

## Mean-field treatment of arbitrary anisotropic ferromagnetic spin Hamiltonians

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The solution of the infinite-range magnetization equation for arbitrary anisotropic spin Hamiltonians is discussed. The procedure presented is applied to the determination of the phases and phase transitions in a family of systems involving a quadratic as well as a quartic anisotropy.

### I. INTRODUCTION

Spin-Hamiltonians of increasing complexity keep appearing in a large variety of contexts.<sup>1-3</sup> While neither the isotropic Heisenberg Hamiltonian nor its extreme anisotropic counterparts, the three-dimensional Ising and XY models, can be treated exhaustively, an impressive number of approaches to the treatment of systems including higher than bilinear terms in the spin operators, as well as to certain classes of anisotropic spin Hamiltonians, have been developed. The spectrum of methods applied to the treatment of anisotropic Hamiltonians with multicomponent order parameters ranges from Landau theory<sup>4-9</sup> to the renormalization-group (RG) technique.<sup>4,10,11</sup>

The significance of the Landau theory analysis was recently discussed by Galam,<sup>8</sup> who pointed out that the terms of order higher than fourth in the Landau expansion, which are irrelevant in the RG sense, can give rise to new phases and affect the nature of the various phase transitions.

Microscopic mean-field theory, while being very close to Landau theory in both its basic assumptions and general consequences, differs from it by taking into account the specific nature of the order parameters involved and by retaining a clear distinction between the role of the internal energy and that of the entropy. Technically, an expansion of the entropy is avoided, so that this approach is more suitable for a discussion of first-order phase transitions as well as second-order transitions between ordered phases (i.e., involving large values of some components of the order parameter). Thus, while clearly exhibiting the difference in the form of the entropy terms in systems involving order parameters of different nature (e.g., spin systems with different elementary spins, various types of orientational and translational ordering in liquid crystals and of structural ordering in ferroelectrics), mean-field formalism explicitly takes into account the fact that for systems of a given nature the form of the entropy is independent of the concrete form of the Hamiltonian.

The equivalence between mean-field theory and the exact treatment of the appropriate infinite-range Hamiltonian enabled the study of spin Hamiltonians more complex than the isotropic Heisenberg Hamiltonian. The general isotropic spin Hamiltonian was considered in Ref. (12). Lee and co-workers<sup>13-16</sup> studied the static and dynamic properties of the infinite-range anisotropic Heisenberg Hamiltonian of uniaxial symmetry. The general anisotropic

Heisenberg Hamiltonian was investigated by Gilmore.<sup>17</sup>

These studies were extended in Refs. 18 and 19 to the general infinite-range uniaxial Hamiltonian

$$\mathcal{H} = NH(\hat{s}_x^2, \hat{s}_z^2).$$

Reference 18 contains a derivation of the corresponding magnetization equation. By analyzing the possible solutions of this equation it was found that three types of ordered phases have to be considered, namely Ising-like, intermediate, and XY-like. A generalization of the magnetization equation to an arbitrary anisotropic spin Hamiltonian was derived in Ref. 20. The general infinite-range Hamiltonian is written in the form

$$\mathcal{H} = NH(\hat{s}_x, \hat{s}_y, \hat{s}_z), \quad (1)$$

where  $N$  is the number of particles and

$$\hat{s}_i = \sum_{j=1}^N \hat{s}_{ij} / N, \quad i = x, y, z.$$

The axes  $x, y, z$  are the principal axes of the tensor of coefficients in the quadratic terms in the spin operators.

The magnetization equation for the Hamiltonian in Eq. (1) was shown to be

$$\mathbf{s} = - \frac{\nabla_s \mathbf{H}}{|\nabla_s \mathbf{H}|} \sigma B_\sigma(\beta \sigma |\nabla_s \mathbf{H}|). \quad (2)$$

Here,  $s_i = \langle \hat{s}_i \rangle$  is the thermal average of  $\hat{s}_i$ ,

$$\nabla_s \mathbf{H} = \mathbf{i} \frac{\partial H}{\partial s_x} + \mathbf{j} \frac{\partial H}{\partial s_y} + \mathbf{k} \frac{\partial H}{\partial s_z},$$

$\sigma$  is the elementary spin,  $B_\sigma$  is Brillouin's function, and  $\beta = 1/k_B T$ .

