 Ising model for a simple cubic lattice and the Hamiltonian

$$H_s = -\frac{1}{2}I_1 \sum S_{\mathbf{g}r}^z S_{\mathbf{g}'r'}^z. \quad (2)$$

Here  $\mathbf{g}$  denotes the two-dimensional position vectors of spin belonging to a given monoatomic layer. The summations always run over different sites. The substrate is divided into monoatomic layers parallel to the planes (100) of a simple cubic lattice. The position of each layer is given by the number  $r = 1, 2, \dots$

Let us assume that the interaction of the nanoscopic pyramid with the bulk substrate is described by the Heisenberg Hamiltonian of the form

$$H_l = -\frac{1}{2}I_2 \sum_{\mathbf{fg}} \mathbf{S}_{fl=4} \cdot \mathbf{S}_{\mathbf{g}r=1}. \quad (3)$$

In order to take into account the interaction of the nanoscopic pyramid with the bulk substrate we shall apply ERSO [6] suitable for description of a physical situation similar to the one we are concerned with. In the derivation of this particular form of ERSO we use the fact that although the Universe as a whole is in the pure state, its arbitrary multiparticle parts are inevitably in mixed states. This is purely a quantum effect following from the holistic properties of the quantum theory, formally related to the fact that the Universe (according to the quantum cosmology postulates) has one vector of state common for all systems and, in the case of interactions among them, we are not able to specify the vector of state for individual subsystem (it may not be the case for the interac-

tions of the effective field type or classical ones). Such a situation does not occur in the classical description because we may know classical trajectories of each particular molecule irrespective of their interactions. The holistic features of the quantum theory imply the use of the formalism of ERSO in description of multiparticle systems. For this reason the interaction (3) of the nanoscopic pyramid with the bulk substrate cannot be described by the Ising Hamiltonian.

In our case ERSO takes the following form [6]:

$$d = \exp[\beta(F - H - H'(\beta))], \quad (4)$$

where  $F$  is the free energy and

$$H'(\beta) = \text{Tr}_s[H_l(1 + K) \exp(\beta(F_s - H_s))] \quad (5)$$

is an effective term describing the interaction between the pyramid and the bulk substrate,  $\text{Tr}_s[\dots]$  is the partial trace over the substrate states,  $K$  is the correlation operator,  $\beta = (k_B T)^{-1}$ , and

$$F_s = -\frac{1}{\beta} \ln \text{Tr}_s[\exp(-\beta H_s)] \quad (6)$$

is the free energy of the substrate. Substituting expression (3) into Eq. (5) with the assumption that

$$|\langle S_{\mathbf{g}r}^x \rangle|, |\langle S_{\mathbf{g}r}^y \rangle| \ll |\langle S_{\mathbf{g}r}^z \rangle|, \quad I_2 \ll I_1, \quad (7)$$

and  $K \approx 0$ , we arrive at

$$H'(\beta) = -\frac{1}{2}I_2 \langle S_{r=1}^z \rangle \sum_{\mathbf{f}} S_{fl=4}^z, \quad (8)$$

where

$$\langle S_{r=1}^z \rangle = \text{Tr}_s[S_{\mathbf{g}r=1}^z \exp(\beta(F_s - H_s))] \quad (9)$$

is the average bulk substrate spin moment in the layer  $r = 1$ .

## 3. Gaussian fluctuation of the molecular field

As a starting point to GFA we choose the following decomposition of the Hamiltonian (1)

$$H = (H - H_1) + H_1 = H_0 + H_1, \quad (10)$$

where the perturbative part  $H_1$  is defined by the transformation

$$H \rightarrow H_1 = H(S_{fl}^z \rightarrow \delta S_{fl}^z), \quad (11)$$

where

$$\delta S_{fl}^z = S_{fl}^z - \langle S_{fl}^z \rangle \quad (12)$$

is the fluctuation operator of the z-component of the spin and

$$\langle S_{fl}^z \rangle = \text{Tr}[S_{fl}^z e^{\beta(F - H - H'(\beta))}], \quad \beta = (k_B T)^{-1} \quad (13)$$

and  $F$  is the free energy.

