Nature of a phase transition and low-temperature phase in cubic ferromagnets

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A $S=2$ ferromagnet with a crystalline field of the cubic symmetry is studied. It is shown that in the case of the three easy axes [100] the model which contains only the Heisenberg bilinear interaction and single-ion anisotropy terms exhibits at a finite temperature the phase transition to a nonmagnetic phase with a long-range quadrupolar order. It is also pointed out if the recent result that in three dimensions the cubic fixed point is stable for the $(n=3)$-component cubic spin model (Kleinert, Thoms, and Schulte-Frohlinde) is correct, all cubic ferromagnets with three easy axes, for example Fe, should undergo a first-order (discontinuous) phase transition. [S0163-1829(99)02806-4]

I. INTRODUCTION

The nature of a phase transition in the cubic ferromagnets is an old topic which has been studied since the early 1970s.[1] But there is still a notable defect in our understanding of the effect of cubic anisotropy on a system with isotropic exchange interaction. The most efficient approach to the problem of the critical behavior of such a system is the momentum space renormalization group (RG) which starts with the so-called Landau-Ginzburg-Wilson (LGW) Hamiltonian. For the n-component cubic vector model in d dimensions the corresponding LGW Hamiltonian reads[1]

$$H(\boldsymbol{\phi})=\int d^d x\left[-|\nabla\boldsymbol{\phi}|^2-r\sum_{\alpha=1}^{n}\phi_\alpha^2 -\sum_{\alpha\beta}(u+v\,\delta_{\alpha\beta})\phi_\alpha\phi_\alpha\phi_\beta\phi_\beta\right]. \quad (1)$$

Within the molecular-field approximation (MFA) the existence of the continuous phase transitions in the model (1) is defined by the conditions

$$u+v>0 \quad\text{and}\quad 3u+v>0, \quad (2)$$

whereas in the RG approach the stability of the isotropic or cubic fixed point depends on the value of n. For $n<n_c$, cubic symmetry-breaking fields are irrelevant and the isotropic fixed point is stable. For $n>n_c$ the cubic fixed point is stable, and for $d=3$, n_c is probably close to 3. Estimates using calculations up to three loops[1] indicated that $n_c>3$. Recently Kleinert, Thoms and Schulte-Frohlinde[2] (KTS) using a rather sophisticated method have concluded that $n_c<3$ and for the most interesting case $d=3$ and $n=3$, ''the critical behavior of the magnetic phase transition in anisotropic crystals with cubic symmetry is governed by the cubic, not by isotropic fixed point. Unfortunately, the result is only of fundamental interest'' and cannot be experimentally confirmed because the differences in the critical exponents for isotropic and cubic universality classes are too small.

However, there may be a chance to check the prediction of KTS, taking into account not only stability of the cubic fixed point but also its accessibility. It was found, within the ϵ expansion[1] that for $n<n_c$ the isotropic fixed point is accessible (continuous phase transition is to be expected) if the initial Hamiltonian lies in the region given by the two following inequalities:

$$u>0 \quad\text{and}\quad v>\frac{v_c}{u_c}u, \quad (3)$$

where $u_c(v_c)$ denote the cubic fixed-point values of $u(v)$ and v_c is negative. For $n>n_c$ the cubic fixed point represents a continuous phase transition and it can be reached for the initial Hamiltonians with

$$u>0 \quad\text{and}\quad v>0. \quad (4)$$

Initial Hamiltonians outside of these regions will flow away to negative values of $v(u)$ and it is believed that the transition becomes discontinuous. Thus, if the result of KTS is correct all systems described by the ϕ^4 Hamiltonian (1) with $n=3$ and $v<0$ in three dimensions (d=3) should undergo discontinuous phase transition.

In the remainder of the paper, we will point out a consequence of the result of KTS for the critical behavior of the cubic ferromagnets with three easy axes and then we will investigate a nonmagnetic long-range order in this kind of ferromagnet described by the Hamiltonian with only the Heisenberg interaction and single-ion anisotropy.

II. CUBIC FIXED POINT

In the ''magnetic'' language the sign of v determines whether the easy axis is along an edge of a cube $(v<0)$—three easy axes [100], or along a main diagonal $(v>0)$—four easy axes [111]. If $n_c>3$ in both cases $v>0$ and $v<0$ a continuous or discontinuous phase transition can occur in cubic ferromagnets because the stable fixed point is accessible also from the region with negative v Eq. (3). It is true that usually one assumes $u>0$ and then all magnets with a [111] easy axis should have continuous phase transition but in the systems with cubic symmetry the phase with magnetization along the [111] direction can be stable also for u negative.

