

**Microscopic approach to orientational order of domain walls**Daniel G. Barci<sup>1</sup> and Daniel A. Stariolo<sup>2,\*</sup><sup>1</sup>*Departamento de Física Teórica, Universidade do Estado do Rio de Janeiro, Rua São Francisco Xavier 524, 20550-013 Rio de Janeiro, Brazil*<sup>2</sup>*Departamento de Física, Universidade Federal do Rio Grande do Sul and National Institute of Science and Technology for Complex Systems, CP 15051, 91501-970 Porto Alegre, RS, Brazil*

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We develop a fully microscopic, statistical mechanics approach to study phase transitions in Ising systems with competing interactions at different scales. Our aim is to consider orientational and positional order parameters in a unified framework. In this paper, we consider two-dimensional stripe-forming systems, where nematic, smectic, and crystal phases are possible. We introduce a nematic order parameter in a lattice, which measures orientational order of interfaces. We develop a mean-field approach that leads to a free energy, which is a function of both the magnetization (density) and the orientational (nematic) order parameters. Self-consistent equations for the order parameters are obtained and the solutions are described for a particular system, the dipolar frustrated Ising ferromagnet. We show that this system has an Ising-nematic phase at low temperatures in the square lattice, where positional order (staggered magnetization) is zero. At lower temperatures, a crystal-stripe phase may appear. In the continuum limit, the present approach connects to a Ginsburg-Landau theory, which has an isotropic-nematic phase transition with breaking of a continuous symmetry.

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**I. INTRODUCTION**

Systems with competing interactions are a rule in nature. Competing tendencies are usually responsible for the complex behavior of natural systems, leading to slow dynamics, metastability, and complex free-energy landscapes, as observed in spin glasses and many other frustrated systems. Competing interactions at different scales may give rise to complex phases and patterns, such as stripes, lamellae, bubbles, and others.<sup>1</sup> Examples range from solid-state systems, such as ultrathin ferromagnetic films<sup>2,3</sup> and strongly correlated electron systems,<sup>4,5</sup> to soft-matter systems, such as Langmuir monolayers,<sup>6</sup> block copolymers,<sup>7,8</sup> colloids, and soft-core systems.<sup>9-11</sup> Aside from the intrinsic interest raised by the complexity of the phase behavior in these systems, their detailed knowledge may be critical for understanding basic phenomena such as high-temperature superconductivity, and also for technological applications such as soft-matter templates for nanoscale systems and future spintronic devices.

Systems with competing interactions at different scales, e.g., a short-range attraction and a long-range repulsion, as present in magnetic films and low-dimensional electronic systems, have a tendency to form *microphases*, i.e., to phase separate at mesoscopic scales, due to the frustration usually present as a consequence of competing effects. These microphases can show at least two types of ordering: positional ordering of the microscopic degrees of freedom and also orientational order of interfaces or domain walls, due to the presence of interfaces at mesoscopic scales. The presence of orientational order allows for an analogy with liquid-crystal phenomenology, where nematic, smectic, and crystal phases can in principle be characterized. Nevertheless, a complete characterization of the phases present in these systems is rare, and usually only positional order parameters are computed and the corresponding phase diagrams are well known.<sup>12-16</sup> In stripe-forming systems, such as the ones studied in the previous references, these mean-field approaches give access

only to modulated crystalline phases, or stripe crystals, where both positional and orientational long-range order are simultaneously present. In fact, the morphologies of domains observed in real films, such as the magnetization patterns in ultrathin ferromagnetic films,<sup>2,3,17</sup> for example, give clear indications that orientational order sets in well before any positional order can appear. It is necessary to go beyond the usual mean-field approaches in order to describe the complete ordering process. To our knowledge, there have been very few attempts to compute full phase diagrams with simultaneous and independent consideration of orientational and positional orders. One of these few works is the phase diagram for ultrathin ferromagnetic films with square lattice anisotropy anticipated by Abanov and co-workers.<sup>18</sup> These authors have analyzed the phases of a Heisenberg spin system in the square lattice with competing exchange and dipolar interactions by means of a mixture of microscopic calculation with phenomenological assumptions. Their analysis gave a very rich phase diagram, with isotropic, Ising-nematic, and smectic phases. Phenomenological approaches are common ground for the analysis of orientational order. Starting from an elastic energy, which assumes a crystalline ground state, specific fluctuations of the ground state can be studied perturbatively. A complete analysis of two-dimensional smectic elasticity was done 30 years ago by Toner and Nelson.<sup>19</sup>

Another interesting approach to study the phase transitions in systems with isotropic competing interactions was due to Brazovskii.<sup>20</sup> By analyzing a generic Ginsburg-Landau model, the main characteristic of which is the presence of a minimum in the spectrum of Gaussian fluctuations at a nonzero wave vector, he showed, in the context of a self-consistent field approximation, that the model has a first-order transition to a modulated phase, induced by fluctuations. While conceptually important, this result suffers from the same problems as other mean-field approaches, namely, it describes phases where both orientational (modulated) and

positional order sets in simultaneously and has very strong transversal fluctuations in two dimensions.<sup>21</sup> In a recent work, we have gone beyond the Brazovskii approximation by considering other terms in the Ginsburg-Landau free energy of two-dimensional systems, which are dictated by the symmetry of the interactions.<sup>22,23</sup> Interestingly, pure symmetry considerations lead to terms in the free energy that encode orientational order parameters. We were able to find a nematic phase at temperatures above the original Brazovskii modulated phase. By analyzing fluctuations of the mean-field nematic solution, we further found that the isotropic-nematic phase transition in the continuum system with isotropic interactions is of the Kosterlitz-Thouless (KT) type.<sup>23</sup> These works set the stage for a complete renormalization-group treatment of this kind of system. Nevertheless, universality in two dimensions is a delicate matter, and it would be important to get contact with the phenomenological theory from more microscopic, interaction-specific, models.

