Theory of helical spin crystals: phases, textures and properties

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Motivated by recent experiments on the itinerant helimagnet MnSi, we study general properties of helical spin crystals - magnetic structures obtained by superposing distinct spin spirals. An effective Landau description of helical spin crystals is introduced and simple rules for stabilizing various spin crystal structures over single spirals are established. Curious properties of the magnetic structures so obtained, such as symmetry stabilized topological textures and missing Bragg reflections are pointed out. The response of helical spin crystals to crystalline anisotropy, magnetic field and non-magnetic disorder are studied, with special reference to the bcc1 spin structure, a promising starting point for discussing the 'partial order' phases seen at high pressure in MnSi. Similar approaches may be applied to other crystallization problems such as Larkin-Ovchinnikov-Fulde-Ferrel states in spin-imbalanced superconductors.

I. INTRODUCTION

Long period helical spin order resulting from Dzyaloshinskii-Moriya (DM) spin-orbit couplings in non-centrosymmetric itinerant magnets (e.g. MnSi, Fe$_x$Co$_{1-x}$Si, FeGe with crystal structure B20) has been intensively studied in the past. One material of this family, MnSi, has attracted recent interest because of its puzzling behavior under applied hydrostatic pressure. A state with non-Fermi liquid transport properties is obtained over a wide range of pressures above a critical threshold $p_c$. Beginning at the same critical pressure, but over a smaller pressure range, magnetic 'partial order' is observed in neutron scattering experiments. While usual helical order gives rise to sharp Bragg peaks in neutron scattering (corresponding to the periodicity of the helical spin-density wave), 'partial order' is characterized by a neutron scattering signal which is smeared over a wavevector sphere rather than localized at discrete points in reciprocal space.

Recent theoretical work on electronic properties, critical fluctuations and collective modes of helimagnets are in Refs. Theoretical proposals for the high pressure state of MnSi have invoked proximity to a quantum multi-critical point or magnetic liquid-gas transitions. Closest in spirit to our approach are the skyrmion-like magnetic patterns studied in Refs. Recently, we have proposed a novel kind of magnetic order, the helical spin crystal, as a promising starting point for a theory of 'partial order'. Helical spin crystals are magnetic patterns, which are obtained by superposition of several helical spin-density waves which propagate in different directions. There is a subtabital resemblance to multi-k magnetic structures (also known as multiple-q or multiple spin density wave states). But in contrast to most other magnetic multi-k systems, in helical spin crystals the ordering wavevectors are selected from an infinite number of degenerate modes lying on a sphere in reciprocal space - a process analogous to the crystallization of liquids.

In this work, we present a detailed theory of such structures. The stability, structure and distinctive properties of such states are described, and the consequences of coupling to non-magnetic disorder is discussed.

The paper is organized as follows. First, we review the standard theory of helimagnetism in Section finishing with a short remark about more general helical magnetic states. Then, the theory of helical spin crystals is developed. The requirements to energetically stabilize helical spin crystal states are investigated in Section III. The analysis works in two directions. First, we establish a phase diagram in terms of natural parameters which tune the interaction between helical modes and second, we give simple rules to construct model interactions which stabilize a large class of helical spin crystals. The remaining parts of the paper are dedicated to extracting testable consequences of these novel magnetic states. In Section IV, we give a description of the most prominent spin crystals which emerge from our energetic analysis in terms of their symmetry. It is shown that the symmetry of the magnetic state may stabilize topological textures like merons and anti-vortices which are otherwise not expected to be stable in the present context, given the order parameter and dimensionality of the system. Symmetry also determines, which higher-harmonics Bragg peaks these structures would produce. We subsequently study the response of helical spin crystals with respect to different perturbations in Section V. For example, sub-leading spin orbit coupling (crystal anisotropy) locks the magnetic crystal to the underlying atomic lattice and thus determines the location of magnetic Bragg peaks. We also study the response to an external magnetic field which, apart from producing a uniform magnetic moment, also leads to distinctive distortions of the helical magnetic structure, which could be observable by neutron scattering. Finally, in Section VI we investigate the implications of non-magnetic impurities, which are expected to destroy long-range magnetic order and produce diffuse scattering.
II. LANDAU-GINZBURG THEORY OF HELIMAGNETISM

