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Phase diagram of the two-dimensional Ising model with random competing interactions

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ABSTRACT

An Ising model with ferromagnetic nearest-neighbor interactions J_1 ($J_1 > 0$) and random next-nearest-neighbor interactions [$+J_2$ with probability p and $-J_2$ with probability $(1-p)$; $J_2 > 0$] is studied within the framework of an effective-field theory based on the differential-operator technique. The order parameters are calculated, considering finite clusters with $n = 1, 2$, and 4 spins, using the standard approximation of neglecting correlations. A phase diagram is obtained in the plane temperature versus p, for the particular case $J_1 = J_2$, showing both superantiferromagnetic (low p) and ferromagnetic (higher values of p) orderings at low temperatures.

Many magnetic compounds are well-described in terms of theoretical models characterized by a competition of nearest-neighbor and next-nearest-neighbor interactions. As typical examples, one has $Eu_xSr_{1-x}S$ [1,2] and $Fe_xZn_{1-x}F_2$ [3], which may present various low-temperature magnetic orderings, depending on its parameters, like the strength of these interactions and the concentration of magnetic ions x. The simplest model for such systems consists of an Ising model with competing uniform interactions, ferromagnetic J_1 (or antiferromagnetic $-J_1$) ($J_1 > 0$) on nearest-neighbor, and antiferromagnetic $-J_2$ ($J_2 > 0$) on next-nearest-neighbor spins (to be denoted hereafter as J_1–J_2 Ising model).

The J_1–J_2 Ising model on a square lattice has attracted the attention of many workers, being investigated through several approximation methods [4–28]. The various possible phases are the paramagnetic (**P**), for high temperatures, whereas for low temperatures one may have the ferromagnetic (**F**), antiferromagnetic (**AF**) and superantiferromagnetic (**SAF**) phases; the later is characterized by alternate ferromagnetic rows (or columns) of oppositely oriented spins. In the absence of a magnetic field, it can be shown that one has a symmetry with respect to the sign of the nearest-neighbor interactions, i.e., the ferromagnetic and antiferromagnetic states are equivalent [5]. At zero temperature (ground state), one has two ordered states depending on the value of the frustration parameter $\alpha = (J_2/J_1)$, namely, the **F** ($0 < \alpha < 1/2$) (in an equivalent way, an **AF** state occurs instead of the **F** one, if the sign of nearest-neighbor interactions is changed) and the **SAF** ($\alpha > 1/2$) phases. On the other hand, its phase diagram for finite temperatures has been the object of some controversies. A continuous critical frontier between the **P** and **F** phases, for $0 < \alpha < 1/2$, with the critical temperature $T_c(\alpha)$ decreasing and approaching zero for $\alpha \to 1/2$, is well-accepted nowadays. However, some characteristics of this phase diagram for $\alpha > 1/2$ are polemic; in particular, the presence of a first-order transition between the **P** and **SAF** phases, as well as a tricritical point characterized by the coordinates (α_t, T_t), beyond which a continuous critical frontier occurs, has been the object of some debate. Most of the works indicate that within the range $1/2 < \alpha < \alpha_t$ one should get a first-order phase transition between the **P** and **SAF** phases, whereas for $\alpha > \alpha_t$ this transition should become continuous. Some studies using cluster-variation methods [12–14] claim a first-order line in the range $0.5 < \alpha < 1.2$, i.e., $\alpha_t = 1.2$. However, recent results

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of Monte Carlo simulations [24,26], analysis of zeros of the partition function [25], and effective-field theory (EFT) [27] suggest that $\alpha_t < 1$, in such a way that a continuous transition between the **P** and **SAF** phases should occur for $\alpha = 1$.

In the present work we introduce a disordered version of the J_1–J_2 Ising model on a square lattice. It is important to mention that due to the symmetry with respect to the sign of the nearest-neighbor interactions, for which a change of sign takes **F** $\leftrightarrow$ **AF**, a disorder that produces a simple change of sign in J_1 is not expected to bring any novel physical properties [5]. A disordered version of the J_1–J_2 Ising model has already been defined in Refs. [29,30] by considering the same bond disorder, $P(J_{ij}) = (1/2)[\delta(J_{ij} - J_1) + \delta(J_{ij} - J_2)]$, for both nearest- and next-nearest-neighbor pairs of spins, which were introduced with the same sign in the corresponding Hamiltonian. In this analysis, the authors have investigated particular choices for the ratio J_2/J_1, through numerical simulations of the model on a square lattice, and have found as its main results: (i) An interesting saturation, with respect to the size of the system, of the specific-heat maxima; (ii) Critical exponents following the weak-universality scenario, according to which critical exponents change with the disorder in a certain way to keep some ratios (e.g., β/ν and γ/ν) unchanged at their corresponding pure values, similarly to what happens for other disordered magnetic models. Herein, we introduce a different disordered version of the J_1–J_2 Ising model, by defining a disorder only in the next-nearest-neighbor interactions, in such a way that we shall consider the Hamiltonian,

