Supersolid Phases in a Realistic Three-Dimensional Spin Model

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Supersolid phases, in which a superfluid component coexists with conventional crystalline long range order, have recently attracted a great deal of attention in the context of both solid helium and quantum spin systems. Motivated by recent experiments on $2H$-$AgNiO_2$, we study the magnetic phase diagram of a realistic three-dimensional spin model with single-ion anisotropy and competing interactions on a layered triangular lattice, using classical Monte Carlo simulation techniques, complemented by spin-wave calculations. For parameters relevant to experiment, we find a cascade of different phases as a function of magnetic field, including three phases which are supersolids in the sense of Liu and Fisher. One of these phases is continuously connected with the collinear ground state of $AgNiO_2$, and is accessible at relatively low values of magnetic field. The nature of this transition, and its possible observation, are discussed.

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Solids and liquids are very different. Placed under stress, a liquid will flow, while a solid resists deformation. The idea of a supersolid, a state which combines the properties of a solid with those of a perfect, nondissipative superfluid, seems therefore to fly in the face of common sense. None the less, the proposal that a supersolid might occur through the Bose-Einstein condensation of vacancies in a quantum crystal [1], was propelled to the center of debate by recent experiments on ^{4}He [2].

A radically different approach to supersolids was initiated by Liu and Fisher [3], who realized that quantum magnets could support states which break the translational symmetry of the lattice (and are therefore solids) while *simultaneously* breaking spin-rotational symmetry within a plane, a form of order analogous to a superfluid. It is now well established that models of two-dimensional frustrated magnets with anisotropy can support such supersolid states [4]. Moreover, since a spin-$1/2$ quantum magnet is in one-to-one correspondence with hard-core bosons, these supersolids might also be realized using cold atoms on optical lattices. Nonetheless, candidates for supersolid states among real, three-dimensional magnets remain scarce. An interesting system in this context is the triangular easy-axis magnet, $2H$-$AgNiO_2$ [5].

$AgNiO_2$ is a very unusual material, built of stacked, two-dimensional nickel-oxygen planes, held together by silver ions. It combines metallicity and magnetism, with the magnetic ions in each plane forming a perfect triangular lattice, nested within a honeycomb network of conducting sites [5]. In the absence of magnetic field $AgNiO_2$ supports a stripelike collinear antiferromagnetic ground state, illustrated in Fig. 1(a). Recently, $AgNiO_2$ has been shown to undergo a complicated set of phase transitions as a function of magnetic field [6]. Of particular interest is the transition out of the collinear ground state at low temperatures.

In applied magnetic field, collinear antiferromagnets with easy-axis anisotropy typically undergo a first-order ‘‘spin-flop’’ transition into a canted state, at a critical field which is broadly independent of temperature. However, the low-field transition in $AgNiO_2$ is accompanied by a relatively broad feature in specific heat, does not exhibit marked hysteresis, and occurs at progressively higher fields as temperature increases. None of these features resemble a typical spin-flop transition, and together they raise the question of whether a novel type of magnetic order is realized in $AgNiO_2$ under field.

In this Letter we explore the different phases that occur as a function of magnetic field in a simple effective spin model already shown to provide excellent fits to inelastic neutron scattering spectra for $AgNiO_2$ [7]. We show that the collinear ground state of this model does not undergo a conventional spin flop, but rather a Bose-Einstein condensation of magnetic excitations which converts it into a state that is a supersolid in the sense of Liu and Fisher. We also identify two magnetization plateaux, and two further supersolid phases at high field.

The model we consider is the Heisenberg model on a layered triangular lattice, with competing antiferromagnetic first- and second-neighbor interactions J_1 and J_2,

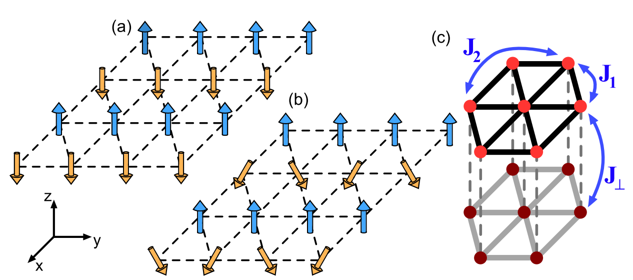

FIG. 1 (color online). (a) Low-field collinear stripe phase with spins aligned along the magnetic easy axis (z axis). (b) Related supersolid phase for magnetic field parallel to the easy axis: down spins cant into the plane perpendicular to field, while up spins remain aligned with the field. (c) First neighbor J_1, second neighbor J_2, and interlayer interactions $J_\perp$ for a stacked triangular lattice.

