Half-magnetization plateau in a Heisenberg antiferromagnet on a triangular lattice

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Half-magnetization plateau in a Heisenberg antiferromagnet on a triangular lattice

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We present the phase diagram of a 2D isotropic triangular Heisenberg antiferromagnet in a magnetic field. We consider spin-S model with nearest-neighbor (J_1) and next-nearest-neighbor (J_2) interactions. We focus on the range of $1/8 < J_2/J_1 < 1$, where the ordered states are different from those in the model with only nearest neighbor exchange. A classical ground state in this range is infinitely degenerate in any field. The actual order is then determined by quantum fluctuations via "order from disorder" phenomenon. We argue that the phase diagram is rich due to competition between competing quantum states which break either orientational or sublattice symmetry. At small and high fields, the ground state is a canted stripe state, which breaks orientational symmetry, but at intermediate fields the ordered states break sublattice symmetry. The most noticeable of such states is "three up, one down" state in which spins in three sublattices are directed along the field and in one sublattice opposite to the field. In such a state magnetization is quantized at exactly one half of the saturation value. We identify gapless states, which border the "three up, one down" state and discuss the transitions between these states and the canted stripe state.

Introduction Recent experimental and theoretical advances renewed the interest in frustrated spin systems. In many of these systems the classical ground state is infinitely degenerate, and the actual ground state spin configuration is selected by quantum fluctuations (the "order from disorder" phenomenon). The resulting ground state is often rather unconventional and in several cases displays a non-monotonic behavior of magnetization in an applied field, with kinks, jumps, and plateaus [1–7]. The most known example of such behavior is in the case of a two-dimensional (2D) quantum antiferromagnet on a triangular lattice with nearest-neighbor exchange J_1 [8, 9]. Classically, all spin configurations, which satisfy $\boldsymbol{S}_r + \boldsymbol{S}_{r+\boldsymbol{\delta}_1} + \boldsymbol{S}_{r+\boldsymbol{\delta}_2} = \boldsymbol{h}S/(3J_1)$ for each triad of neighboring spins, have the same ground state energy. Quantum fluctuations lift the degeneracy and select a set of three coplanar configurations, between which the system transforms upon increasing field. The middle configuration, which exists at h around 1/3 of the saturation field $h_{sat} = 9J_1$, is a collinear state with two spins up (U) and one spin down (D) in every elementary triangle (an UUD state). In such a state only a discrete $\mathbb{Z}_3$ symmetry is broken (one spin in a triad is selected to be antiparallel to a field). As a result, all excitations are gapped and the magnetization has a plateau at exactly one-third of the saturation value [8–14]. This plateau has been observed in Cs_2CuBr_4 [15–19] and in $Ba_3CoSb_2O_9$ [20]. An UUD state survives in a finite range of perturbations, like the spatial anisotropy of the exchange interaction [10–12, 21–24], multiple-spin ring exchange [25], and the next nearest neighbor exchange [26]. What replaces the UUD state at larger perturbations has been the subject of intensive research over the last several years [10–14, 22–24, 27–32].

In this communication we study spin-S Heisenberg antiferronmagnet on a triangular lattice with nearest (J_1) and second-nearest (J_2) exchange interaction. Previous studies have found that the UUD phase and other two

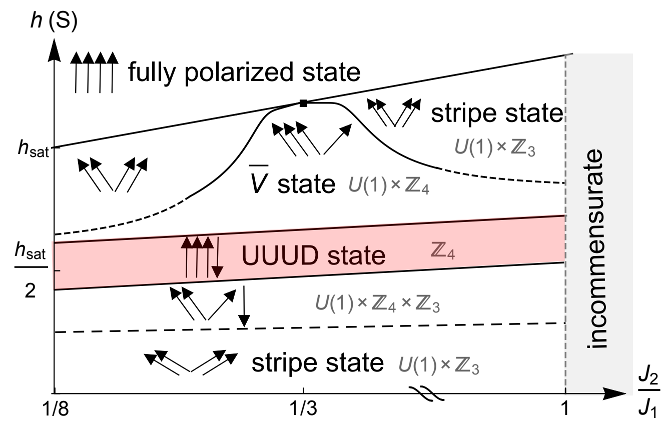

FIG. 1. Schematic semiclassical phase diagram of a spin-S $J_1 - J_2$ antiferromagnet on a triangular lattice at $1/8 < J_2/J_1 < 1$. Solid (dotted) lines are second-order (first-order) phase transitions, which we identified and analyzed in this work. Dashed line is a first-order transition, which we expect to hold, but didn't analyze. Arrows indicate magnetic order in the four-sublattice representation, and symbols like $U(1) \times Z_3$ indicate the broken symmetry in each state. The physics in a narrow range (at order 1/S) of J_2/J_1 near $J_2/J_1 = 1/8$ and $J_2/J_1 = 1$ is not analyzed in this work.