In many real magnets the isotropic short-range exchange is associated with single-ion (crystal field) terms which break full rotational invariance and can be origin of the cubic anisotropy. For a lattice of cubic symmetry in the lowest order such a system can be described by the Hamiltonian

$$H=-J\sum_{\langle ij\rangle}\sum_{\alpha}S_i^{\alpha}S_j^{\alpha}+\frac{1}{3}K\sum_i\sum_{\alpha}(S_i^{\alpha})^4, \tag{5}$$

where S^{α} is the α component of the spin operator $\mathbf{S}$ ($S\geqslant 2$). In this quantum model the sign of K determines whether the spins tend to align along the cubic axes ($K<0$), or along a main diagonal ($K>0$). One can use Eq. (5) to find the Landau free energy and then the LGW Hamiltonian.[3] For spin $S=2$ the LGW parameters were found in the form

$$v=\frac{24\varkappa}{K^4}\frac{\varkappa(\varkappa-1)e^{\varkappa}-2\varkappa\cosh\varkappa+3\sinh\varkappa}{4\cosh\varkappa+e^{\varkappa}}, \tag{6}$$

$$u=\frac{4\varkappa}{K^4}\left[2\varkappa\left(\frac{4\sinh\varkappa+\varkappa e^{\varkappa}}{4\cosh\varkappa+e^{\varkappa}}\right)^2-\frac{(\varkappa^3+18\varkappa^2-6\varkappa)e^{\varkappa}+6(1-2\varkappa)\sinh\varkappa}{3(4\cosh\varkappa+e^{\varkappa})}\right], \tag{7}$$

where $\varkappa=K\beta$ ($\beta=1/k_BT$) and we have assumed $Jz_1=1$ (z_1 denotes the number of the nearest-neighbor spin pairs). It is easy to check that $\mathrm{sgn}(v)=\mathrm{sgn}(K)$ but u is negative for $2.38/\beta<K<7.14/\beta$. Thus, if $n_c>3$ the conditions for the accessibility of the isotropic fixed point Eq. (4) can be fulfilled or not for both cases $K>0$ and $K<0$. It means that within the RG approach the system with $S=2$ is predicted to have a continuous or discontinuous phase transition in dependence on the value of K both to the phases with magnetization along [111] and [100] axes. The similar estimation for $S=5/2$ leads to the conclusion that in this latter case the system should undergo a continuous phase transition to the phase [111] (if $K>0$) and a discontinuous one to the phase [100] (if $K<0$). So if $n_c>3$, the kind of the phase transition in cubic ferromagnets depends on the ratio of an anisotropy to exchange interaction and value of the spin. If $n_c<3$ the situation is qualitatively different, the stable *cubic* fixed point is not accessible for the initial Hamiltonian with $v<0$. It means that if the KTS result , $n_c<3$, is correct then the phase transition in cubic ferromagnets with three easy axes [100] should be a discontinuous one.

III. LOW-TEMPERATURE PHASE

As mentioned in the previous sections the character of the phase transition in the cubic ferromagnets with three easy axes is still unsettled. The earlier results suggested that this transition can be continuous or discontinuous in dependence on the values of the model parameters, however, recently[2] it has been concluded that these systems can undergo only a discontinuous phase transition. Apart from the character of the phase transition another feature of the model described by the Hamiltonian (5)—a feature of the low-temperature phase—should also be clarified.

Within the MFA (Refs. 3 and 4) one obtains that for $K<-4zJ$ (z denotes the number of nearest neighbors) a system described by the Hamiltonian (5) does not exhibit any magnetic order for an arbitrary temperature. In the ground state the magnetization disappears continuously with K decreasing from 0 to $-4zJ$ and for $K<-4zJ$ the nonmagnetic phase is observed. In this approximation it means that the models under consideration have no long-range order for sufficiently small, negative, anisotropy constant K and no phase transitions in such systems exist. The natural question is if there is some other kind of long-range order in the cubic magnets with ferromagnetic interaction and large negative anisotropy omitted by the MFA. In our recent paper,[5] where mainly the ground state has been considered, it has been shown by using the perturbation theory for $J/|K|\ll 1$ that in opposition to the MFA prediction there is a quadrupolar long-range order in such a system. In the present paper we consider the model at $T\neq 0$ to show that as a consequence of the quantum character of spins there is possibly a finite-temperature phase transition to the quadrupolar, nonmagnetic state in the $S=2$ magnet without a quadrupolar type of interaction.