single-ion anisotropy D and interlayer coupling $J_\perp$

$$\mathcal{H} = J_1 \sum_{\langle ij\rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ij\rangle_2} \mathbf{S}_i \cdot \mathbf{S}_j + J_\perp \sum_{\langle ij\rangle_\perp} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i (\mathbf{S}_i^z)^2 - h \sum_i \mathbf{S}_i^z. \quad (1)$$

[cf. Fig. 1(c)]. For concreteness, we set $J_1 = 1$, $J_2 = 0.15$, $J_\perp = -0.15$ and $D = 0.5$, measuring magnetic field h and temperature T in units of J_1. These are ratios of parameters comparable to those used to fit inelastic neutron scattering spectra for $AgNiO_2$ [7]. Like $AgNiO_2$, in the absence of magnetic field, this model exhibits a collinear stripelike magnetic ground state, illustrated in Fig. 1(a). The stripes have three possible orientations, and so break a $\mathbb{Z}_3$ rotational symmetry of the lattice. The collinear stripe state also breaks translational symmetry in the direction perpendicular to the stripes. But in the presence of a magnetic easy axis, it *does not* break spin-rotation symmetry.

This collinear stripe state supports two branches of spin-wave excitations, which are gapped, and degenerate in the absence of magnetic field. However, for parameters relevant to $AgNiO_2$, the dispersion minimum does *not* occur at the magnetic ordering vector M, as would be expected, but rather at points M' related by the broken $\mathbb{Z}_3$ rotational symmetry [7].

Applying a magnetic field parallel to the easy axis lifts the degeneracy of the two spin-wave branches, and reduces the gap at M'. Within linear spin-wave theory, neglecting dispersion in the out-of-plane direction and expanding about a stripe state with ordering vector $M = (0, 2\pi/\sqrt{3})$, we find

$$\epsilon_\pm(\mathbf{k}) = 4S[\{J_1 \cos^2(k_x/2) + J_2 \cos^2(\sqrt{3}k_y/2) + D/2\}^2 - \{\cos(\sqrt{3}k_y/2) \times [J_1 \cos(k_x/2) + J_2 \cos(3k_x/2)]\}^2]^{1/2} \pm h,$$

and the spin gap at $M' = (\pm\pi, \pi/\sqrt{3})$ closes completely at a critical field $h = 2DS$. The resulting dispersion is shown in Fig. 2(a).

As in the celebrated example of $TlCuCl_3$, the closing of this spin gap leads to Bose-Einstein condensation of spin-wave excitations (magnons) [8]. This Bose-Einstein condensate breaks a $U(1)$ spin-rotation symmetry in the S^x-S^y plane, and so has superfluid character. Since the resulting state inherits the broken $\mathbb{Z}_2$ translational and $\mathbb{Z}_3$ rotational symmetries of the collinear ground state, it is a supersolid. This quantum phase transition can also be understood at a mean-field level—instead of undergoing a spin-flop, the down spins cant, while the up spins remain aligned with the field. The nature of the new magnetic supersolid is illustrated in Fig. 1(b).

These arguments establish the possibility of a supersolid state in $AgNiO_2$, but tell us nothing about its thermodynamic properties. If the low-field transition in $AgNiO_2$ is

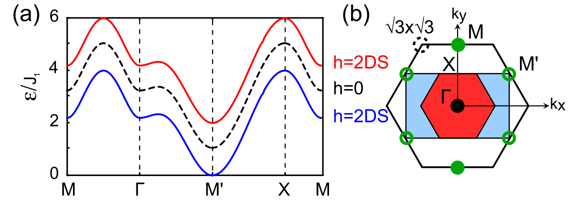

FIG. 2 (color online). (a) Linear spin-wave dispersion of the collinear stripe phase of Eq. (1) for $h = 0$ (dashed line) and $h = 2DS$ (solid lines) in the $k_z = 0$ plane, for $S = 1$, $J_1 = 1$, $J_2 = 0.15$, $J_\perp = -0.15$ and $D = 0.5$. (b) First Brillouin zone for a triangular lattice, showing the ordering vector M, and related symmetry points M'. The magnetic Brillouin zones for the collinear stripe phase and associated supersolid are shown by a blue rectangle and a red hexagon, respectively.

into a supersolid, why does the critical field increase with increasing temperature? What might the experimental signatures of this new phase be? What other states might occur at higher magnetic field, and how do they evolve with temperature?

