

Free energy and metastable states in the square-lattice J_1 - J_2 Ising model

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Abstract

We calculate the free energy as a function of polarization for the square-lattice J_1 - J_2 Ising model for $J_2 < |J_1|/2$ using the Random local field approximation (RLFA) and Monte Carlo (MC) simulations. Within RLFA, a metastable state with zero polarization is present in the ordered phase. Moreover, the free energy calculated within RLFA indicates a geometric slab-droplet phase transition at low temperature, which cannot be predicted by the mean field approximation. In turn, free energy calculations by definition for finite-size samples using MC simulations reveal metastable states with a wide range of polarization values, the origin of which we discuss. The calculations also reveal additional slab-droplet transitions (at $J_2 > 0.25$). These findings enrich our knowledge of the J_1 - J_2 Ising model and the RLFA as a useful theoretical tool to study phase transitions in spin systems.

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1 Introduction

The square-lattice J_1 - J_2 Ising model is one of the minimal extension of the standard Ising model, in which the coupling J_1 between nearest neighbors is complemented by the coupling J_2 between diagonally next-nearest neighbors. The properties of this model are of both fundamental and practical interest, in particular, since its quantum Heisenberg counterpart is relevant to the antiferromagnetism in the parent compounds of the cuprate and pnictide families of high-temperature superconductors [1–3]. Indeed, recent state-of-the-art numerical calculations [4–14] confirm earlier findings [15–22] that diagonal interactions are important in describing the available experimental data for these compounds. Magnetic frustration due to the J_2 coupling leads to the quasi-degeneracy of the ground state [6, 19, 20] and possibly to a quantum spin liquid state at J_2 close to $|J_1|/2$ [23, 24].

We recently highlighted the existence of metastable states with arbitrary polarization in the square-lattice J_1 - J_2 Ising model for $J_2 \in (0, |J_1|)$ using Monte Carlo (MC) simulations, which was further supported by simple microscopic energy considerations [25]. For the ferromagnetic ground state, i.e. for $J_1 < 0$ and $J_2 < |J_1|/2$, these states are rectangles with polarization opposite to the surrounding, briefly considered much earlier in [26, 27]. Significantly, the Random local field approximation (RLFA) [28], also applied in [25], points to a metastable state with zero polarization in the same J_2 coupling range, thus reflecting the appearance of microscopic metastable states, which seems impossible for mean field approximations (MFA). Note that the above states differ from the metastable states of the standard Ising model, consisting of straight stripes, into which a system with zero polarization, when applying the single-spin flip MC algorithm and periodic boundary conditions, relaxes after quenching only in about 30% of cases and only in the absence of an external field [29–31].

Polarization-dependent (called restricted or Landau) free energy $F(\mathbf{m})$, considered in the framework of Landau’s phenomenological theory of phase transitions [32], also provides information on metastable states (including those in an external field) and can be used to calculate the relaxation rate of the system to the ground state via the Landau-Khalatnikov equation [33] (see, e.g., [34] for such calculations in ferroelectrics). It should be noted, however, that for short-range interactions, the restricted free energy obtained within the MFA differs qualitatively from the free energy calculated exactly or using the MC method for finite-size samples [35–37]. In the former case, below the phase transition temperature, the free energy takes into account only homogeneous states inside the two-phase (spins up and down) coexistence region and, as a consequence, is a double-well shaped function of polarization. In the latter case, all inhomogeneous states contribute to $F(\mathbf{m})$. Thus, at a temperature close to zero, the barrier between two minima with opposite polarization is determined by the interface energy between two large domains and is proportional to the sample size L . Relative to the total energy, proportional to the number of spins $N = L^2$, it vanishes in the thermodynamic limit [38, 39]. It was shown that, despite the loss of detailed information about microscopic spin configurations, $F(\mathbf{m})$ can be harnessed to well reproduce the MC polarization dynamics of the Ising model in good agreement with the droplet theory [40]. These ideas were further developed in [41–44]. The temperature dependence of the free energy barrier in the J_1 - J_2 Ising model, but only in three dimensions, was analytically estimated in [27] in connection with domain growth and shrinking after low-temperature quenching.

Here we calculate the restricted free energy $F(\mathbf{m})$ for the square-lattice J_1 - J_2 Ising model in the framework of RLFA, exactly for a square sample size $L = 6$ and using the MC method for $L = 10$. We pay special attention to the metastable states, which appear in this model and were studied earlier in [25], and explore how they are reflected in the free energy. The features of the geometric slab-droplet phase transition in the free energy calculated by both methods are also briefly discussed.