$$\mathcal{H} = -J_1 \sum_{\text{nn}} \sigma_i \sigma_j - \sum_{\text{nnn}} J_{ij}^{(2)} \sigma_i \sigma_j \quad (\sigma_i = \pm 1), \tag{1}$$

where the summations are over nearest-neighbor (nn) and next-nearest-neighbor (nnn) pairs of spins, with ferromagnetic ($J_1 > 0$) and random $J_{ij}^{(2)}$ interactions, respectively. The later follow a bimodal probability distribution,

$$\mathcal{P}\big(J_{ij}^{(2)}\big) = p\delta\big(J_{ij}^{(2)} - J_2\big) + (1-p)\delta\big(J_{ij}^{(2)} + J_2\big) \quad (J_2 > 0). \tag{2}$$

The particular case $p = 0$ of this model corresponds to the pure J_1–J_2 Ising model, described above and has been studied in the literature by many authors [4–28]. The introduction of the above type of disorder allows for the possibility of a richer critical behavior, characterized by a competition between different types of orderings as one varies p, for a fixed ratio of the interaction strengths, $\alpha = (J_2/J_1)$. In addition to that, from the point of view of physical realizations, the parameter p, which is usually associated with the concentration of a given chemical element, is much easier to be varied experimentally than the ratio of interaction strengths, α. In the present analysis we restrict ourselves to the case $J_2 = J_1$, i.e., $\alpha = 1$.

Herein, we treat this model by using the EFT method, which has been shown recently to be very useful in the pure case (i.e., $p = 0$) [27]. The EFT technique is based on rigorous correlation identities, as a starting point, and applies the differential-operator technique developed by Honmura and Kaneyoshi [31]. Within this method, the averages of a general function $\mathcal{A}(\{n\})$, involving spin variables, is obtained by

$$\big\langle \mathcal{A}(\{n\}) \big\rangle = \left\langle \frac{\text{Tr}_{\{n\}}\, \mathcal{A}(\{n\}) e^{-\beta \mathcal{H}_{\{n\}}}}{\text{Tr}_{\{n\}}\, e^{-\beta \mathcal{H}_{\{n\}}}} \right\rangle, \tag{3}$$

where the partial trace $Tr_{\{n\}}$ is calculated exactly over the set $\{n\}$ of spin variables that belong to the finite cluster specified by the multisite spin Hamiltonian $\mathcal{H}_{\{n\}}$ and $\langle \cdots \rangle$ indicates the usual canonical thermal average over the surrounding of the cluster. In the case $n = 2$ (see Fig. 1) the trace $Tr_{\{n\}}$ is calculated over spins S_1 and S_2, whereas $\langle \cdots \rangle$ is to be considered over $\sigma_1, \sigma_2, \ldots, \sigma_{10}$.

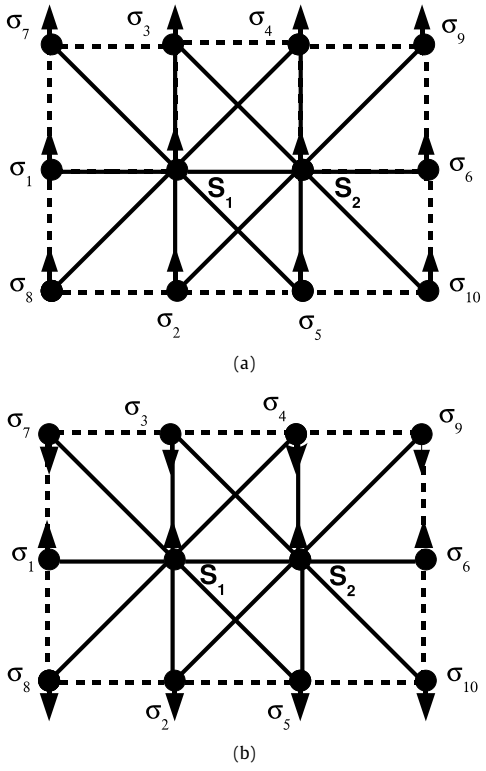

Fig. 1. A cluster with $n = 2$ (represented by spins S_1 and S_2) and its surrounding (spins $\sigma_1, \sigma_2, \ldots, \sigma_{10}$), for the model defined in Eq. (1), in its ferromagnetic (a) and superantiferromagnetic (b) ground states. The full lines represent the interactions of spins S_1 and S_2, which are taken into account exactly in Eq. (3), whereas the dashed lines represent the usual square-lattice structure.