three-sublattice coplanar ground states in a field are immune to J_2 up to $J_2/J_1 < 1/8$. At larger J_2, however, the set of classical ground states changes discontinuously from three-sublattice configurations to four sublattice ones, in which four spins on two neighboring triads satisfy $\boldsymbol{S}_r + \boldsymbol{S}_{r+\boldsymbol{\delta}_1} + \boldsymbol{S}_{r+\boldsymbol{\delta}_2} + \boldsymbol{S}_{r+\boldsymbol{\delta}_3} = \boldsymbol{h}S/(2(J_1 + J_2))$ (see Fig. 3(a)). This condition does not uniquely specify spin order, even at zero field. The selection of the order by quantum fluctuations at $h = 0$ has been analyzed by various means [33–36], and the consensus is that for $1/8 < J_2/J_1 < 1$ the winner is the stripe order with ferromagnetic alignment of spins along one of three prin-

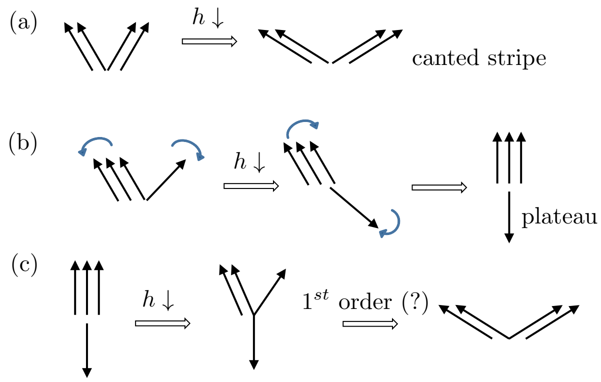

FIG. 2. (a), (b) – Two candidate quantum four-sublattice ground states upon decreasing of the magnetic field h towards a half of saturation value. (a) A Z_3 breaking canted stripe state. As field goes down, the angle between two pairs of parallel spins increases. (b) $\bar{\text{V}}$ and UUUD states. Both break Z_4 sublattice symmetry by selecting one sublattice with a different spin orientation compared to the other three. (c) Evolution from the UUUD state to the canted stripe state as h decreases below $h_{sat}/2$.

ciple axes on a triangular lattice and antiferromagnetic along the other two. The same order (the canted stripe state, see Fig. 2(a)) is selected by quantum fluctuations near the saturation field, and semiclassical (large S) spin-wave analysis shows [26] that this state remains stable at all fields. It would seem natural to conjecture that this state, with monotonic magnetization $M(h)$, is the true quantum ground state for $1/8 < J_2/J_1 < 1$ in all fields.

We argue that the phase diagram of $J_1 - J_2$ model in a field is actually rather complex, with multiple phases (see Fig. 1), and the stripe order is the ground state configuration only in some range of fields and of J_2/J_1. For other values of h and J_2/J_1 the ground state configurations are the co-planar states, similar to those at small J_2. In particular, around $h = h_{sat}/2$, the ground state is the UUUD state, in which spins in three sublattices are aligned along the field and in the forth sublattice opposite to the field. This spin order breaks $\mathbb{Z}_4$ sublattice symmetry, but doesn't break any continuous symmetry. As a result, spin-wave excitations are gapped, and the magnetization has a plateau at exactly 1/2 of the saturation value. We argue that the UUUD state exists for all J_2 in the interval $1/8 < J_2/J_1 < 1$. We also analyze the proximate states to the UUUD state. Above the upper critical field h_u, the UUUD state becomes unstable towards a state in which three up-spins rotate in one direction from the direction of $\boldsymbol{h}$, and the down-spin rotates in the opposite direction (see Fig. 2(b)). Below the lower critical field h_l, we found, at large S, a particular coplanar state, in which down-spin does not move, while three up-spins again rotate, but now one of these three spins splits from the other two (see Fig. 2(c)). A non-coplanar, chiral umbrella state [25, 37] is close in energy and may be the ground state near h_l at smaller S (see Fig. 4).