A. Effective Hamiltonian

Because there is not a quadrupolar type of interaction in the Hamiltonian (5) it is easy to see that any kind of the MFA cannot lead to the multipolar ordering in the phase without magnetic (dipolar) long-range order. On the other hand, there is no method beyond MFA which allows us to study efficiently the $S=2$ model in $d>1$ (for the case $d=1$ the density-matrix renormalization group theory has been applied[6] to study the ground-state properties of the model under consideration). So we propose some kind of renormalization procedure to find an effective Hamiltonian which includes the quadrupolar interaction caused by the quantum fluctuations.

The Hamiltonian (5) is divided into two parts

$$H(\mathbf{S})=H_0(\mathbf{S})+H_1(\mathbf{S}), \tag{8}$$

where H_0 contains all single-ion terms. H_0 can be diagonalized exactly and be removed from the problem, which leads to the renormalized interaction between effective spins $\boldsymbol{\sigma}$.

The renormalized Hamiltonian $H_{\mathrm{eff}}(\boldsymbol{\sigma})$ is defined by the operator equation

$$\exp[H_{\mathrm{eff}}(\boldsymbol{\sigma})]=\mathrm{Tr}_{\mathbf{S}}P(\boldsymbol{\sigma},\mathbf{S})\exp[H(\mathbf{S})], \tag{9}$$

with the projection operator $P(\boldsymbol{\sigma},\mathbf{S})$ which couples the original spin ($\mathbf{S}$) space, and effective spin $\boldsymbol{\sigma}$ space and due to the translational invariance can be written in the form

$$P(\boldsymbol{\sigma},\mathbf{S})=\prod_i p(\boldsymbol{\sigma}_i,\mathbf{S}_i), \tag{10}$$

where i denotes ith site.

The most general rotationally invariant projection operator for the spin-2 model is

$$p(\boldsymbol{\sigma}_i,\mathbf{S}_i)=\sum_{n=0}^{4}a_n(\boldsymbol{\sigma}_i\mathbf{S}_i)^n. \tag{11}$$

However, in this paper we confine ourselves to the simpler form

$$p(\boldsymbol{\sigma}_i,\mathbf{S}_i)=a_0+a_1(\boldsymbol{\sigma}_i\mathbf{S}_i)+a_2(\boldsymbol{\sigma}_i\mathbf{S}_i)^2, \tag{12}$$

which is sufficient to find the appropriate effective Hamiltonian.

The projection operator p should satisfy the following condition:

$$\mathrm{Tr}_{\boldsymbol{\sigma}}p=1, \tag{13}$$

which insures that the partition function of the original and effective spin problem are the same, and

$$\mathrm{Tr}_{\mathbf{S}}S^{\alpha}p=\sigma^{\alpha}, \quad \mathrm{Tr}_{\mathbf{S}}(S^{\alpha})^2p=(\sigma^{\alpha})^2, \tag{14}$$

which insures that Eq. (9) transforms the original system in itself for $K=0$ (isotropic case). It is easy to see that the conditions (13) and (14) are fulfilled if

$$a_0=-\frac{13}{35}, \quad a_1=\frac{13}{105}, \quad a_2=\frac{1}{21}. \tag{15}$$

In order to solve Eq. (9) one has to use some approximation, for example, the cumulant expansion. To take into account that H_0 and H_1 do not commute, we apply the identity

$$\exp[H_0+H_1]=\exp[H_0]\Upsilon\exp\left[\int_0^1 d\lambda H_1(\lambda)\right], \tag{16}$$

where

$$H_1(\lambda)=\exp[-\lambda H_0]H_1\exp[\lambda H_0], \tag{17}$$

and Υ is the time-ordering operator with respect to λ. The transformation (9) can then be written as

$$H_{\mathrm{eff}}(\boldsymbol{\sigma})=\ln \mathrm{Tr}_{\mathbf{S}}P(\boldsymbol{\sigma},\mathbf{S})\exp[H_0(\mathbf{S})] + \ln\left[\sum_{n=0}^{\infty}\int_0^1 d\lambda_1 \ldots \int_0^{\lambda_{n-1}} d\lambda_n \times\langle H_1(\lambda_1)\ldots H_1(\lambda_n)\rangle\right], \tag{18}$$

