

Metastable states in the $J_1 - J_2$ Ising model

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Competing interactions of nearest and next-nearest-neighbor electron spins on a lattice with coupling constants J_1 and J_2 , respectively, give the system some interesting and unusual properties and may even underlie high temperature superconductivity according to the resonating valence bond theory. Here we study the $J_1 - J_2$ Ising model on the square lattice using the Random local field approximation (RLFA) and Monte Carlo (MC) simulations for various values of the ratio $p = J_2/|J_1|$. We show that the accuracy of RLFA is comparable to the mean field cluster approximation, but, in addition, and most surprisingly, RLFA predicts metastable states with zero polarization at low temperature for $p \in (0, 1)$. This is confirmed by our MC simulations, in which the system reaches these states after the energy relaxation of initially randomly oriented spins. Furthermore, in a moderate external field, the MC simulations reveal the existence of still other metastable states with nonzero polarizations. These states also appear when the initial state before relaxation has a nonzero polarization. We list compounds appropriate for experimental verification of our predictions, with high-temperature superconductors being the most suitable. These findings could be crucial for explaining the magnetic and electric properties of materials, in particular, under ultrafast nonequilibrium conditions.

In recent years, many compounds have been discovered in which electron spins-1/2 form a two-dimensional square lattice and interact with their nearest and diagonal next-nearest neighbors via isotropic exchange interaction with the constants J_1 and J_2 , respectively (Fig. 1) [1, 2]. This also includes the parent compound of the cuprate high-temperature superconductors La_2CuO_4 [3] and is likely relevant to iron-based superconductors [4]. The corresponding Heisenberg model has been studied extensively by a variety of methods, see e.g. [5] and references therein. For the values of the ratio $p = J_2/|J_1|$ near $p_0 = 1/2$, where two different ordered low energy states have the same energy, the quantum spin liquid ground state was predicted [1, 6, 7], which may be the key to solving the problem of high-temperature superconductivity according to the resonating valence bond theory [8]. This ground state has recently been observed experimentally in several compounds [2, 9], and another exotic nematic ground state at high magnetic field has also been reported [10].

The $J_1 - J_2$ Ising model, in which spins can only point in two directions, up and down, has also been thoroughly studied theoretically using cluster mean field theory [11–13], Monte Carlo (MC) simulations [11, 14–16], and tensor network simulations [17]. The nematic phase was predicted for moderate external fields [18, 19]. Although its implementations seem less common in nature, the easier to study the $J_1 - J_2$ Ising model nonetheless is interesting in its own right, and can also shed light on some properties of its more complex Heisenberg counterpart. This is especially true for the Ising model in a transverse field, where quantum fluctuations induce gap between two phases around p_0 [13, 19–21] with the valence-bond-

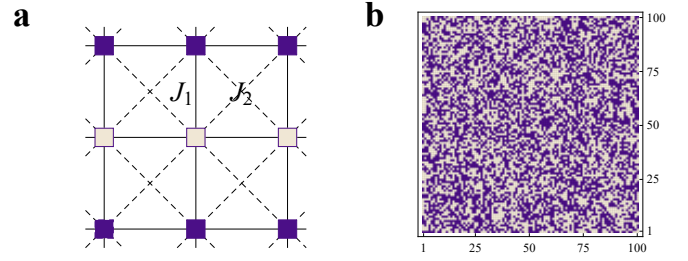


Fig. 1 | The $J_1 - J_2$ Ising model scheme. **a**, Square lattice of Ising spins (white - up, blue - down) with the interaction constant J_1 between nearest neighbors along horizontal and vertical (solid) lines and J_2 between next-nearest-neighbors along (dashed) diagonals. **b**, Random (very high temperature) spin configuration; the number of spins along each side of the square sample is $L = 100$.

solid state predicted [20]. Indeed, the phase diagrams of both models have a lot in common [1, 2, 7, 13].

At present, MC simulations can be used to calculate model properties with very high accuracy, nevertheless, easy-to-implement, adequate and efficient approximations are highly desirable to quickly and reliably determine the most salient system features. Here we use the random local field approximation (RLFA) [22] to study the phase transition in the $J_1 - J_2$ Ising model on the square lattice. We show that RLFA reveals metastable states with zero polarization at low temperature in zero field. We then confirm this finding with our MC simulation and show that, in fact, metastable states can have an arbitrary polarization value.