A cascade of field-induced magnetic transitions at fields below $h_{sat}/2$ has been observed in $2H-\text{AgNiO}_2$ [38, 39]. It has been argued [40] that in this material Ni^{2+} ions are localized and form a $S = 1$ triangular lattice antiferromagnet with $J_2 = 0.15 J_1$, single-ion easy axis anisotropy D, weak ferromagnetic exchange between layers. And Classical Monte-Carlo calculations for this model have found [41] the region of UUUD phase, whose width at $T = 0$ scales with D. We show that in a quantum model the UUUD phase is stable in a finite range of h already at $D = 0$. We expect that future measurements of the magnetization in $2H-\text{AgNiO}_2$ at higher fields will be able to detect the UUUD phase and also the cascade of phases above $h_{sat}/2$. The analysis of the high-field phases will allow one to distinguish whether UUUD order is stabilized predominantly by quantum fluctuations or by single-ion anisotropy [42]

Model and the high field phase diagram The $J_1 - J_2$ Heisenberg antiferromagnet on a triangular lattice is described by

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - S\boldsymbol{h} \cdot \sum_i \boldsymbol{S}_i \quad (1)$$

where $\langle i,j \rangle$ and $\langle\langle i,j \rangle\rangle$ run over all the nearest and next nearest neighbor bonds. Due to the global spin-rotational symmetry, the direction of $\boldsymbol{h}$ does not matter. We choose $\boldsymbol{h} = h\,\hat{e}_z$, and consider the range of $1/8 < J_2/J_1 < 1$.

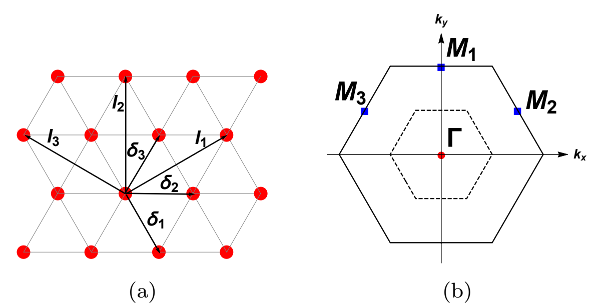

FIG. 3. (a) The nearest-neighbor ($\boldsymbol{\delta}_i$) and next-nearest-neighbor ($\boldsymbol{l}_i$) bonds on a triangular lattice. (b) Solid (dashed) line: Single-sublattice (four-sublattice) Brillouin zone. The points labeled as M_i are relevant to our discussion of spin-wave excitations near h_{sat}. The spin-wave excitations of the four-sublattice UUUD state soften at Γ point.

The first indication that the stripe phase is not the only ground state in a field comes from the Ginzburg-Landau analysis of the order immediately below the saturation field at $J_2 \approx J_1/3$. Spin-wave excitations soften at $h = h_{sat}$ at three points in the Brillouin zone - M_1, M_2, M_3, see Fig. 3(b). To understand the order below h_{sat} one needs to introduce three condensates Φ_1, Φ_2, Φ_3. The ground state energy in terms of Φ is:

$$E_\Phi/N = -\mu \sum_{i=1,2,3} |\Phi_i|^2 + \frac{1}{2}\Gamma_1 \sum_{i=1,2,3} |\Phi_i|^4 + \Gamma_2(|\Phi_1|^2|\Phi_2|^2 + |\Phi_1|^2|\Phi_3|^2 + |\Phi_2|^2|\Phi_3|^2) + \Gamma_3(\Phi_1^2\Phi_2^2 + \Phi_2^2\Phi_3^2 + \Phi_3^2\Phi_1^2 + h.c.) \quad (2)$$