where the angular brackets denote a partial expectation value defined for some operator $\mathbf{A}$ by

$$\langle\mathbf{A}\rangle=\mathrm{Tr}_{\mathbf{S}}\exp[H_0(\mathbf{S})]\mathbf{A}P(\boldsymbol{\sigma},\mathbf{S})/\mathrm{Tr}_{\mathbf{S}}\exp[H_0(\mathbf{S})]P(\boldsymbol{\sigma},\mathbf{S}). \tag{19}$$

In the first-order cumulant expansion the effective Hamiltonian contains only a spin-spin interaction between nearest neighbors

$$H_{\mathrm{eff}}^{(1)}=-J_p\sum_{\langle i,j\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j, \tag{20}$$

where

$$J_p=\frac{8\varkappa e^{-2\varkappa}-2e^{-4\varkappa}-4e^{-2\varkappa}+\varkappa+6}{\varkappa(3+2e^{-2\varkappa})^2}. \tag{21}$$

In the second-order approximation for the cubic lattice there come into play nine interactions with six different coupling parameters ($J_1\ldots J_6$):

$$\begin{aligned}H_{\mathrm{eff}}^{(2)}=&-J_1\sum_{\langle i,j\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_j-J_2\sum_{\langle i,k\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_k-2J_2\sum_{\langle i,k'\rangle}\boldsymbol{\sigma}_i\cdot\boldsymbol{\sigma}_{k'}-J_3\sum_{\langle i,j\rangle,\alpha}(\sigma_i^{\alpha})^2(\sigma_j^{\alpha})^2-J_4\sum_{\langle ij\rangle,\alpha\beta}\sigma_i^{\alpha}\sigma_i^{\beta}\sigma_j^{\alpha}\sigma_j^{\beta}\\&-J_5\sum_{\langle ijk\rangle,\alpha}\sigma_i^{\alpha}\sigma_k^{\alpha}(\sigma_j^{\alpha})^2-J_6\sum_{\langle ijk\rangle,\alpha\beta}\sigma_i^{\alpha}\sigma_k^{\beta}\sigma_j^{\alpha}\sigma_j^{\beta}-J_5\sum_{\langle ijk'\rangle,\alpha}\sigma_i^{\alpha}\sigma_{k'}^{\alpha}(\sigma_j^{\alpha})^2-J_6\sum_{\langle ijk'\rangle,\alpha\beta}\sigma_i^{\alpha}\sigma_{k'}^{\beta}\sigma_j^{\alpha}\sigma_j^{\beta},\end{aligned} \tag{22}$$

where j, k', and k denote nearest, second, and third neighbors of the site i, respectively. In Fig. 1 the coupling parameters J_1 and J_3 as functions of single-ion anisotropy constant K are presented. Thus, due to the interaction of the spins with the crystal field, between others, the effective quadrupole-quadrupole interactions (J_3,J_4) appear. These interactions can, of course, cause the existence of the quadrupolar ordering even in the nonmagnetic phase.

B. Phase diagram

In the second-order cumulant expansion the effective Hamiltonian is much more complicated and no more tractable than the original one. However, we regard the Hamiltoninan (22) as a better starting point for the approximation. It is easy to see that even within the simplest MFA using Eq. (22), one can expect to obtain a quadrupolar ordering with-

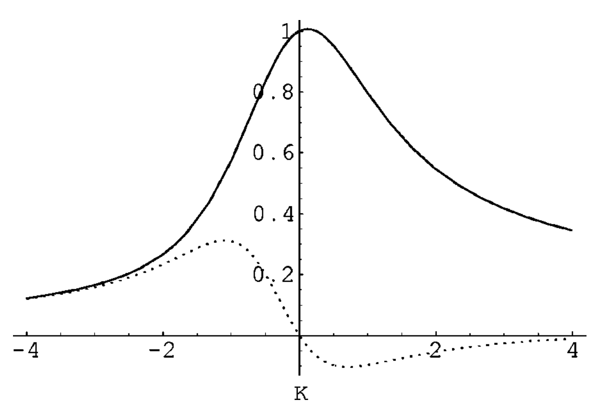

FIG. 1. The coupling parameters J_1 (full line) and J_3 (dotted line) as functions of cubic single-ion anisotropy.

