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Magnetic Vortex Induced by Non-magnetic Impurity in Frustrated Magnets

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We study the effect of a non-magnetic impurity inserted in a 2D frustrated ferromagnet above its saturation magnetic field H_{sat} for arbitrary spin S . We demonstrate that the ground state includes a magnetic vortex that is nucleated around the impurity over a finite range of magnetic field $H_{\text{sat}} \leq H \leq H_{\text{sat}}^I$. Upon approaching the quantum critical point at $H = H_{\text{sat}}$, the radius of the magnetic vortex diverges as the magnetic correlation length: $\xi \propto 1/\sqrt{H - H_{\text{sat}}}$. These results are derived both for the lattice and in the continuum limit.

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Introduction. It is known that magnets with competing interactions can support *metastable* skyrmion solutions [1]. Magnetic skyrmions are particle-like spin textures of topological origin, which can be found in non-centrosymmetric (chiral) magnets with competing ferromagnetic (FM) exchange and Dzyaloshinskii-Moriya interaction [2, 3], or in frustrated (non-chiral) magnets with competing exchange interactions [4, 5]. In the light of these results, it is natural to ask if similar topological structures can be rendered *thermodynamically stable* by introducing impurities. This question can be addressed by considering the case of frustrated magnets near a Lifshitz transition between incommensurate spiral ordering (ordering wave vector $\mathbf{Q} \neq 0$) and $\mathbf{Q} = 0$ FM ordering. As we demonstrate in this Letter, a simple non-magnetic impurity is enough to nucleate a magnetic vortex above the saturation field H_{sat} required to fully polarize the spins of the clean system. The vortex state emerges below the local saturation field around the impurity, $H_{\text{sat}}^I > H_{\text{sat}}$, and its radius is equal to the magnetic correlation length, ξ , which diverges as $\xi \propto 1/\sqrt{H - H_{\text{sat}}}$ upon approaching the quantum critical point (QCP) at $H = H_{\text{sat}}$. The mean field exponent $\nu = 1/2$ is characteristic of QCP's in the Bose-Einstein universality class, where the driving parameter (H) couples to a conserved quantity [6].

We present a general theory based on a semi-classical analysis of the continuum theory, which is complemented by an exact solution of the problem on a lattice. We start by considering a spin S triangular lattice (TL) Heisenberg model with FM nearest-neighbor (NN) exchange, $J_1 < 0$, and AFM third NN exchange J_3 :

$$\mathcal{H} = J_1 \sum_{\langle j,l \rangle} \mathbf{S}_j \cdot \mathbf{S}_l + J_3 \sum_{\langle\langle j,l \rangle\rangle} \mathbf{S}_j \cdot \mathbf{S}_l - H \sum_j S_j^z. \quad (1)$$

The relative position vectors of the NNs in the TL are $\pm \mathbf{e}_\nu$, with $\mathbf{e}_1 = \hat{\mathbf{x}}$, $\mathbf{e}_2 = -\hat{\mathbf{x}}/2 + \sqrt{3}\hat{\mathbf{y}}/2$ and $\mathbf{e}_3 = -\hat{\mathbf{x}}/2 - \sqrt{3}\hat{\mathbf{y}}/2$. The main role of J_1 and J_3 is to produce a momentum space interaction, $J(\mathbf{q}) = \sum_{j=1,3} (J_1 \cos \mathbf{q} \cdot \mathbf{e}_j + J_3 \cos 2\mathbf{q} \cdot \mathbf{e}_j)$ with global minima at finite wave-vectors $\pm \mathbf{Q}_\nu$, of magnitude $Q = |\mathbf{Q}_\nu| = 2 \arccos \left[\frac{1}{4} \left(1 + \sqrt{1 - \frac{2J_1}{J_3}} \right) \right]$, which are connected by the C_6 symmetry group of the TL. The system is a ferromagnet for $J_3 < -J_1/4$. The onset of a fi-

nite ordering wave-vector above $J_3 = -J_1/4$ signals a Lifshitz transition from FM to incommensurate AFM ordering. Correspondingly, H_{sat} also becomes finite for $J_3 > -J_1/4$: $H_{\text{sat}} = -SJ(\mathbf{Q}_\nu) + 3S(J_3 + J_1)$. Right above $J_3 = -J_1/4$, we can expand in the small parameter $\delta = 3(J_1 + 4J_3)/(2J_3)$:

$$H_{\text{sat}} \simeq 6S\delta^2 J_3^2 / (J_1 + 32J_3) \quad \text{and} \quad Q \simeq \frac{2}{3}\sqrt{\delta}, \quad (2)$$

which implies that $H_{\text{sat}} \propto Q^4$ near the Lifshitz point.