where $\mu \sim S(h_{\rm sat} - h)$. The type of spin order that minimizes E_Φ depends on the interplay between the quartic coefficients Γ_i. In the classical limit, $\Gamma_1 = \Gamma_2 = 8(J_1 + J_2)$, $\Gamma_3 = 0$, i.e., any state from the manifold $|\Phi|_1^2 + |\Phi|_2^2 + |\Phi|_3^2 \equiv \mu/\Gamma_1$ is the ground state. Quantum fluctuations lift the degeneracy. To leading order in $1/S$ we found [26], near $J_2 = J_1/3$,

$$\Gamma_2 - \Gamma_1 = \frac{24\sqrt{3}J_1}{\pi} \left(\frac{J_2}{J_1} - 1/3\right)^2 \frac{|\log(h_{\rm sat} - h)|}{S} - \beta_1/S$$
$$\Gamma_3 = -\beta_2/S \quad (3)$$

where $\beta_{1,2} > 0$ are numbers of order one. The logarithm $|\log(h_{\rm sat} - h)|$ is present because of quadratic dispersion near M-points. Because of the logarithm, $\Gamma_2 > \Gamma_1$, A straightforward analysis then shows that only one Φ_i is then non-zero because it costs extra energy to develop simultaneously condensates from different valleys. One can check [26] that the resulting order is the stripe state. However, the prefactor for the logarithm in $\Gamma_2 - \Gamma_1$ in Eq. 3 vanishes at $J_2 = J_1/3$, when $\omega_{\boldsymbol{k}}$ becomes isotropic ($\alpha = 0$). For this J_2/J_1, the sign of $\Gamma_2 - \Gamma_1$ is determined by regular $1/S$ terms, along with the sign of Γ_3. We computed these terms and found $\Gamma_2 - \Gamma_1 < 0$, $\Gamma_3 < 0$. As a result, at $J_2/J_1 = 1/3$, all three condensates emerge with equal amplitudes and relative phases 0 or π (because $\Gamma_3 < 0$). The four choices for (Φ_1, Φ_2, Φ_3) are (Φ, Φ, Φ), $(\Phi, -\Phi, -\Phi)$, $(-\Phi, \Phi, -\Phi)$, $(-\Phi, -\Phi, \Phi)$. In each of these states spins in three sublattices tilt to one direction from the field, and in one sublattice tilt to the opposite (see Fig. 1). We label such a state $\bar{\rm V}$ by analogy with the corresponding V state [43] at $J_2 < J_1/8$ [9–12]. The $\bar{\rm V}$ state breaks $U(1)$ spin-rotational symmetry in the plane perpendicular to the field, and also breaks a $\mathbb{Z}_4$ sublattice symmetry by selecting a sublattice in which spin direction is different from that in other three sublattices.

Immediately below h_{sat}, the $\bar{\rm V}$ state is stable in the infinitesimally small range around $J_2 = J_1/3$, at $(J_2/J_1 - 1/3)^2 < 1/|\log(h_{sat} - h)|$. As h decreases, the width grows and becomes $\mathcal{O}(1)$ at $h_{\rm sat} - h = \mathcal{O}(1)$. The $\bar{\rm V}$ and the stripe state break different discrete symmetries ($\mathbb{Z}_4$ and $\mathbb{Z}_3$, respectively), hence the transition between the two states is likely first order. The increase of the width of the $\bar{\rm V}$ state with decreasing field can be understood as a generic consequence of the fact that this state is favored by regular $1/S$ terms, i.e., by quantum fluctuations at short length scales, while the stripe phase is favored by $|\log(h_{\rm sat} - h)|$, which comes from long-wavelength fluctuations. As the magnitude of the transverse order increases with decreasing field, long wavelength fluctuations are suppressed, and $\bar{\rm V}$ state becomes more favorable.