In this paper, we give a step further in the description of nematic order in systems with competing interactions, and develop a fully microscopic statistical mechanical approach, which allows us to consider positional as well as orientational order parameters on equal grounds. We study Ising systems with arbitrary competing interactions on a square lattice. We introduce a nematic order parameter on the lattice by analogy with the order parameter studied in the framework of the Ginsburg-Landau model.<sup>23</sup> Then, we develop a mean-field approach, which includes both positional (magnetization) and orientational (nematic) order parameters in the free energy. Due to the two-body nature of the orientational order parameter, it is necessary to go beyond one-site approximations in order to compute the mean-field free energy. A set of self-consistent equations for the order parameters, equivalent to the Bragg-Williams approximation for the Ising model, is obtained and the solution for a model system presented. We computed the phase diagram of the dipolar frustrated Ising ferromagnet (DFIF), a well-known and very studied model for ultrathin magnetic films with perpendicular anisotropy.<sup>24–27</sup> We show that this model has an Ising-nematic phase in the square lattice, where only orientational order is present. At lower temperatures, a further transition to a modulated phase with positional order, the stripe phase, is possible. In the continuum limit, our approach leads to the Ginsburg-Landau model discussed before.<sup>22</sup> It is also shown that, when considering the nematic order parameter, the magnetic (spin-spin) susceptibility diverges at a higher temperature compared with the usual mean-field calculation, in which the orientational terms in the free energy are absent.

The paper is organized as follows: In Sec. II, we introduce the nematic and the Ising-nematic order parameters. In this section, we also develop a mean-field approach to deal with general competing interactions in the square lattice. In Sec. III, we apply our technique to the dipolar frustrated Ising ferromagnet. Finally, we discuss our results and conclusions in Sec. IV.

## II. MICROSCOPIC NEMATICLIKE ORDER PARAMETER

Figure 1 shows a typical pattern of magnetic domains in an ultrathin ferromagnetic film of Fe/Cu(001) with strong

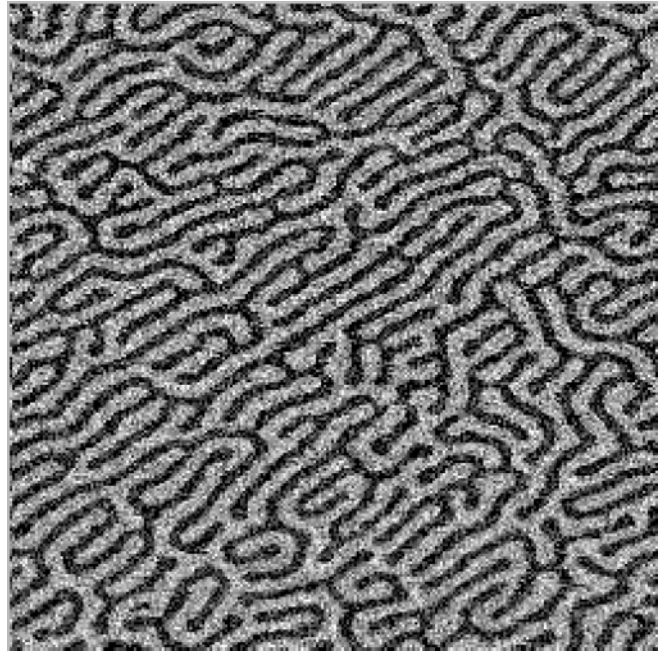


FIG. 1. Image of domain patterns in a Fe/Cu(001) ultrathin film, taken by scanning electron microscopy with polarization analysis (SEMPA) (courtesy of D. Pescia, ETH, Zurich).

perpendicular anisotropy. In this system, the competing interactions are the short-range exchange interaction and the long-range dipolar interaction. It is clear that this pattern does not present positional order, but it is possible to distinguish some degree of orientational order in the striplike pattern. We want to define a suitable order parameter for this kind of order and quantify it.

Domain walls are observed at the transitions between positive and negative values of the perpendicular magnetization. In an appropriate mesoscopic scale, it is possible to define a continuum magnetization density  $\Phi(\vec{x})$ . The gradient of this quantity

$$\mathbf{N}(\vec{x}) \equiv \nabla\phi(\vec{x}) = (\partial_x\phi, \partial_y\phi) \quad (1)$$

naturally defines a director that quantifies the degree of orientation of domain walls.

However, the kind of order we are looking for is insensitive to the vector orientation, as shown in Fig. 2. Therefore, in analogy with the nematic order parameter of liquid crystals, it is possible to define a local tensor order parameter

$$Q_{ij}(\vec{x}) \equiv \phi(\vec{x})(\partial_i\partial_j - \frac{1}{2}\partial^2\delta_{ij})\phi(\vec{x}), \quad (2)$$

where  $i, j = \{x, y\}$ . This symmetric and traceless tensor has, in two dimensions, only two independent elements, which essentially represent the mean orientation of domain walls and the strength of the orientational order. Similarly to the vector order parameter, a nonzero value of  $Q_{ij}$  represents a breakdown of the rotational symmetry  $O(2)$ . However, differently from a vector, this order parameter is invariant under rotations by  $\pi$  characterizing a nematic symmetry.