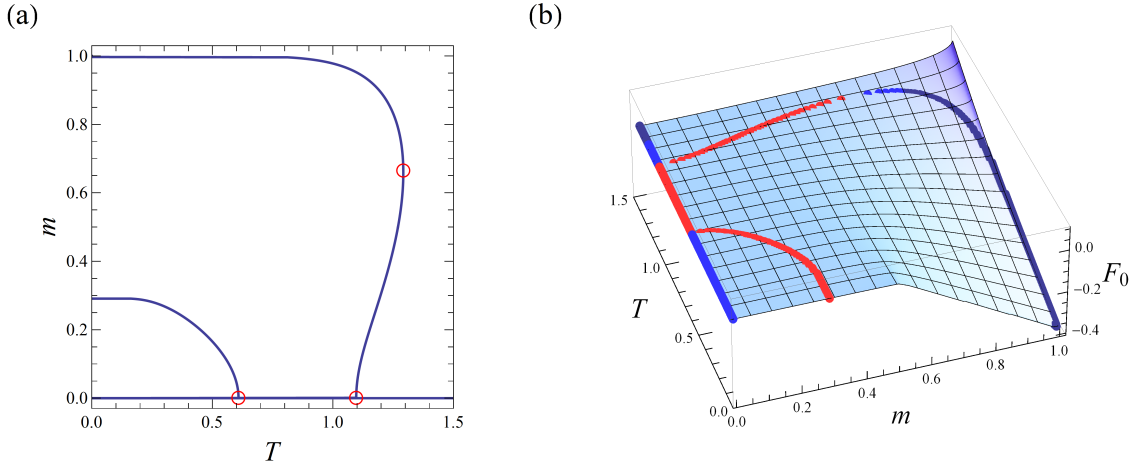


Figure 1: (a) The RLFA solution for $J_2 = 0.3$ (see also Fig. 2 in [25] for different values of J_2). Red circles define temperatures $T_0 < T_1 < T_2$. (b) Restricted free energy $F_0(m)$ within RLFA for $J_2 = 0.3$ as a function of temperature. Red points correspond to the local maximum of $F_0(m)$ at each temperature (a barrier), dark blue points correspond to its global minimum (stable states), and light blue points correspond to its local minimum (metastable states), which are zoomed in on in Fig. 2a. Metastable states with zero polarization appear at temperatures from zero to $T_0 \approx 0.6$ and from $T_1 \approx 1.1$ to $T_c < T_2$ ($T_2 \approx 1.26$), at which a first order phase transition occurs.

62 2 Model

63 The square-lattice J_1 - J_2 Ising model Hamiltonian reads

$$H = J_1 \sum_{\langle i,j \rangle} s_i s_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} s_i s_j - \sum_i h_i s_i, \quad (1)$$

64 where each spin takes the value plus or minus 1. The sums are over nearest $\langle i,j \rangle$ and diag-
 65 onal next-nearest $\langle\langle i,j \rangle\rangle$ neighbors, as well as over each spin coupled to the external field h_i
 66 at its position. In what follows, we set the values of the coupling constants values $J_1 = -1$
 67 and $J_2 < 1/2$, which correspond to the ferromagnetic ground state (the case $J_2 > 1/2$ with
 68 a striped antiferromagnetic ground state is similar in many aspects, but has a more complex
 69 spin topology and will be considered separately). Note that the model is invariant with respect
 70 to the simultaneous change of the sign of J_1 and the replacement of homogeneous polariza-
 71 tion with Néel checkerboard one, corresponding to the antiferromagnetic order of the parent
 72 compounds of cuprate superconductors [45].

73 3 Random local field approximation

74 RLFA is based on the exact formula for the average spin [28, 46]:

$$\langle s_i \rangle = \langle \tanh \beta (h_i^s + h_i) \rangle, \quad (2)$$

75 where $\beta = 1/T$ is the inverse temperature in energy units. The local field, $h_i^s = -\sum_j J_{ij} s_j$,
 76 acting on the spin s_i is caused by all spins s_j coupled with it.

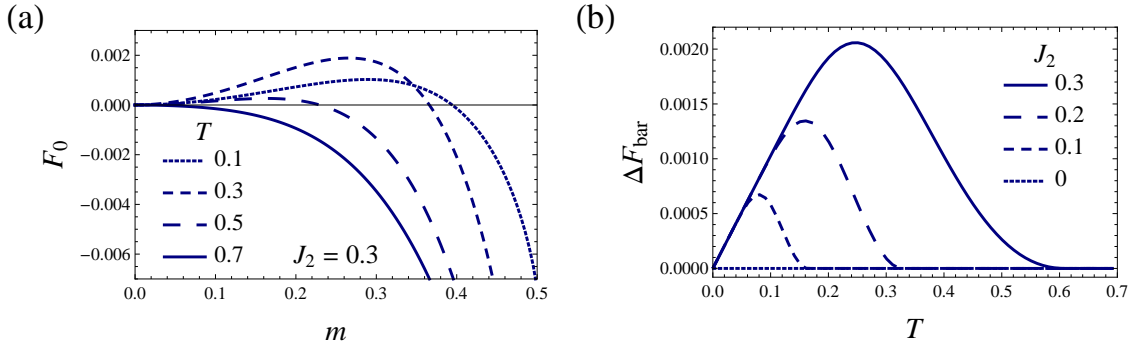


Figure 2: (a) Restricted free energy $F_0(m)$ within RLFA for $J_2 = 0.3$ and polarization limited by $m \in (0, 0.5)$ to show the appearance at low temperature of a barrier at $m \neq 0$ whose height first increases and then decreases as the temperature approaches zero. (b) The barrier height $\Delta F_{\text{bar}} = F_{\text{bar}} - F(0)$ for the metastable state at $m = 0$, which appears in the restricted free energy $F(m)$ calculated within RLFA, as a function of temperature T . Only $J_2 < 0.31$ are considered when $T_0 < T_1$ (for temperatures definition see Fig. 1a), since these two temperatures become undefined at larger J_2 [25].