For a cubic magnet without a center of inversion, the Landau-Ginzburg free energy to quadratic order in the magnetization \( \mathbf{M}(\mathbf{r}) \) is

\[
F_2 = \langle r_0 \mathbf{M}^2 + J (\partial_a M_\beta)(\partial_a M_\beta) + 2DM \cdot (\nabla \times \mathbf{M}) \rangle, \tag{1}
\]

where \( \langle \ldots \rangle \) indicates sample averaging, \( r_0, J, D \) are parameters \( (J > 0) \) and Einstein summation is understood. The last term of Eq. (1) is the DM interaction, which is odd under spatial inversion and originates in spin-orbit coupling. Fourier transformation, \( \mathbf{M}(\mathbf{r}) = \sum_q \mathbf{m}_q e^{-i\mathbf{q} \cdot \mathbf{r}} \) with \( \mathbf{m}_q = \mathbf{m}_q^* \), leads to

\[
F_2 = \sum_q \left[ (r_0 + Jq^2) |\mathbf{m}_q|^2 + 2Dm_q^* \cdot (iq \times \mathbf{m}_q) \right]. \tag{2}
\]

Clearly, the energy is minimal for circularly polarized spiral modes, where \( \nabla \times \mathbf{M} \) points in the direction of \(-D\mathbf{M}\). For such modes,

\[
F_2 = \sum_q r(q) |\mathbf{m}_q|^2, \tag{3}
\]

where \( r(q) = r_0 - Jq^2 + J(q - Q)^2 \) with \( Q = |D|/J \). The Gaussian theory thus determines both the chirality of low-energy helical modes and their wavelength \( \lambda = 2\pi/Q \). The latter is typically long (between 180° in MnSi and 2300° in FeCoSi, reflecting the smallness of spin-orbit coupling effects compared to exchange. However, no preferred spiraling direction is selected by Eq. (1), since \( F_2 \) is rotation-invariant. Cubic anisotropy terms which break this invariance are of higher order in the spin-orbit interaction and therefore small. We neglect them for the moment and reintroduce them later.

The isotropic Gaussian theory leaves us with an infinite number of modes which become soft as \( r(Q) \to 0 \). They consist of helical spin-density waves with given chirality (determined by the sign of \( D \)), whose wave-vectors lie on a sphere \(|\mathbf{q}| = Q\) in reciprocal space. Each of these helical modes is determined by an amplitude and a phase. Hence, for each point \( \mathbf{q} \) on the sphere, we define a complex order parameter \( \psi_\mathbf{q} \) (with \( \psi_{-\mathbf{q}} = \psi_\mathbf{q}^* \)) through

\[
\mathbf{m}_q = \frac{1}{2} \psi_\mathbf{q} (\xi_q + i\xi_q^*); \tag{4}
\]

where \( \xi_q, \xi_q^* \) and \( \hat{q} \) are mutually orthogonal unit vectors (with a defined handedness, given by the sign of \( D \)). Obviously, changing the phase of \( \psi_\mathbf{q} \) is equivalent to rotating \( \xi_q \) and \( \xi_q^* \) around \( \hat{q} \). The phase of \( \psi_\mathbf{q} \) is thus only defined relative to some initial choice of \( \xi_q \). The neutron scattering intensity is proportional to \(|\hat{q} \times \mathbf{m}_q|^2 = 1/2|\psi_\mathbf{q}|^2\), independent of the phase. Changing the phase of \( \psi_\mathbf{q} \) is also equivalent to translating \( \mathbf{M}(\mathbf{r}) \) along \( \hat{q} \).

In the following, we study minima of the free energy in the ordered phase \( |r(Q)| < 0 \). These depend on the interactions between degenerate modes (i.e., free energy contributions, which are quartic or higher order in \( \mathbf{M} \)). We only consider interactions which, as \( F_2 \), have full rotation symmetry and we will include the weak crystal anisotropy last. The most general quartic term which has full rotation symmetry (transforming space and spin together) is of the form

\[
F_4 = \sum_{\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3} U(q_1, q_2, q_3) (m_{q_1} \cdot m_{q_2}) (m_{q_3} \cdot m_{q_4}), \tag{5}
\]

with \( q_4 = -(q_1 + q_2 + q_3) \).

