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Field induced ordering in highly frustrated antiferromagnets

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We predict that an external field can induce a spin order in highly frustrated classical Heisenberg magnets. We find analytically stabilization of collinear states by thermal fluctuations at a one-third of the saturation field for kagome and garnet lattices and at a half of the saturation field for pyrochlore and frustrated square lattices. This effect is studied numerically for the frustrated square-lattice antiferromagnet by Monte Carlo simulations for classical spins and by exact diagonalization for $S = 1/2$. The field induced collinear states have a spin gap and produce magnetization plateaus.

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Frustrated magnets, classical and quantum, exhibit spectacular and often unexpected behaviors [1]. A few of them do not order at any $T > 0$. Extensive residual entropy in the classical ground state in such cases prevents the realization of the usual order by disorder scenario [2]. An external magnetic field changes the degeneracy and topology of the ground state manifold stabilizing, eventually, the (nondegenerate) saturated state at $H > H_{\rm sat}$. If the order by disorder effect occurs in a finite field and suppresses residual entropy, then, such an effect can be used for practical applications. During a demagnetization process a spin system has to regain its entropy and, therefore, the whole crystal will cool down. Examples of geometrically frustrated AFMs on kagome [3], pyrochlore [4], and garnet [5] lattices include magnetic compounds with rather small exchange constants opening the way for experimental tests of their finite field behavior.

In this Letter, we predict that fluctuations stabilize collinear spin configurations at rational values of the field $H/H_{\rm sat} = 1/2$ or $1/3$. We present an analytical proof of the field induced ordering driven by thermal fluctuations and suggest a similar role for quantum fluctuations on the basis of spin-wave and numerical results. In fact, an ordered spin phase in a finite field has already been observed in gadolinium garnet $\mathrm{Gd_3Ga_5O_{12}}$, though dipolar anisotropy plays a crucial role in this material [5].

To be specific we use as an example the frustrated square-lattice antiferromagnet (FSAFM) [6] in an external field:

$$\hat{\mathcal{H}} = J\sum_{\rm n.n.} \mathbf{S}_i\cdot\mathbf{S}_j + J'\sum_{\rm n.n.n.} \mathbf{S}_i\cdot\mathbf{S}_j - H\sum_i S_i^z \ . \qquad (1)$$

First, we present analytical arguments, making them as general as possible, in order to include the other highly frustrated AFMs. Second, we consider numerical results for classical and quantum FSAFMs in a magnetic field, which confirm our predictions.

Let us briefly discuss magnetically ordered phases of the FSAFM. For small diagonal exchange $J' < 0.5J$, classical spins form the Néel state in zero field. At $J' > 0.5J$ the classical ground state consists of two $\sqrt{2}\times\sqrt{2}$ interpenetrating antiferromagnets, which are locked by fluctuations in a striped AFM state described by a single wave-vector $(\pi, 0)$ or $(0, \pi)$ [2]. An applied magnetic field cants the spins and creates an easy plane for the AFM sublattices. Thus, both the Néel and striped states have a transverse spin order in a finite field. Thermal and quantum fluctuations destroy the transverse order in the vicinity of the highly frustrated point $J' = 0.5J$. As an illustration we present in Fig. 1 the phase diagram for the spin-1/2 model at $T = 0$ obtained in the linear spin-wave theory. This phase diagram agrees with the previous zero-field studies [6], which suggest a quantum disordered ground state for $S = \frac{1}{2}$ near $J' = 0.5J$. Since the disordered singlet phase has a spin gap, it becomes unstable above a finite critical field.

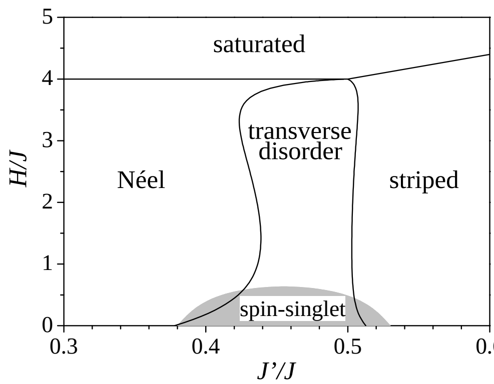

FIG. 1. Zero temperature phase diagram of the spin-1/2 FSAFM in external field obtained from the linear spin-wave theory. The shaded region indicates the stability region of the quantum disordered state with a spin gap.