A comprehensive description of the physical meaning of this order parameter can be found in Ref. 28. Since the tensor is symmetric, it can be diagonalized and, in the principal axis

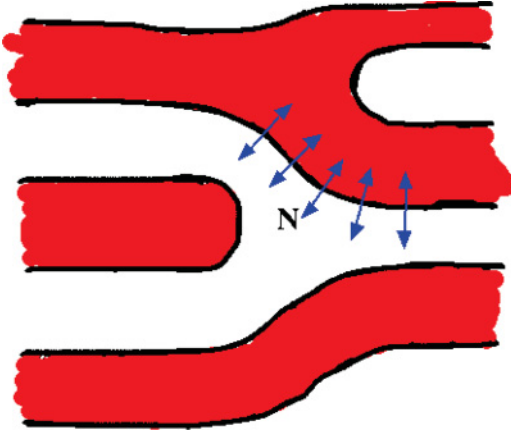


FIG. 2. (Color online) Schematic representation of a stripe phase with a dislocation. The arrows represent the director field, which is a local measure of orientation of domain walls.

base, it has only one independent component, which can be written as

$$\langle Q \rangle = \int d^2k k^2 \cos(2\theta) C(\vec{k}), \quad (3)$$

where  $C(\vec{k})$  is the structure factor of the system and  $\vec{k} = k(\cos\theta, \sin\theta)$ . This expression makes possible the computation of  $\langle Q \rangle$  from experimental or numerical simulation data, since the structure factor is a usual observable. In this way, the orientational order parameter quantifies the degree of anisotropy of the domain pattern [i.e., the anisotropy in  $C(\vec{k})$ ].

In Ref. 22, it was shown that this nematiclike order parameter emerges naturally from symmetry considerations in the Landau expansion of the free energy of a system with isotropic competing interactions. At mean-field level plus Gaussian fluctuations,<sup>23</sup> the phase transition turns out to be in the Kosterlitz-Thouless universality class. This seems to be a robust result, and was originally obtained in a somewhat different context by Toner and Nelson.<sup>19</sup>

Due to the ubiquity of orientational phases, intermediate between crystal and fluid ones, in a great variety of condensed matter systems, it is important to have a unified microscopic description. This would also allow us to make closer contact with experiments and simulations. With this motivation, and also trying to clarify the nature of the nematiclike interaction term in the Ginsburg-Landau free energy analyzed in Refs. 22 and 23, in this paper we develop a general classic statistical mechanics approach to obtain the phase diagram of generic competitive systems with Ising variables.

Consider the following Ising Hamiltonian on a square lattice:

$$\mathcal{H} = \frac{1}{2} \sum_{i,j} J_{ij} S_i S_j - \sum_i B_i S_i, \quad (4)$$

where  $\{S_i = \pm 1, i = 1, \dots, N\}$  are  $N$  Ising variables, the sum runs over all  $i, j$  pairs in the lattice, and  $B_i$  is an external magnetic field. The interaction matrix  $J_{ij}$  is assumed to have a ferromagnetic part  $J_{ij} = J < 0$  if  $i, j$  are nearest-neighbor sites (NNS) and an antiferromagnetic part  $J_{ij} > 0$  if  $i, j$  are not NNS. These are the only constraints in order to apply the method.

Consider a discrete version of the nematic order parameter defined in (2) coupled to a conjugate external nematic field through

$$h_k^{ab} \left( \Delta_a S_k \Delta_b S_k - \frac{\delta_{ab}}{2} (\Delta S_k)^2 \right), \quad (5)$$

where  $k$  is the lattice index,  $a, b = x, y$ , and  $\Delta_a$  are lattice derivatives. The field  $h_k^{ab}$  has the same symmetries as the tensor order parameter, then we can choose the coordinate system along the principal axes and write the conjugate field  $h_k^{ab}$  as

$$h_k^{ab} = \begin{pmatrix} h_k & 0 \\ 0 & -h_k \end{pmatrix}. \quad (6)$$

The coupling term of Eq. (5) can be written as

$$\frac{1}{2} \sum_{ij} h_i S_i K_{ij} S_j, \quad (7)$$

where the matrix  $K$  is given by

$$K_{ij} = \begin{cases} +1 & \text{if } j = i \pm \hat{x}, \\ -1 & \text{if } j = i \pm \hat{y}, \end{cases} \quad (8)$$

$\hat{x}$  and  $\hat{y}$  are unit vectors along the  $x$  and  $y$  axis of the square lattice. In this way, the Hamiltonian now reads as

$$\mathcal{H} = \frac{1}{2} \sum_{i,j} (J_{ij} - h_i K_{ij}) S_i S_j - \sum_i B_i S_i. \quad (9)$$

The local orientational order parameter is defined as

$$\langle N_i \rangle = \frac{1}{\beta} \frac{\partial \ln Z}{\partial h_i} = \langle S_i S_{i+\hat{x}} - S_i S_{i+\hat{y}} \rangle, \quad (10)$$

where  $Z = \sum_{\{S\}} \exp\{-\beta\mathcal{H}\}$  is the canonical partition function, with  $\beta = 1/k_B T$ . This order parameter has only one component and not two, as the one of Eq. (2). If  $\langle N_i \rangle$  is positive, the director points along the  $x$  direction, while if it is negative, the director mainly points in the  $y$  direction. These are the only possible directions of the director. For this reason, if  $\langle N_i \rangle \neq 0$ , the resulting anisotropic phase is called Ising Nematics since it breaks the rotational point group of the lattice and it is invariant under rotations by  $\pi$ . Along the paper, for brevity, we generally use the term “nematic” to refer to this phase, however, whenever we deal with a square lattice model, “Ising Nematic” should be understood.

The global order parameter can be written as

$$Q = \frac{1}{2} \sum_{ij} K_{ij} \langle S_i S_j \rangle. \quad (11)$$

Equation (11) is completely analogous to the continuous version given by Eq. (3). The anisotropic matrix  $K$  for the Ising-Nematic phase plays the same role of  $k^2 \cos 2\theta$  for the nematic one. Thus, in the same way that Eq. (3) measures the degree of anisotropy of the structure factor  $C(\vec{k})$ , the order parameter (11) describes the degree of anisotropy in the nearest-neighbor correlation functions between the  $x$  and  $y$  directions of the lattice. A slightly different form of this order parameter has been used before in simulations of stripe-forming systems.<sup>26,29,30</sup> In this work, we attempt to compute it in a statistical mechanics framework.