According to the rules of the thermodynamic perturbation expansion we can write

$$\langle S_{fl}^z \rangle = \langle S_{fl}^z e^{-\beta H_1} \rangle_0 / \langle e^{-\beta H_1} \rangle_0, \quad (14)$$

where

$$\langle \dots \rangle_0 = \text{Tr}[\dots d_0] \quad (15)$$

and

$$d_0 = \{\text{Tr}[\exp(-\beta(H_0 + H'(\beta)))]\}^{-1} \exp(-\beta(H_0 + H'(\beta))). \quad (16)$$

The right-hand side of expression (13) can be expanded into a series with respect to the perturbing term  $H_1$ . From such an infinite series we now choose a certain partial sum which can be represented graphically in the following way [10]

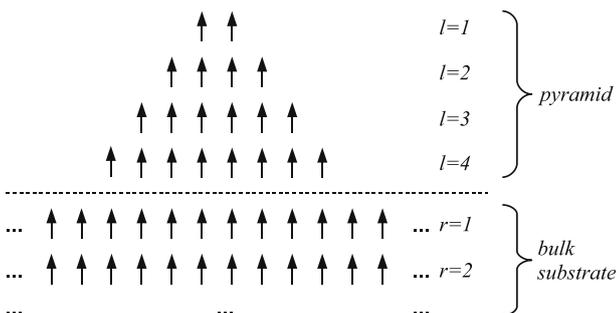


Fig. 1. Schematic diagram of ferromagnetic nanoscopic pyramid deposited on a ferromagnetic bulk substrate.

$$\langle S_{fl}^z \rangle = \boxed{\text{---}}, \quad (17)$$

where

$$\boxed{\text{---}} = \boxed{\text{---}} + \boxed{\text{---}} + \boxed{\text{---}} + \dots \quad (18)$$

and symbols --- denote the renormalized interaction line,

$$\text{---} = \sum_{f'l'} \left( \dots \boxed{\text{---}} \dots \right), \quad (19)$$

$$\underbrace{\boxed{\text{---}}}_{m} = \frac{d^m}{dW_{fl}^m} L(W_{fl}), \quad (20)$$

$$fl \dots f'l' = \beta l \sum_{f'l'}, \quad (21)$$

$$L(W_{fl}) = \ln(2 \cosh(W_{fl}/2)), \quad (22)$$

$$W_{fl} = \frac{\beta}{2} \left( l \sum_{f'l'} \langle S_{f'l'}^z \rangle + I_2 \delta_{l,4} \langle S_{r-1}^z \rangle \right), \quad (23)$$

where we assumed  $\hbar = 1$  and  $\sum_{f'l'}$  denotes summation over nearest-neighbors of the  $fl$  spin.

As a result of calculating the infinite sum (18) and (19) we obtain

$$\langle S_{fl}^z \rangle = \frac{1}{2\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} \tanh(W_{fl} + u\delta W_{fl}) du, \quad (24)$$

where

$$\delta W_{fl} = \frac{\beta}{2} \left( \frac{1}{2\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} \left[ l^2 \sum_{f'l'} (1 - \tanh^2(W_{f'l'} + u\delta W_{f'l})) + I_2^2 \delta_{l,4} (1 - \tanh^2(V_r + u\delta V_r)) \right] du \right)^{1/2}. \quad (25)$$

Similarly, using Eq. (9) for  $r = 1, 2, \dots$ , the molecular field  $V_r$  of the bulk substrate in the GFA satisfies the equation

$$V_r = \frac{\beta l_1}{2} \left( \sum_{\mathbf{g}} \langle S_{\mathbf{g}r}^z \rangle + \sum_{\mathbf{g}r'} \langle S_{\mathbf{g}r'}^z \rangle \right), \quad (26)$$

where

$$\langle S_r^z \rangle = \frac{1}{2\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} \tanh(V_r + u\delta V_r) du, \quad (27)$$

$$\delta V_r = \frac{\beta l_1}{2} \left( \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} [4(1 - \tanh^2(V_r + u\delta V_r)) + (1 - \delta_{r,1})(1 - \tanh^2(V_{r-1} + u\delta V_{r-1})) + (1 - \tanh^2(V_{r+1} + u\delta V_{r+1}))] du \right)^{1/2} \quad (28)$$

and  $\delta W_{fl}$ ,  $\delta V_r$  are the mean Gaussian fluctuations of the molecular fields  $W_{fl}$  and  $V_r$ , respectively.

