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Field induced ordering in the frustrated square-lattice antiferromagnet

O. A. Petrenko,[1,2] A. Honecker,[3] and M. E. Zhitomirsky[4]
[1] *ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, OX11 0QX, United Kingdom*
[2] *University of Warwick, Department of Physics, Coventry, CV4 7AL, United Kingdom*
[3] *Institut für Theoretische Physik, TU Braunschweig, Mendelssohnstr. 3, D–38106 Braunschweig, Germany*
[4] *Theoretische Physik, ETH Hönggerberg, CH–8093 Zürich, Switzerland*
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We investigate, analytically and numerically, the behavior of the frustrated square-lattice antiferromagnet with quantum and classical spins in an external magnetic field. In the vicinity of the strongly frustrated point, $J' \approx 0.5J$, an applied magnetic field induces a transition from a gapless spin-liquid state with finite magnetization into a commensurate gapped state. The latter state has an up-up-up-down spin order and is characterized by the appearance of a magnetization plateau at a half of the saturated magnetization.

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Frustrated magnets, classical and quantum, exhibit spectacular and often unexpected behaviors at zero field [1]. In some cases they remain disordered down to $T = 0$. This phenomenon is attributed to a huge degeneracy in the classical ground state, which is only partially lifted by thermal or quantum fluctuations. Finite field properties of frustrated magnets are not well understood at present. Previous studies have mainly focused on the magnetization process of spin systems with long-range order in their zero field ground state, see, e.g., [1]. Here, we consider the field evolution of a strongly frustrated magnet, which has a disordered ground state at $H = 0$. One motivation for our study is the disordered frustrated three-dimensional antiferromagnet $Gd_3Ga_5O_{12}$ ($S = 7/2$, *i.e.* essentially classical spins), which only orders in finite magnetic fields [2]. The other motivation is the recently discovered 2D spin-gap material $SrCu_2(BO_3)_2$ (quantum spins, $S = 1/2$), which exhibits several plateaus on the magnetization curve [3]. The origin of these plateaus and the nature of the corresponding spin states are presently under intense debate [4]. It is argued that some of the plateaus in $SrCu_2(BO_3)_2$ might correspond to certain types of ordering. In this Letter we demonstrate that field induced ordering and related magnetization plateaus represent a general phenomenon in disordered frustrated magnets with *arbitrary* spin.

Specifically, we consider the competition between exchange interactions and an applied magnetic field for the frustrated square-lattice antiferromagnet (SAFM). This model has attracted a lot of attention during the last decade as an example of the melting of long-range magnetic order due to quantum fluctuations. The Hamiltonian of the model is

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

At zero magnetic field, and for small diagonal exchange $J' < 0.5J$, spins are ordered antiferromagnetically in the Néel state. At $J' > 0.5J$ the classical ground state consists of two $\sqrt{2} \times \sqrt{2}$ independent interpenetrating antiferromagnets. This degeneracy is lifted by quantum or thermal fluctuations which select a so called collinear state described by a single wave-vector $(\pi, 0)$ or $(0, \pi)$. The critical point $J' = 0.5J$ is special in that any spin configuration with total zero spin per elementary plaquette is a classical ground state [5,6]. In the vicinity of this point, linear spin-wave theory [5] and exact diagonalization studies [6] suggest a spin-liquid ground state for the spin-1/2 Hamiltonian (1). The nature of this singlet state is still subject to debate [7]. Here, we consider high-field ground states with finite magnetization, created when the triplet gap is closed by the external field.

We study the finite field properties of the frustrated SAFM with an arbitrary spin S by a combination of analytical and numerical techniques. First, we consider the stability of the two ordered phases in an external field in the linear spin-wave theory (LSWT). Then, we study the high-field magnetization process of the classical model at and around the critical point $J' = 0.5J$ by Monte Carlo (MC) simulations at finite temperatures. Finally, we consider the quantum spin-1/2 model by means of exact diagonalization. The last two methods provide strong evidence for field-induced long-range ordering and a magnetization plateau well below the saturation field H_{sat}.