The technical problem of computing the order parameter reduces to the calculation of nearest-neighbor correlation functions in a proper approximation. In this paper, we describe a mean-field-like approximation based on the use of a Hubbard-Stratonovitch (HS) transformation.

Introducing a real variable on the lattice ( $\Phi_i$ , where  $i$  is the lattice index) by means of a HS transformation on the original Hamiltonian (4) (see, e.g., Ref. 31) and exactly summing up the Ising variables  $S_i$ , we obtain an effective Hamiltonian given by

$$\mathcal{H}_{\text{eff}}[\{\Phi\}] = \frac{1}{4} \sum_{ij} \Phi_i J_{ij} \Phi_j - \frac{1}{2} \sum_i B_i \Phi_i - \frac{1}{\beta} \sum_i \log \cosh \left( \beta \sum_j J_{ij} \Phi_j \right). \quad (12)$$

By using the general relation between correlations of the original discrete and continuous variables<sup>31</sup>

$$\langle S_i S_j \rangle = -\frac{1}{2\beta} J_{ij}^{-1} + \frac{1}{4} \langle \Phi_i \Phi_j \rangle \quad (13)$$

for the cases of interest in this work, of an isotropic interaction matrix  $J_{ij}$ , the order parameter reads as

$$Q = \frac{1}{8} \sum_{ij} K_{ij} \langle \Phi_i \Phi_j \rangle. \quad (14)$$

### A. Mean-field approximation for the order parameter $Q$

To compute  $Q$  given by Eq. (14), we begin by considering the partition function

$$Z = \mathcal{N} \int \mathcal{D}\Phi e^{-\beta H_{\text{eff}}(\Phi)}, \quad (15)$$

where  $\mathcal{N}$  is a normalization constant and  $H_{\text{eff}}(\Phi)$  is given by Eq. (12). We want to introduce an order parameter with nematic symmetry. For this purpose, we introduce a symmetric traceless tensor  $Q_k^{a,b}$ , where  $k$  is the lattice index and  $a, b = x, y$  and write the partition function as

$$Z = \mathcal{N}' \int \mathcal{D}\Phi e^{-\beta H_{\text{eff}}(\Phi)} \int \mathcal{D}Q e^{-\beta \sum_k \text{Tr}(Q_k^2)}, \quad (16)$$

where  $\mathcal{D}Q = \prod_k dQ_k^{a,b}$ . Note that, in the square lattice, the introduction of a fully symmetric traceless tensor of rank two is redundant, since the Ising-Nematic order parameter has only one independent component. However, we prefer to use this notation to stress that the method is general and can be also used to treat continuous systems with  $O(2)$  symmetry, where the full tensor is needed to describe the phase transition.

The Gaussian integral in  $Q$  is simply a constant that can be absorbed in the normalization factor  $\mathcal{N}'$ . Shifting  $Q_k \rightarrow Q_k - N_k(\Phi)$ , keeping the measure  $\mathcal{D}Q$  invariant, we have

$$Z = \mathcal{N}' \int \mathcal{D}\Phi e^{-\beta H_{\text{eff}}(\Phi)} \int \mathcal{D}Q e^{-\beta \sum_k \text{Tr}(Q_k - N_k)^2}, \quad (17)$$

where the symmetric traceless tensor  $N_k(\Phi)$  will be defined later.

At mean-field level,  $Q_k^{ab}$  is given by the saddle-point equation

$$\frac{\partial \ln Z}{\partial Q_k^{ab}} = 0, \quad (18)$$

which reads as

$$Q_k^{ab} = \langle N_k^{ab} \rangle. \quad (19)$$

The average is given by

$$\langle N_k^{ab} \rangle = \frac{1}{Z(Q)} \int \mathcal{D}\Phi N_k^{ab}(\Phi) e^{-\beta [H_{\text{eff}}(\Phi) + \sum_k \text{Tr}(Q_k - N_k)^2]}. \quad (20)$$

Choosing the coordinate system along the  $Q_k^{ab}$  principal axes

$$Q_k^{ab} = Q_k \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (21)$$

and

$$N_k^{ab} = N_k(\Phi) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (22)$$

and defining  $\sum_k N_k = \frac{1}{8} \sum_{ij} K_{ij} \Phi_i \Phi_j$ , the effective Hamiltonian takes the form

$$\begin{aligned} H[\{\Phi_i, Q_i\}] &= \frac{1}{4} \sum_{ij} \Phi_i J_{ij} \Phi_j - \frac{1}{\beta} \sum_i \log \cosh \left( \beta \sum_j J_{ij} \Phi_j \right) \\ &\quad + 2 \sum_i (N_i)^2 + 2 \sum_i Q_i^2 - 4 \sum_i Q_i N_i, \end{aligned} \quad (23)$$

where the external magnetic field  $B_i$  has been set to zero. This is a nonquadratic Hamiltonian in the variables  $\Phi_i$  in the presence of a ‘‘mean-field’’ orientational order parameter  $Q$ . The next step is to integrate the partition function in  $\Phi_i$  considering quadratic fluctuations of  $\Phi_i$  around a saddle-point approximation.

Thus, the expansion of  $H$  up to quadratic order in the  $\Phi$  fluctuations reads as

$$H(\Phi) = H(\Phi_{SP}) + \frac{1}{2!} \sum_{ij} H''_{ij} |_{\Phi=\Phi_{SP}} \delta\Phi_i \delta\Phi_j, \quad (24)$$

where  $\Phi_{SP}$  is the saddle-point solution  $H'(\Phi_{SP}) = 0$ ,  $H''_{ij} = \frac{\partial^2 H}{\partial \Phi_i \partial \Phi_j}$ , and  $\delta\Phi = \Phi - \Phi_{SP}$ .