Half-magnetization plateau As the field decreases towards $h_{sat}/2$, the $\bar{\rm V}$ state evolves: the spin in one sublattice continuously rotates away from the field direction towards the direction antiparallel to $\boldsymbol{h}$. The spins in three other sublattices remain parallel to each other and first rotate away from the field, and then rotate back. Eventually, near $h = h_{sat}/2$, spins in the three sublattices become parallel to $\boldsymbol{h}$ and spins in the fourth sublattice become antiparallel to $\boldsymbol{h}$ (see Fig. 2(b)). Once this happens, the system enters into the new, UUUD phase. In this phase, $U(1)$ symmetry is restored (there is no sublattice spin component transverse to the field), but Z_4 symmetry is still broken. To obtain the boundaries of the UUUD phase, we compute its excitation spectrum. For this, we introduce four sets of Holstein-Primakoff (H-P) bosons and do spin-wave calculations to order $1/S$. In the classical, $S \to \infty$ limit, the spin-wave excitations are stable only at $h = h_{sat}/2$, where the spectrum consists of one gapped spin wave branch (in-phase precession of all spins around the field), and three gapless branches, with zero modes at Γ point of the four-sublattice Brillouin zone (see Fig. 3(b)). Quantum $1/S$ correction to spectrum, however, make it stable in a finite range of h around $h_{sat}/2$. Namely, all spin-wave branches become gapped (and positive) in a range $h_l < h < h_u$, where $h_l = h_{sat}/2 - \delta_1$ and $h_u = h_{sat}/2 + \delta_2$. We show the details of the calculations in the Supplementary Material (SM) and present the results for δ_1 and δ_2 in Table I. We found, somewhat unexpected, that the stability width of the UUUD phase is finite for *all* J_2 in the interval $1/8 < J_2/J_1 < 1$. We further computed the ground state energy of the UUUD phase to order $1/S$ (classical energy plus $1/S$ corrections from zero point fluctuations), and compared with that of the stripe phase. We found that for all J_2 the energy of the UUUD state is lower. Because of this and because the UUUD state naturally emerges from the $\bar{\rm V}$ state, we argue that the UUUD state is the true ground state near $h = h_{sat}/2$ for all $1/8 < J_2/J_1 < 1$. As all excitations in the UUUD state are gapped, this state has magnetization fixed at exactly 1/2 of the saturation value.

We also verified that at the upper critical field of the UUUD state, it becomes unstable towards $\bar{\rm V}$ state. Namely, at $h = h_u$ one of the spin-wave branches condenses, and the condensate leads to $\langle S_x \rangle = a$ for spins on three up-spin sublattices, and $-3a$ for the spins on the down-spin sublattice. This result in turn implies that the $\bar{\rm V}$ state, which started at a point $J_2 = J_1/3$ at $h = h_{sat}$, extends over the whole range of J_2 near $h_{sat}/2$ (see Fig. 1).

At the lower boundary of the UUUD phase, two other spin-wave modes become unstable at the Γ point. To

J_2/J_1	1/8	1/4	1/3	1/2	1
$\delta_1(1/S)$	0.46	0.15	0.11	0.11	0.28
$\delta_2(1/S)$	1.2	0.80	0.75	0.75	1.09

TABLE I. Results for the boundaries of UUUD state for different J_2/J_1 (see SM for details of calculations). The UUUD state is stable in the range $h_l < h < h_u$, where $h_l = h_{sat}/2 - \delta_1$ and $h_u = h_{sat}/2 + \delta_2$.

determine the the spin order below h_l, we again perform Landau Free energy analysis in terms of the corresponding two complex order parameters Δ_1 and Δ_2. We present the details in the SM. The Free energy has the form [44, 45]:

$$E_\Delta/N = -\mu(|\Delta_1|^2 + |\Delta_2|^2) + \frac{1}{2}\Gamma(|\Delta_1|^2 + |\Delta_2|^2)^2 + \frac{1}{2}K|\Delta_1^2 + \Delta_2^2|^2 \tag{4}$$

Classically, $\Gamma = h_{sat}/4$, $K = 0$. Then $|\Delta_1|^2 + |\Delta_2|^2 \equiv \mu/\Gamma$, i.e. different ordered states are degenerate. Quantum fluctuations lift the degeneracy, and the result depends on the sign of K. If $K > 0$, $\Delta_1 = \pm i\Delta_2$. It can be checked that this gives rise to a non-coplanar umbrella state, in which the down-spin remains intact, and three up-spins split out and form a cone. Such a state breaks $U(1) \times \mathbb{Z}_4 \times \mathbb{Z}_2$ symmetry. If $K < 0$, the relative phase between Δ_1 and Δ_2 is either 0 or π, and the order is coplanar (see Fig. 4).