We now focus on the classical critical point $J' = 0.5J$, where the Hamiltonian (1) can be written up to a constant term as a sum over edge-sharing plaquettes:

$$\hat{\mathcal{H}} = \frac{1}{2^n}\sum_\alpha \left\{ J\,|\mathbf{L}_\alpha|^2 - HL_\alpha^z \right\} , \qquad (2)$$

where $\mathbf{L}_\alpha = \sum_{i\in\alpha} \mathbf{S}_i$ is the total spin in plaquette α and $n = 2$. The block form of the spin Hamiltonian (2) is common to all highly frustrated spin models. For AFMs on kagome and garnet lattices the blocks are triangles and for a pyrochlore AFM they are tetrahedra with the corner-sharing arrangements and $n = 1$ in all three cases.

A spin configuration minimizes the energy (2) at $H = 0$ provided $\mathbf{L}_\alpha = 0$ for each plaquette. This constraint can be satisfied for many classical states. We estimate the number of continuous degrees of freedom in the ground state of the N-site FSAFM as $\sim N^{1/2}$. For pyrochlore and kagome AFMs the degeneracy is larger and the dimensionality of the ground state manifold scales with N [3,4,7]. Thus, zero-field properties are not universal for disordered frustrated AFMs. We now show that a universal behavior does appear in a magnetic field.

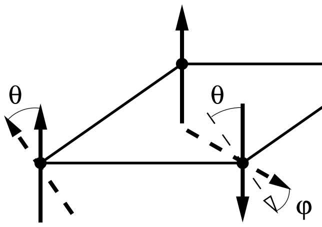

FIG. 2. The collinear spin structure for a four-spin block at $H = 0.5H_{\rm sat}$. Rotations by angles θ and φ parameterize the zero-energy and soft modes in this state.

In a finite field the classical energy (2) is minimized for spin configurations with the plaquette magnetization $L^z_\alpha = H/(2J)$. There are many degenerate classical states which satisfy this constraint below the saturation field $H_{\rm sat} = 8JS$ (pyrochlore and FSAFM) or $6JS$ (kagome and garnet). Generally, all these states are noncollinear as, e.g., canted Néel and striped phases for FSAFM. Collinear spin arrangements appear only at special rational values of the applied field: at $H_c = \frac{1}{2}H_{\rm sat}$ for 4-spin blocks (FSAFM, pyrochlore), the up-up-up-down (uuud) structure, see Fig. 2, and at $H_c = \frac{1}{3}H_{\rm sat}$ for 3-spin blocks (kagome, garnet), the up-up-down structure. The distribution of down-spins on the lattice in a collinear state is not unique: it only obeys the one down-spin per plaquette condition.

Zero-energy modes correspond to continuous distortions of a given classical ground state, which do not violate the magnetization constraint. To construct a zero mode for the collinear states described above one has to draw an (open) line through the lattice points with adjacent sites on a line occupied by antiparallel spins with no plaquettes crossed more than once. A zero mode consists of a simultaneous rotation of all spins along the line by an angle θ (Fig. 2). The complete set of zero modes is constructed when a set of such lines goes through all upspins. Zero modes connect a uuud state to noncollinear ground state configurations. Hence, the collinear states are singular points on the ground state manifold. There are no other collinear states in the local neighborhood of a selected collinear state.

Moessner and Chalker [7] have recently discussed the order by disorder phenomenon in zero field for classical frustrated models in terms of a local topology of the ground state manifold. They reached definite conclusions only for an XY magnet on the pyrochlore lattice and for a Heisenberg kagome lattice AFM. We show that a similar analysis predicts stabilization of collinear states near special rational values of the applied field. Let $\mathbf{x}$ denote the coordinates on the ground state manifold and $\mathbf{y}$ be the transverse directions, which span the rest of the configuration space. Low-energy excited states are described by the quadratic Hamiltonian $\mathcal{H}_2 = \sum_l \epsilon_l(\mathbf{x})y_l^2$ resulting in a probability distribution

$$Z(\mathbf{x}) = \int dy_l e^{-\beta\mathcal{H}_2} \propto \prod_l [T/\epsilon_l(\mathbf{x})]^{1/2} \qquad (3)$$

over the ground state manifold at low temperatures. At a special point $\mathbf{x}_0$ some of the stiffnesses $\epsilon_l(\mathbf{x}_0)$ may vanish making $Z(\mathbf{x}_0)$ divergent. The corresponding coordinates y_l describe soft modes. The appearance of the order by disorder depends on a number of soft modes, which exist for a given classical ground state $\mathbf{x}_0$.