77 The brackets in Eq. (2) correspond to thermal averaging, which is performed with proba-
 78 bility distribution [28, 47]:

$$P = \prod_j (1 + m_j s_j) / 2, \quad (3)$$

79 where the product is taken over all spins s_j coupled to s_i , and $m_j = \langle s_j \rangle = m e^{i\mathbf{q}\cdot\mathbf{r}_j}$ is the
 80 thermally averaged polarization at position \mathbf{r}_j , the variation of which in space is determined
 81 by the propagation vector \mathbf{q} . Here we consider only homogeneous polarization m and external
 82 field \mathbf{h} , which corresponds to $\mathbf{q} = (0, 0)$. Note that Eq. (3) implies that, within RLFA, the
 83 fluctuations of each spin are considered independent.

84 Eq. (2) follows from equating to zero the derivative of the restricted free energy $F(m)$ [40],
 85 which corresponds to thermodynamic equilibrium at a fixed value of polarization m [48]. To
 86 obtain the correct dependence of $F(m)$ on the external field \mathbf{h} , we rewrite Eq. (2) in the
 87 form $\partial F / \partial m = f(m) - \mathbf{h}$, integration of which yields $F(m) - F(0) = \int_0^m f(m) dm - \mathbf{h}m$.
 88 Although $F(0)$ depends on temperature, this is of little interest to us and for convenience we
 89 choose $F(0) = 0$ at each temperature and define $F_0(m) = \int_0^m f(m) dm - \mathbf{h}m$.

90 The RLFA solution and the restricted free energy $F_0(m)$ calculated in this way for $J_2 = 0.3$
 91 are shown in Fig. 1. The calculated free energy indeed points to the metastable state with
 92 $m = 0$, discussed in [25], which we have zoomed in on in Fig. 2a. With decreasing temper-
 93 ature, a barrier appears at $T_0 \approx 0.6$ near $m = 0$, see Fig. 1a. Then its height first increases
 94 and its position shifts up to $m \approx 0.29$, after which, at a temperature slightly less than J_2 , the
 95 height begins to decrease linearly in T to zero, see Fig. 2b. The maximum barrier height of
 96 about 0.002 is close to the estimate in [25] based on the value of the coercive field. In Fig. 2b,
 97 the barrier height for various values of J_2 is shown. At $J_2 \approx 0.31$ we have $T_0 = T_1$, where T_1 is
 98 the highest temperature at which $F(m)$ is maximum at $m = 0$, see Fig. 1. For larger J_2 , these
 99 two temperatures are not defined [25], and the unstable RLFA solution $m = 0$ extends from 0
 100 to the critical temperature $T_c < T_2$, where T_2 is the temperature below which the minimum
 101 of $F(m)$ at $m \neq 0$ appears. It should be noted that within RLFA the transition turns out to
 102 be first order for $0.25 \lesssim J_2 \lesssim 1.25$ [25], while recent more accurate calculations narrow this
 103 interval to a small region around $J_2 = 0.5$ [49–52].

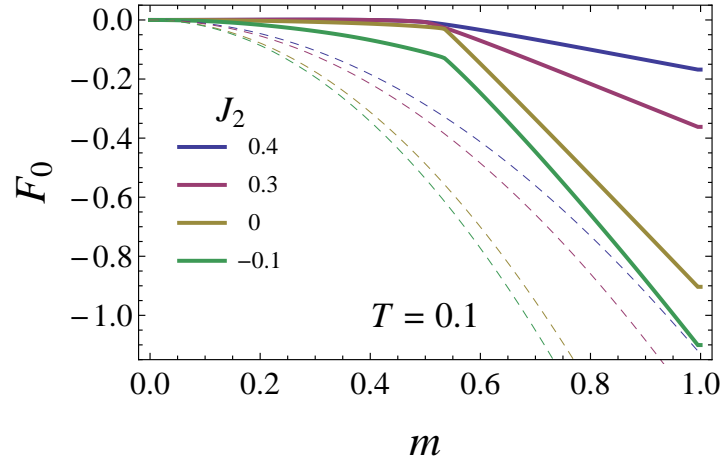


Figure 3: Restricted free energy $F_0(m)$ calculated within RLFA (solid lines) for several values of J_2 at temperature $T = 0.1$, which shows a kink at magnetization around $m = 0.5$. The dashed lines are the MFA free energy.