Zero-point fluctuations in the two ordered states can be accounted for in LSWT, which has been applied before to the frustrated SAFM at zero external field [5]. We extend this analysis to finite fields by taking into account the sublattice canting and applying the Holstein-Primakoff transformation in a local coordinate frame for each sublattice. In finite fields both Néel and collinear states have a transverse spin order, which determines a choice of the criterion for a transition into a disordered state. At $H = 0$, the natural requirement is a vanishing of the sublattice magnetization: $\langle S \rangle = 0$ [5]. This criterion does not work at $H \neq 0$ because, at the point where the transverse components vanish and the long-range order disappears, there is still a nonzero component of spins along the field direction. Therefore, we choose for the transition point the condition $\langle S \rangle = M$, where M is a per site magnetization. Apart from this our calculations are standard and we present only the final diagram calculated for $S = 1/2$ in Fig. 1. Generally, at finite fields the intermediate disordered region on the phase diagram becomes narrower but never disappears completely. The only condition imposed on the spin configuration at $J' = 0.5J$ is that the magnetization of each elementary plaquette is $M_{\rm plaq} = H/(2J)$. Such degeneracy results in extra "accidental" zero-energy modes for the Néel and collinear states, which yield diverging first-order spin-wave corrections at $J' = 0.5J$ in the whole field range $0 < H < H_{\rm sat}$. The nature of the intermediate phase and, in particular, whether new types of order appear between the two ordered phases cannot be determined from these calculations.

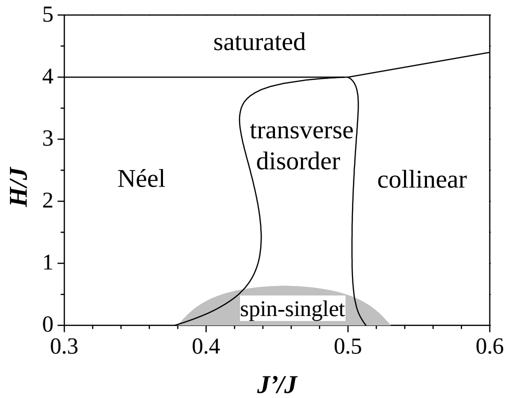

FIG. 1. The zero temperature phase diagram of the spin-1/2 frustrated SAFM in an external magnetic field, obtained from LSWT.

Apart from an infinite number of continuously varying classical states there are spin configurations, which can exist only at certain special values of the applied field. For example, at $H = 4JS$ (one half of the saturation field at $J' = 0.5J$) we can have an up-up-up-down (uuud) state on each four-spin plaquette. The magnetization of the 'uuud' state is $m = M/S = \frac{1}{2}$ and satisfies the above plaquette condition only at $H = 0.5H_{\rm sat}$. For all other fields this state is unstable. If mapped to a boson Hubbard model [8], the 'uuud' state corresponds to a charge-density wave or a solid phase, which becomes stable only at a rational filling (magnetization). The periodic solid of the 'uuud' state is a superposition of spin harmonics with wave-vectors $(0,0)$, (π,π), $(\pi,0)$ and $(0,\pi)$. There are four different classical spin-wave modes in the magnetic Brillouin zone with energies

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

where $\eta_{\mathbf{k}} = (1-\cos k_x)(1-\cos k_y)$. Three of these modes become negative at $H \neq 0.5H_{\rm sat}$, which signals an instability. However, because of its singular position in the manifold of the classical ground states we expect that thermal and quantum fluctuations can increase the stability region for such a commensurate solid: Fluctuations produce a finite gap in the excitation spectrum (2) which gives rise to a plateau on the magnetization curve in line with general considerations (see e.g. [9]). A similar effect is indeed seen for an ordered triangular lattice antiferromagnet [10], where a plateau appears at $m = \frac{1}{3}$. The peculiar feature of the $m = \frac{1}{2}$ plateau in our problem is that we have a spin-liquid phase above and below the plateau, whilst the plateau-state itself is ordered. In the language of a boson-Hubbard model this corresponds to a normal liquid-solid transition. To check the validity of such a scenario for the 'uuud' state at the $m = \frac{1}{2}$ plateau we now resort to numerical calculations.