A special feature of the collinear states with finite magnetization is that each zero mode generates exactly one soft mode. They are constructed in the following way. First, all spins along a zero mode line are rotated by θ about the axis perpendicular to the field and, second, all down spins on the same line are rotated by an angle φ about the field direction, see Fig. 2. The total spin of each plaquette in a deformed state is $\mathbf{L}_\alpha \approx S(0, \theta\varphi, 2)$ for small θ and φ and contributes to an energy increase $\sim \theta^2\varphi^2$. Thus, we identify x_l with θ and y_l with φ.

To allow the order by disorder selection, not only has the probability density $Z(\mathbf{x}_0)$ to diverge, but the statistical weight $\int Z(\mathbf{x})d\mathbf{x}$ must be concentrated entirely near the collinear spin states. We check this by integrating $Z(\mathbf{x})$ in the D-dimensional neighborhood of a collinear state parameterized by zero modes. Since each zero mode has one soft mode with $\epsilon_s(\mathbf{x}) \sim x^2$, the integral

$$\int Z(\mathbf{x})d\mathbf{x} \sim \int x^{-D} d^D x \qquad (4)$$

diverges independently of the actual value of D. There is a vanishingly small probability of finding a spin system in one of the noncollinear states surrounding a given collinear configuration. Hence, the order by disorder selection occurs: thermal fluctuations stabilize a discrete set of collinear states. This result relies only on the special block structure (2) of the spin Hamiltonian and the symmetry properties of the collinear states. Therefore, the order by disorder in an external field is a universal effect and occurs for all frustrated Heisenberg AFMs: apart from the FSAFM the uuud states at $H = \frac{1}{2}H_{\rm sat}$ are stabilized for a pyrochlore AFM and the uud states appear for Heisenberg AFMs on kagome and garnet lattices at $H = \frac{1}{3}H_{\rm sat}$. A subsequent selection between collinear states with different patterns of down-spins is made to maximize D in Eq. (4) and corresponds in the case of FSAFM to the most symmetric $\mathbf{q} = 0$ uuud state. The above proof of the field induced ordering is based on the special property of the collinear states with finite magnetization. Namely, an equal number of zero and soft modes. For a Heisenberg magnet on the pyrochlore lattice in zero field the same equality holds only approximately: in the leading order in the number of spins N. Therefore, no analytical conclusion has been reached in a zero field case, while numerical simulations indicated no ordering [7].

For quantum models in the limit $S \gg 1$ we check the relative stability of different states by comparing their zero-point oscillation energies: $\frac{1}{2}\sum_{\mathbf{k}}\omega_{\mathbf{k}}$. The magnon spectrum of the $\mathbf{q} = 0$ uuud state has four modes:

$$\omega_{1,2} = JS\sqrt{\eta_{\mathbf{k}}}\ , \quad \omega_{3,4} = 2JS \pm JS\sqrt{4-\eta_{\mathbf{k}}} \qquad (5)$$

with $\eta_{\mathbf{k}} = (1-\cos k_x)(1-\cos k_y)$. Zero-point contributions to the energies of the $\mathbf{q} = 0$ uuud state, the Néel state, and the striped AFM at $J' = 0.5J$ and $H = \frac{1}{2}H_{\rm sat}$ are $0.57JS$, $0.69JS$, and $0.81JS$, respectively. (They have the same classical energy.) Thus, this comparison suggests that quantum fluctuations also select the collinear states in a magnetic field due to their large number of soft modes. Collinear states preserve the $O(2)$-rotational symmetry. Hence, their renormalized magnon spectrum becomes gapped and plateaus arise on the magnetization curve. The spin-density wave of the $\mathbf{q} = 0$ uuud state in FSAFM is a superposition of spin harmonics with wave-vectors $(0,0)$, (π,π), $(\pi,0)$ and $(0,\pi)$.