A. Single-spiral state

For example, if \( U(q_1, q_2, q_3) \) is a constant, then \( F_4 \propto \langle \mathbf{M}^4 \rangle \). If the interaction depends only on the local magnetization amplitude, i.e., in general if \( F = F_2 + \langle f(M^2) \rangle \) for some function \( f \), then the absolute minimum of \( F \) is given by a single-spiral state (also known as helical spin density wave) \( \mathbf{M}(\mathbf{r}) = \mathbf{m}_q e^{i\mathbf{k} \cdot \mathbf{r}} + \mathbf{m}_q^* e^{-i\mathbf{k} \cdot \mathbf{r}} \), where a single pair of opposite momenta \( \pm \mathbf{k} \) is selected. To proof this, we write \( F \) as

\[
\sum_q [r(q) - r(Q)] |\mathbf{m}_q|^2 + \langle r(Q)M^2 + f(M^2) \rangle. \tag{6}
\]

In the single-spiral state, \( \mathbf{M}^2 \) is constant in space and it minimizes the first and the second term of Eq. (6) independently. Therefore, no other magnetic state can be lower in energy.

Because \( Q \) is small, the relevant wavevectors entering Eq. (6) are also small and \( U(q_1, q_2, q_3) \) is effectively close to a constant. Therefore, the single-spiral state, as observed in FeCoSi, reflecting the most natural helical magnetic order from the point of view of Landau theory.

B. Linear superpositions of single-spiral states

Motivated by the phenomenology of ‘partial order’, we will now extend the theory beyond this standard solution. We speculate that \( U(q_1, q_2, q_3) \) is not constant, such that \( F_4 \) favors a linear superposition of multiple spin-spirals with different wave-vectors on the sphere of degenerate modes \(|\mathbf{q}| = Q\).

One may first speculate about magnetic patterns whose Fourier transform is non-zero everywhere on the wave-vector sphere and peaked infinitely sharply perpendicular to the sphere, i.e.

\[
|\psi_\mathbf{q}|^2 \propto \delta(|\mathbf{q}| - Q) \tag{7}
\]

[see Eq. (4)]. This idea turns out to be complicated for at least two reasons.

The first complication is that there is no continuous way of attributing a finite-amplitude spiral mode to each
point on the wavevector sphere. This can be seen by noting that $\hat{r}_q$ [Eq. (4)] is a tangent vector field on the sphere. Thus, it cannot be continuous (impossibility of combing a hedgehog). Thus there is no “uniform” superposition of helical modes on the sphere. The problem of singularities can be avoided if one assumes a $\psi_q$ with point nodes on the sphere.

The second complication is that higher harmonics would result in a broadening of the delta-function in Eq. (7). This is seen as follows. Consider three momenta $\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3$ on the wavevector sphere and $\mathbf{q}_4$ which is off the sphere. The non-vanishing modes $m_{\mathbf{q}_1}, m_{\mathbf{q}_2}, m_{\mathbf{q}_3}$ couple linearly to $m_{\mathbf{q}_4}$ via Eq. (5) and thus induce a higher harmonic “off-shell” mode $m_{\mathbf{q}_4} \neq 0$. Since this happens for every point away from the sphere, the effect is an intrinsic broadening of the peak in $|\psi_q|^2$, in contradiction with the initial assumption of Eq. (7).

III. ENERGETICS OF HELICAL SPIN CRYSTALS

In the following, we study magnetic structures which are superpositions of a finite number of degenerate helical modes $\psi_q$ with wavevectors $\pm \hat{k}_j$, $j = 1, \ldots, N$. We call the resulting states helical spin crystals, because of the analogy with weak crystallization theory of the solid-liquid transition.

A. Structure of the quartic interaction

We assume that $F_4$ is small, and that its main effect is to provide an interaction between the modes which are degenerate under $F_2$. Thus, the relevant terms of $F_4$ are those with $|\mathbf{q}_1| = |\mathbf{q}_2| = |\mathbf{q}_3| = |\mathbf{q}_4| = Q$. This phase-space constraint and rotational symmetry implies that the coupling function $U$ depends only on two relative angles between the momenta

$$U(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)|_{\mathbf{q}_4} = U(\theta, \phi),$$

where we have chosen the following parameterization:

$$2\theta = \arccos(\hat{q}_1 \cdot \hat{q}_2)$$

$$\phi/2 = \arccos \left[ \frac{(\hat{q}_2 - \hat{q}_1) \cdot \hat{q}_3}{\hat{q}_1 \cdot \hat{q}_2} \right].$$