Thus, we consider the Hamiltonian

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

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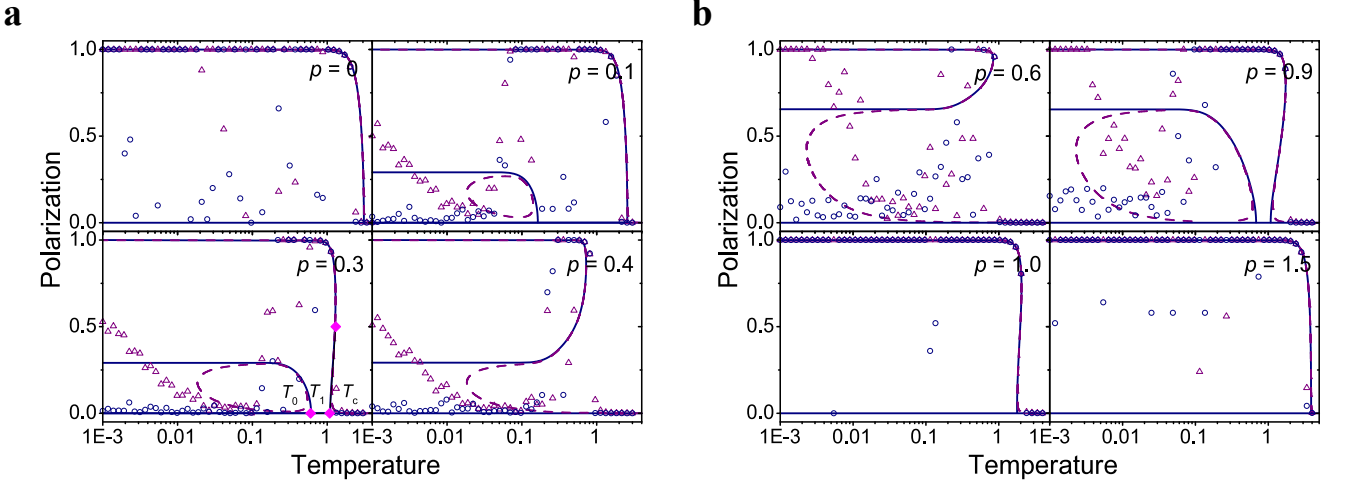


Fig. 2 | Polarization as a function of temperature obtained using the RLFA analytical approach and MC simulations. **a**, Solution of the RLFA equation (lines) and MC results (markers) for uniform polarization in a uniform field, $p < p_0$, and **b**, for staggered polarization in a staggered field, $p > p_0$. The magnitude of the field is $h = 0.001$ (purple dashed line and triangles) and $h = 0$ (blue solid line and circles). Each data point is derived from a single MC run with a random initial spin configuration at each temperature. The magenta diamonds in the bottom left panel (a) indicate the temperatures $T_0 < T_1 < T_c$.

where the sums are over nearest $\langle i, j \rangle$ and next-nearest (diagonal) $\langle\langle i, j \rangle\rangle$ neighbors (see Fig. 1a), as well as over each spin coupled to the external field h_i at its position; each s_i can take values ± 1 . In what follows, we set ferromagnetic (FM) $J_1 = -1$ and antiferromagnetic (AFM) $J_2 = p > 0$ coupling constants, providing together the frustration of the system. For this choice of couplings, the ground state is FM at $p < p_0$ (being AFM with Néel checkerboard order for $J_1 > 0$) and striped or super-AFM at $p > p_0$; at $p = p_0$ the ground state is not ordered.

The starting point of RLFA is the exact formula for the average spin value [22, 23]:

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

where $\beta = 1/T$ is the inverse temperature (in energy units), $h_i^s = \sum_j J_{ij} s_j$ is the local field acting on the spin s_i due to neighboring spins s_j ; the brackets stand for the thermal averaging.

With RLFA the fluctuations of each spin are considered as independent, and averaging in Eq. (2) is carried out with the product of the probability distributions for each spin [22, 24]:

$$P(s_i) = (1 + m_i s_i)/2, \quad (3)$$

where $m_i = \langle s_i \rangle = m e^{i\mathbf{q}\cdot\mathbf{r}_i}$ is the thermally averaged polarization at the position $\mathbf{r}_i$ determined by the propagation vector $\mathbf{q}$. The uniform polarization corresponds to $\mathbf{q} = (0, 0)$, while the staggered polarization refers to the vectors $(0, \pi)$ and $(\pi, 0)$. The same applies to the spatial dependence of the external field h_i .

Eqs. (2) and (3) together constitute the essence of RLFA. Eq. (2), which is a seventh degree polynomial in m according to the eight neighbor spins in the model, can be solved numerically.