out a dipolar one. The effective Hamiltonian can be applied to study the whole phase diagram of the cubic ferromagnets but then one should apply a more powerful approximation than the second-order cumulant expansion to get this Hamiltonian (the approximation used in this paper is valid only in high temperature for $J/T<1$), and consider the eight-dimensional order-parameter space (three components of the magnetization and five quadrupolar parameters).[7]

Taking into account the symmetry of the problem, in order to check the possibility of the existence of the finite-temperature quadrupolar phase in cubic ferromagnets with three easy axes ($K<0$), we can confine ourselves to consider only the phase transitions to the magnetic phase with order parameter $m=\langle S^{\alpha}\rangle \neq 0$, and the quadrupolar phase with order parameter $q=\langle (S^{\alpha})^2-2\rangle \neq 0$, and $\langle S^{\alpha}\rangle =0$ ($\alpha=x$ or y or z). The appropriate free energy in the MFA reads

$$F_{\mathrm{MFA}}=A-T\ln\left[2e^{-4C/T}\cosh\frac{2B}{T}+2e^{-C/T}\cosh\frac{B}{T}+1\right]-12J_3z_1, \tag{23}$$

where

$$A=[J_1z_1+J_2(z_2+2z_2')+4J_5(z_3+z_3')]m^2+6J_3z_1q+\frac{3}{2}J_3z_1q^2+2J_5(z_3+z_3')m^2q,$$

$$B=-2[J_1z_1+J_2(z_2+2z_2')+2J_5(z_3+z_3')]m-2J_5(z_3+z_3')mq,$$

$$C=-3J_3z_1q-J_5(z_3+z_3')m^2, \tag{24}$$

and for the cubic lattice, $z_1=z_2=z_3=3$, $z_2'=6$, $z_3'=12$.

The limit of the existence of the disordered phase is defined by the conditions:

$$\frac{\delta^2F_{\mathrm{MFA}}}{\delta m^2}\Big|_{(m=0,q=0)}=0, \tag{25}$$

or

$$\frac{\delta^2F_{\mathrm{MFA}}}{\delta p^2}\Big|_{(m=0,q=0)}=0. \tag{26}$$

Conditions (25) and (26) indicate a transition temperature to the magnetic (dipolar) phase with $m\neq 0$ and $q\neq 0$ or quadrupolar phase with $m=0$ and $q\neq 0$. Of course, the system undergoes the transition at the higher of these two temperatures. It is easy to see that condition (25) can be fulfilled only for $K>-3.5$ and for $K<-3.5$ the transition temperature is defined by the condition (26). The expectation values of order parameters m and q can be determined from the set of equations

$$\frac{\delta F_{\mathrm{MFA}}}{\delta m}=0 \quad \text{and} \quad \frac{\delta F_{\mathrm{MFA}}}{\delta p}=0. \tag{27}$$

From Eq. (27) one finds that the solution $m\neq 0$ and $q\neq 0$ exists only for $K>K_c$ where $K_c\approx -7.6$ and for $-7.6<K<0$ the system undergoes the transition to the magnetic

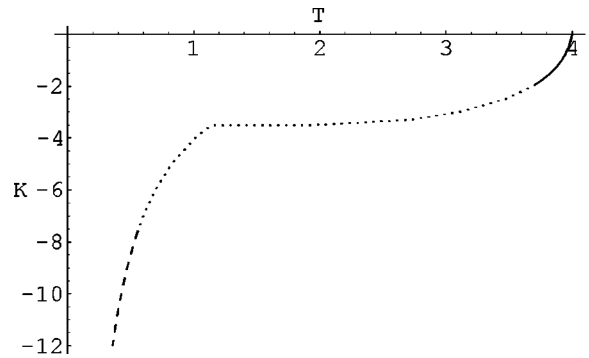

FIG. 2. Phase diagram of the cubic ferromagnet with three easy axes [100]. The full and dotted lines denote the continuous and discontinuous phase transitions to the magnetic phase, respectively. The dashed line denotes the phase transition to the quadrupolar phase.

phase, whereas for $K<-7.6$ the system should undergo the transition to the quadrupolar phase without magnetic order. Similarly as in the previous papers,[34] we have found that for a negative but small enough anisotropy constant in our approximation $K>-1.94$ the cubic three axial ferromagnet should undergo the continuous phase transition. Of course, if the recent result of KTS's is correct the fluctuations can change this transition into a discontinuous one. The MFA phase diagram in the plane (K,T) is presented in Fig. 2. The dependence of the order parameters m and q on the single-ion anisotropy K for a given temperature is shown in Fig. 3.