We have applied the EFT method for clusters of with $n = 1, 2$, and 4 spins (to be denoted hereafter EFT-n) and below we describe the case of EFT-2, illustrated in Fig. 1. The Hamiltonian for this cluster is given by

$$\mathcal{H}_2 = -J_1 S_1 \cdot S_2 + S_1 a_1 + S_2 a_2, \tag{4}$$

with

$$a_1 = -J_1 \sum_{i=1}^{3} \sigma_i - \sum_{j=4,5,7,8} J_{1j}^{(2)} \sigma_j, \tag{5}$$

and

$$a_2 = -J_1 \sum_{i=4}^{6} \sigma_i - \sum_{j=2,3,9,10} J_{2j}^{(2)} \sigma_j. \tag{6}$$

We have divided the square lattice into two sublattices A and B, each of them defined by alternating lines (or columns) in such a way that in Fig. 1 one has $m_A = \langle\langle S_1 \rangle\rangle_{J^{(2)}} = \langle\langle S_2 \rangle\rangle_{J^{(2)}} = \langle\langle \sigma_i \rangle\rangle_{J^{(2)}}$ $(i = 1, 6)$ and $m_B = \langle\langle \sigma_i \rangle\rangle_{J^{(2)}}$ $(i = 2, 3, 4, 5, 7, 8, 9, 10)$, where $\langle \cdots \rangle_{J^{(2)}}$ denotes an average over the disorder; in the **F** (**SAF**) phase one has that $m_A = m_B = m$ ($m_A = -m_B = m$). At zero temperature ($m = 1$), the ground state of the Hamiltonian of Eq. (1) is ferromagnetic for $p = 1$ [see Fig. 1(a)], whereas for $p = 0$ and $\alpha = 1$ one gets the superantiferromagnetic state [shown in Fig. 1(b)]. In each case, a continuous phase transition occurs at a finite temperature between the corresponding low-temperature state and the paramagnetic state. To the best of our knowledge, theoretical works to investigate the critical behavior of this model for $0 < p < 1$ have never been carried and this represents the purpose of the present work.

The parameter m may be obtained by evaluating the average magnetization per spin in sublattice A, $m_A = \langle\langle \frac{1}{2}(S_1 + S_2) \rangle\rangle_{J^{(2)}}$, cal-

culating the inner trace in Eq. (3) over spins $S_1, S_2 = \pm 1$, which for the Hamiltonian of Eq. (4) yields

$$m = \left\langle\!\!\left\langle \frac{\sinh(\tilde{a}_1 + \tilde{a}_2)}{\cosh(\tilde{a}_1 + \tilde{a}_2) + \exp(-2K_1)\cosh(\tilde{a}_1 - \tilde{a}_2)} \right\rangle\!\!\right\rangle_{J^{(2)}}, \tag{7}$$

where $\tilde{a}_1 = -\beta a_1$, $\tilde{a}_2 = -\beta a_2$, and $K_1 = \beta J_1$. Using the identity $\exp(aD_x + bD_y)g(x, y) = g(x+a, y+b)$, where $D_\mu = \frac{\partial}{\partial \mu}$ $(\mu = x, y)$ is the differential operator, Eq. (7) becomes

$$m = \left\langle\!\!\left\langle \exp(\tilde{a}_1 D_x + \tilde{a}_2 D_y) \right\rangle\!\!\right\rangle_{J^{(2)}} g(x, y)\big|_{x,y=0}, \tag{8}$$

with

$$g(x, y) = \frac{\sinh(x + y)}{\cosh(x + y) + \exp(-2K_1)\cosh(x - y)}. \tag{9}$$

Applying the van der Waerden identity for the exponentials containing Ising variables, i.e., $\exp(\alpha\sigma_i) = \cosh(\alpha) + \sigma_i \sinh(\alpha)$ $(\sigma_i = \pm 1)$, the right-hand side of Eq. (8) can be written *exactly* in terms of multiple spin correlation functions; however, it is clear that if one tries to treat exactly all correlation functions, the problem becomes unmanageable. In this work, we use a decoupling procedure that ignores all high-order correlations on the right-hand side of Eq. (8), i.e.,