To test these predictions we have performed Monte Carlo (MC) simulations for the model Eq. (1) with unit vector spins. Fig. 3, top panel shows the magnetization curve at $T = 0.1J$ obtained with 2×10^5 MC steps per spin for each point. The field derivative of the magnetization has a dip around $H = 4J = \frac{1}{2}H_{\rm sat}$, which indicates the presence of a new plateau phase. A jump in the magnetization and a hysteresis of about $\Delta H \simeq 0.3J$ suggests a first-order transition to a high-field phase. To determine the nature of the spin state at the plateau we calculated different components of the static structure factor $S^{\alpha\beta}(q) = \frac{1}{N^2}\sum_{r,x} e^{i\mathbf{q}\cdot\mathbf{x}}\langle S^\alpha_r S^\beta_{r+x}\rangle$. The Néel and the collinear states have nonzero transverse components $S^{xx} = S^{yy}$ at $\mathbf{q} = (\pi,\pi)$ and $\mathbf{q} = (\pi,0)$ or $(0,\pi)$, respectively. The uuud state has nonzero longitudinal components $S^{zz}(q)$ at all the above vectors simultaneously. The static structure factor presented in Fig. 3, bottom panel was obtained by averaging 50 'instant shots' separated by 10^3 MC steps. In the region of weak (strong) diagonal exchange J' the data clearly support the Néel (striped) type of spin correlations. Nonzero harmonics in the longitudinal structure factor both at $\mathbf{q} = (\pi,0)$ and $\mathbf{q} = (\pi,\pi)$ exist only for $0.494 < J'/J < 0.508$. We have also checked that neither a lower field of $H = 2J$, nor a higher field of $H = 6J$ induces a nonzero value of $S^{zz}(q)$ at these points in the Brillouin zone. Thus, these results unambiguously identify the spin configuration on the plateau as the $\mathbf{q} = 0$ uuud state.

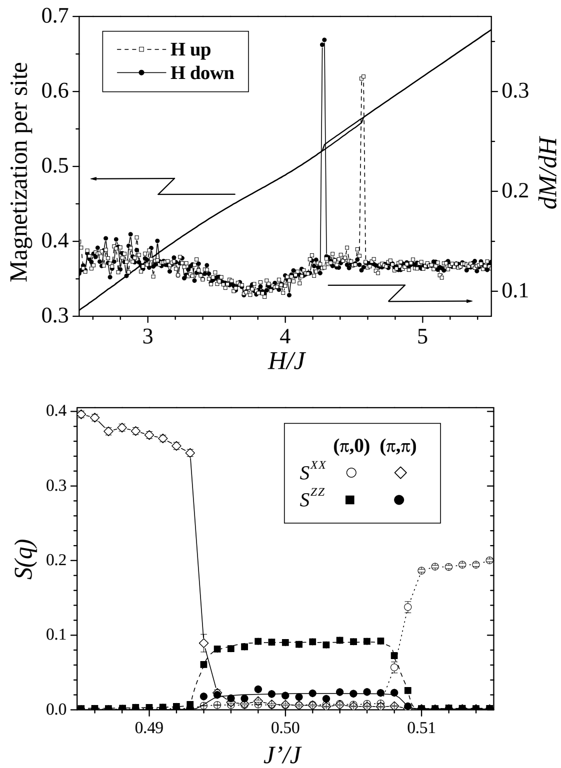

FIG. 3. Monte Carlo results for the classical FSAFM: the magnetization curve for $J'/J = 0.5$ (top panel) and the static structure factor $S^{\alpha\beta}(q)$ vs. the frustration parameter for $H = 4J$ (bottom panel) on a 24×24 lattice at $T = 0.1J$.

The uuud phase breaks the translational symmetry in such a way that the wave-vectors belonging to different irreducible representations of the space group are mixed: (π,π) vs. $(\pi,0)$ and $(0,\pi)$. Hence, the spin structure of the uuud phase is a mixture of different order parameters. In such a case the phase transition to a disordered spin liquid state at the high-field end of the plateau, Fig. 3, must go either via a first order transition or in several steps with an intermediate supersolid state.