In order to address these questions, we have performed classical Monte Carlo simulations of Eq. (1). We employed a parallel tempering Monte Carlo scheme [9], combined with successive over-relaxation sweeps [10]. Simulations of 48–128 replicas were performed for rhombohedral clusters with periodic boundary conditions, of $3L \times 3L \times L = 9L^3$ spins, where $L = 4, 6, 8, 10$ counts the number of triangular lattice planes. Typical simulations involved 4×10^6 steps, half of which were discarded for thermalization. Each step combines one local-update sweep of the lattice and two over-relaxation sweeps, with replicas at different temperatures exchanged every 10 steps. We set $|\mathbf{S}| = S = 1$ throughout.

The results of these simulations are summarized in Fig. 3. For the parameters used, we find a total of six distinct ordered phases as a function of increasing field: (i) a collinear stripe ground state with a 2-site unit cell; (ii) a supersolid phase with a 4-site unit cell; (iii) a collinear one-third magnetization plateau state with a 3-site unit cell; (iv) a second supersolid, formed by a 2:1:1 canting of spins, with the same unit cell as (ii); (v) a collinear half-magnetization plateau state, with the same unit cell as (ii); and (vi) a third supersolid, formed by a 3:1 canting of spins approaching saturation, with the same unit cell as (ii). Phase transitions were identified using peaks in the relevant order-parameter susceptibilities. These transitions are generically first order, except between collinear and supersolid phases with the same unit cell. All of these phases can also be found in mean-field theory at $T = 0$, and transitions between them are shown by open symbols on the h axis of Fig. 3.

This phase diagram shows some intriguing similarities with experimental work on $AgNiO_2$ [6]. In particular, the topology of the low-field phases is correctly reproduced, with the low-field supersolid phase contained entirely within the envelope of the collinear stripe phase. The phase

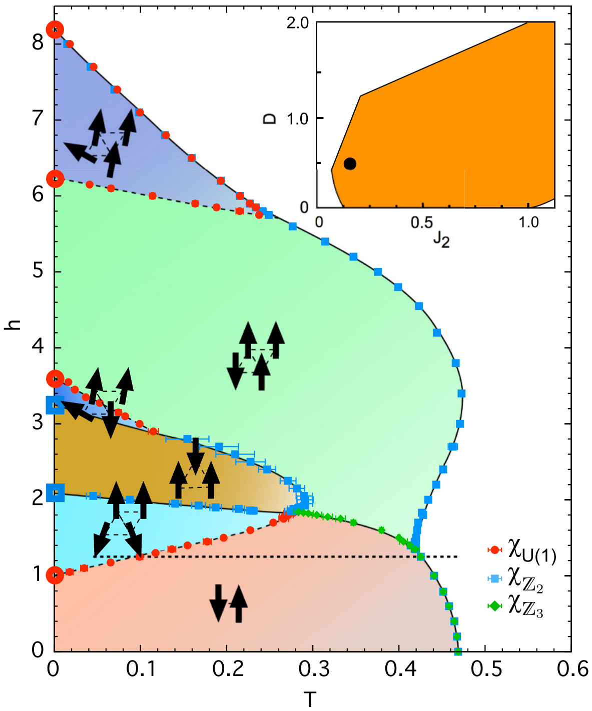

FIG. 3 (color online). Magnetic phase diagram obtained from classical Monte Carlo simulation of Eq. (1) for a cluster of $24 \times 24 \times 8$ spins with $J_1 = 1$, $J_2 = 0.15$, $J_\perp = -0.15$, $D = 0.5$. Temperature T and magnetic field h are measured in units of J_1. Phase boundaries are determined from peaks in order-parameter susceptibilities. Phase transitions are first order, except where shown with a dashed line. A dotted black line shows the cut at $h = 1.25$ used in Figs. 4(a), 4(b), and 4(d) and Fig. 5. Inset shows the range of parameters for which a supersolid arises as the first instability of the stripe phase in magnetic field, as determined by mean-field calculations for $J_\perp = -0.15$.

transition between these two phases is continuous, and the critical field increases with increasing temperature [11]. For this reason we now concentrate on the low-field properties of the model, leaving the rich physics at higher field for discussion elsewhere. We note, however, that the one-third magnetization plateau (iii) is well known from studies of easy-axis triangular lattice antiferromagnets [12], and that states analogous to the half-magnetization plateau (v) and high-field supersolids (iv) and (vi) also occur in models of Cr spinels [13].