After introducing the following reduced magnitudes

$$X_{fl} = 2\langle S_{fl}^z \rangle, y_r = 2\langle S_r^z \rangle, t = \frac{4}{\beta l}, a = \frac{I_2}{l}, b = \frac{l_1}{l}, \quad (29)$$

Eqs. (24)–(26) can be written in a compact form,

$$X_{fl} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} \tanh(W_{fl} + u\delta W_{fl}) du, \quad (30)$$

where

$$W_{fl} = \frac{1}{t} \left( \sum_{f'l'} X_{f'l'} + a\delta_{l,4} y_{r-1} \right), \quad (31)$$

$$\delta W_{fl} = \frac{2}{t} \left( \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} [1 - \tanh^2(W_{fl} + u\delta W_{fl}) + \sum_{f'l'} (1 - \tanh^2(W_{f'l'} + u\delta W_{f'l'})) + a^2 \delta_{l,4} \times (1 - \tanh^2(V_r + u\delta V_r))] du \right)^{1/2}, \quad (32)$$

$$V_r = \frac{b}{t} (4y_r + y_{r+1} + (1 - \delta_{r,1})y_{r-1}), \quad (33)$$

$$y_r = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} \tanh(V_r + u\delta V_r) du, \quad (34)$$

$$\delta V_r = \frac{2b}{t} \left( \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} e^{-u^2/2} [4(1 - \tanh^2(V_r + u\delta V_r)) + (1 - \delta_{r,1})(1 - \tanh^2(V_{r-1} + u\delta V_{r-1})) + (1 - \tanh^2(V_{r+1} + u\delta V_{r+1}))] du \right)^{1/2}. \quad (35)$$

Eqs. (30)–(35) were solved numerically. In order to carry out the calculations we have to specify the magnetic lattices pyramid and substrate as a simple cubic one. Results are presented graphically.

Fig. 2 shows the spatial distributions of magnetization  $X_4$  in the  $l = 4$  layer of the nanoscopic pyramid for temperature  $t = 4.0$  and  $a = 0.5$ ,  $b = 1.0$  in both MFA and GFA.

Figs. 3–5 present spatial distributions of Gaussian fluctuations of molecular field  $\delta W_{fl}$  for  $l = 2, 3, 4$ , respectively, temperature  $t = 4.0$  and  $a = 0.5$ ,  $b = 1.0$ . In the tip layer adjacent to the substrate the magnitude of fluctuations is largest in the second row of atoms, counting from the outside. Comparing Figs. 4 and 5 we see that maximum fluctuations occur along the edges of the pyramid. Smaller fluctuations of magnetic moments in the center of the base of the apex is an effect of the substrate.

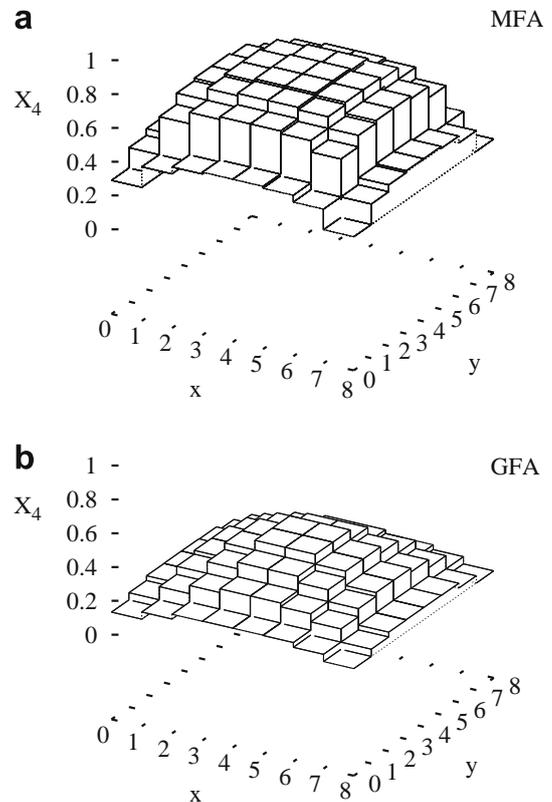


Fig. 2. Spatial distribution of magnetization in nanoscopic pyramid for  $l = 4$ , temperature  $t = 4.0$ , and  $a = 0.5$ ,  $b = 1$  in MFA [11] and GFA.