We will assume that $\delta > 0$ and $H > H_{\text{sat}}$. We want to know the effect of replacing a spin by a non-magnetic impurity. It has been shown recently that non-magnetic impurities lead to non-coplanar spin structures in the TL Heisenberg model with only NN AFM interaction [7–10]. In contrast, here we will consider the effect of non-magnetic impurities on frustrated magnets with competing FM and AFM interactions. Given the FM nature of J_1 , H_{sat}^I turns out to be higher than H_{sat} , implying that the spins should cant away from the z -axis around the non-magnetic impurity for $H_{\text{sat}} \leq H < H_{\text{sat}}^I$. The spin texture far away from the non-magnetic impurity can be obtained by taking the continuum limit $Q \ll 1$. In this limit, \mathcal{H} can be reexpressed as $\int dr^2 [\mathcal{H}^{\text{iso}}(\mathbf{r}) + \mathcal{H}^{\text{ani}}(\mathbf{r})]$, with the Hamiltonian densities

$$\mathcal{H}^{\text{iso}} = -\frac{\delta}{2} J_3 (\nabla \mathbf{S})^2 + \frac{3}{64} (J_1 + 16J_3) (\nabla^2 \mathbf{S})^2 - \mathbf{H} \cdot \mathbf{S},$$

$$\mathcal{H}^{\text{ani}} = A [11 (\partial_x^3 \mathbf{S})^2 + 15 (\partial_x^2 \partial_y \mathbf{S})^2 + 45 (\partial_x \partial_y^2 \mathbf{S})^2 + 9 (\partial_y^3 \mathbf{S})^2],$$

up to sixth order in a gradient expansion and $A = (J_1 + 64J_3)/7680$. Given that the leading order amplitude of the C_6 anisotropy is Q^6 , we can neglect \mathcal{H}^{ani} for $Q \ll 1$. We will rescale our energy, length and magnetic field units in order to normalize the Hamiltonian coefficients [5]:

$$\mathcal{H}^{\text{iso}} = \int \left[-\frac{\delta}{2} (\nabla \mathbf{S})^2 + \frac{1}{2} (\nabla^2 \mathbf{S})^2 - \mathbf{H} \cdot \mathbf{S} \right] dr^2. \quad (3)$$

In the new units we have $H_{\text{sat}} = S\delta^2/4$ and $Q = \sqrt{\delta}/\sqrt{2}$. Equation (3) is the universal theory for centrosymmetric frustrated magnets near a Lifshitz point.

The non-magnetic impurity can be modeled by modifying the stiffness $-\delta$: δ is enhanced near the origin because of the suppression of J_1 on the bonds connecting the non-magnetic

impurity and its NNs. To allow for an analytical solution we consider a step-like modulation of the stiffness:

$$\tilde{\delta}(r) = \delta[1 + \Delta_0 \Theta(r_0 - r)], \quad (4)$$

where $\Theta(x)$ is the Heaviside step function, r_0 is the range over which the local stiffness is modified by the presence of the impurity, and Δ_0 is a positive dimensionless parameter. The choice of the step function is arbitrary because details of this non-universal function do not affect the asymptotic behavior of the solution for $r \gg r_0$.