In this approximation,  $\langle \delta\Phi_i \delta\Phi_j \rangle = \frac{1}{4\beta} (H''_{ij})^{-1}$  and then the mean-field equation for  $Q$  [Eq. (14)] reduces to

$$Q = \frac{1}{32\beta} \sum_{ij} K_{ij} (H''_{ij})^{-1}. \quad (25)$$

In order to get the desired self-consistent equation for  $Q$ , it is necessary to find an explicit expression for  $H''(\Phi_{SP})$ . After a lengthy but straightforward calculation, we obtain for the Hessian

$$\begin{aligned} H''_{lm} |_{SP} &= \frac{1}{2} J_{lm} - \beta \sum_i \frac{J_{il} J_{im}}{\cosh^2(\beta \sum_k J_{ik} \Phi_k^{SP})} - Q K_{lm} \\ &\quad + \frac{1}{16} \sum_k (\Phi_k^2)_{SP} K_{kl} K_{km}. \end{aligned} \quad (26)$$

By inserting this result in (25), a self-consistent equation for  $Q$  is obtained. It depends on the field  $\Phi^{SP}$  and its square, which can be computed within the same mean-field approximation.

The previous result allows, in principle, the calculation of positional ( $\Phi$ ) and orientational ( $Q$ ) order parameters. We expect that a purely orientational phase, with  $Q \neq 0$  and  $\Phi^{SP} = 0$ , which will be identified with a nematic phase with orientational order of domain walls, but without positional order, may be present for a class of systems with competing interactions. Considering this situation, the simplest self-consistent approximation for the nematic order parameter reads, in matrix notation, as

$$Q = \frac{1}{16\beta} \text{Tr} \left\{ \frac{K}{J - 2\beta J^2 - 2QK} \right\}. \quad (27)$$

This self-consistent equation for  $Q$  is the analog of the Curie-Weiss approximation for the magnetization in the Ising model. This is one of the main results of our paper. If Eq. (27) has a nontrivial solution  $Q \neq 0$ , then the system exhibits an anisotropic but homogeneous phase with nematic symmetry. The presence, or not, of this phase depends of the detailed structure of the competing interactions, coded in the explicit form of the matrix  $J$ .

Guided by the results of the continuum Ginsburg-Landau model,<sup>22</sup> we search for a critical point signaling an isotropic-nematic transition: Defining  $A = J - 2\beta J^2$  and expanding the right-hand side of (27) in  $Q \text{Tr}(K/A) \ll 1$ , we have

$$16\beta Q \approx \text{Tr} \left( \frac{K}{A} \right) + 2Q \text{Tr} \left( \frac{K}{A} \right)^2 + 4Q^2 \text{Tr} \left( \frac{K}{A} \right)^3 + 8Q^3 \text{Tr} \left( \frac{K}{A} \right)^4. \quad (28)$$

If  $A$  is rotationally invariant (invariant under discrete rotations in the square lattice), then  $\text{Tr}(\frac{K}{A}) = \text{Tr}(\frac{K}{A})^3 = 0$ . Then,  $Q = 0$  is always a solution of the saddle-point equations. If  $Q \neq 0$ ,

$$\left[ 16\beta - 2 \text{Tr} \left( \frac{K}{A} \right)^2 \right] Q - 8Q^3 \text{Tr} \left( \frac{K}{A} \right)^4 = 0, \quad (29)$$

and then, for  $Q \ll 1$ ,

$$Q \approx \frac{1}{2} \left[ \frac{8\beta - \text{Tr} \left( \frac{K}{A} \right)^2}{\text{Tr} \left( \frac{K}{A} \right)^4} \right]^{1/2}. \quad (30)$$

This result implies a continuous, second-order *isotropic-nematic* transition at a critical temperature given by

$$\beta_c = \frac{1}{8} \text{Tr} \left( \frac{K}{A(\beta_c)} \right)^2. \quad (31)$$

The existence of a solution of Eq. (31) depends on the detailed structure of the matrix  $A$ . In the next section, we will show an explicit calculation on a model system in which this transition is present.

### III. DIPOLAR FRUSTRATED ISING FERROMAGNET

The dipolar frustrated Ising ferromagnet (DFIF) is a simple model for studying the thermodynamic phases of ultrathin ferromagnetic films with strong perpendicular anisotropy.<sup>2,3,25-27</sup>

In the strong anisotropy limit, the anisotropic term of the classical dipole-dipole interactions is zero because the dipolar moments point perpendicular to the lattice plane, and the system degrees of freedom are well represented by Ising variables pointing perpendicular to the plane of the lattice. Furthermore, the strong uniaxial (perpendicular) anisotropy renders the energy symmetric with respect to rotations on the lattice. The Hamiltonian with competition between short-range exchange and long-range dipolar interactions can be written as<sup>26</sup>

$$\mathcal{H} = -\frac{J}{2} \sum_{(i,j)} S_i S_j + \frac{g}{2} \sum_{(i,j)} \frac{S_i S_j}{r_{ij}^3}. \quad (32)$$

The first sum runs over all pairs of nearest-neighbor spins, and the second one over all pairs of spins of the lattice;  $r_{ij}$  is the distance, measured in lattice units, between sites  $i$  and  $j$ . The relevant parameter is the ratio between the exchange  $J > 0$  and the dipolar  $g > 0$  intensities. Then, we fix  $g = 1$  without losing generality. Note that the ferromagnetic, short-range exchange interaction is frustrated by the long-range, antiferromagnetic dipolar interaction. The possibility of an Ising nematic phase in these systems has been anticipated theoretically by Abanov *et al.*,<sup>18</sup> and numerical evidence from Monte Carlo simulations has been reported in Ref. 26. Several experimental works have reported results that show domain patterns that could be identified with a nematic phase,<sup>2,3</sup> but up to now, its characterization and properties have not been discussed. Also, several theoretical, mainly numerical works have shown results of orientational order parameters,<sup>32-34</sup> but no quantitative characterization or distinction between, e.g., stripe, smectic, and nematic phases have been attempted so far.