104 At low temperature, the RLFA restricted free energy shows a kink at a polarization value
 105 $m_c \approx 0.5$ for $J_2 = 0.3$, see Fig. 1b. This kink corresponds to a polarization for which the
 106 most likely configuration changes from a slab (for $m < m_c$) to a droplet (for $m > m_c$) [40].
 107 For $J_2 = 0$, the RLFA predicted critical polarization $m_c \approx 0.53$ is close to the exact value
 108 $m_c = 0.5$ [53], see Fig. 3. In general, this effect is called geometric phase transition and
 109 is present in finite-size systems when periodic boundary conditions are used in the simula-
 110 tion [53, 54]. For the two-dimensional Ising model, it was thoroughly studied by the Monte
 111 Carlo method used to calculate the free energy in [55, 56]. We emphasize that RLFA is able
 112 to predict the geometrical phase transition, in contrast to MFA. We have also checked that
 113 even the four-spin cluster approximation, formulated as in [57], does not predict this transi-
 114 tion, despite the good accuracy of the approximation in describing ferroelectric phase transi-
 115 tions [58–61]. It is also worth noting that the RLFA free energy becomes flat, signifying the
 116 absence of a phase transition, in the limit $J_2 \rightarrow 0.5$ [25].

117 4 Exact values of restricted free energy

118 Now we want to compare the above results with the free energy obtained by definition,

$$F(M) = -T \log \sum_E g(M, E) \exp(-E/T), \quad (4)$$

119 where the sum is over all possible energies E and $g(M, E)$ is the density of states with total
 120 spin $M = mN$ and energy E for N spins.

121 For small samples, the sum in (4) can be computed exactly. For a square sample of size
 122 $L = 6$, yielding the total number of spins $N = 36$, the result for $J_2 = 0.3$ is shown in Fig. 4.
 123 In all calculations we apply periodic boundary conditions. The critical temperature for $L = 6$
 124 is equal to $T_c = 1.67$, while for $L = 100$ (which practically corresponds to an infinite sample
 125 size) it is $T_c = 1.26$ [25].

126 Configurations that contribute to the free energy $F(M)$ at zero temperature (i.e. have the
 127 lowest energies) for different values of M are shown in Fig. 5. At zero temperature, at
 128 $m \lesssim 0.5$, the free energy is flat for $0 < J_2 < 0.5$, Fig. 6a. The pits, also mentioned in [40], are
 129 due to spin configurations with completely flat interface between two slabs with opposite spin

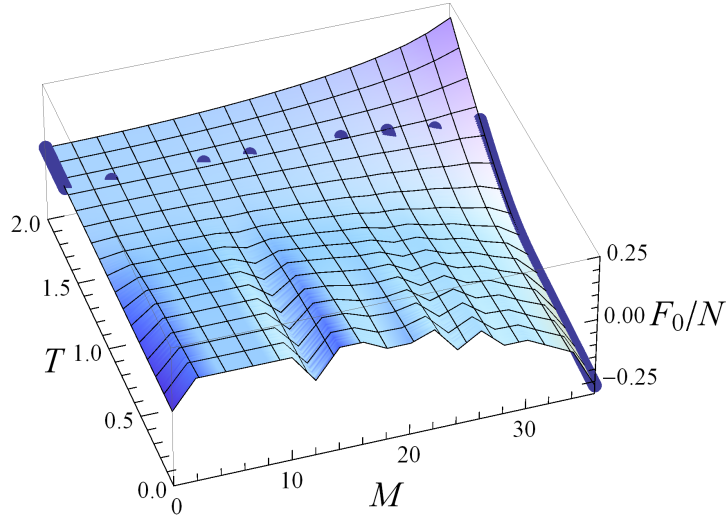


Figure 4: Restricted free energy per spin, F_0/N , calculated exactly by Eq. (4) at $J_2 = 0.3$ for sample size $L = 6$ as a function of total spin M and temperature T . The free energy is defined only for integer even values of M and linearly interpolated between them. For ease of comparison with the RLFA results in Fig. 1, we set here $F = 0$ at $M = 0$ at each temperature. Dark blue points correspond to the global minimum of F_0/N at each temperature.

130 directions, see configurations with $M = 0$ (M_0) and $M = 12$ (M_{12}) in Fig. 5. When a spin flips
 131 on this interface (configurations M_2 and M_{14} in Fig. 5) the energy increases by $4J_1$. When
 132 the last spin in the row flips, the energy decreases by this value (see transition $M_{10} \rightarrow M_{12}$ in
 133 Fig. 5 and Fig. 6a). The distance between two neighbor pits of $F(M)$ at $m \lesssim 0.5$ is equal to
 134 $2L$, since any spin flip changes the total moment by 2.