H_{sat}^I corresponds to the magnetic field value at which the lowest energy mode of the fully polarized (FP) state becomes gapless. The nature of the instability below $H = H_{\text{sat}}^I$ is determined from the structure of the lowest energy mode. We will obtain this mode from a semi-classical expansion of \mathcal{H}^{iso} , which turns out to be exact for arbitrary spin S . To describe the small oscillations around the ground state configuration, we introduce the bosonic field $\varphi(\mathbf{r})$ via the Holstein-Primakoff transformation: $S_{\mathbf{r}}^+ = S_{\mathbf{r}}^x + iS_{\mathbf{r}}^y = \sqrt{2S - \varphi^\dagger(\mathbf{r})\varphi(\mathbf{r})} \varphi(\mathbf{r})$ and $S_{\mathbf{r}}^z = S - \varphi^\dagger(\mathbf{r})\varphi(\mathbf{r})$. By expanding $\mathcal{H}_{J_1-J_3}^{\text{iso}}$ up to quadratic order in $\varphi(\mathbf{r})$, we obtain the spin-wave Hamiltonian density :

$$\mathcal{H}_{J_1-J_3}^{\text{sw}} = H\varphi^\dagger\varphi - \tilde{\delta}(r)S\nabla\varphi^\dagger\nabla\varphi + S\nabla^2\varphi^\dagger\nabla^2\varphi. \quad (5)$$

The resulting equation of motion for $\varphi(\mathbf{r})$ is:

$$-i\partial_t\varphi = H\varphi - \tilde{\delta}(r)S\nabla^2\varphi + S\nabla^4\varphi. \quad (6)$$

In absence of the impurity, the eigenmodes are magnons with a dispersion relation,

$$\omega_{\mathbf{k}} = -\delta S k^2 + S k^4 + H = S \left(k^2 - \frac{\delta}{2} \right)^2 + H - \frac{S\delta^2}{4}. \quad (7)$$

This expression shows explicitly that $H_{\text{sat}} = S\delta^2/4$, i.e., the magnon gap is $\Delta_s = H - S\delta^2/4$. H enters as an additive constant that shifts the whole spectrum because it couples to the conserved quantity $S_T^z = \int S_{\mathbf{r}}^z dr^2$.

In presence of the impurity, the spatially uniform stiffness $-\delta$ must be replaced by the non-uniform stiffness, $-\tilde{\delta}$, of Eq. (4). The eigenmodes, $\psi^\dagger|0\rangle$, are now created by operators of the form $\psi^\dagger = \int \psi^*(\mathbf{r})\varphi^\dagger(\mathbf{r})dr^2$, where $\psi(\mathbf{r})$ is an eigenfunction of the operator $H - S\tilde{\delta}(r)\nabla^2 + S\nabla^4$. Given that rotational symmetry around the origin is still preserved, it is convenient to introduce polar coordinates $\mathbf{r} = r(\cos\phi, \sin\phi)$ and work in the basis of eigenstates of $\mathcal{H}_{J_1-J_3}^{\text{sw}}$ with well defined angular momentum l . These propagating circular waves are described by the function $J_l(kr)e^{il\phi}$, where $J_l(kr)$ is the l -th Bessel function of the first kind:

$$\nabla^2[J_l(kr)e^{il\phi}] = -k^2 J_l(kr)e^{il\phi}. \quad (8)$$

For $r \leq r_0$, the stiffness is constant and equal to $-\delta(1 + \Delta_0)$, implying that the eigenmodes in this region are linear combinations of two circular waves

$$\psi(r \leq r_0) = A_1 J_l(q_+ r) e^{il\phi} + B_1 J_l(q_- r) e^{il\phi}, \quad (9)$$