#### A. Nematic phase

We have numerically solved Eq. (31) for the DFIF model (32) in the thermodynamic limit where the linear size of the system  $L \rightarrow \infty$ . In reciprocal space, Eq. (31) reads as

$$\beta_c = \frac{1}{8} \int \frac{d^2k}{(2\pi)^2} \left[ \frac{K(\vec{k})}{J(\vec{k})[1 - 2\beta_c J(\vec{k})]} \right]^2, \quad (33)$$

where  $J(\vec{k})$  and  $K(\vec{k})$  are the Fourier transforms of the matrices  $J_{ij}$  and  $K_{ij}$ . The anisotropic function  $K(\vec{k})$  in the square lattice is given by

$$K(\vec{k}) = 2(\cos k_x a - \cos k_y a), \quad (34)$$

where  $a$  is the lattice spacing. The Fourier transform of the interaction function is given by

$$J(\vec{k}) = -J L_{nn}(\vec{k}) + g L_{dip}(\vec{k}), \quad (35)$$

where the the Fourier transform of the nearest-neighbor interaction  $L_{NN}(\vec{k}) = 2J(\cos k_x a + \cos k_y a)$  and the dipolar interaction for small  $k$  is approximated<sup>35</sup> by  $L_{dip}(\vec{k}) \approx 1 - ka + (ka)^2/4$ .

We have solved Eq. (33) in two different limits: keeping the full structure of the functions  $K(\vec{k})$  and  $J(\vec{k})$  where the lattice symmetry is preserved, and keeping only the leading-order

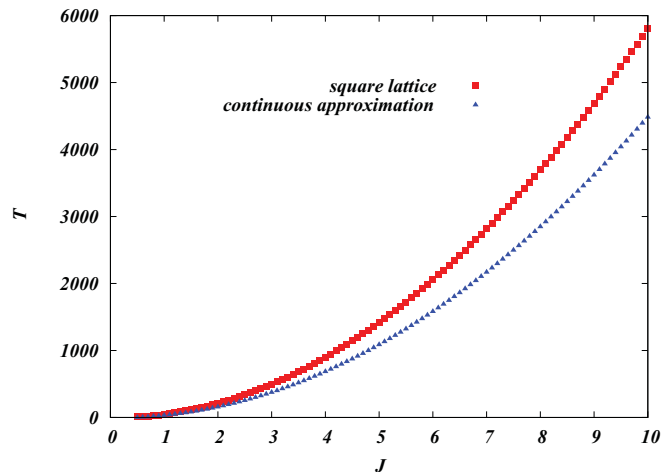


FIG. 3. (Color online) Isotropic-nematic transition line in the dipolar frustrated Ising ferromagnet.

terms in  $ka \ll 1$ , where the effects of the lattice are suppressed (continuum approximation). The phase diagram in the  $T, J$  plane is shown in Fig. 3 for the two cases considered. Both cases give similar results in this mean-field approach. The temperature scale depends on the value of the cutoff needed to get convergence of the integrals. This is a well-known limitation of the HS transformation<sup>36</sup> when the interaction matrix is not positive definite, as in this case. In any case, as the approximation is of a mean-field nature, we only expect that the phase diagram be qualitatively correct.

An isotropic-nematic transition without positional order,  $\langle \Phi \rangle = 0$ , is obtained in both cases. It is important to stress that, in the full lattice calculation, the low-temperature phase is in fact an Ising nematic, while in the continuum limit, the phase is a nematic one. This last case corresponds to the result found in Ref. 22 from a phenomenological Ginsburg-Landau model. A fundamental difference between both cases arises in the nature of the transitions. While in the continuum case the transition is of the KT type, in the first case, the universality class is probably Ising (see Ref. 18 for a discussion of different scenarios when lattice anisotropy is considered), but this remains to be proved for this model. The present microscopic approach for the nematic phase has the Ginsburg-Landau Hamiltonian studied in Refs. 22 and 23 as the continuum limit. Note that the continuum limit of Eq. (27) corresponds to Eq. (12) of Ref. 22. Interestingly, orientational order develops at higher temperatures for systems where the competing interaction is weak, i.e., for weak frustration. This corresponds to the experimental situation in ultrathin ferromagnetic films, where visual inspection of the domains formed point to the existence of a nematic phase.<sup>2,37</sup>

In Fig. 4, we show the order parameter  $Q$  near the transition, obtained solving Eq. (30) for the lattice case. It shows the second-order nature of the phase transition. This is an expected result since, in two dimensions, the nematic symmetry only allows even powers of the order parameter (nematic as well as Ising Nematics) in the free energy.<sup>38</sup> This is different from three dimensions where the transition is of first order. However, as was already indicated, fluctuations could probably drive the transition to a different class.

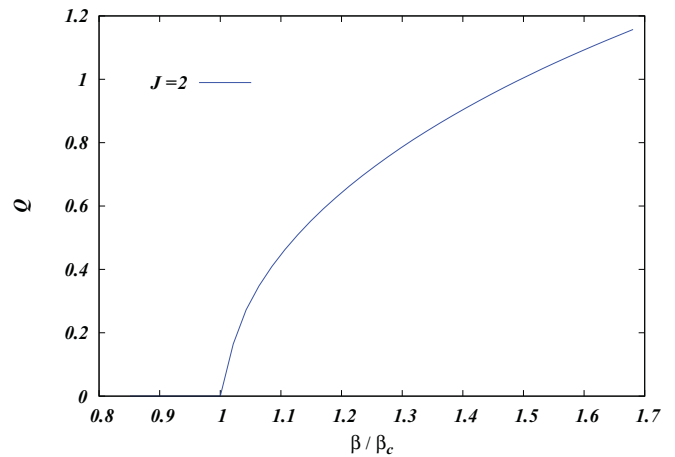


FIG. 4. (Color online) Mean-field nematic order parameter for  $J = 2$ .