IV. CONCLUSION

A magnetic state has always a quadrupole moment which is, of course, not generally true for a paramagnetic state. However, if there is a quadrupolelike interaction in the system one can expect some quadrupolar order also in the paramagnetic phase. At first glance if no higher-order coupling exists the system should simply have the magnetic phase transition, and no quadrupolar order without the magnetic one should occur. In the present paper we have shown that for the $S=2$ cubic ferromagnet the quadurpolar order can exist, although there is no quadrupolar coupling and there is no interaction of the spins with other dergrees of freedom like phonons or itinerant electrons which could mediate such

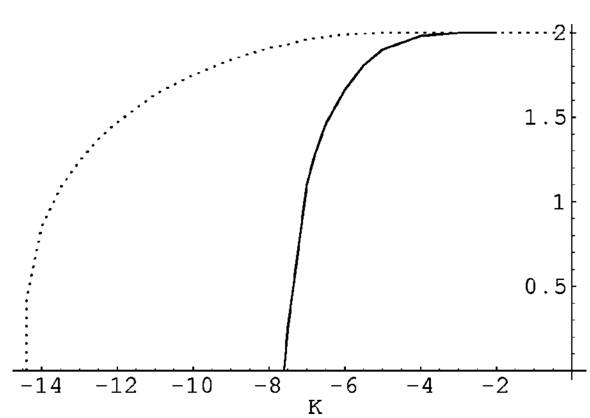

FIG. 3. The dependence of the magnetic (full line) and quadrupolar (dotted line) order parameters on the single-ion cubic anisotropy for a given value of temperature.

a coupling. The quadrupolar phase transition is driven by the interaction of the quantum spins with the crystalline field of cubic symmetry and could not be observed in the model of the classical spins.

Using a kind of renormalization procedure we have transformed the original Hamiltonian containing the bilinear (dipole) interaction and single-ion cubic anisotropy terms into an effective Hamiltonian with multi-ion and multipolar interactions. This effective Hamiltonian can be used as a starting point for some approximation to find the whole phase diagram of the cubic ferromagnet. In this paper we have found the effective Hamiltonian in the second order of the cumulant expansion in order to analyze in the MFA the possibility of the phase transition to the magnetic phase with the order parameter $\langle S_z \rangle$ and to the quadrupolar phase with the order parameter $\langle S_z^2 - 2 \rangle$.

It has been shown that for sufficiently large and negative values of the cubic anisotropy $K < K_c \approx -7.6$ the system undergoes the quadrupolar phase without magnetic order. On this basis and taking into account our previous results[5] from perturbation theory we conclude that the $S=2$ cubic ferromagnets described by the Hamiltonian (5) with $K < K_c$, for $T < T_c$ exhibit the long-range quadrupolar order in the paramagnetic state. For $K > K_c$ the system should have the ferromagnetic phase transition with the tricritical point at $K \approx -1.94$ and $T \approx 3.71$ (for $K > -1.94$ one observes the continuous phase transition, while for $K < -1.94$ discontinuous one). It is obvious that this molecular field location of the tricritical point would be considerably changed if one would take into account the fluctuations.

We have also pointed out that if the recent result of KTS[2] is correct and the critical dimensionality of the cubic model order parameter n_c is less than 3, for $d=3$, then one should observe the discontinuous phase transitions in all systems described by the phenomenological LGW Hamiltonian (1) with $v < 0$. This conclusion is independent of the microscopic Hamiltonian of the model and it means that all cubic ferromagnets with three easy axes [100], for example Fe, should undergo the discontinuous (first-order) phase transition.

To be concrete we have presented the LGW Hamiltonian parameters for the $S=2$ cubic ferromagnets with single-ion anisotropy. If $n_c < 3$,[2] this model in dependence of the value of the cubic anisotropy would exhibit the continuous or discontinuous phase transition to the phase [111] (Ref. 3) and always discontinuous transition to the phase [100]. The tricritical behavior of the system would be controlled by the isotropic (Heisenberg) fixed point at $K=0$.

ACKNOWLEDGMENT

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