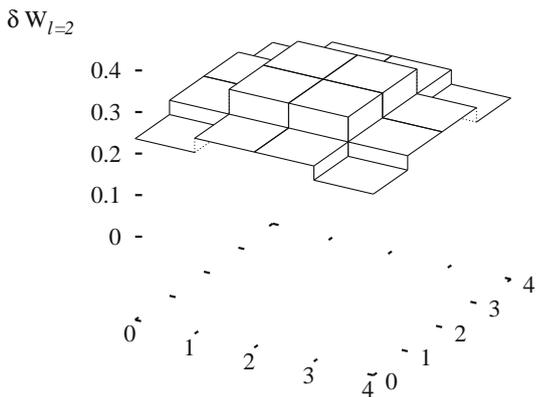


Fig. 3. Spatial distribution of Gaussian fluctuations of the molecular field in pyramid layer  $l = 2$  at temperature  $t = 4.0$  and  $a = 0.5, b = 1$ .

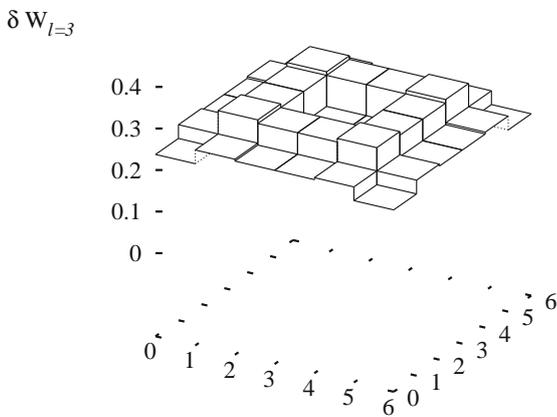


Fig. 4. Spatial distribution of Gaussian fluctuations of the molecular field in pyramid layer  $l = 3$  at temperature  $t = 4.0$  and  $a = 0.5, b = 1$ .

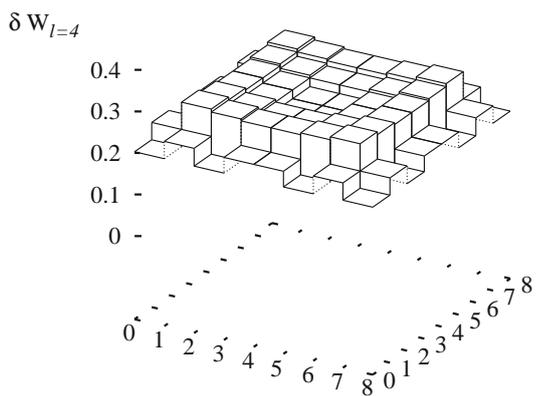


Fig. 5. Spatial distribution of Gaussian fluctuations of the molecular field in pyramid layer  $l = 4$  at temperature  $t = 4.0$  and  $a = 0.5, b = 1$ .

Fig. 6 shows the mean value of magnetization

$$X_l = \frac{\sum_f X_{fl}}{\sum_f} \quad (36)$$

in each monoatomic layer of pyramid  $l = 1, 2, 3, 4$  in GFA as a function of temperature  $t$  for  $a = 0, b = 1$  and  $a = 0.5, b = 1.0$ .

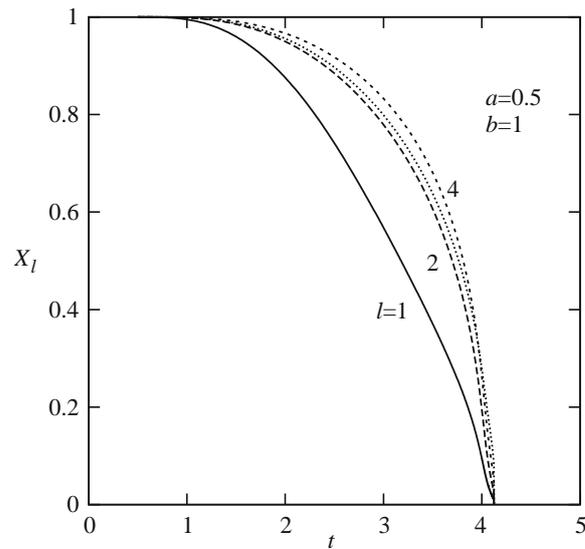


Fig. 6. Mean value of magnetization,  $X_l = \sum_f X_{fl} / \sum_f$  in each monoatomic layer of pyramid,  $l = 1, 2, 3, 4$ , in GFA as a function of temperature  $t$  for  $a = 0.5, b = 1.0$ .