### B. Positional order

At lower temperatures, aside from the orientational order represented by the nematic phase, positional order may also emerge. Within the present framework, the positional order parameter is given by the density field  $\Phi_i$ , which can be computed together with  $Q(T)$  from Eqs. (23) and (26). We have not pursued this in this work, which is devoted to the description of the nematic phase. Instead, it is interesting to analyze the behavior of the two-point spin correlation function in the nematic phase, which gives information of the possible growth of positional order at high temperatures.

The spin-spin correlation function [Eq. (13)] in reciprocal space reads as

$$G(\vec{k}) = \frac{1}{J(\vec{k}) - 2\beta J(\vec{k})^2 - 2QK(\vec{k})} - \frac{1}{J(\vec{k})}. \quad (36)$$

This equation must be solved self-consistently with Eq. (27). If the term proportional to  $Q(T)$  in the denominator is disregarded, we end with the usual, mean-field approach to the computation of modulated phases. In Fig. 5, we show the function  $G(\vec{k})$  for  $Q = 0$  for a temperature slightly above the mean-field critical temperature, which in this case is given by  $T_c = \max_{\vec{k}} J(\vec{k})$ . The high-temperature profile of  $G(\vec{k})$  is isotropic and a phase transition to a modulated phase with characteristic wave vector  $k_0 \neq 0$  takes place through a breaking of a continuous rotational symmetry. This is the ‘‘Brazovskii’s scenario’’ for the isotropic-stripes phase transition in systems with nearly isotropic competing interactions.<sup>20</sup> Instead, upon inclusion of the nematic order parameter correction, the spectrum above the low-temperature modulated phases changes in an essential way. In fact, because the nematic transition breaks rotational symmetry, the structure factor is anisotropic in this phase, as shown in Fig. 6, again for a characteristic temperature just above the transition to a modulated phase with positional order (divergence of a staggered magnetic susceptibility). Note that the spectrum has a broad support in the plane of wave vectors, a clear indication that there is no positional order in the nematic phase. Nevertheless, it shows two well-defined maxima at characteristic wave vectors  $\pm \vec{k}_0$ , which show the  $\pi$ -rotation

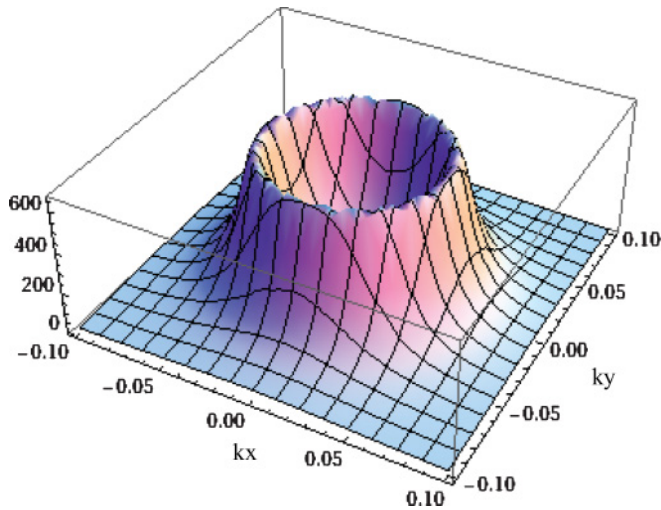


FIG. 5. (Color online) Structure factor of the DFIF near the paramagnetic-stripes phase transition, without considering the nematic correction term, computed from Eq. (36) for  $J = 10$ .

symmetry of the nematic phase. Furthermore, both peaks are on the  $x$  axis, a consequence of the breaking of continuous symmetry already imposed by the square lattice. This is the structural signature of an Ising-nematic phase.

Another interesting question regards the (in)commensurability of the characteristic wave vectors. In this solution, as expressed, e.g., by Eq. (36), it is clear that the wave vector that maximizes the structure factor in the nematic phase depends continuously on temperature. Then, at least in the nematic phase, the characteristic wave vectors are incommensurate with the lattice.

A very different situation arises in short-ranged interaction models such as anisotropic next-nearest-neighbor Ising<sup>39</sup> (ANNNI) or the biaxially next-nearest-neighbor Ising model (BNNNI).<sup>40</sup> This class of models could be similarly treated

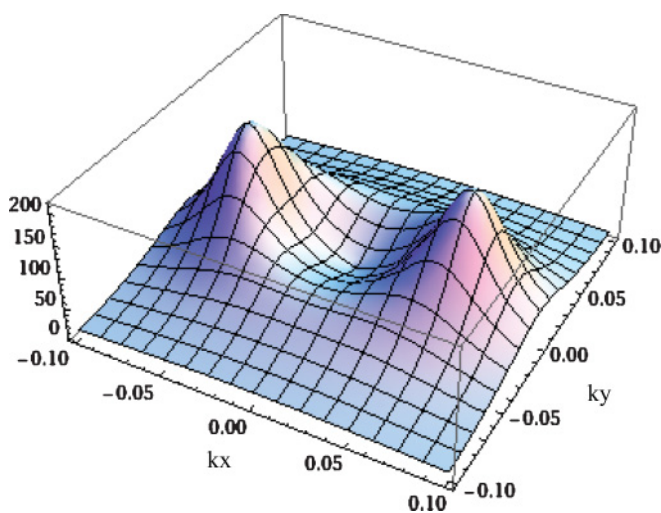


FIG. 6. (Color online) Structure factor of the DFIF in the nematic phase near the nematic-stripes phase transition for  $J = 10$ .