In the present paper we discuss the procedure for the concrete application of the formalism derived in Ref. 20. A certain class of anisotropic spin Hamiltonians containing both quadratic and quartic terms is analyzed and the corresponding phase diagrams are constructed. The types of phases and the location and nature of the phase transitions are determined. An elementary derivation of the magnetization equation for an arbitrary anisotropic spin Hamiltonian in the classical limit ( $\sigma = \infty$ ) is presented in the Appendix.

## II. THE GENERAL PROCEDURE

The magnetization equation for an anisotropic spin Hamiltonian, Eq. (2), is actually a set of three coupled nonlinear equations for the spin components. The following types of solutions are possible. (a) Paramagnetic,  $s_x = s_y = s_z = 0$ ,  $\nabla_s \mathbf{H} = 0$ . (b) Ising-like (say,  $z$  type): Only one Cartesian component of the magnetization,  $s_i$ , does not vanish;  $\nabla_s \mathbf{H} = \mathbf{u}_i \partial H / \partial s_i$ , where  $\mathbf{u}_i$  is a unit vector in the direction  $i$ . (c)  $XY$ -like: Two Cartesian components of the magnetization,  $s_i$  and  $s_j$ , do not vanish. In general

$$s_i \neq s_j, \quad \nabla_s \mathbf{H} = \mathbf{u}_i \frac{\partial H}{\partial s_i} + \mathbf{u}_j \frac{\partial H}{\partial s_j}.$$

(d) Generalized intermediate (or  $xyz$ ): All three components of the magnetization are nonvanishing.

Altogether, there are seven types of ordered phases ( $x, y, z, xy, yz, zx, xyz$ ). For each phase the magnetization equation enables the determination of the magnetization components as functions of the temperature. These, in turn, can be used to evaluate the free energy in order to establish the identity of the thermodynamic ground state for a given temperature. In many cases involving second-order phase transitions the sequence of phases and values of the transition temperature can easily be determined by the intersections of the resultant magnetization curves,  $s(T)$ , for the various phases, without explicitly evaluating the free energy.

From Eq. (2) it follows that the resultant magnetization  $s = |\mathbf{s}|$  satisfies the equation

$$s = \sigma B_\sigma (\beta \sigma |\nabla_s \mathbf{H}|), \quad (3)$$

which is of the same form as that for an isotropic spin Hamiltonian,<sup>13</sup> with  $|\nabla_s \mathbf{H}|$  replacing  $-dH/ds$ . In addition we have in the  $xyz$  phase

$$\frac{1}{s_x} \frac{\partial H}{\partial s_x} = \frac{1}{s_y} \frac{\partial H}{\partial s_y} = \frac{1}{s_z} \frac{\partial H}{\partial s_z}, \quad (4a)$$

in the  $xy$  phase

$$\frac{1}{s_x} \frac{\partial H}{\partial s_x} = \frac{1}{s_y} \frac{\partial H}{\partial s_y}, \quad s_z = 0 \quad (4b)$$

and in the  $z$  phase

$$s_z = s, \quad s_x = s_y = 0. \quad (4c)$$

Using Eqs. (4) and the relation  $s^2 = s_x^2 + s_y^2 + s_z^2$  we can express  $s_x$ ,  $s_y$ , and  $s_z$  as functions of  $s$ . Consequently,  $|\nabla_s \mathbf{H}|$ , whose concrete form is different for each type of phase, can be expressed in terms of  $s$ . Equation (3) can now be used to determine  $s$  as a function of  $T$ . Finally, the expressions for the spin components in terms of  $s$  can be used to determine their temperature dependence.