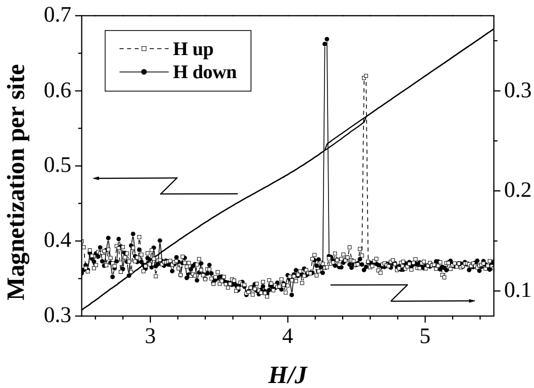

FIG. 2. The field dependence of the magnetization and susceptibility $\chi = dM/dH$ for a 24×24 lattice at $T = 0.1J$ with $J'/J = 0.5$. 2×10^5 MCS were performed for each point.

We start with the classical model assuming that the $\mathbf{S}_i$ in Eq. (1) are unit vectors. MC simulations have been performed for lattice sizes $N = L\times L$, with $L = 10$, 16, 24 and 36. Up to 4×10^6 MC steps per spin (MCS) were performed at the lowest temperatures, using a standard Metropolis algorithm. Fig. 2 presents the magnetization curve from MC calculations at $T = 0.1J$ and $J'/J = 0.5$. The magnetization depends linearly on the applied field and deviates from a straight line only in a limited field range around $H = 4J = 0.5H_{\rm sat}$. There is no perfectly horizontal plateau here because of thermal excitations across the gap, which is itself produced by thermal fluctuations. At low temperatures the plateau width becomes narrower vanishing at $T = 0$. A sudden jump of the magnetization at $H \approx 4.4J$ indicates a first-order transition to a high-field phase. This transition is accompanied by a significant hysteresis of about $\Delta H \simeq 0.3J$ at $T = 0.1J$.

To determine the nature of the spin state at the plateau we calculated different components of the static structure factor $S^{\alpha\beta}(q) = \frac{1}{N^2}\sum_{r,x} e^{i\mathbf{q}\cdot\mathbf{x}}\langle 0|S^\alpha_r S^\beta_{r+x}|0\rangle$. 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 must have nonzero longitudinal components $S^{zz}(q)$ at all the above vectors simultaneously. The static structure factor presented in Fig. 3 was obtained for a 24×24 lattice by averaging 50 'instant shots' separated by 10^3 MCS. The top panel shows that at $H = 4J$ nonvanishing harmonics in the longitudinal structure factor both at $\mathbf{q} = (\pi, 0)$ and $\mathbf{q} = (\pi, \pi)$ exist only for $0.494 < J'/J < 0.508$. 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. The lower panel of Fig. 3 presents the transverse structure factor. In the region of weak (strong) diagonal exchange J' the data clearly support the Néel (collinear) type of spin configuration, though real long-range ordering does not appear at finite T. At $H = 0.5H_{\rm sat}$ the corresponding peaks jump to zero at exactly the same values of J'/J, where the peaks in the longitudinal structure factor appear. These results unambiguously identify the spin configuration at the plateau as the 'uuud' state.