Fig. 7 presents temperature dependence of the mean pyramid magnetization

$$X = (4X_1 + 16X_2 + 36X_3 + 64X_4) / 120 \quad (37)$$

obtained in GFA for  $a = 0$  and  $a = 0.5$  and  $b = 0.5, 1.0, 2.0$ . Fig. 8 shows magnetization of individual layers of the pyramid and the substrate. The influence of the pyramid extends several layers into the substrate. Similar conclusion was reached in an earlier study [12] of the interaction between a magnetic nanotip and a magnetic surface using the tight-binding model. The tight-binding calculation for a Fe tip on a Fe surface shows that only the first four layers of the tip support are affected. The inclusion of fluctuations in our work increase the depth of this influence as  $t \rightarrow t_c$ , see Fig. 8.

Fig. 9 presents the mean Gaussian fluctuation of molecular field

$$\delta W_l = \frac{\sum_{fl} \delta W_{fl}}{\sum_{fl}} \quad (38)$$

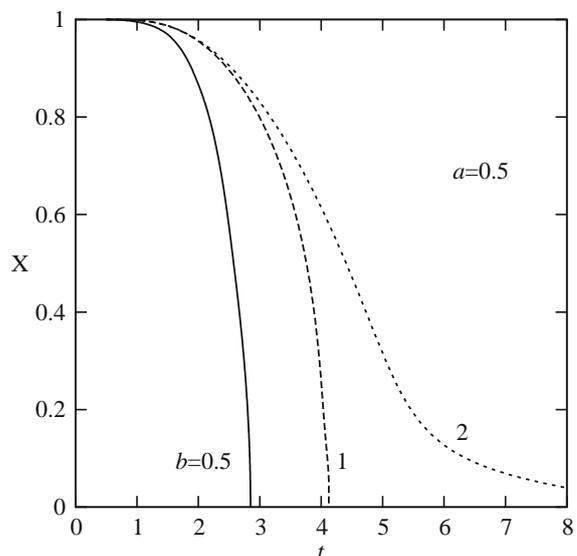


Fig. 7. Temperature dependence of the mean pyramid magnetization  $X = (4X_1 + 16X_2 + 36X_3 + 64X_4) / 120$  for  $a = 0.5$  and  $b = 0.5, 1, 2$ .

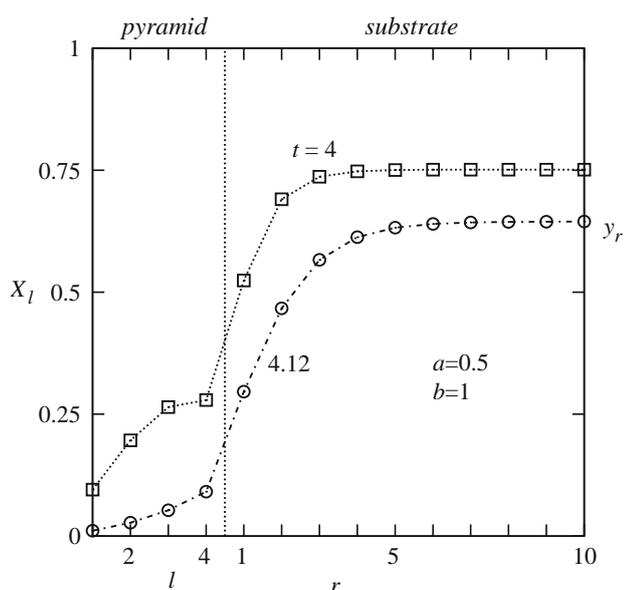


Fig. 8. Magnetization in individual layers of the pyramid  $X_l$  and the substrate  $y_r$  for  $a = 0.5$ ,  $b = 1.0$  and  $t = 4.0, 4.12$ .