We note in passing that, at least for  $\sigma = \frac{1}{2}$ , Eq. (3) can be written in the inverted form

$$T = |\nabla_s \mathbf{H}| / \ln \left[ \frac{1+2s}{1-2s} \right]. \quad (5)$$

This form can enable a noniterative determination of the magnetization curve, as mentioned for isotropic spin

Hamiltonians in Ref. 21.

For future reference we write down the equilibrium free energy in the form

$$A = H + s |\nabla_s \mathbf{H}| - k_B T \ln \left[ \frac{\sinh[\beta |\nabla_s \mathbf{H}| (\sigma + \frac{1}{2})]}{\sinh(\beta |\nabla_s \mathbf{H}| / 2)} \right], \quad (6)$$

which, for  $\sigma = \frac{1}{2}$ , reduces to

$$A = H - k_B T \left\{ \ln 2 - \frac{1}{2} [(1+2s) \ln(1+2s) + (1-2s) \ln(1-2s)] \right\}. \quad (7)$$

## III. THE GENERAL ANISOTROPIC QUARTIC SPIN HAMILTONIAN

The family of Hamiltonians to be considered in more detail is of the form

$$H = \frac{1}{2} (a_x s_x^2 + a_y s_y^2 + a_z s_z^2) + \frac{1}{4} (b_x s_x^4 + b_y s_y^4 + b_z s_z^4) + \frac{1}{2} (c_x s_y^2 s_z^2 + c_y s_z^2 s_x^2 + c_z s_x^2 s_y^2). \quad (8)$$

Note that

$$\frac{1}{s_x} \frac{\partial H}{\partial s_x} = a_x + b_x s_x^2 + c_y s_z^2 + c_z s_y^2, \dots$$

Equation (4a), corresponding to the  $xyz$  phase, becomes

$$\begin{aligned} a_x + b_x s_x^2 + c_y s_z^2 + c_z s_y^2 &= a_y + b_y s_y^2 + c_x s_z^2 + c_z s_x^2 \\ &= a_z + b_z s_z^2 + c_x s_y^2 + c_y s_x^2, \end{aligned} \quad (9)$$

$$s_x^2 + s_y^2 + s_z^2 = s^2,$$

which can be solved for  $s_x^2$ ,  $s_y^2$ , and  $s_z^2$  in terms of  $s^2$ . One obtains

$$s_i^2 = \alpha_i s^2 + \beta_i, \quad i = x, y, z \quad (10)$$

where

$$\begin{aligned} \alpha_x &= \frac{1}{\Delta} [c_x(3c - 2c_x) - \mathbf{c} \cdot \mathbf{b} + c_x b_x + b_y b_z], \\ \beta_x &= \frac{1}{\Delta} [c_x(4a_x - 3a) - 3a_x b - \mathbf{a} \cdot \mathbf{c} \\ &\quad + a_x b_x + a_y(b_z + c_z) + a_z(b_y + c_y)], \end{aligned} \quad (11)$$

⋮

and

$$\Delta = \frac{3b_x b_y b_z}{\tilde{b}} + \frac{6c_x c_y c_z}{\tilde{c}} - (c_x^2 + c_y^2 + c_z^2) - 2\mathbf{b} \cdot \mathbf{c}.$$

Here,

$$a = (a_x + a_y + a_z)/3,$$

$$b = (b_x + b_y + b_z)/3,$$

$$c = (c_x + c_y + c_z)/3,$$

$$\frac{3}{\tilde{b}} = \frac{1}{b_x} + \frac{1}{b_y} + \frac{1}{b_z},$$

and

$$\frac{3}{\bar{c}} = \frac{1}{c_x} + \frac{1}{c_y} + \frac{1}{c_z}.$$

Note that  $\alpha_x + \alpha_y + \alpha_z = 1$  and  $\beta_x + \beta_y + \beta_z = 0$ . This solution is relevant provided that  $s_i^2 \geq 0$ ,  $i = x, y, z$ . The vanishing of any one of these components, say  $z$ , signals a transition into an  $xy$ -type phase. If the corresponding transition is of second order, this is actually the transition