The solution of the RLFA equation for uniform (at $p < p_0$) and staggered (at $p > p_0$) polarization is shown in Fig. 2. This solution corresponds to the zero value of the Landau potential derivative with respect to m and, therefore, corresponds to its local minimum (stable or metastable state), local maximum or inflection point (unstable states). In the absence of an external field, zero polarization is always a solution to the equation, and it is unique and stable at high temperatures. At zero temperature, there is always (except in the case of $p = p_0$) another solution $m = 1$, which corresponds to full polarization and supposed to be stable. And there is still a third solution about $m \approx 0.29$ for $p \in (0, p_0)$ and $m \approx 0.65$ for $p \in (p_0, 1)$. It is natural to assume that it corresponds to a local maximum of the Landau potential separating two local minima at $m = 1$ and $m = 0$, the first of which is a stable solution, and the second is a metastable one. In an external field, the metastable state exists only in a certain temperature window (dashed purple lines in Fig. 2) and completely disappears at high fields.

The phase diagram for the $J_1 - J_2$ Ising model obtained using RLFA is shown in Fig. 3a. The critical temperatures obtained from the maxima of the dielectric susceptibility in the MC simulation (see Methods) are given for comparison and are in good agreement with the literature data [14, 16]. Within RLFA, the transition turns out to be of the first order for p from about 0.25 up to 1.25, while recently it has been shown to be of the second order everywhere using the tensor network

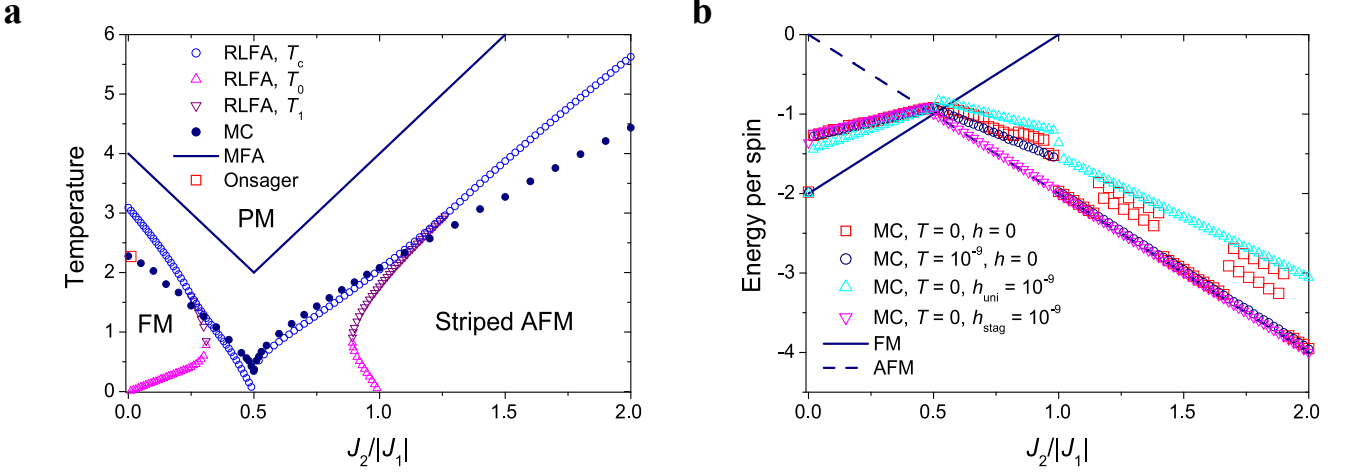


Fig. 3 | Critical temperatures and energies in the $J_1 - J_2$ Ising model as functions of $p = J_2/|J_1|$. **a**, Phase diagram obtained using RLFA, MFA and MC. Solid dark blue curves correspond to MFA, open blue circles, magenta up-triangles and purple down-triangles are T_c , T_0 and T_1 obtained within RLFA (Fig. 2a). The red square is the exact Onsager’s solution for the 2D Ising model. Dark blue filled circles are calculated using the MC method. **b**, Energy per dipole obtained using MC simulation at zero temperature in the absence of a field (red squares), at a temperature $T = 10^{-9}$ and a field $h = 0$ (blue circles) and $T = 0, h = 10^{-9}$ (cyan up-triangles for uniform and magenta down-triangles for staggered field). Each data point is averaged over 100 samples (see Methods). Standard deviations of the energy distribution histogram over 100 samples for each data point are smaller than the markers (see Extended Data Fig. 4). Blue solid and dashed lines correspond to the energy of the FM and AFM states, respectively.

simulation technique [17], which apparently resolves the long-standing dispute about the order of the phase transition at $p > p_0$ [15]. The agreement between the RLFA and MC results is good for $p > p_0$. At these points, the rise of polarization is steep enough, resulting in smaller fluctuations, which are neglected by RLFA. However, at p just below p_0 , the discrepancy turns out to be significant, with the critical temperature determined by RLFA going almost linearly to zero, where it should have a finite value. In Fig. 3a, we also traced the temperatures T_0 and T_1 (see Fig. 2a, left bottom panel), which indicate the range of the zero-polarization metastable state and the temperature of the first-order phase transition in the absence of an external field. The overall accuracy of RLFA turns out to be comparable to the commonly used cluster mean field approximation (MFA) [11–13].