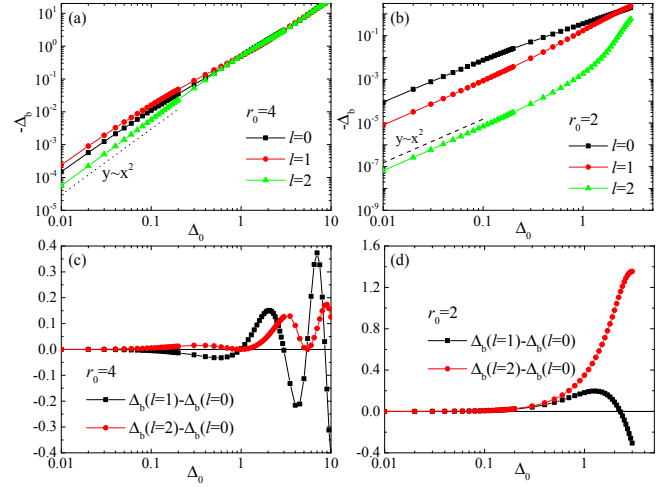


FIG. 1. (color online) Binding energies Δ_b^l for $l = 0, 1, 2$ as a function of Δ_0 for (a) $r_0 = 2$ and (b) $r_0 = 4$. Panels (c) and (d) show the difference between the energies of the $l = 1, 2$ and the $l = 0$ bound states for $r_0 = 2$ and $r_0 = 4$, respectively.

with $q_{\pm} = \frac{\delta(1+\Delta_0) \pm \sqrt{\delta^2 - 4H/S - 4\omega/S + 2\delta^2\Delta_0 + \delta^2\Delta_0^2}}{\sqrt{2}}$. The bound states must decay exponentially for $r \rightarrow \infty$. Therefore, for $r > r_0$ we need to use the modified Bessel functions of the second kind, which satisfy:

$$\nabla^2[K_l(kr)e^{il\phi}] = k^2 K_l(kr)e^{il\phi}. \quad (10)$$

Once again, the eigenfunction for $r > r_0$ is a linear combination of two functions

$$\psi(r \geq r_0) = A_2 K_l(k_+ r) e^{il\phi} + B_2 K_l(k_- r) e^{il\phi}, \quad (11)$$

with momenta $k_{\pm} = \frac{-\delta \pm \sqrt{\delta^2 - 4H/S - 4\omega/S}}{\sqrt{2}}$, which produce an exponential decay for $r \rightarrow \infty$. We note that $\delta^2 - 4H/S - 4\omega/S > 0$ because bound states must lie below the magnon gap $\Delta_s = H - S\delta^2/4$. The last part of the calculation is to impose continuity at $r = r_0$ of the the eigenmodes and their derivatives up to third order. This condition arises from the fourth order nature of the differential equation (6).

The problem is analogous to the 2D quantum mechanical problem of a single particle with an effective mass that depends on the distance to the origin. The only important difference is that the manifold of kinetic energy minima is a ring of radius $k = Q$ instead of a point at the origin [see Eq. (7)], implying that the density of single-magnon states, $\rho(\omega)$, has the same Van Hove singularity as a 1D system when ω approaches the bottom of the single-magnon dispersion $\omega = \Delta_s$: $\rho(\omega) \propto 1/\sqrt{\omega - \Delta_s}$ for $\omega \rightarrow \Delta_s$. This behavior leads to the formation of a bound state for an infinitesimal attractive potential well. As shown in Figs. 1 (a) and (b), the 1D divergence in $\rho(\omega)$ produces a binding energy, $\Delta_b = \omega_b - \Delta_s \propto -\Delta_0^2$, in contrast to the milder essential singularity of the usual 2D problem with a single minimum.

The effective attraction produced by an increase in $|\delta(r)|$ for $r < r_0$ acts on all the circular waves with different angular