Geometrically, $\phi/2$ is the angle between the two planes spanned by $(\mathbf{q}_1, \mathbf{q}_2)$ and $(\mathbf{q}_3, \mathbf{q}_4)$ (Fig. 1). In the special case $\mathbf{q}_1 + \mathbf{q}_2 = 0$, it becomes the angle between $\mathbf{q}_2$ and $\mathbf{q}_3$. This mapping allows $\theta$ and $\phi$ to be interpreted as the polar and azimuthal angles of a sphere and the coupling $U(\theta, \phi)$ is a function on that sphere. Since it describes an effective coupling between modes on the wavevector sphere, the coupling $U(\theta, \phi)$ has a status similar to that of Fermi liquid parameters in the theory of metals.

![FIG. 1: The set of quartets $(\mathbf{q}_1, \ldots, \mathbf{q}_4)$ satisfying $|\mathbf{q}_i| = Q$ and $\mathbf{q}_1 + \ldots + \mathbf{q}_4 = 0$, modulo global rotations, may be parameterized by two angles $\theta$ and $\phi$ as shown in this figure.]

The Landau free energy $F = F_2 + F_4$ of helical spin crystal states is calculated as follows. Obviously,

$$F_2 = r(Q) \sum_j |\psi_j|^2.$$  \hspace{1cm} (10)

The quartic term, Eq. (5), may be split into three distinct contributions $F_4 = F_s + F_p + F_m$. The first term, $F_s$, is the self-interaction of each spiral mode with itself:

$$F_s = U_s \sum_j |\psi_j|^4,$$  \hspace{1cm} (11)

where $U_s = U(\theta = \pi/2, \phi = 0)$. A minimum requirement for stability of the theory is $U_s > 0$.

The next term, $F_p$, stems from pairwise interactions between modes. It is of the form

$$F_p = 2 \sum_{i<j} V_p(\theta_{ij}) |\psi_i|^2 |\psi_j|^2,$$  \hspace{1cm} (12)

where $2\theta_{ij} = \arccos(\hat{k}_i \cdot \hat{k}_j)$ and

$$V_p(\theta) = U\left(\frac{\pi}{2}, 4\theta\right) + \sin^4\theta U(\theta, 0) + \cos^4\theta U\left(\frac{\pi}{2}, -\theta, 0\right).$$  \hspace{1cm} (13)

Since $\lim_{\theta \to 0} V_p(\theta) = 2U_s$ is large and positive, $F_p$ provides an efficient repulsion of modes which are too close on the wavevector sphere. Energetically stable superpositions of spirals require substantially smaller values of $V_p(\theta)$ and therefore big-enough angles between them (Section III C). This “mode repulsion” suggests that a continuous distribution of Fourier modes on the wavevector sphere, as discussed in the Section III B, is difficult to stabilize energetically by slowly varying coupling functions $U(\theta, \phi)$. However, singular behavior of the coupling
Finally, the non-trivial quartic term $F_{nt}$ stems from quartets of modes, whose wavevectors sum up to zero. The geometry of four equal-length wavevectors summing to zero is depicted in Fig. 1. A quartet-contribution occurs for $0 < 2\theta < \pi$ and $0 < \phi/2 < \pi$. In this case, the eight wavevectors $\pm k_j$ form the vertices of a cuboid. Hence,

$$F_{nt} = \sum_{j_1 < \ldots < j_4} F_{j_1,j_2,j_3,j_4},$$

where the summation is over such quartets. For example, if $k_{j_1} + k_{j_2} + k_{j_3} + k_{j_4} = 0$, there is a term $F_{j_1,j_2,j_3,j_4} \propto \psi_{j_1}\psi_{j_2}\psi_{j_3}\psi_{j_4}$. The algebra of these terms, which depend on the relative phases of modes, is rather lengthy and obviously depending on the phase convention used to define the $\psi$-variables.