It should be noted that neither staggered for $p \in (0, p_0)$ nor uniform for $p \in (p_0, 1)$ polarizations with $m = 1$ are solutions to the RLFA equation, although they are very close to it. At the same time, these states are metastable at zero temperature. Indeed, any spin flip in these states leads to an increase in energy $8J_2$ and $-8(J_1 + J_2)$, respectively.

To further explore the metastable states, we perform MC simulations with single-spin-flip dynamics (see Methods), making a deep quench from a (high-temperature) random spin configuration. It was previously shown that under these conditions a 2D Ising system (corresponding to $p = 0$) on the square lattice at zero temperature reaches not only a completely polarized ground state, but sometimes a frozen stripe state with a probability

of about ≈ 0.3 [25, 26]. Later, this behavior and the probability of the occurrence of a metastable state were explained by revealing a deep connection between the zero-temperature coarsening with critical continuum percolation [27, 28].

In the absence of an external field, MC simulations do indeed show metastable states with nearly zero polarization for $p \in (0, 1)$, although at temperatures several times lower than follows from RLFA (Fig. 2). Typical spin configurations of these states at zero temperature are shown in Figs. 4b, 4f. For $p \in (0, p_0)$ the real-space correlation function of these states is exponential and the correlation length is about $l_c \approx 1.8$ with no apparent dependence on p . The result is the same when the initial state at each temperature is AFM for $p \in (0, p_0)$ or FM for $p \in (p_0, 1)$. Furthermore, when we start relaxation from a random spin configuration with nonzero polarization at low temperatures, the resulting state also has a nonzero polarization, slightly higher than the initial one (Extended Data Fig. 1). This proves the existence of metastable states with an arbitrary polarization value in the $J_1 - J_2$ Ising model, in contrast to the case $p = 0$, when there are only metastable states with zero polarization [25, 26]. Next, we discuss quenching from random spin configurations with zero polarization or close to it.

For a small external field (uniform for $p < p_0$ and staggered for $p > p_0$) at low temperature, the metastable states do not relax to the ground state, as it would be according to RLFA, but get stuck instead in other metastable states (Fig. 2). At zero temperature, these metastable states appear already in an infinitesimally low