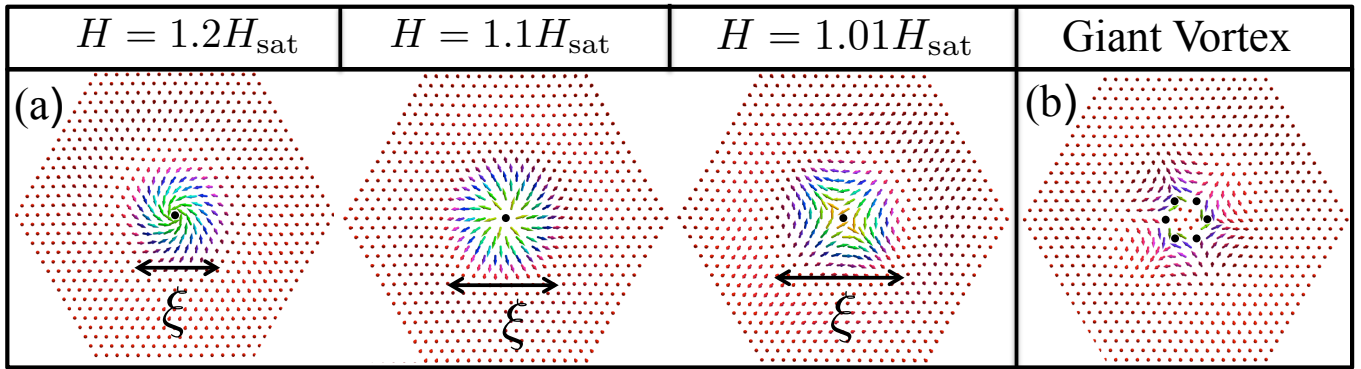


FIG. 2. (color online) Vortex solutions for $H_{\text{sat}} < H < H_{\text{sat}}^I$ obtained from numerical simulations of \mathcal{H} in the classical limit ($S \rightarrow \infty$). (a-c) Vortex bound to a single-site non-magnetic impurity for different field values. The vortex helicity is arbitrary due to the U(1) symmetry of \mathcal{H} . (d) Giant vortex solution ($l = \pm 2$) obtained after removing the spins from the six sites indicated with black dots, for $J_3 = -0.2777J_1$ and $Q = 2\pi/12$. The saturation field is $H_{\text{sat}} = 0.02235|J_1|$, and $H_{\text{sat}}^I = 0.04725|J_1|$.

momenta l . Given that the singularity in $\rho(\omega)$ does not depend on l , a bound state must appear in each l -channel. A lowest energy bound state with finite l implies that a vortex solution with winding number l should emerge right below H_{sat}^I . For weak attraction, $\Delta_0 \ll 1$, the amplitude of the attractive interaction in each channel is

$$g_l(r_0) = Q^3 S \Delta_0 \int_0^{r_0} J_l^2(Qr) dr. \quad (12)$$

Given the asymptotic form of the Bessel functions for small argument, $J_l(z) \simeq (z/2)^l / \Gamma(l+1)$ for $0 \ll z \ll \sqrt{l+1}$, g_l is maximized for $l = 0$ if $r_0 \ll 1$. However, as shown in Fig. 1(b), this is not necessarily true for $r_0 Q \gtrsim 1$. In particular, g_l acquires its maximum value for $l = \pm 1$ when $r_0 Q$ becomes of the order or bigger than the first root of $J_1(z)$. Moreover, a generalized impurity that increases $|\delta(r)|$ in the ring $R - r_0/2 < r < R + r_0/2$ will maximize g_l for $|l|$ -values, which increase monotonically with R . This type of impurity corresponds to removing spins from a ring of sites, instead of the single-site ($R = r_0/2$) that we are currently considering.

Figures 1(a) and (b) show the binding energies of the bound states in the $l = 0, 1, 2$ channels as a function of Δ_0 for $r_0 = 2$ and for $r_0 = 4$. Interestingly enough, the $l = \pm 1$ bound state becomes the ground state above a critical value of the impurity potential Δ_0 for $r_0 = 2$, while it is already the ground state for arbitrarily small values of Δ_0 if $r_0 = 4$. According to this result, the leading instability around an impurity in a frustrated magnet can be a magnetic vortex with winding number $l = \pm 1$. Moreover, strong impurity potentials can produce lowest energy bound states with even higher l values.

Our semiclassical analysis of the continuum theory implies that the FP state is unstable towards a new ground state in which the spins near the non-magnetic impurity are canted away from the field axis. The spin component perpendicular to the field axis must exhibit a finite winding number l (vortex state) under quite general conditions. Finding the actual distortion below H_{sat}^I for arbitrary spin S requires solving a complex many-body problem. However, this problem can be