$$\begin{aligned} &\langle\!\langle \sigma_i \cdot \sigma_j \cdots \sigma_l \cdot \sigma_p \rangle\!\rangle_{J^{(2)}} \\ &\quad \simeq \langle\!\langle \sigma_i \rangle\!\rangle_{J^{(2)}} \cdot \langle\!\langle \sigma_j \rangle\!\rangle_{J^{(2)}} \cdots \langle\!\langle \sigma_l \rangle\!\rangle_{J^{(2)}} \cdot \langle\!\langle \sigma_p \rangle\!\rangle_{J^{(2)}}, \end{aligned} \tag{10}$$

where $i \neq j \neq \cdots \neq l \neq p$. The present procedure neglects correlations between different spins, but takes into account relations such as $(\sigma_i)^2 = 1$, whereas in the usual mean-field approximation both self- and multi-spin correlation functions are neglected. Applying the approximation of Eq. (10) in Eq. (8), one gets the equations of state,

$$m = \sum_{k=0}^{\tilde{k}} A_{2k+1}^{(\Gamma)}(T, p) m^{2k+1}, \tag{11}$$

where, due to the symmetry of the Hamiltonian, the even powers of m do not occur. In the equation above, the coefficients $A_{2k+1}^{(\Gamma)}(T, p)$ depend on the boundary conditions shown in Fig. 1, through the index $\Gamma = \mathrm{F}, \mathrm{SAF}$; in the present EFT-2 analysis one has that the highest power occurring on the right-hand side is defined by $\tilde{k} = 4$. In general, the coefficients $A_{2k+1}^{(\Gamma)}(T, p)$ depend on the size n of the cluster used in EFT-n, getting more complicated for larger clusters; in addition to that, $\tilde{k}$ increases with n, e.g., $\tilde{k} = 3$ for $n = 1$ and $\tilde{k} = 5$ for $n = 4$, in such a way that the procedure may get very tedious for large clusters.

The borders of continuous phase transitions between the ordered (**F** and **SAF**) phases and the high-temperature disordered (**P**) phase may be obtained by considering the limit $m \to 0$ in Eq. (11), which leads to

$$A_1^{(\Gamma)}(T_c, p) = 1. \tag{12}$$

By solving Eq. (12) numerically, one finds $T_c(p)$ for both phases **F** and **SAF**, as shown in the phase diagram of Fig. 2. For $p = 0$ one has a continuous transition between the **SAF** and **P** phases at $(k_B T_c(0)/J_1) \cong 2.263$, which is in good agreement with the value $(k_B T_c(0)/J_1) \cong 2.083$ estimated from Monte Carlo simulations [10,24], as well as from a computation of the zeros of the partition function [25]. The critical temperature associated with the **SAF** phase decreases as p increases, in such a way that one gets $T_c(p_{c1}) = 0$, for $p_{c1} \cong 0.075$. Due to strong frustration effects, the system does not present long-range order in the interval $p_{c1} < p < p_{c2}$, where $p_{c2} \cong 0.471$. For $p > p_{c2}$ one gets that the

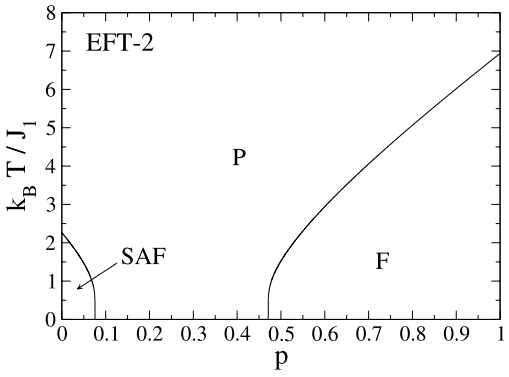

Fig. 2. Phase diagram of the model defined in Eqs. (1) and (2) for $J_2 = J_1$ within EFT-2. The phases are the paramagnetic (**P**), ferromagnetic (**F**), and superantiferromagnetic (**SAF**), as described in the text.

critical temperature associated with the **F** phase increases monotonically, leading to $(k_B T_c(1)/J_1) \cong 6.937$. Besides the results calculated within EFT-2 shown in Fig. 2, we have also computed these critical frontiers within EFT-1 and EFT-4. Only in the EFT-1 case, the critical frontier **SAF**–**P** comes as a first-order transition; we believe this to be a spurious result (attributed to the smallness of the cluster), since it is in contrast with those obtained from EFT-2 and EFT-4 (which yield continuous phase transitions), as well as with those of Monte Carlo simulations for the particular case $p = 0$ [10,24]. From EFT-4 one gets that $(k_B T_c(0)/J_1) \cong 2.310$ and $p_{c1} \cong 0.088$, indicating an enlargement of the **SAF** phase, whereas for the **F** phase one has $p_{c2} \cong 0.474$ and $(k_B T_c(1)/J_1) \cong 6.750$. It should be mentioned that our estimates for $(k_B T_c(1)/J_1)$, from EFT-1, EFT-2, and EFT-4, suggest a slow convergence towards those from low- and high-temperature series expansions, which yield $(k_B T_c(1)/J_1) \cong 5.260$ and $(k_B T_c(1)/J_1) \cong 5.257$, respectively [32], or to the more recent one, obtained from an analytical expression for the interface free energy, $(k_B T_c(1)/J_1) \cong 5.376$ [22]. In a similar way, our results suggest a decrease in the gap $(p_{c2} - p_{c1})$, although it is not possible to conclude whether the phases **SAF** and **F** should meet at zero temperature.