As in some previously studied models [4,14], two finite-temperature phase transitions separate the low-field supersolid phase from the paramagnet. The first of these is a first-order transition into the collinear stripe state at a temperature $T \approx 0.42$. The second is a continuous transition at a critical temperature which varies approximately linearly with magnetic field from $T = 0$ ($h = 1$) to $T \approx 0.3$ ($h \approx 2$). Both translational and rotational lattice symmetries are broken at the upper transition. To study this it is convenient to introduce a two-component order parameter based on an irreducible representation of the $C_3 \cong \mathbb{Z}_3$ rotation group, which measures the orientation of the "stripes" in the plane

$$\psi_{2s} = \frac{1}{\sqrt{6}N} \sum_i 2S_i^z S_{i+\delta_1}^z - S_i^z S_{i+\delta_2}^z - S_i^z S_{i-\delta_1-\delta_2}^z,$$

$$\psi_{2a} = -\frac{i}{\sqrt{2}N} \sum_i S_i^z S_{i+\delta_2}^z - S_i^z S_{i-\delta_1-\delta_2}^z,$$

where $\delta_1 = (1, 0)$ and $\delta_2 = (1/2, \sqrt{3}/2)$ are the primitive vectors of the triangular lattice. Figures 4(c) and 4(d) show the behavior of this order parameter, Binder cumulants for energy, and related susceptibility for $h = 1.25$, $T \approx 0.42$. The transition is clearly first order; we have checked explicitly that the $\mathbb{Z}_2$ symmetry associated with translations is broken at the same temperature.

Spin-rotation symmetry in the S^x-S^y plane is broken at the lower phase transition into the supersolid state. This can be measured by constructing a $U(1)$ order parameter which measures the difference between S^x and S^y components of the canted down spins, as illustrated in Fig. 4(f). Figures 4(a) and 4(b) show the behavior of this order parameter, its Binder cumulants, and related susceptibility for $h = 1.25$, $T \approx 0.1$. The phase transition remains con-

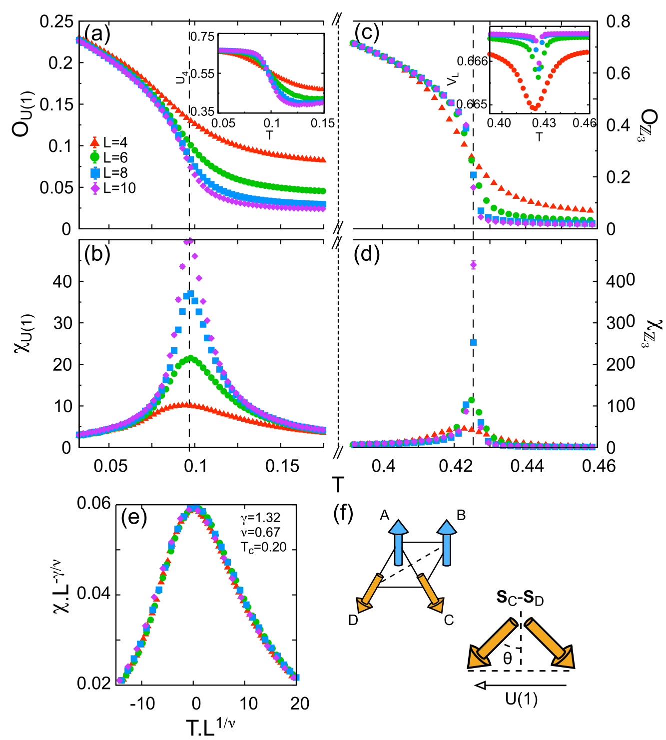

FIG. 4 (color online). (a) $U(1)$ order parameter showing onset of supersolid phase for $h = 1.25$, $T \approx 0.1$ (inset: crossing of associated Binder cumulants). (b) Related order-parameter susceptibility $\chi_{U(1)}$. (c) $\mathbb{Z}_3$ order parameter showing onset of collinear stripe phase for $h = 1.25$, $T \approx 0.42$ (inset: Binder cumulants for energy, showing a dip indicative of a bimodal distribution). (d) Related order-parameter susceptibility $\chi_{\mathbb{Z}_3}$. Results from simulations of clusters of $3L \times 3L \times L$ spins, with $L = 4, 6, 8, 10$, for parameters identical to Fig. 3. (e) Finite-size scaling of order-parameter susceptibility at transition into supersolid for $h = 1.5$, $T \approx 0.2$. (f) Graphical representation of $U(1)$ order parameter as a vector in the S^x-S^y plane.