solved in the classical limit by minimizing the Hamiltonian energy functional given in Eq. (1). This is done by numerically solving the Landau-Lifshitz-Gilbert equation of motion, $\partial_t \mathbf{S} = -\mathbf{S} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{S} \times \partial_t \mathbf{S}$, where α is the Gilbert damping parameter and $\mathbf{H}_{\text{eff}} \equiv -\delta \mathcal{H} / \delta \mathbf{S}$ is the effective magnetic field acting on each spin. The non-magnetic impurity is introduced by setting the spin at the origin to 0: $S_0 = 0$. Consistently with our previous analysis, a vortex is nucleated around the impurity once the system is allowed to relax from an initial FP state. The result is independent of the value of α . As shown in Figs. 2 (a), (b) and (c), the linear vortex size increases upon approaching $H = H_{\text{sat}}$. Indeed, by solving the Euler-Lagrange equations of the continuum model of Eq. (3) for $H_{\text{sat}} < H < H_{\text{sat}}^I$, one can verify that the vortex amplitude (tilting of the spins away from the z -axis) decays exponentially over the magnetic correlation length ξ , which diverges as $\xi \propto 1/\sqrt{H - H_{\text{sat}}}$ upon approaching the bulk saturation field H_{sat} [11]. The vortex radius then diverges at the critical point $H = H_{\text{sat}}$, meaning that the exponential decay is replaced by an algebraic decay $1/\sqrt{r}$. [11], which signals a second order transition into a conical single- \mathbf{Q} magnetic ordering. Finally, Fig. 2 (d) shows a giant vortex solution ($l = \pm 2$) obtained by removing the spins from the ring of six sites indicated with black dots. As it can be anticipated from Eq. (12), such a ring favors values of l which maximize $J_l(z)$ in the region $R - r_0/2 < z < R + r_0/2$.

Lattice. The continuum theory is only valid in the long wave-length limit. A similar calculation valid for any wave-length can be done on the lattice for arbitrary spin S . The modes for $H > H_{\text{sat}}^I$ are obtained by exact diagonalization of \mathcal{H} in the $S_{\bar{r}}^z = NS - 1$ sector, i.e., in the subspace of states with a single-spin flip ($S \rightarrow S - 1$) relative to the FP ground state. The flipped spin can be regarded as a single particle moving in the central potential generated by the impurity at the origin. If the impurity consists of a smaller magnetic moment S' , the flipped spin has a lower energy, $\epsilon_1 = J_1(S - S')$ ($J_1 < 0$) when sitting on the first hexagon of NN sites around the impurity (potential well) and a higher

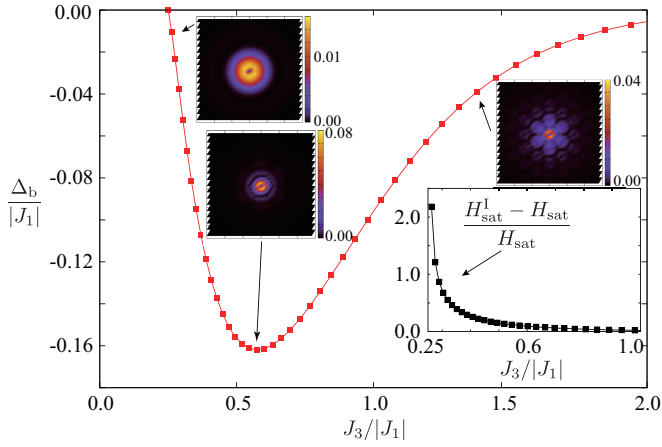


FIG. 3. (color online) Binding energy, Δ_b , of the $l = \pm 1$ bound state around a non-magnetic impurity inserted in the TL model described by \mathcal{H} . $|\Delta_b|$ is equal to the difference between the saturation field around the impurity, H_{sat}^I , and the bulk saturation field H_{sat} . The lower inset shows the ratio $(H_{\text{sat}}^I - H_{\text{sat}})/H_{\text{sat}}$ as a function of $J_3/|J_1|$. The contour plots correspond to the amplitude of the ground state wave-function for selected values of $J_3/|J_1|$.

energy $\epsilon_3 = J_3(S - S')$ (potential barrier) when sitting on the six third-NNs. The hopping amplitude between a pair of sites j and l away from the origin is $J_{jl}S$, while the hopping between the impurity and a different site l is $J_{0l}\sqrt{SS'}$. It is then clear that the total spin S is an overall scaling factor for the effective single-particle Hamiltonian. In other words, the normal modes depend only on the ratio $\alpha = S'/S$, implying that for the case of a non-magnetic impurity ($S' = 0$) the normal modes are exactly the same all the way from $S = 1/2$ to the classical limit $S \rightarrow \infty$.