$$\begin{aligned} \gamma &= \frac{1}{\Delta} [b_x b_y b_z + 2c_x c_y c_z - (b_x c_x^2 + b_y c_y^2 + b_z c_z^2)], \\ \delta &= \frac{1}{\Delta} [(a_x + a_y)(c_x c_y - b_z c_z) + (a_y + a_z)(c_y c_z - b_x c_x) + (a_z + a_x)(c_z c_x - b_y c_y) \\ &\quad + a_x b_y b_z + a_y b_z b_x + a_z b_x b_y - (a_x c_x^2 + a_y c_y^2 + a_z c_z^2)]. \end{aligned} \quad (13)$$

Equation (3) can now be used to determine  $s$  versus  $T$ .

To study the  $xy$ -type solution we write Eq. (4b) in the form

$$\begin{aligned} a_x + b_x s_x^2 + c_x s_y^2 &= a_y + b_y s_y^2 + c_x s_x^2, \\ s_x^2 + s_y^2 &= s^2, \end{aligned}$$

and obtain

$$\begin{aligned} s_x^2 &= [(b_y - c_x)s^2 + a_y - a_x] / \Delta, \\ s_y^2 &= [(b_x - c_z)s^2 + a_x - a_y] / \Delta, \\ -|\nabla_s \mathbf{H}| &= s [s^2(b_x b_y - c_z^2) + a_x b_y \\ &\quad + a_y b_x - (a_x + a_y)c_z] / \Delta, \end{aligned} \quad (14)$$

$$\Delta = b_x + b_y - 2c_z.$$

In analogy with the  $xyz$  solution, this solution is possible provided that both  $s_x^2$  and  $s_y^2$  as given by Eqs. (14) are non-negative, the vanishing of, say,  $s_x^2$ , signaling a transition into the  $y$ -type phase. Finally, for the  $z$ -type solution

$$\begin{aligned} s_x^2 &= s^2, \\ -|\nabla_s \mathbf{H}| &= s [b_z s^2 + a_z]. \end{aligned} \quad (15)$$

#### IV. PHASE DIAGRAMS FOR ANISOTROPIC QUARTIC SPIN HAMILTONIANS

Leaving the systematic study of the possible phase diagrams for Hamiltonians of various symmetries for the future, we shall now consider a few special cases which exhibit some typical phenomena associated with the system specified by Eq. (8).

$$\text{A. } b_x = b_y = b_z = b, \quad c_x = c_y = c_z = 0$$

In this case the  $xyz$  solution satisfies

$$s_x^2 = \frac{1}{3}s^2 + \frac{a - a_x}{b}, \dots,$$

where  $a = (a_x + a_y + a_z)/3$ . Assuming that  $a_x < a_y < a_z$

point. Otherwise, the temperature corresponding to this point is the lowest metastability temperature of the  $xy$ -phase, the actual (first-order) transition occurring at some higher temperature. To obtain the temperature dependence of the magnetization we write

$$-|\nabla_s \mathbf{H}| = s(\gamma s^2 + \delta), \quad (12)$$

where

$< 0$  and  $b > 0$ , it follows that  $s_x^2 > s_y^2 > s_z^2$  and that  $(a - a_z)/b < 0$  so that the condition  $s_z^2 \geq 0$  is only satisfied for

$$s^2 > s_{xyz}^2 \equiv \frac{3(a_z - a)}{b}.$$

For the  $xy$  solution

$$s_x^2 = \frac{s^2}{2} + \frac{a_y - a_x}{2b}, \quad s_y^2 = \frac{s^2}{2} - \frac{a_y - a_x}{2b}$$

so that  $s_y^2 \geq 0$  provided that

$$s^2 \geq s_{xy}^2 \equiv \frac{a_y - a_x}{b}.$$

These results imply that the transition from the paramagnetic phase, if it is of second order, has to be into a  $z$ -type solution. For  $s^2$  sufficiently large, both the  $z$ -type solutions and the  $xy$ -type solutions are possible, the ground state depending on the free energy. Finally, at yet higher  $s^2$ , the  $xyz$  solution is possible.