135 At $m \gtrsim 0.5$, the configurations that contribute to the free energy $F(M)$ at zero temperature
 136 correspond mainly to the droplet (starting from configuration M_{18} in Fig. 5), but depend on
 137 J_2 , as indicated in the bottom row of Fig. 5. The latter also affects the dependence of the
 138 energy barrier height, $\Delta F_M = F_M - F_{M-2}$, on J_2 (Fig. 6a). As shown in Fig. 7a, for $M = 22$
 139 at $J_2 < 0.33$ and for $M = 26$ at $J_2 < 0.25$, the barrier height is $4J_2$ and is determined by the
 140 spin flip at the corner of the droplet. Note that the exact values of J_2 are equal to $1/3$ and $1/4$
 141 and follow from the energy ratio of the different configurations M_{22} , M_{24} , and M_{26} in Fig. 5.
 142 At larger values of J_2 for both values of M , there is a reverse transition to the slab phase and
 143 then back again, as can be seen in the bottom row of Fig. 5. When a spin flips on a side of the
 144 droplet whose length $D > 1$, the energy does not change until the last spin on the side flips,
 145 then the energy decreases by $\Delta E = 4(J_1 + J_2)$ (see transitions $M_{22} \rightarrow M_{24}$ at $J_2 < 0.25$ and
 146 $M_{26} \rightarrow M_{28}$ at $J_2 < 0.33$ in Fig. 5 and Fig. 6a). For $D = 1$, $\Delta E = 4(J_1 + 2J_2)$, which is valid
 147 for transitions $M_{30} \rightarrow M_{32}$ and $M_{32} \rightarrow M_{34}$ in Fig. 6a. At $J_2 \leq 0$, we see only decrease in free
 148 energy with M for the droplet phase in Fig. 6a.

149 At higher temperatures, other higher energy configurations in addition to those shown in
 150 Fig. 5 contribute to the partition function in Eq. (4) for each value of M . This affects the
 151 dependence of the above discussed energy barriers on temperature, which for $J_2 = 0.3$ is
 152 shown in Fig. 8a. The metastable state barrier at $M = 22$ disappears at $T \approx 0.65$, which is
 153 close to the corresponding temperature $T_0 \approx 0.6$ from the RLFA solution (see Fig. 1). Note
 154 that at this value of J_2 the barrier at $M = 26$ is determined by the slab-droplet transition and
 155 not by the metastable state (see Fig. 5 and Fig. 7a).

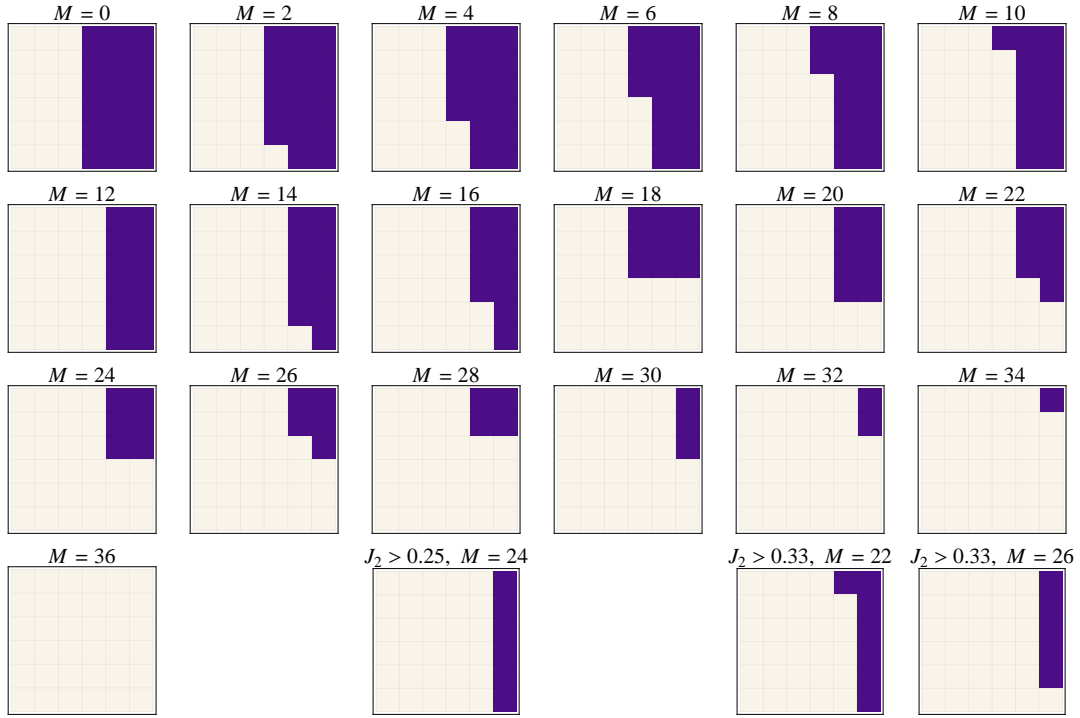


Figure 5: Configurations that contribute to the restricted free energy $F(M)$ at zero temperature, i.e. have minimal energies, for all possible values of the total spin M at $L = 6$. The configurations that change for $J_2 > 0.25$ and $J_2 > 0.33$ are shown separately in the bottom row (they affect the dependence of the energy barrier ΔF_M on J_2 shown in Fig. 7).