The ground state of the single spin-flip for \mathcal{H} plus a non-magnetic impurity at the origin ($S' = 0$) is doubly degenerate with quasi-angular momenta $l = \pm 1$ and binding energy Δ_b (see Fig. 3). This ground space is the precursor of the vortex state that appears around the impurity right below the field $H = H_{\text{sat}}^I$, at which the energy of the $l = \pm 1$ bound state becomes equal to the energy of the FP state: $H_{\text{sat}}^I = H_{\text{sat}} - \Delta_b$. $|\Delta_b|$ reaches its maximum value near $J_3/J_1 \simeq -0.6$ and decreases upon approaching the other incommensurate-commensurate transition ($J_3/J_1 \rightarrow \infty$), as expected from the potential well disappearance ($\epsilon_1 \rightarrow 0$) for $J_1 \rightarrow 0$. [12] The inset of Fig. 3 shows that the window of magnetic field values where the non-magnetic impurity is expected to bind a vortex is a sizable fraction of H_{sat} for $J_3 \lesssim J_1$.

While we have used a particular Hamiltonian \mathcal{H} for describing the creation of magnetic vortices by non-magnetic impurities above H_{sat} , our conclusion is valid for a larger family of frustrated Hamiltonians exhibiting Lifshitz transitions. The FM interaction, represented by J_1 in our model, is necessary to have $H_{\text{sat}}^I > H_{\text{sat}}$, i.e., to have a bound state around the impurity. We note, however, that other types of impurities, such as a strain-induced local enhancement of the exchange interactions, can also produce a local saturation field $H_{\text{sat}}^I > H_{\text{sat}}$

in frustrated magnets with spiral ordering induced by two or more competing AFM interactions, such as $\text{Ba}_3\text{Mn}_2\text{O}$ [13–17]. Moreover, the underlying lattice does not need be C_6 invariant. Our continuum theory analysis indicates that vortex states should also appear in tetragonal systems. This is confirmed by an explicit calculation for non-magnetic impurity inserted on a J_1 - J_3 square lattice model [11].

NiGa_2S_4 is a possible realization of \mathcal{H} with $-J_1/J_3 = 0.2(1)$. [18] Unfortunately this small ratio produces an extremely narrow field window above H_{sat} for observing the vortex-impurity bound state. NiBr_2 provides an alternative realization of \mathcal{H} with $-J_3/J_1 = 0.26$. [19] Non-magnetic impurities can be introduced by replacing Ni with Zn. [20] We predict that $\text{Zn}_x\text{Ni}_{1-x}\text{Br}_2$ should exhibit magnetic vortices bounded to the Zn-impurities right above H_{sat} . $\text{CeRhAl}_4\text{Si}_2$ and CeIrAl_4Si are alternative examples of tetragonal frustrated magnets with competing FM and AFM interactions, which exhibit incommensurate intra-layer magnetic ordering at zero field and low enough temperature. [21, 22] Non-magnetic impurities should also nucleate magnetic vortices above the saturation field of these materials.

Finally, the NN FM exchange also leads to the formation of two-magnon bound states above H_{sat} [23–25], implying that the bulk saturation field is in general higher than the value associated with a single-magnon condensation [see Eq. (2)]. The attractive magnon-magnon interaction can produce a continuous transition into some form of multipolar ordering (e.g., nematic ordering for the condensation of magnon pairs) [26–31] or a discontinuous transition. In any case, the magnon-magnon interaction is of order one, while the attractive interaction between the magnon and a non-magnetic impurity is proportional to S , implying that H_{sat}^I remains higher than H_{sat} for large enough S . Another factor that makes the magnon-impurity binding energy larger than the magnon-magnon binding energy is the static nature of the impurity: the reduced mass for the two-magnon problem is half of the single magnon mass relevant for the magnon-impurity problem.

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