We have also studied the quantum spin-1/2 model (1) at $T = 0$ by Lanczos diagonalizations of finite clusters. Fig. 4 presents the magnetization $m = M/S$ vs. field at $J'/J = 0.6$. There are no magnetization plateaus in the thermodynamic limit for $m > 1/2$. To determine whether the plateau at $m = \frac{1}{2}$ remains after finite size scaling we show its width in the inset for three cluster sizes as a function of J'/J. If a plateau disappears in the thermodynamic limit, its width should decrease as $\Delta m = 1/N$. Therefore, the ratio of the plateau-widths of the 4×4 and 6×6 clusters should be 16/36 or less if the magnetization curve has a nonzero slope for $N\to\infty$. The ratio 16/36 is exceeded for $m=1/2$ only in the interval $0.49 \lesssim J'/J \lesssim 0.66$, where we would expect to have a plateau on an infinite lattice.

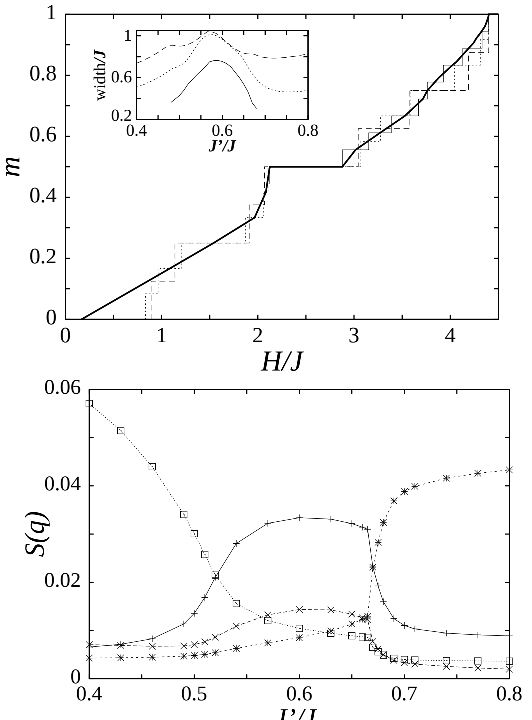

FIG. 4. Exact diagonalization results for the spin-1/2 FSAFM. Top panel: the magnetization curve for $J'/J=0.6$. Inset: width of the finite-size plateau with $m=\frac{1}{2}$ as a function of J'/J. The lines are for lattices of size 4×4 (dashed), 6×4 (dotted), and 6×6 (full). The bold line is an extrapolated curve. Bottom panel: static structure factor as a function of J'/J at $m=\frac{1}{2}$ on a 6×6 cluster. $S^{zz}(0,\pi)$ is shown by '+', $S^{zz}(\pi,\pi)$ by '$\times$', $S^{xx}(0,\pi)$ by '$\ast$', and $S^{xx}(\pi,\pi)$ by squares.

Quantum fluctuations have a strong effect on the stability of the $m=\frac{1}{2}$ plateau. Both, the width of the plateau for the spin-1/2 system and the range where it appears are larger than for the classical model. In addition, the parameter range for the plateau is shifted asymmetrically around the classical critical point $J'=0.5J$. The plateau is most pronounced for $J'\approx0.6J$. This is the value we used for the presented magnetization curve. The bold line in Fig. 4 was obtained by connecting the mid-points of the steps for the largest available systems size, except for the plateau at $m=\frac{1}{2}$, for which the corners of the 6×6 cluster data were used. The value of the spin gap (*i.e.* the boundary of the $m=0$ plateau) was taken from Kotov *et al.* [6]. Fig. 4 also shows the peaks of the static structure factor vs. J'/J for the $m=\frac{1}{2}$ plateau. The peaks in $S^{zz}(q)$, which indicate the presence of the uuud state, exist for $0.51\lesssim J'/J\lesssim0.67$.

The discussed mechanism for the magnetization plateaus in highly frustrated AFMs has to be contrasted with that of weakly coupled 1D spin systems (see, e.g., [8]). There, magnetization plateaus correspond to disordered states with a gap, that are stable under a weak higher-dimensional coupling, while intermediate gapless regions are immediately ordered at sufficiently low temperatures once a higher-dimensional coupling is switched on. Here, we found that an external field induces a long-range collinear ordering with an excitation gap on the plateau, whereas intermediate gapless regions remain disordered. Further exploration of such possibilities should also be important for the general problem of classifying magnetization plateaus in two dimensions [9].

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