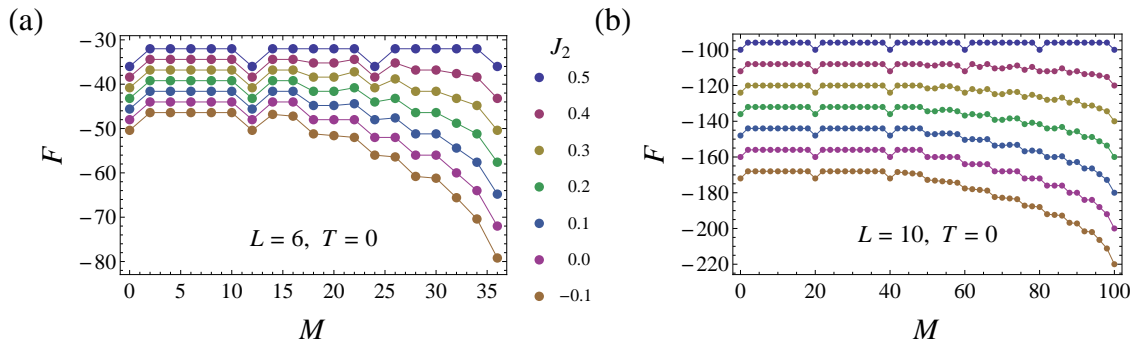


Figure 6: Restricted free energy F as a function of total spin $M = mL^2$ at zero temperature $T = 0$, calculated according to Eq. (4) for several values of J_2 (listed in the legend, valid for both plots), (a) exactly for the sample size $L = 6$, (b) using Monte Carlo method for $L = 10$. The solid lines provide guides to the eye.

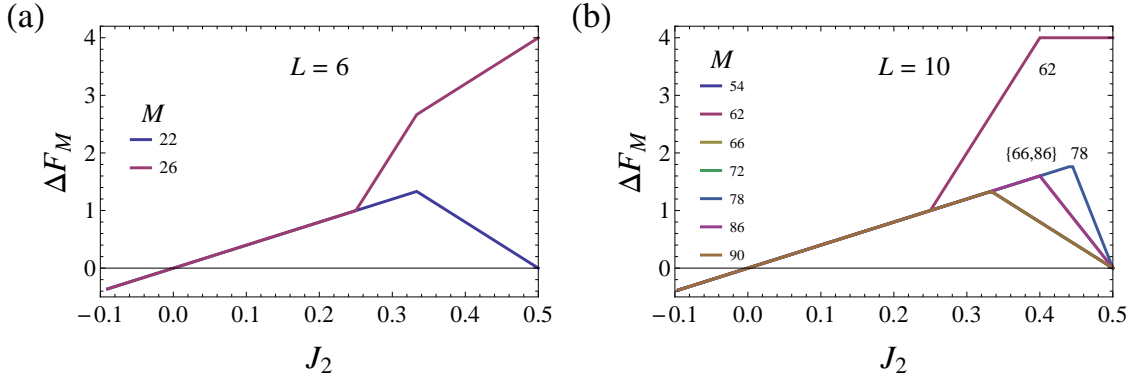


Figure 7: Restricted free energy barrier height, $\Delta F_M = F_M - F_{M-2}$, as a function of J_2 for several values of the total spin M at zero temperature. (a) The sample size is $L = 6$. (b) The sample size is $L = 10$. Lines for different values of M overlap. The lines corresponding to $M = 62, 78$ and overlapping $\{66, 86\}$ are marked separately.

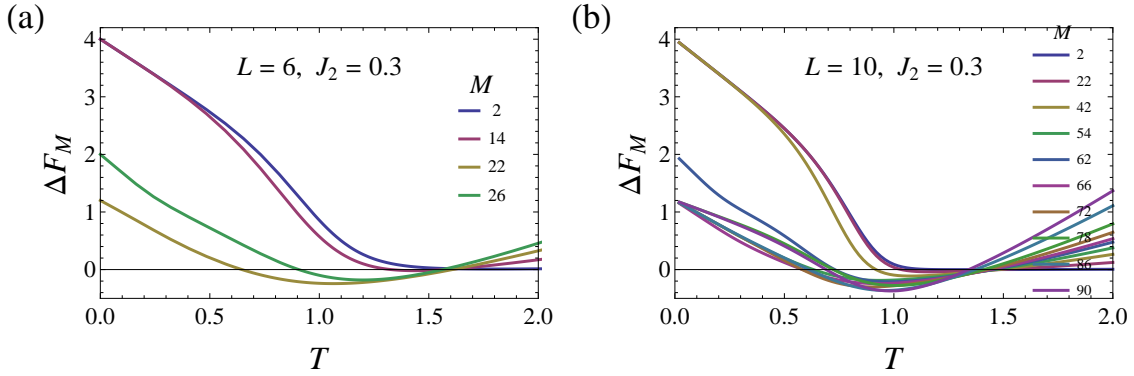


Figure 8: Restricted free energy barrier height, $\Delta F_M = F_M - F_{M-2}$, as a function of temperature for several values of the total spin M and $J_2 = 0.3$. (a) The sample size is $L = 6$. (b) $L = 10$. The three upper curves correspond to $M = 2, 22$ and 42 , and in the middle is $M = 62$.