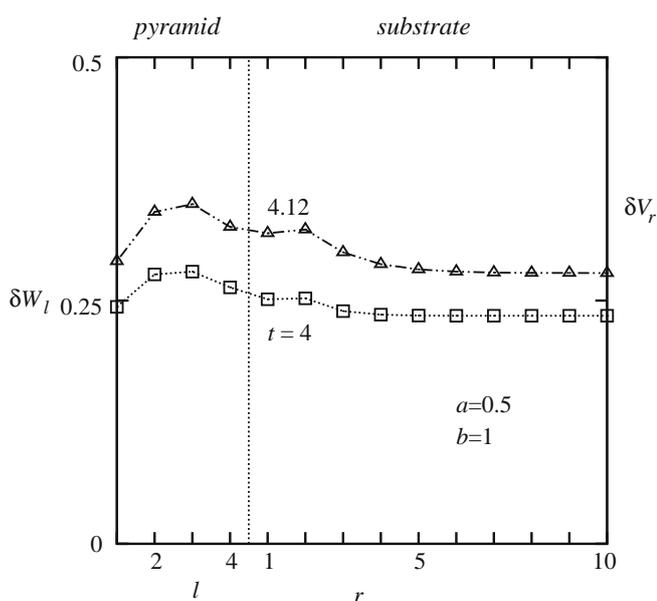


Fig. 9. Gaussian fluctuation of the molecular field in individual layers of the pyramid and the substrate for  $a = 0.5$ ,  $b = 1.0$  and  $t = 4.0, 4.12$ .

and  $\delta V_r$  for pyramid and the substrate, respectively.

Finally, Fig. 10 shows Gaussian fluctuation of the molecular field as a function of temperature.

In Table 1 the Curie temperature  $t_c$  of the ferromagnetic nanoscopic pyramid is given in relative units for different values of parameters  $a$  and  $b$  obtained in MFA and GFA.

We can see that for a nanopyramid the ratio of the Curie temperature in GFA to that obtained in MFA is 0.783. For an infinite simple cubic lattice this ratio is equal to 0.856 and for a square lattice Ising monolayer it has a value of 0.799 [8]. The ratio of the Curie temperature of a monolayer obtained in GFA to the exact result is 1.409. The same quantity for the sc bulk Ising ferromagnet is approximately 1.138.

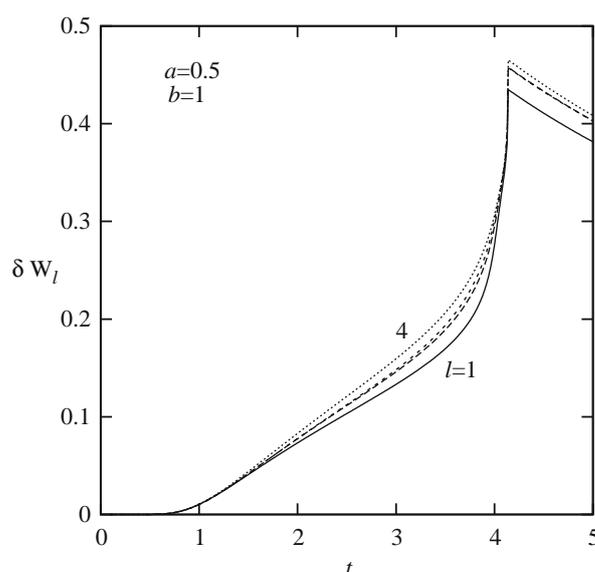


Fig. 10. Temperature dependence of Gaussian fluctuations of molecular field in individual layers of the pyramid  $\delta W_l$  for  $a = 0.5$  and  $b = 1.0$ .

Table 1

Curie temperature  $t_c$  (in relative units) for the nanoscopic pyramid.