Note that for the  $z$ -type phase  $-|\nabla_s \mathbf{H}| = s(a_x + bs^2)$ . Since  $b > 0$  the transition from the paramagnetic to the  $z$ -type phase will always be of second order. The same can be shown to apply to the transition from the  $z$  to the  $xy$  type and from the  $xy$  to the  $xyz$  phase.

As an illustration, Eq. (3) was solved for  $\sigma = \frac{1}{2}$ ,  $a_x = -3$ ,  $a_y = -2$ ,  $a_z = -1$ , and  $b = 15$ . The temperature dependence of the resultant magnetization is presented in Fig. 1, and that of the three components, in Fig. 2. The results indicate a monotonic increase of the resultant magnetization upon lowering the temperature. The magnetization is directed along the  $x$  axis for  $T_{xy} < T < T_x$ , rotates in the  $xy$  plane in the temperature range  $T_{xyz} < T < T_{xy}$ , and finally rotates in space upon further reduction of the temperature. A similar phase, with two nonzero components of the order parameter, whose relative magnitude is temperature dependent, was obtained by Galam and Birman<sup>6,7</sup> who studied the cubic  $xy$  model with eighth-order anisotropy, as well as in a mean-field treatment of the three-component spin system with uniax-

ial symmetry.<sup>17</sup> Galam and Birman<sup>7,8</sup> discussed the relevance of this behavior to the interpretation of the experimentally observed temperature dependence of the ferroelectric ordering in  $\text{Tb}_2(\text{MoO}_4)_3$ .<sup>22,23</sup>

$$\text{B. } a_x < a_y < a_z < 0, \quad b_x, b_y, b_z > 0, \quad c_x = c_y = c_z = 0$$

In this case we readily obtain for the  $xyz$  phase

$$s_x^2 = \frac{\tilde{b}}{3b_x} \left[ s^2 + \left( \frac{a_y - a_x}{b_y} + \frac{a_z - a_x}{b_z} \right) \right], \dots$$

This immediately implies that apart from quantitative differences, this case is very similar to the previous one. The transition between the  $xyz$  and the  $xy$  phases will occur upon vanishing of  $s_z^2$ , when

$$s_{xyz}^2 = \frac{a_z - a_x}{b_x} + \frac{a_z - a_y}{b_y}.$$

As one can readily check by noting that the coefficient of  $s^3$  in  $-|\nabla_s \mathbf{H}|$  is positive, all the transitions will be of second order. Note that whereas the quartic anisotropy associated with  $\mathbf{b}$  is qualitatively irrelevant, the suppression of the quadratic anisotropy, i.e., taking  $a_x = a_y = a_z = a$  results in  $s_x^2 = (\tilde{b}/3b_x)s^2, \dots$ . In the latter case the  $xyz$  phase will be the only stable ordered phase. It is obtained from the paramagnetic phase at  $T_c = \sigma(\sigma + 1)a/3$ . In this case the magnetization direction is temperature independent, the direction cosines being  $(\tilde{b}/3b_i)^{1/2} i = x, y, z$ . This is a generalization of the diagonal phase ( $s_x = s_y = s_z$ ) in the case of cubic anisotropy.<sup>5</sup> A similar phase with a frozen direction in the plane was obtained for the cubic  $xy$  model by Galam and Birman.<sup>6,8</sup>

$$\text{C. } a_x < a_y < a_z < 0, \quad \mathbf{b} = 0, \quad c_x = c_y = 0, \quad c_z < 0$$

In this case the interesting phases are the  $x$  and  $xy$  phases. For the latter,

$$s_x^2 = \frac{1}{2}s^2 + \frac{a_x - a_y}{2c_z}, \quad s_y^2 = \frac{1}{2}s^2 + \frac{a_y - a_x}{2c_z}.$$