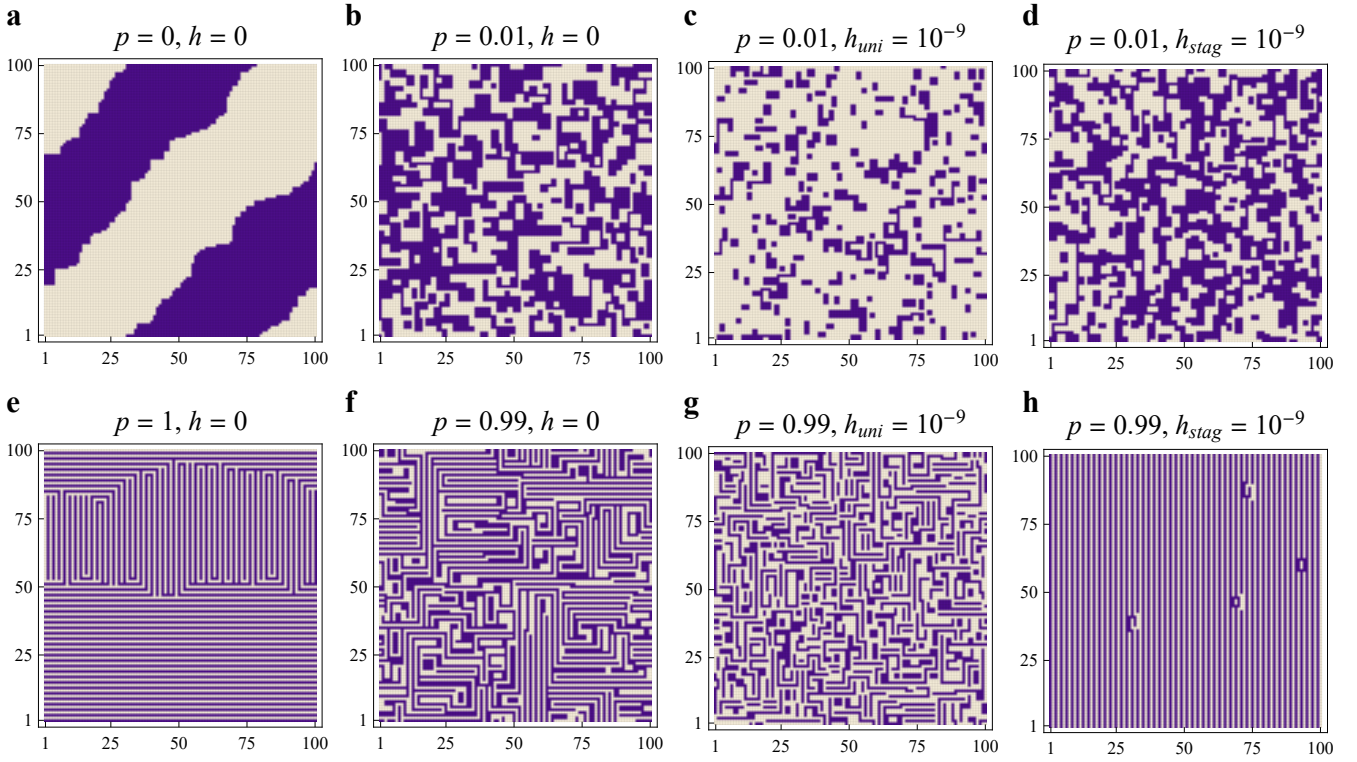


Fig. 4 | Spin configuration in relaxed states at zero temperature. Examples of spin configurations of final absorbing states after energy relaxation, starting from a random spin configuration, has been performed in the absence or in a very small uniform or staggered external field h for the values of $p = J_2/|J_1|$ equal to 0, 0.01, 0.99, and 1.

external field (down to 10^{-9}) with their typical configurations shown in Figs. 4c, 4h. The polarization of these states is about $m \approx 0.5$ for $p < p_0$, and very close to $m = 1$ for $p > p_0$. It first decreases with increasing temperature, and then increases to $m = 1$ in both cases in accordance with the RLFA solution. When a uniform field is applied for $p > p_0$, which is an experimentally relevant situation, the system relaxes into even other metastable states, see Fig. 4g.

We note that some data with an intermediate polarization value in Fig. 2 actually correspond to incompletely relaxed FM and AFM states divided into slowly relaxing large domains (Figs. 4a, 4e). At the same time, the energy of these states does not differ much from a completely ordered state, but it is significantly higher for truly disordered states (Extended Data Fig. 2). Thus, we plot the energy per dipole at zero temperature as a function of p (Fig. 3b), where the metastable states at $p \in (0, 1)$ (red squares) are clearly visible. For some values of $p > p_0$, the energy of metastable states in Fig. 3b appears to be slightly higher and goes above the general trend. However, it suffices to apply an infinitely small temperature of $T \sim 10^{-9}$ for these fragile metastable states to quickly relax into robust metastable states for $p \in (p_0, 1)$ or stable states for $p > 1$. Similar to the 2D Ising model ($p = 0$) [25, 26], relaxation of robust metastable states into the ground FM or AFM states

is apparently determined by the activation energy E_a as $\tau \simeq L^3 \exp(-E_a/T)$ (Extended Data Fig. 3). For $p \in (0, p_0)$, where the ground state is FM, metastable states consist of rectangles with more than two spins on each side, surrounded by spins with the opposite direction. These rectangles are then interconnected, making up the whole picture (Figs. 4b - 4d). The energy cost for a spin flip in the corner of the rectangle is $4J_2$, on its side is $4J_1$, and in the middle of a long line of one spin wide is $8J_2$. Thus, the activation energy in this case is determined by the minimum of these energies, $E_a = 4J_2$. At the same time, the external field sufficient for spin flip in the metastable state is half this value. For $p = 0.05$, for example, it is $h_{\text{bar}} = 0.1$ in agreement with the MC simulation results in Extended Data Fig. 3. For $p \in (p_0, 1)$, the excitations are rings with opposite polarization inside (Figs. 4f - 4h) and the activation energy $-4(J_1 + J_2)$.

According to the results of the MC simulation, the energy landscape of the $J_1 - J_2$ Ising model resembles a hedgehog when mapped onto an imaginary two-dimensional configuration space with radially increasing polarization. Different spin configurations with a certain polarization have different energies, but their average energy decreases with increasing polarization. At sufficiently low temperatures, relaxation of the system to the ground state with polarization $m = 1$ becomes impossible because of the traps due to metastable states

lying on its way there.