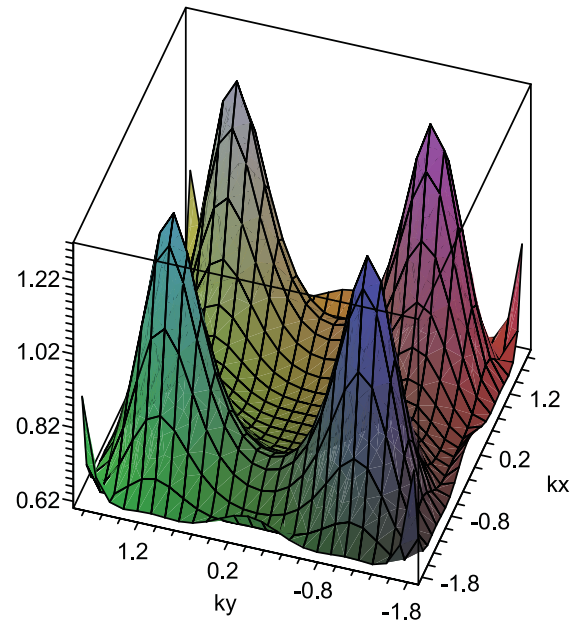


FIG. 7. (Color online) Structure factor of the BNNNI model near the paramagnetic-stripes phase transition, computed from Eqs. (36) and (37).  $\delta = 1.8$  and  $\beta = 1/13$ .

within our formalism. For instance, the Fourier transform of the interaction matrix  $J_{ij}$  for the BNNNI model reads as

$$J(k) = 2\delta[\cos(k_x a) + \cos(k_y a)] - 2[\cos(2k_x a) + \cos(2k_y a)], \quad (37)$$

where  $a$  is the lattice constant and  $\delta$  measures the competition between first- and second-neighbor interactions. In Fig. 7, we show the structure factor of this model, computed from Eq. (36) in the high-temperature phase. We observe the presence of four peaks obeying the lattice symmetry. The peaks' weights grow as the temperature is lowered, signaling the onset of positional order. However, the position of the peaks in reciprocal space is given by  $\cos(k_x a) = \cos(k_y a) = \delta/2$ , which does not depend on temperature. This fact allows a possible incommensurate as well as a commensurate positional order at low temperatures. Concerning the main focus of this paper (the nematic phase), we have computed the self-consistent equation (31) for the nematic critical temperature and we have found no solution for this case. This is consistent with the general belief that it is necessary to have a macroscopically degenerate number of ordering wave vectors to produce a pure orientational ordered phase (compare Fig. 5 with Fig. 7).

#### IV. CONCLUSIONS

We have developed an approach to study phase transitions in two-dimensional Ising systems with competing interactions that may show orientational and positional orders. A nematic order parameter in the square lattice has been defined, suitable to quantify the degree of orientational order of interfaces, useful in systems that show microphase separation. A mean-field approach has been developed, which leads to a set of self-consistent equations for orientational and positional order parameters, similar in spirit to the Bragg-Williams

approximation. The approach is very general, and can be applied in principle to any Ising system with competing interactions. Nematic, smectic, and stripe-crystal phases can be studied within this framework. We solved the self-consistent equations for the dipolar frustrated Ising ferromagnet. This model for ultrathin ferromagnetic films with perpendicular anisotropy is known to have a striped low-temperature phase, and the ground state is striped for arbitrary small dipolar interaction. Within the present approach, we have gone beyond the usual mean-field approximation for the stripe phase, showing that an isotropic-nematic phase transition takes place at a higher temperature than the mean-field isotropic-stripe transition.

Comparing with experimental evidence, as discussed in the Introduction, a nematic phase without positional order is probably present in ultrathin ferromagnetic films, although it has not been characterized already.<sup>2,3</sup> Our results indicate that the nematic phase is more robust for higher values of the ratio between the exchange and dipolar interactions  $J/g$ , as is the case in experiments where the intensity of the dipolar term is two or three orders of magnitude weaker than the exchange interaction.

Comparison with numerical results from computer simulations is still difficult for several reasons. The first one is that our calculation is of mean-field character, and then our results can only be qualitatively correct. A second problem is that, up to now, there have been very few attempts to quantify and characterize orientational order in systems of the type considered in this work, even in computer simulations.<sup>30,32–34,41</sup> To our knowledge, the most detailed simulational study of the phase transitions in the DFIF presented so far is the work by Cannas *et al.*,<sup>26</sup> where evidence was shown of an intermediate nematic phase between a paramagnetic and a stripe-crystal phase. In that reference, small values of the ratio  $J/g$  were considered. In fact, the nematic phase was reported for a ratio  $J/g = 2$ , which was the larger value considered in that work. For that case, the nematic phase was observed in a narrow temperature interval, in qualitative agreement with our phase diagram of Fig. 3.

Note, also, that for  $J/g = 1$ , no evidence of nematic phase was reported in that work, again in agreement with our results, which point to the absence of nematic phase for  $J \leq 1$ . It is still a major challenge for computer simulations to attain ratios in the experimental range  $J/g \sim 10^2 - 10^3$ . To our knowledge, the largest values attained in simulations have been around  $J/g \sim 10$ , which can only yield a narrow nematic phase.<sup>34,41</sup>

As already said, the present approach is valid for arbitrary microscopic interactions, which enter only in the computation of the final self-consistent equations. Because of its generality, we expect it can be useful for facing some important yet unsolved problems regarding the behavior of systems with competing interactions. One of these problems is the role of the range of interactions in producing pure orientational phases. The necessity of long-range interactions is frequently invoked, but the actual influence of the relative range of the competing interactions is still an open problem.

Some points have to be addressed in order to turn the approach quantitative. The computation of the nematic order parameter is equivalent to the computation of nearest-neighbor correlation functions. Then, better approximations for the computation of correlation functions will turn the results quantitatively reliable. Another point is the formal problem with the nonpositive character of the quadratic form, necessary for applying the Hubbard-Stratonovich transformation. Some previous works have already addressed this question,<sup>11,42</sup> which will be a subject of future work.

Although we have not tried to get quantitative results, we hope that the present approach will be useful to describe in full the phase transitions in two-dimensional systems, which show microphase separation and nematiclike orientational phases originated from competing interactions.

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