It is important to mention that the EFT method, which replaces averages over products of spins by the respective products of their averages, neglects correlations and, as a consequence, the associated critical exponents are mean-field-like. Therefore, this method is not suitable for investigations of possible changes in the critical exponents with respect to variations of important parameters of the problem (like R and p), which represents a major question in its $p = 0$ particular case, the J_1–J_2 Ising model. Furthermore, since the EFT-n procedure consists in treating a cluster of n spins exactly, whereas the interaction of this cluster with its surrounding are treated as an average (mean-field-like), the method becomes unmanageable for increasing values of n. However, this technique presents the advantage of providing phase diagrams that may be qualitatively correct as a whole, and in some cases it may yield critical points with a good accuracy, in spite of considering clusters with small n, like in the following examples: (i) The tricritical point in the phase diagram of the J_1–J_2 Ising model on the square lattice, which was estimated to occur for $(J_2/J_1) < 1$ within EFT [27], is in agreement with other methods, like Monte Carlo simulations [24,26] and analysis of zeros of the partition function [25]; (ii) The present estimate of the critical temperature for $p = 0$, $(k_B T_c(0)/J_1) \cong 2.263$, yields a relative discrepancy of typically 8% with respect to the value $(k_B T_c(0)/J_1) \cong 2.083$, estimated from Monte Carlo simulations [10,24] and zeros of the partition function [25]. In the present problem, a full phase diagram was obtained for the case $(J_2/J_1) = 1$, where an elimination of the **SAF** phase occurs as one increases the concentration of second-

neighbor ferromagnetic bonds. Therefore, according to the present EFT approach, only for low values of p one gets a sufficient concentration of second-neighbor antiferromagnetic bonds in order to yield the **SAF** ordering at low temperatures. As the concentration p increases, the competition between ferromagnetic couplings (both from the nearest-neighbor pairs and second-neighbor pairs with probability p) and the second-neighbor antiferromagnetic ones [with probability $(1-p)$] destroys the **SAF** ordering at the concentration p_{c1}. Above this value, the competition between ferro- and antiferromagnetic interactions leads to a sufficient amount of frustrations that could, in principle, favor a type of spin-glass ordering at low temperatures. However, similarly to many other frustrated two-dimensional systems, which do not exhibit a spin-glass phase for finite temperatures, herein the paramagnetic phase dominates up to zero temperature, in the interval $p_{c1} < p < p_{c2}$. For $p > p_{c2}$, one has sufficient ferromagnetic bonds leading to the **F** ordering. Obviously, the present estimates of p_{c1} and p_{c2} are expected to change under other approximation techniques (e.g., Monte Carlo simulations) and one cannot rule out the possibility of a critical frontier separating the phases **SAF** and **F** at finite temperatures.

To conclude, we have introduced an Ising model characterized by ferromagnetic nearest-neighbor interactions J_1 and random $\pm J_2$ next-nearest-neighbor interactions [$+J_2$ with probability p and $-J_2$ with probability $(1-p)$]. This model, which generalizes the J_1–J_2 Ising model (herein corresponding to the case $p = 0$) is expected to be relevant for some diluted magnetic compounds, characterized by a competition between nearest- and next-nearest-neighbor interactions. We have studied the phase diagram of this model in the particular case $J_1 = J_2$ within the framework of an effective-field theory based on the differential-operator technique, considering finite clusters with $n = 1, 2$, and 4 spins. We have found a low-temperature sublattice ordering associated with a superantiferromagnetic phase for small p ($0 < p < p_{c1}$, where $p_{c1} \cong 0.088$ in the case $n = 4$), as well as a ferromagnetic phase for larger values of p ($p > p_{c2}$, where $p_{c2} \cong 0.474$ in the case $n = 4$), whereas in the interval $p_{c1} < p < p_{c2}$ the system does not present long-range order. Other approximation methods, such as renormalization-group and Monte Carlo simulations, should be used to obtain further information on the critical behavior of this model.

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