This phase will only exist for

$$s^2 \geq s_{xy}^2 = \frac{a_x - a_y}{c_z}.$$

The interesting point is that the transition between the  $x$  and  $xy$  phases will be of first order if  $|c_z|$  is large enough. This is presented in Fig. 3 for  $\sigma = \frac{1}{2}$ ,  $a_x = -3$ ,  $a_y = -2$ , and  $c_z = -16$ . The transition temperature is obtained by evaluating the free energy in both phases, using Eq. (7). In order to determine the value of  $c_z$  at which the transition becomes of first order, we write the magnetization equation for  $\sigma = \frac{1}{2}$  in the inverted form

$$T = [-(a_x + a_y)s - c_z s^3] / \ln \left[ \frac{1 + 2s}{1 - 2s} \right]$$

and note that at  $s = s_{xy}$  we should have  $ds/dT = \infty$  or  $dT/ds = 0$ . Using the magnetization equation and the value of  $s_{xy}$  obtained above we obtain the transcendental equation

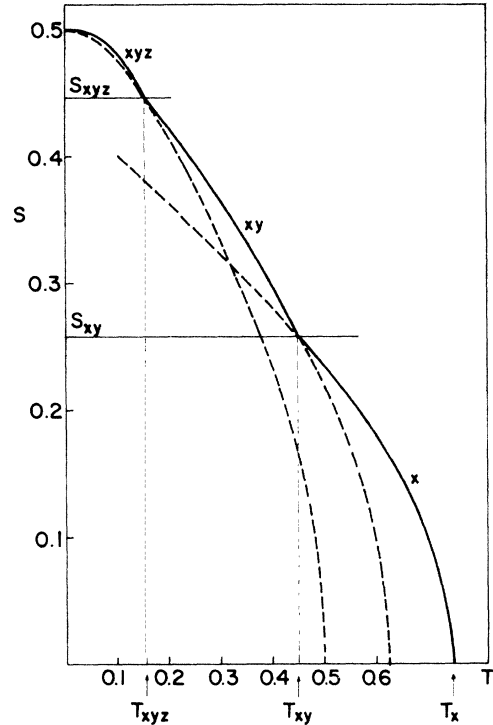


FIG. 1. Resultant magnetization for  $\sigma = \frac{1}{2}$ ,  $a_x = -3$ ,  $a_y = -2$ ,  $a_z = -1$ ,  $b_x = b_y = b_z = 15$ , exhibiting the sequence of transitions  $x \rightarrow xy \rightarrow xyz$  upon lowering the temperature. The solid curve denotes the stable phases and the dashed curves present the unstable solutions.

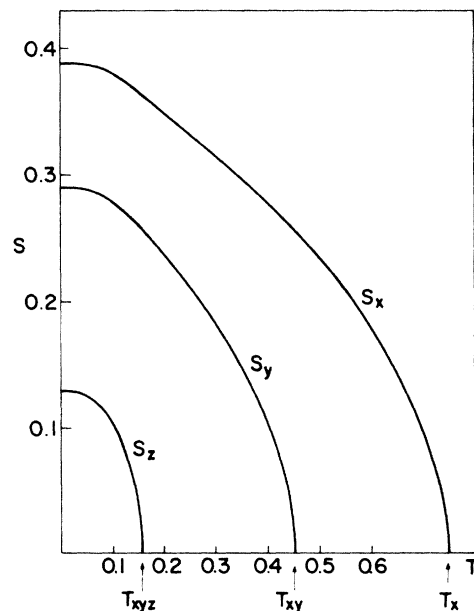


FIG. 2. Components of the magnetization for the case presented in Fig. 1.