The term $F_{nt}$ fixes the relative phases of $\psi_j$ to minimize the energy. If there is no frustration between minimizing each individual quartet-term, the phases arrange in such a way that all $F_{j_1,j_2,j_3,j_4} \leq 0$. It follows that $F_{nt} \leq 0$, after minimization for the $\psi$-phases. A special case is the quartet of modes, whose wavevectors $\pm k_j$ form the vertices of a cube (e.g. modes along $\langle 111 \rangle$). In this case, $F_{j_1,j_2,j_3,j_4} = 0$ independently of the coupling function $U(\theta, \phi)$ as a consequence of rotational symmetries. This is the reason why no such quartet term appears in the theory of Bak and Jensen. As a consequence, a superposition of four modes along all $\langle 111 \rangle$ directions is necessarily unstable towards shifting the wavevectors away from the perfect cubic configuration in order to gain energy from $F_{nt}$ (see description of fcc* below).

### B. Phase diagram

If $U(\theta, \phi)$ is only slowly varying, it is justified to expand it in spherical harmonics ($Y_{lm}$). That is,

$$U(\theta, \phi) = U_0 + U_{11} \sin \theta \cos \phi + U_{20}(3 \cos^2 \theta - 1) + U_{22} \sin^2 \theta \cos 2\phi,$$

where we retained all terms with $l \leq 2$, which satisfy the relation $U(\theta, \phi) = U(\pi - \theta, \phi) = U(\theta, 2\pi - \phi)$.

Different quartic expressions in terms of the real-space magnetization $M(r)$ may lead to the same projected coupling $U(\theta, \phi)$. For example, the three terms

$$\langle W M^4 + W' [\nabla (M^2)]^2 + W'' (\partial_\alpha M_\beta)(\partial_\alpha M_\beta)M^2 \rangle$$

lead to $U(q_1, q_2, q_3) = W + W'(q_1 + q_2)^2 - W'' q_1 \cdot q_2$. When projected onto the wavevector sphere, they generate only two spherical harmonics, namely

$$U_0 = W + \frac{1}{3}(4W' + W'')Q^2,$$

$$U_{20} = \frac{2}{3}(2W' - W'')Q^2.$$

The terms $W$ and $W'$ have intuitive interpretations: $W$ restricts the magnetization amplitude, whereas $W'$ favors or disfavors modulations of $M^2$, depending on the sign of $W''$. In the following, we set $W'' = 0$ and use the four parameters $W, W', U_{11}$ and $U_{22}$ to tune the interaction $U(\theta, \phi)$.

It is hard to find the exact global minimum of $F_2 + F_4$ for a general coupling function $U(\theta, \phi)$. We therefore restrict ourselves to a certain variational class of magnetic states and determine the minimum within this class. Recently, we studied only states which can be obtained by superposition of all six modes which propagate along the $\langle 111 \rangle$ directions (6-mode model). Here, we explore a much broader class of states. We include

1. any superposition of those 13 spin-spirals which propagate along the directions $\langle 111 \rangle$, $\langle 110 \rangle$ and $\langle 100 \rangle$ (13 complex variables $\psi_j$ as variational parameters),

2. any superposition of up to four spirals with arbitrary propagation direction (four variables $\psi_j$ and five independent angles between wavevectors as variational parameters).

Within these constraints, we have computed a phase diagram as a function of the coupling parameters $W, W', U_{11}$ and $U_{22}$, shown in Figs. 3 and 2.

All magnetic ground states are equal-amplitude superpositions of 1,2,3,4 or 6 spiral modes. The body-centered cubic (bcc) states are superpositions of all $\langle 110 \rangle$-modes. bcc1 and bcc2 differ by the relative phases of the six interfering helical modes (see Section 4). The simple cubic (sc) crystal consists of three mutually orthogonal spirals (e.g. along all $\langle 100 \rangle$ directions). A face-
FIG. 3: Same as Fig. 3 with \( Q^2 W' = -0.5W \). The point \( U_{11} = U_{22} = 0 \) is now at the phase boundary between spiral order and simple cubic.