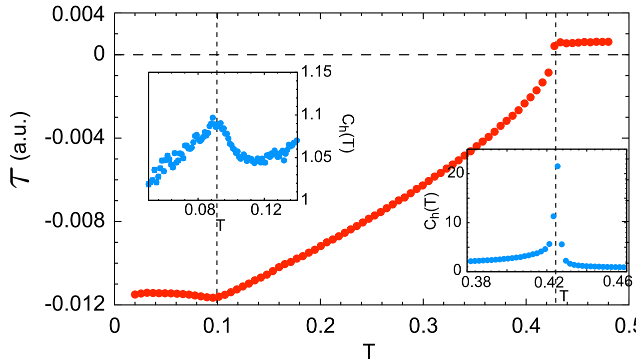

FIG. 5 (color online). Magnetic torque $\tau = \mathbf{m} \times \mathbf{h}$ as a function of temperature, for a magnetic field $h = 1.25$ at an angle of 5° to the easy axis (natural units). Torque changes sign abruptly at the first-order transition from paramagnet to collinear stripe phase for $T \approx 0.42$. A change in slope for $T \approx 0.1$ signals the continuous transition from stripe phase to magnetic supersolid. Insets show the heat capacity anomalies at each of these transitions.

tinuous at finite temperature, with Binder cumulants for different system size crossing at a single temperature. A good collapse of susceptibility data is obtained using susceptibility and correlation length exponents $\gamma = 1.32$ and $\nu = 0.67$ for the 3D XY universality class, as shown in Fig. 4(e).

While the relative extent of each phase and details of critical fields and temperatures are different, $AgNiO_2$ exhibits a similar double transition on cooling: a transition from paramagnet to a collinear stripe phase at $T \approx 20$ K accompanied by a sharp feature in specific heat and then, for fields greater than 13.5 T, a continuous or very weakly first-order transition from the collinear stripe phase into an unknown low temperature magnetic state. The transition field into this phase increases with temperature, suggesting that the stripe phase has higher entropy than the competing high-field phase, as found in our simulations. Is the high-field phase in $AgNiO_2$ then a supersolid?

Direct confirmation of the magnetic order at high field by elastic neutron scattering is challenging, since no large single crystals are presently available. None the less, it should be possible to observe the closing of the spin gap on entry to the supersolid phase [7]. Moreover, both transport and thermodynamic measurements clearly resolve magnetic phase transitions [6]. In Fig. 5 we present predictions for magnetic torque, $\tau = \mathbf{m} \times \mathbf{h}$, and heat capacity C_h, spanning supersolid, stripe and paramagnetic phases for $h = 1.25$. For the ratios of parameters used, with $J_1 = 1.32$ meV (cf. [7]), this translates into a field of 12.5 T, with the supersolid transition occurring at 1.5 K. The heat capacity anomalies at both transitions strongly resemble those observed in $AgNiO_2$ [6], and the smooth evolution of torque entering the supersolid should be contrasted with earlier work on a spin-flop transition [15].

In summary, we have studied the magnetic phase diagram of a realistic three-dimensional spin model with single-ion anisotropy and competing interactions on a layered triangular lattice, identifying three phases which are magnetic supersolids in the sense of Liu and Fisher [3]. We find that these supersolids are continuously connected with parent collinear phases through the Bose-Einstein condensation of magnons. The model studied was motivated by the metallic triangular lattice antiferromagnet $2H$-$AgNiO_2$, and is known to describe its magnetic excitations in zero field [7]. Since the model does not take itinerant charge carriers into account, it cannot pretend to be a complete theory of $AgNiO_2$. Nonetheless, it motivates a reexamination of the low-field transitions seen in $AgNiO_2$, where a magnetic supersolid may already have been observed [6].

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