Experimentally, strongly nonequilibrium conditions equivalent to quenching can be achieved in ultrafast light-pump experiments, such as [29]. Note that metastable states have recently been reported in incipient ferroelectric SrTiO_3 under high-intensity THz pumping [30, 31] and were predicted from *ab-initio* calculations in the antiferroelectric NaNbO_3 [32]. The nonequilibrium conditions can also be created by applying an external field to bring the system out of the fully polarized state with $m = 1$ and turning off the field abruptly. The switch-off time in this case must be less than the spin relaxation time, while in electrical circuits it is limited to hundreds of microseconds for magnetic fields [33], and hundreds of picoseconds for electric fields [34]. However, in experiments with laser pumping, it can be short enough in both cases [35–38]. We also note that in some cases metastable states can be achieved with slowly varying external fields. Indeed, as described in the previous section, a field less than $h_{\text{bar}} = 2J_2$ does not cause a transition between two metastable (or metastable and stable) states. However, for $p > p_0$, a field larger than $h_{\text{tr}} = E_{\text{AFM}} - E_{\text{FM}} = -4J_2 - 2J_1$, see Fig. 3, can make the FM state stable, i.e. the ground state. Therefore, by cooling in the field $h_{\text{tr}} < h < h_{\text{bar}}$ and then turning the field off, we can trap the system in a metastable state. This is possible when $h_{\text{tr}} < h_{\text{bar}}$, i.e. for $p \in (p_0, 2/3)$.

The metastable states discussed here should also exist in a moderate transverse external field, and therefore they could be equally expected in the Heisenberg quantum model relating to magnetism in high-temperature superconductors [4, 39], depending on p , however. In La_2CuO_4 with the Néel AFM ground state, the values of $J_1 \approx 1300$ K [3] and later about 1700 K [40] were obtained from fitting inelastic neutron scattering data in general agreement with *ab-initio* calculations [41–44]. However, the value and the sign of J_2 differ in different sources with $p \approx -0.1$ reported in [3, 43] and ranging from about 0.2 [40, 42, 44] to 0.8 in [41]. At the same time, in Sr_2IrO_4 , similar to cuprates in many aspects, apart from superconductivity, $J_2 < 0$ [45–47] and, consequently, with no frustration, metastable states are not

expected.

For iron-based superconductors, which have a striped AFM ground state in their parent compounds [39, 48], it was shown that biquadratic coupling together with isotropic in-plane coupling constants explain many of the observed features [49, 50]. For CaFe_2As_2 , for example, experimental data are well fitted for $J_1 = 102$ K and $p = 0.86$ [49]. We also mention LaFeAsO , where p is very slightly more than one, as calculated in [51], while it was claimed about 0.71 in [52, 53]. In both compounds $J_1 > 0$ and they could be tested as well as La_2CuO_4 for metastable states.

Other suitable magnetic compounds corresponding to the $J_1 - J_2$ Heisenberg model also include VOMoO_4 with $J_1 = 100 - 150$ K and $p \simeq 0.2$, and the Néel temperature $T_N = 42$ K [54, 55]. In $\text{BaCdVO}(\text{PO}_4)_2$, the ground state is striped AFM with $J_1 = -3.6$ K and $J_2 = 3.2$ K [10, 56], which gives $p \simeq 0.9$. However, the expected temperature of metastable states is approximately two orders of magnitude lower (Fig. 2) than the already low phase transition temperature $T_N = 1.05$ K [10], which may complicate its experimental study. In PbVO_3 , where $J_1 \approx 190 - 200$ K and $p \approx 0.2 - 0.4$ is close to the gap in the phase diagram around p_0 , there is no long-range magnetic ordering down to 1.8 K [57]. The solid solution $\text{Sr}_2\text{Cu}(\text{Te}_{1-x}\text{W}_x)\text{O}_6$ is unique for studying frustrated square-lattice antiferromagnetism as it can be tuned from the Néel ($x = 0$, $J_1 \approx 83$ K, $p \approx 0.03$) to the striped AFM order ($x = 1$, $J_1 \approx 14$ K, $p \approx 7.92$) by varying the composition [58]. Thus, this compound may also be a preferred choice for studying metastable states.

In conclusion, using RLFA, we predict the existence of metastable states with zero polarization in the $J_1 - J_2$ Ising model at low temperature. Our MC simulation confirm this predication but also indicate metastable states with an arbitrary polarization value. We point to some antiferromagnets, including known high-temperature superconductors, where these states can be revealed at low temperature. These findings may be crucial for explaining the magnetic and electric properties of some materials and may directly manifest themselves, in particular, under the nonequilibrium conditions of modern experiments with high-power ultrashort light pumping.