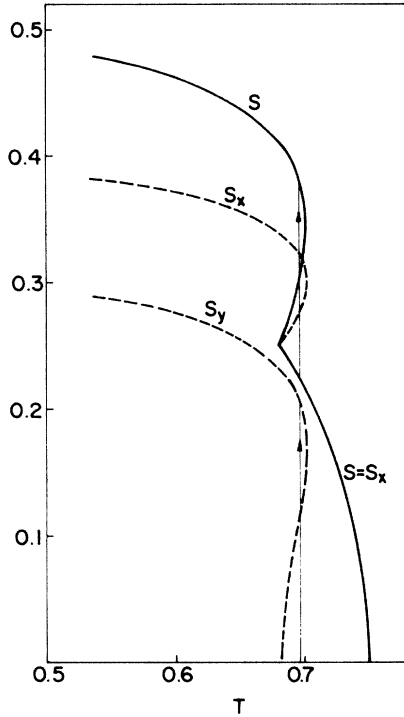


FIG. 3. The magnetization and its components for  $\sigma = \frac{1}{2}$ ,  $a_x = -3$ ,  $a_y = -2$ ,  $a_z = -1$ ,  $c_x = -16$ , exhibiting a first-order phase transition between the  $x$  and  $xy$  phases.

$$\ln \left( \frac{1 + 2s_{xy}}{1 - 2s_{xy}} \right) = \frac{3s_{xy}}{1 - 4s_{xy}^2},$$

which can be solved numerically to yield  $s_{xy} \approx 0.29433$ . For  $a_x = -3$ , and  $a_y = -2$  this corresponds to  $c_x \approx -11.54$ .

## V. CONCLUSIONS

The magnetization equation for arbitrary anisotropic spin Hamiltonians, derived in Ref. 12, was shown to lead to a straightforward procedure of analysis. This procedure was applied to the quartic spin Hamiltonian, Eq. (8). While the possible ordered phases ( $x, xy, xyz$ ) were identified, and the most typical sequences of phase transitions ( $x \rightarrow xy \rightarrow xyz$ ; second or first order) exemplified, a comprehensive search of all the possible sequences remains to be carried out.

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## APPENDIX: DERIVATION OF THE MAGNETIZATION EQUATION IN THE CLASSICAL LIMIT

Using the procedure presented in Ref. 24, the magnetization equation for an isotropic spin Hamiltonian obtains, in the classical limit ( $\sigma = \infty$ ), the form

$$s = \langle \cos\theta \rangle = \frac{\partial \ln Z}{\partial \alpha}, \quad (\text{A1})$$

where

$$Z = \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta e^{-\alpha \cos\theta} = \frac{4\pi}{\alpha} \sinh(\alpha) \quad (\text{A2})$$

and

$$\alpha = \beta \frac{dH}{ds}.$$

Substituting Eq. (A2) in Eq. (A1) we obtain

$$s = \coth(\alpha) - \frac{1}{\alpha} \equiv L(\alpha),$$

where  $L$  is Langevin's function. For an anisotropic spin Hamiltonian, the above procedure results in

$$s_i = \frac{\partial \ln Z}{\partial \alpha_i}, \quad i = x, y, z, \quad (\text{A3})$$

where

$$Z = \int_0^{2\pi} d\varphi \int_0^\pi d\theta \sin\theta e^{-\alpha \hat{s}} \quad (\text{A4})$$

and

$$\alpha = \beta \nabla_s \mathbf{H},$$

$$\hat{s} = (\sin\theta \cos\varphi, \sin\theta \sin\varphi, \cos\theta).$$

Defining

$$\alpha_x = \alpha_0 \cos\varphi_0, \quad \alpha_y = \alpha_0 \sin\varphi_0 \quad (\alpha_0^2 = \alpha_x^2 + \alpha_y^2)$$

we carry out the integration over  $\varphi$ , obtaining

$$\begin{aligned} Z &= 2\pi \int_0^\pi d\theta \sin\theta e^{-\alpha_z \cos\theta} I_0(\alpha_0 \sin\theta) \\ &= 4\pi \sinh(|\alpha|) / |\alpha|, \end{aligned} \quad (\text{A5})$$

where

$$|\alpha| = \beta |\nabla_s \mathbf{H}|.$$

Substituting Eq. (A5) in Eq. (A3) we finally obtain

$$\mathbf{s} = - \frac{\nabla_s \mathbf{H}}{|\nabla_s \mathbf{H}|} L(\beta |\nabla_s \mathbf{H}|), \quad (\text{A6})$$

which is the classical limit of Eq. (2).

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