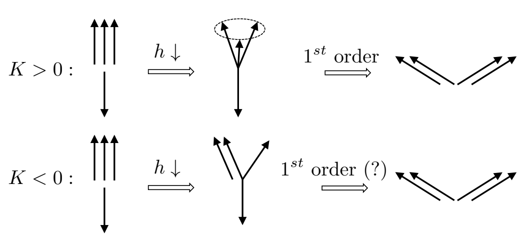

FIG. 4. Evolution of the magnetic order below the UUUD state, depending on sign of the K term in Eq. 4.

We computed K to accuracy $1/S$. The details of calculations are presented in SM, and here we quote the result: K is the sum of logarithmical, $|\log(h_l - h)|/S$, $\log S/S$, and non-logarithmical, $\mathcal{O}(1/S)$ terms, much like Eq. 3. The logarithmical term yields $K < 0$, however the prefactor for the logarithm vanishes at $J_2 = J_1/3$, and at this value of J_2 non-logarithmical terms become relevant. Near $J_2 = J_1/3$, we have

$$K = -\frac{2\sqrt{3}J_1}{\pi}\left(\frac{J_2}{J_1} - 1/3\right)^2\left(\frac{|\log(h_l - h)|}{S} + \beta_\phi\frac{\log S}{S}\right) - \frac{\beta_K}{S}. \tag{5}$$

Where the $|\log(h_l - h)|/S$ term is a contribution from spin wave modes which go as k^2 at $h = h_l$, and $\log S/S$ term comes from another spin wave mode that softens at $h = h_u = h_l + \mathcal{O}(1/S)$. In distinction to the situation near h_{sat}, here we found that K remains negative, even for $J_2/J_1 = 1/3$. This implies that the state below h_l is a co-planar state. An umbrella state is not ruled out, however, for smaller S as we computed β_K in Eq. 5 at $S \gg 1$.

To determine the structure of the coplanar state below h_l more work is actually required because for $K < 0$, the Free energy to order Δ^4 is $E_\Delta/N = -\mu(|\Delta_1|^2 + |\Delta_2|^2) + \frac{1}{2}(\Gamma - |K|)(|\Delta_1|^2 + |\Delta_2|^2)^2$, i.e., the degeneracy is not fully lifted. To select the order, one has to compute $\mathcal{O}(\Delta^6)$ terms in the Free energy. We found (see SM for detail) that sixth-order terms select the order in which of the three up-spins two are tilting in one direction and another in the opposite direction, while the down spin remains intact (see Fig. 4). This state breaks $U(1) \times \mathbb{Z}_4 \times \mathbb{Z}_3$ symmetry. It can potentially transform gradually into the stripe state, which breaks $U(1) \times \mathbb{Z}_3$, if the down spin begins rotating at higher deviations from h_l and match the spin from up-triad, which is separated from the other two. Or, the transition can be first order. Either way, at small fields, the order becomes a stripe. A more complex phase diagram at low fields is expected in the presence of a single-ion anisotropy [38, 46].

Conclusions In this work we analyzed the phase diagram of a Heisenberg antiferromagnet on a triangular lattice, with nearest and second nearest neighbor interactions ($J_1 - J_2$ model), in a magnetic field. We focused on the case $1/8 < J_1/J_2 < 1$, when semiclassical description involves four sublattice representation. We argued that the phase diagram is quite rich and contains several phases. The most substantial result of our study is the identification of the UUUD phase, in which spins in the three sublattices are directed along the field, and spins in the fourth sublattice are directed opposite to the field. Such a state breaks a discrete Z_4 sublattice symmetry, but no orientational and continuous symmetry. As a result, all excitations in the UUUD phase are gapped, and the magnetization is fixed at exactly 1/2 of the saturation value. We demonstrated that this phase is stable in a finite range of fields near $h_{sat}/2$ at all J_2 from the interval $1/8 < J_1/J_2 < 1$. We identified gapless planar states around the UUUD phase. The one at higher fields breaks $U(1) \times \mathbb{Z}_4$ symmetry. The one at lower fields breaks $U(1) \times \mathbb{Z}_4 \times \mathbb{Z}_3$ symmetry. We call for magnetization measurements in quasi-2D triangular-lattice antiferromagnets with $J_2 > J_1/8$, best matetials with $S = 1/2$, as they normally have no single-ion anisotropy, but also $S = 1$ materials, like $2H{-}AgNiO_2$ [38, 39], to verify the existence of the plateau at a half of the saturation value of magnetization and quantum phases above this field.

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