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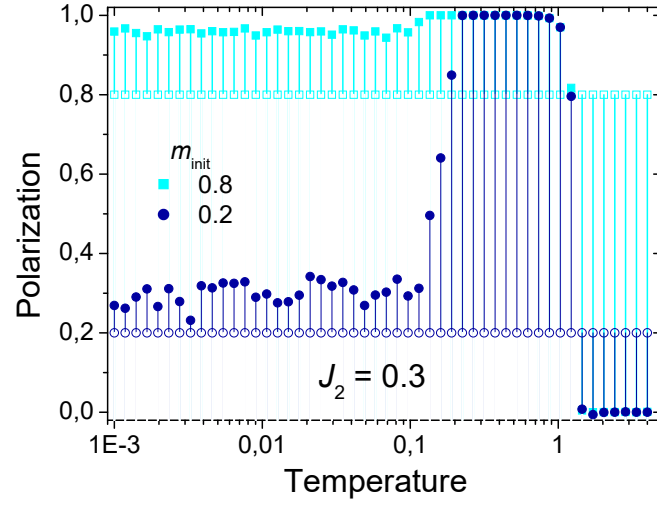
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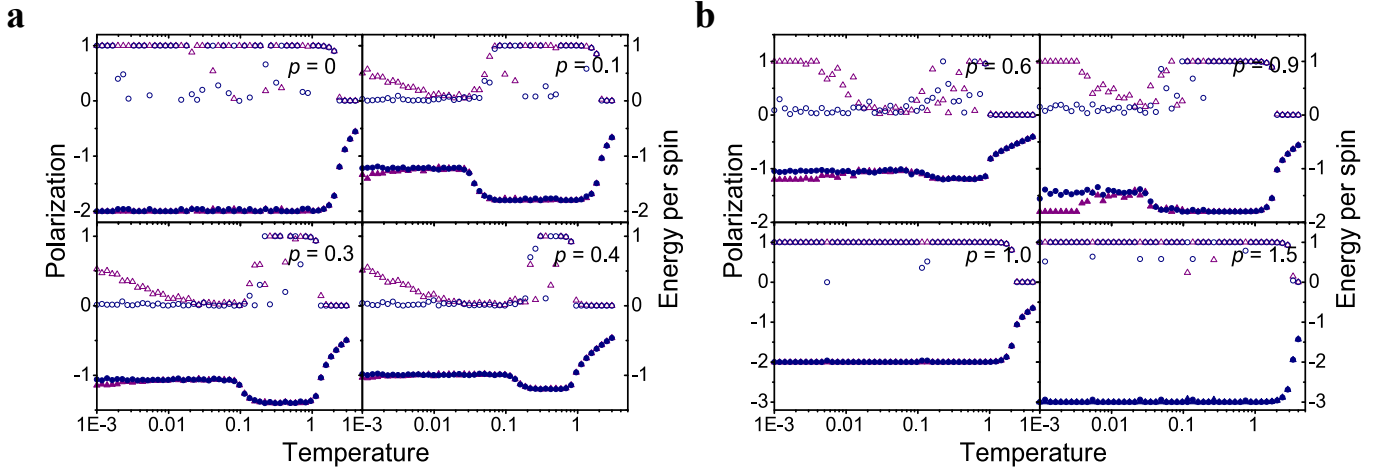
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Methods

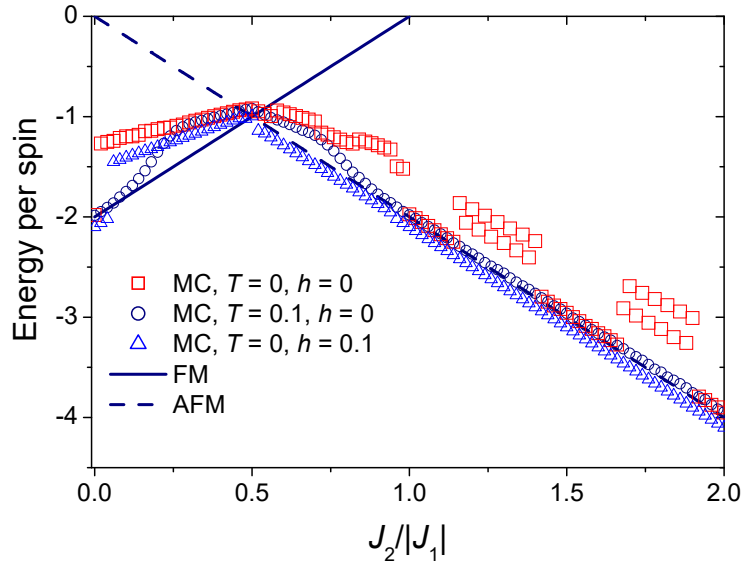
We perform Monte Carlo simulations with single-spin-flip Glauber dynamics at zero temperature and Metropolis dynamics at low temperatures, making a deep quench from a high-temperature (random) spin configuration, according to the following standard algorithm. If a randomly chosen spin flip leads to a negative energy change $\Delta E < 0$, then a new state is accepted. Otherwise, when $\Delta E \geq 0$, the probability of acceptance is $\alpha = \exp(-\Delta E/T)$ for the Metropolis algorithm, and $\alpha/(1 + \alpha)$ for the Glauber dynamics. Both algorithms satisfy the detailed balance criteria and give the same final result. Periodic boundary conditions were used in all calculations and the sample size was equal to $L = 100$. To obtain the data in Fig. 2 and Extended Data Fig. 2, relaxation was performed at each temperature, starting from a random spin configuration, with 10^5 Monte Carlo steps per spin (MCS) used for thermalization and the same number of MCS for subsequent calculations of thermodynamic quantities for each run. For staggered polarization, the largest of the values corresponding to the propagation vectors $\mathbf{q} = (0, \pi)$ and $\mathbf{q} = (\pi, 0)$ is taken. We also calculated the susceptibilities of N interacting spins via fluctuations of the average spin, $s = N^{-1} \sum_{i=1}^N s_i e^{i\mathbf{q}\cdot\mathbf{r}_i}$, with the propagation vector $\mathbf{q}$ as $\chi = NT^{-1}(\langle s^2 \rangle - \langle s \rangle^2)$ and obtained critical temperatures from their maxima (Fig. 3a). When simulating at zero temperature (Fig. 3b), each data point was averaged over 100 samples with a relaxation time of 10^4 MCS (Extended Data Fig. 4).