$a$	$b$	$t_c$	
		MFA	GFA
0	Arbitrary	4.93	3.86
0.1	0.5	4.93	1.88
0.1	1.0	5.99	4.13
0.1	2.0	11.95	9.30
0.5	1.0	5.99	4.13
0.5	2.0	11.95	9.27

#### 4. Conclusions

The results we obtained lead us to the following conclusions:

- The interaction of a ferromagnetic nanoscopic pyramid with its bulk ferromagnetic substrate may have essential influence on the properties of the ferromagnetic pyramid,
- The distribution of Gaussian fluctuations is highly nonuniform,
- $\lim_{t \rightarrow 0} \delta W_{fl} = 0$ , as expected,
- The maximum of molecular field fluctuations  $\delta W_{fl}$  is reached at the Curie temperature  $t_c$ , as expected,
- The Curie temperature  $t_c$  of the pyramid strongly depends on the Curie temperature of the bulk substrate,
- We believe that ERSO (4) may be successfully applied in studies of the influence of the bulk substrate on the thermodynamic properties of nonmagnetic systems and other more complex nanoscopic systems [4,5].

The development of the magnetic exchange force microscopy (ME $\times$ FM) [13] may enable studies of the spatial distribution of magnetization in nanopyrramids deposited on nonmagnetic surface. Such magnetic particles are of interest in magnetic recording techniques.

It would be useful to study the temperature dependence of exchange force between the nanotip and a well-characterized magnetic surface of a material with Curie temperature higher than  $T_c$  of the tip. On cooling the system one should observe the magnetic transition of the tip. The force of the interaction between the tip

and the surface is measured as the shift of frequency of oscillations of the cantilever. As  $T$  decreases, magnetization of the tip increases and so does the exchange interaction with the surface, resulting in a frequency shift. If the ratio of exchange energies  $I_1/I_2$  is large, i.e. when exchange interaction between atoms within the tip is much larger than exchange between the bottom layer of the tip and the first layer of the substrate, or when the substrate is nonmagnetic, fluctuations may lead to a long tail of magnetization as a function of temperature, see Fig. 7. In this case magnetization may saturate at temperatures much smaller than  $T_c$ .

The measurement of the frequency shift as a function of temperature at fixed position above the surface may provide a good estimate of possible magnetic fluctuation effects. The crossover temperature between the tail, see  $b = 2$  curve in Fig. 7, and the saturated part at low  $T$  detected via the oscillation frequency shift should not depend on position above the surface. Temperature scans at different surface positions would result in frequency shifts of different magnitudes occurring at approximately the same temperature.

Recent attempts to map single spins on the surface of antiferromagnetic insulator NiO were only partially successful. The experiment of Kaiser et al. [13] with Fe tip performed at about 10 K in a magnetic field of 5 T showed clear picture of antiferromagnetic structure of the (001) surface of nickel oxide. It cannot be ruled out, however, that the applied magnetic field induces structural changes in NiO. The measurements of Schmid et al. [14] conducted at room temperature, using Co and NiO tips did not show any spin contrast. The ordering temperature of NiO is 525 K. The authors point out that the magnetic ordering temperature of the tip may be significantly reduced relative to the bulk value. Our calculation shows that it is also possible that the spin ordering of the tip is present but is very weak. Performing this experiment at lower temperatures, as authors of Ref. [14] suggest, may yield better results.

Atomically sharp magnetic tips are crucial in achieving lateral high spatial resolution. Therefore the knowledge of spatial distribution of fluctuations and its temperature dependence might improve the calibration of the STM device and help in estimating experimental error of the ME×FM method. For small distances between the tip and the surface it may be necessary to include interaction of more than one atom of the tip or of the surface [15]. In this case our model with nonuniform magnetization of the tip may provide useful insight. The temperature dependence of mean value of magnetization of the individual tip layers shown in Fig. 6

shows that the magnetization in the second layer can be significantly larger than the magnetization of the apex. This implies an increased range of tip-surface separation where the interaction with atoms in the next layer should be taken into account.

The ability to control magnetic properties of nanoclusters [16] is one of the central problems of nanotechnology. Arrays of nanoclusters are also intensively studied [17,18]. The theory presented here may be applied to various geometrical shapes used in experiments [2], e.g. chains of particles, striped and cylindrical nanowires, nanodots, nanojunctions, surface steps. We intend to carry out calculations in a more realistic model of the substrate surface and the interface between the pyramid and the substrate.

Finally let us note that ERSO (4) couples the nanoscopic pyramid with the substrate. This means that the system does not have a finite number of spins. Therefore the use of e.g. Monte Carlo simulations would have little justification.

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