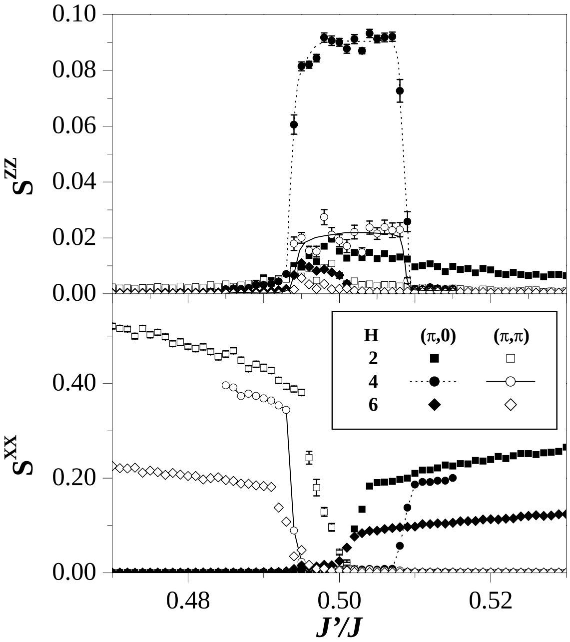

FIG. 3. Longitudinal (top panel) and transverse (bottom panel) components of the static structure factor, $S^{\alpha\beta}(q)$ versus frustration parameter J'/J for $T = 0.1J$ at different magnetic fields. Solid and open symbols correspond to the $S^{\alpha\beta}(q)$ at $\mathbf{q} = (\pi, 0)$ and $\mathbf{q} = (\pi, \pi)$, respectively. If not shown, the error bars do not exceed the size of the symbols.

Results of the calculations of the transverse components at values of the field above and below the plateau indicate that the stability regions of the Néel and collinear phases never reach the strongly frustrated point $J' = 0.5J$. This observation allows us to interpret the first-order transition seen in Fig. 2. The high-field spin-liquid phase possesses both $U(1)$ rotational symmetry in spin space and the translational symmetry of a square lattice. The 'uuud' solid breaks this translational invariance in such a way that wave-vectors belonging to different irreducible representations of the space group are mixed: (π, π) vs. $(\pi, 0)$ and $(0, \pi)$. Therefore, the spin structure of the 'uuud' phase is a mixture of different order parameters and the phase transition from a disordered symmetric state must go either in several steps or via a first order transition.

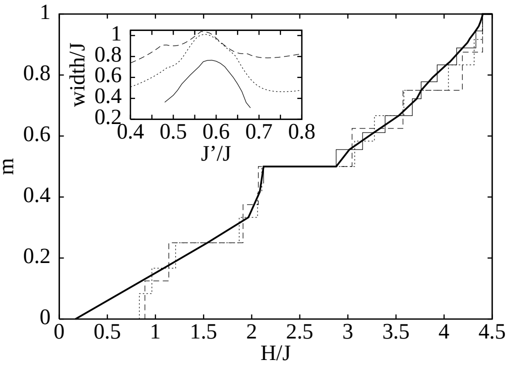

FIG. 4. Zero-temperature magnetization curve of the $S = 1/2$ model at $J'/J = 0.6$. Inset: Width of the finite-size plateau with $m = \frac{1}{2}$ as a function of J'/J. The lines are for lattices of size 4×4 (dashed), 6×4 (dotted), and 6×6 (full). The bold line is an extrapolated curve (see text for details).

Finally, we have studied the quantum spin-1/2 model (1) at $T = 0$ by Lanczos diagonalizations of finite clusters. We did the calculations in the whole range for the magnetization $0 \leq m \leq 1$, but only for the relatively small lattice sizes: 4×4 and 6×4. With the 6×6 cluster we investigated the region $m \geq \frac{4}{9}$, but larger systems were used in the vicinity of $m = 1$. Fig. 4 presents the magnetization curves for a few rectangular clusters at $J'/J = 0.6$. We do not see any 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 on the inset of Fig. 4 its width for three cluster sizes as a function of J'/J (note that the vertical scale of the inset does not include zero). If such 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 $0.495 \lesssim J'/J \lesssim 0.665$, where we would expect to have a plateau on an infinite lattice.

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 of J'/J, 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 $J' \approx 0.6J$, which we chose 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 $m = \frac{1}{2}$, where the corners of the 6×6 cluster data were used [11]. The value of the spin gap (*i.e.* the boundary of the $m = 0$ plateau) was taken from Kotov *et al.* [7].