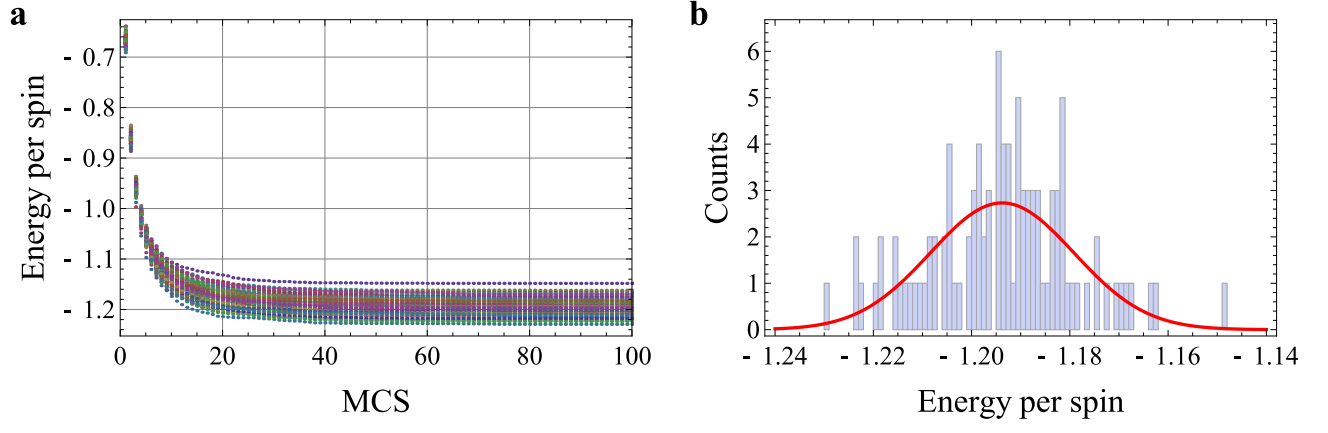
Extended Data Fig. 1 | Polarization versus temperature. Polarization after Monte Carlo relaxation (10^4 MCS) for $J_2/|J_1| = 0.3$ and two values of initial polarization $m_{\text{init}} = 0.2$ and 0.8 .



Extended Data Fig. 2 | MC results for polarization and energy per dipole for different values of $p = J_2/|J_1|$. **a**, Uniform polarization (left y-axis) and energy per dipole (right y-axis) in a uniform field, $p < 1/2$. **b**, Staggered polarization and energy per dipole in a staggered field, $p > 1/2$. Open markers correspond to polarization, filled markers indicate energy. Purple triangles correspond to the field magnitude $h = 0.001$, blue dots correspond to the absence of a field. Each data point is derived from a single MC run with a random initial state at each temperature.



Extended Data Fig. 3 | Energy per dipole obtained by MC simulations for different values of the ratio $p = J_2/|J_1|$. Red squares correspond to zero temperature and no field, dark blue circles and blue triangles correspond to $T = 0.1$, $h = 0$, and $T = 0$, $h = 0.1$, respectively. The applied external field is uniform for $p < 1/2$, and it is staggered for $p > 1/2$. The standard deviations of the energy distribution histogram over 100 samples is smaller than the markers (see Extended Data Fig. 4). Dark blue solid and dashed lines correspond to the energies of the FM and AFM states at $h = 0$. For $T = 0$, $h = 0.1$, the transition between stable and metastable states occurs around $p_{\text{FM}} = 0.05$ and $p_{\text{AFM}} = 0.95$.



Extended Data Fig. 4 | Monte Carlo simulation for $J_2/|J_1| = 0.1$ at zero temperature. **a**, Energy relaxation for 100 samples. **b**, Final energy histogram and superimposed Gaussian function with a calculated mean value of $\langle E \rangle = -1.19$ and a standard deviation of $\sigma_E = 0.015$.