156 5 Monte Carlo simulations

157 For larger square samples, with $L = 7$ and $L = 8$, the free energy from Eq. (4) can only be
 158 calculated using supercomputers, given the large number of 2^N configurations for N spins. Al-
 159 ternatively, it can be calculated approximately with sufficiently high accuracy using the Monte
 160 Carlo method. We use the Wang-Landau algorithm [62–64], which has proven to be very
 161 efficient for this purpose at low temperature. It consists in performing a random walk in po-
 162 larization and energy space to extract an estimate for the density of states $g(\mathbf{M}, \mathbf{E})$ that gives
 163 a flat histogram.

164 Using the Wang-Landau algorithm, we reproduce the exact results for $L = 6$ with high
 165 accuracy and obtain similar result for $L = 10$, see Fig. 6b and Fig. 9, where the free energy
 166 barriers at $m > m_c$ are clearly visible at low temperature and $J_2 = 0.3$. For $J_2 = 0$, the
 167 calculated free energy at zero temperature, Fig. 6b, agrees with [40]. I note that the free
 168 energy for larger samples could also be calculated, as was done, for example, in [40] for
 169 $J_2 = 0$. However, for the purposes of this article, namely to show how metastable states are
 170 reflected in the free energy, the size $L = 10$ seems optimal, and all metastable states are clearly

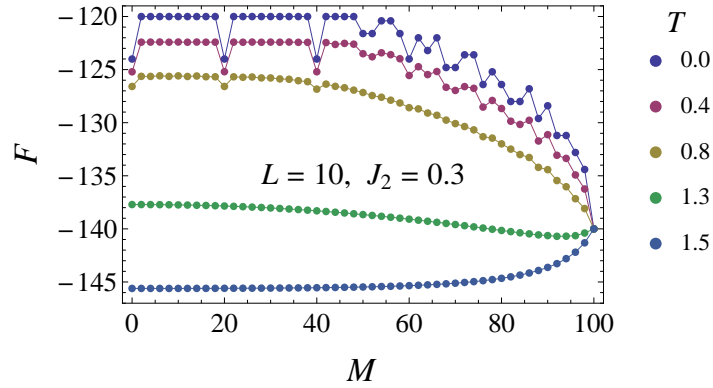


Figure 9: Restricted free energy F as a function of total spin M for $J_2 = 0.3$ calculated by the MC method at several temperatures below and above the phase transition. The sample size is $L = 10$.

171 visible and convenient for analysis.

172 The barrier heights for metastable states at $m > m_c$ correspond to the spin flip at the corner
 173 of the droplet and are equal to $4J_2$ for $J_2 < 0.25$ (Fig. 7b). For $J_2 > 0.25$, this dependence
 174 changes due to additional slab-droplet and vice versa transitions, as in the case of $L = 6$. The
 175 dependence of these barriers on temperature is shown in Fig. 8b. It is almost linear and the
 176 barriers disappear in the temperature range from approximately 0.6 to 0.7, which is close to
 177 the RLFA predicted T_0 in Fig. 1. We verified that the linear dependence of barriers height on
 178 temperature and their disappearance at a temperature close to T_0 , obtained within RLFA, are
 179 also valid for other values of J_2 . Note that the linear dependence on T at low temperature
 180 follows directly from the definition of the free energy $F = U - TS$, where U is the energy and
 181 S is the entropy.

182 6 Discussion

183 The primary goal of this work was to confirm, by calculating the restricted free energy, the
 184 presence of metastable states in the J_1 - J_2 Ising model, recently found in the RLFA solution
 185 and MC simulations of low-temperature quenching [25]. The restricted free energy $F(m)$ as a
 186 function of polarization calculated within RLFA indeed shows local minima at zero polarization
 187 at low temperature for $J_2 > 0$, see Fig. 1 and Fig. 2, thus indicating a metastable state.

188 At the same time, the exact calculations of $F(m)$ for a small sample size $L = 6$ (Fig. 4
 189 and Fig. 6a) and MC simulations for $L = 10$ (Fig. 6b and Fig. 9) indicate local minima corre-
 190 sponding to metastable states with various values of polarization. Some of them, at $m \lesssim 0.5$,
 191 are due to long stripes with an activation energy of $4J_1$ of a spin flip on a flat domain bound-
 192 ary (Fig. 5). In the standard Ising model, the system can become stuck in these states with a
 193 final polarization following a Gaussian distribution after zero-temperature quenching from an
 194 initially random configuration with zero polarization [30].

195 Metastable states at $m \gtrsim 0.5$ are caused by droplet-shaped domains with an activation
 196 energy of $4J_2$ of a spin flip in their corner, at least at $J_2 < 0.25$ for both sample sizes $L = 6$
 197 and $L = 10$ (Fig. 7). It is interesting whether this value of J_2 holds true for larger sample
 198 sizes. At $J_2 > 0.25$, the dependence of the barrier height on J_2 for some metastable states
 199 changes. Our exact energy calculations for a sample size of $L = 6$ show that for $J_2 > 1/4$
 200 and then for $J_2 > 1/3$ the sequence of minimal energy configurations for increasing total spin
 201 M changes (see Fig. 5) and a return occurs into the slab phase at certain values of M , which

202 may be important for some applications. Indeed, the importance of the geometrical slab-
 203 droplet transition for various physical situations, including the dewetting transition between
 204 hydrophobic surfaces, was highlighted in [54].