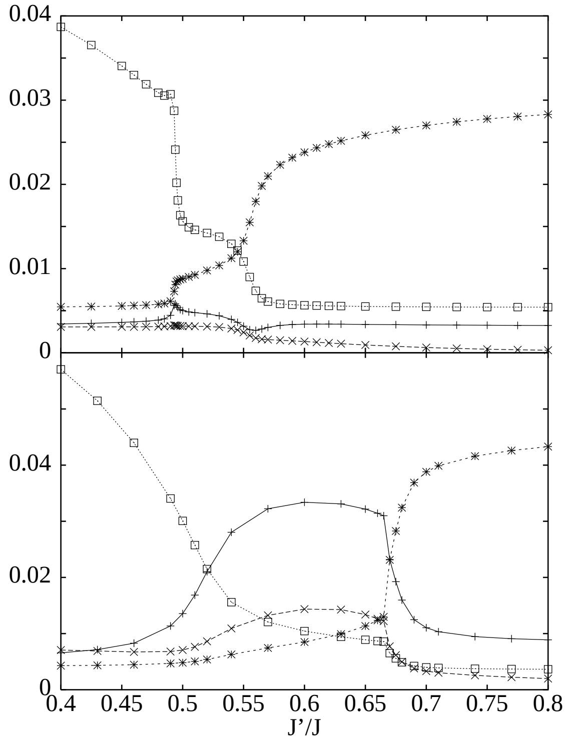

FIG. 5. Static structure factors of the $S = 1/2$ model as a function of J'/J at $m = \frac{7}{9}$ (top) and at $m = \frac{1}{2}$ (bottom) on a 6×6 cluster. The longitudinal structure factor $S^{zz}(0, \pi)$ is shown by '+', $S^{zz}(\pi, \pi)$ by '×' and the transverse structure factor $S^{xx}(0, \pi)$ by '∗' and $S^{xx}(\pi, \pi)$ by open squares.

Fig. 5 shows the J'-dependence of the peaks of the static structure factor for two selected values of m. At $m = \frac{1}{2}$ (lower panel), we have the transverse Néel order for $J'/J \lesssim 0.51$. For $0.51 \lesssim J'/J \lesssim 0.67$, the order parameter of the 'uuud' state develops supporting its appearence on the $m = \frac{1}{2}$ plateau. At $J'/J \approx 0.67$ there is a transition between the 'uuud' plateau state and the collinear state at larger J'. At $m = \frac{7}{9}$ (upper panel) we can identify the Néel order in the transverse components for $J'/J \lesssim 0.5$ and the collinear order for $J'/J \gtrsim 0.56$. This implies in particular that the high-field region of Fig. 4 coincides with the collinear phase, whereas for $0.51 \lesssim J'/J \lesssim 0.56$ the 'uuud' state is replaced by a disordered state at higher fields. Although the small system sizes do not always allow a definite conclusion, all transitions as a function of J'/J could be of first-order, as is also suggested by the previous symmetry arguments.

In conclusion, we have shown that an external magnetic field can induce a Mott-type transition from a gapless spin-liquid state into a commensurate gapped state in strongly frustrated AFMs. An important issue is the order of the transition, which, as we argued, depends on the symmetry of a commensurate solid. In most cases of solid-liquid or solid-superfluid transitions they must be of the first order. The only possibility of a continuous transition is a solid-supersolid transition [8], when the spin state above or below a plateau corresponds to a spin helix in the field plane.

The present ordering scenario is to be contrasted to that of weakly coupled one-dimensional systems (see [12] and references therein): There, magnetization plateaus correspond to disordered states with a gap which are stable under a weak higher-dimensional coupling while the gapless transition regions are immediately ordered at sufficiently low temperatures once a higher-dimensional coupling is switched on. Here we have presented an example of an intrinsically two-dimensional system with an ordered plateau state and disordered transition regimes. The exploration of such possibilities should also be important for the general problem of classifying magnetization plateaus in two dimensions [9,13] as well as explaining those observed in $SrCu_2(BO_3)_2$ [3] in particular.

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