205 Although the zero polarization of the metastable state and the low height of the barrier
 206 proportional to the temperature near zero (see Fig. 2) is not exactly what follows from MC
 207 calculations, where the barrier heights are much higher and decrease with temperature (see
 208 Fig. 8), the fact of even a rough indication of the metastable state by RLFA is very valuable.

209 Another valuable RLFA prediction that turns out to be quite accurate is the geometric slab-
 210 droplet phase transition at low temperature (Fig. 1b and Fig. 3). The reason why RLFA is so
 211 effective in this situation, in our opinion, is that by definition it takes into account the local
 212 field due to all possible configurations of spins interacting with the central spin, not just the
 213 mean field. The probability of these configurations, in turn, is determined by the mean spin.

214 The distance between striped metastable states along the M axis (at $m \lesssim 0.5$) is equal
 215 to $2L$ and, as a result, their number is proportional to L . For droplet metastable states (at
 216 $m \gtrsim 0.5$), the distance is determined by the droplet size, which becomes smaller as M
 217 increases. Thus, one can expect that the number of droplet metastable states scales as L^2 and
 218 they are distributed along the M axis much more densely. This is confirmed by our MC simu-
 219 lations in Figs. 6, 7, and 9. This could be the reason why, during low-temperature quenching
 220 from high temperature in an external field, the system is not captured into striped metastable
 221 states in the standard Ising model [30], but gets stuck in droplet metastable states with finite
 222 polarization when $J_2 \in (0, 1/2)$ [25]. Note that the polarization of such a final state turns out
 223 to be about 0.5 at zero temperature [25], which is close to the slab-droplet phase transition,
 224 from where droplet metastable states begin to appear as M increases (Fig. 9). However, the
 225 polarization after quenching (from high temperature) sharply decreases with final tempera-
 226 ture [25] and does not correspond to the critical polarization m_c of the slab-droplet transition
 227 in the standard Ising model, the temperature dependence of which resembles the equilibrium
 228 polarization [53].

229 It should be noted here that the metastable states into which the system relaxes after low-
 230 temperature quenching in [25] are not exactly the same as shown in Fig. 5 and which deter-
 231 mines the free energy at zero temperature. The energy of the former is much higher, and the
 232 system is more likely to get stuck in them, relaxing in energy during quenching on the way to
 233 thermal equilibrium. Metastable states like in Fig. 5 can in principle be reached after quen-
 234 ching at non-zero temperature after a sufficiently long relaxation time and domain coarsening,
 235 with a higher probability for those closer to the equilibrium polarization. At the same time,
 236 any of these states will be reached inevitably if the total spin is conserved during quenching,
 237 as in the Kawasaki [65] two-spin exchange algorithm, which is relevant for models describing
 238 transport phenomena caused by spatial inhomogeneity such as diffusion, heat conduction, etc.

239 Finally, we will mention some recent advances in the experimental observation of metastable
 240 states using sub-picosecond optical pulses, which we believe can be applied to reveal metastable
 241 states discussed here and in [25]. For instance, in the quasi-two-dimensional antiferromag-
 242 net Sr_2IrO_4 , a long-range magnetic correlation along one direction was converted into a glassy
 243 condition by a single 100-fs-laser pulse [66]. Atomic-scale $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices, coun-
 244 terpoising strain and polarization states in alternate layers, was converted by sub-picosecond
 245 optical pulses to a supercrystal phase in [67]. In a layered dichalcogenide crystal of $1T\text{-TaS}_2$,
 246 a hidden low-resistance electronic state with polaron reordering was reached as a result of a
 247 quench caused by a single 35-femtosecond laser pulse [68]. See also the references to relevant
 248 superconducting and magnetic materials with next-nearest-neighbor interactions mentioned
 249 in Introduction and [25].

7 Conclusion

In conclusion, we calculated the restricted free energy $F(\mathbf{m})$ as a function of polarization \mathbf{m} for the square-lattice J_1 - J_2 Ising model (at $J_2 < |J_1|/2$) within RLFA and using the MC method. Both approaches indicate the appearance of metastable states at low temperature, corresponding to local minima of $F(\mathbf{m})$ along the \mathbf{m} coordinate. The zero-polarization metastable state predicted by RLFA reflects the true metastable states with various polarization values at $\mathbf{m} \gtrsim 0.5$ that appear in our exact calculation and MC simulations of the restricted free energy. We show that RLFA predicts the slab-droplet phase transition for the J_1 - J_2 Ising model as a kink in the polarization dependence of $F(\mathbf{m})$ at low temperature. Exact calculations of $F(\mathbf{m})$ for a sample size of $L = 6$ reveal also additional slab-droplet transitions at $J_2 > 0.25$. We believe, easy-to-use RLFA can help reveal the presence of metastable states and geometrical phase transitions in more complex systems, e.g., with site or bond disorder and spin tunneling in a transverse field.

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