FIG. 4: a) and b) For the locations \( Q \) of Fig. 2, the ratio between the pair interaction \( V \) and the self-interaction \( U \) is plotted as a function of the angle \( 2\theta \) (the angle between propagation directions of modes). A: single-spiral state; B: phase boundary between single-spiral and bcc1; C: bcc1; D: sc; E: \( \triangle \).

centered cubic (fcc) helical spin crystal is obtained by superposing all four (111)-modes. However, the ground state is not fcc but a small distortion of it: fcc*. In fcc*, the wavevectors are shifted slightly away from (111) in order to gain energy from the quartet-term \( F_{nt} \). The symbols “\( \Delta \)” and “□” are used differently here than in our previous paper. Here, “□” stands for a superposition of four modes with wavevectors as shown in Fig. 1 with \( \phi/2 = \pi/2 \). Hence, wavevectors \( \pm k_j \) form a square cuboid and allow for one quartet-term \( F_{nt} \). The angle \( 2\theta \) changes as a function of interaction parameters within the range \( 0.24\pi < 2\theta < 0.38\pi \). Finally, the phase “\( \triangle \)” consists of three modes. The wavevectors \( k_1, k_2, k_3 \) point to the vertices of an equilateral triangle on the sphere, whose size is determined by the requirement that the mutual angle between two wavevectors, \( 2\theta \), minimizes Eq. (13). The angle \( 2\theta \) is parameter-dependent and lies in the range \( 0.14\pi < 2\theta < 0.24\pi \).

As expected, a negative \( W' \) [Eq. (16)] favors multimode spin crystal states with varying magnetization amplitude relative to the spiral state with constant \( M^2 \) (compare Figs. 2 and 3). Positive \( W' \) has the opposite effect, and enhances the region of the spiral phase (not shown). The term \( W' \) alone (i.e., with \( U_{11} = U_{22} = 0 \)) stabilizes sc in the regime \( Q^2 W' < -W/2 < 0 \). However, a small positive \( U_{22} \) is sufficient to favors bcc1 over sc.

In conclusion, we observe that two helical spin crystals, bcc1 and sc, appear adjacent to the single-spiral state and are stable at relatively small values of \( Q^2 W' \). In the following, we study the properties of the bcc1, and sc states since they are the most likely candidates of helical spin crystals from the point of view of energetics.

C. Model interactions with exact ground states

In the preceding Section, we established a variational phase diagram for “natural”, i.e., slowly varying coupling functions \( U(\theta, \phi) \). Most phases in this phase diagram, (“spiral”, sc, bcc1, bcc2, fcc and “\( \triangle \)” can be shown to be the exact global minima for some fine-tuned model interaction, that are constructed below.

Let us consider the toy model \( F_{toy} = F_2 + F_s + F_p \) [Eqs. (10-12)] where the quartet term \( F_{nt} \) is dropped and we replace \( V'(\theta) \) by a constant \( V \). In this model, local minima with \( N \) non-vanishing modes (\( N \geq 1 \)) must have equal amplitudes \( |\psi_j|^2 = |r|/(U_s - V + NV)/2 \) and the minimum energy with \( N \) modes is

\[
F_{toy, N} = -\frac{1}{4} \frac{r^2}{V + U_s - V},
\]

There are three regimes. If \( 0 < U_s < V \), the ground state is the single-mode phase with \( N = 1 \) (all but one \( \psi \) are zero). For \( 0 < V < U_s \), the energy is monotonically decreasing with \( N \). It means that the system includes as many modes as possible to lower its energy. Finally, for \( V < 0 \) or \( U_s < 0 \), \( F_{toy} \) is not bounded from below and therefore unstable.

To make use of this toy model, we must tune the interaction such that \( U(\theta, \phi) \to 0 \) unless \( \theta = \pi/2 \) or \( \phi = 0 \). This removes the quartet term \( F_{nt} \) and we are left with a model similar to \( F_{toy} \) with the difference that \( V'(\theta) \) is not constant.
We now tune the interaction such that $V_\psi(\theta)$ is very big everywhere except at some angle $\theta_{opt}$, where it has a narrow minimum. For such a pair interaction, all arrangements of modes which involve angles other than $2\theta_{opt}$ are excluded. Within this constraint, we are left with our toy model with a constant pair interaction $V = V_\psi(\theta_{opt})$, but the number of modes is restricted to $1 \leq N \leq 3$, since no more than three modes can have equal mutual angles between them. It is clear that in the region $-3U_s/2 < V_\psi(\theta_{opt}) < U_s$, the ground state is a helical spin crystal with three modes, i.e., the state “$\Delta$” or sc (in the case $2\theta_{opt} = \pi/2$).

A different class of exact ground states is obtained, if the interaction is tuned such that

1. $V_\psi(\theta)$ is very big in the region $0 < 2\theta < 2\theta_c$ and

2. $V_\psi(\theta) = V$ (constant) in the region $2\theta_c \leq 2\theta \leq \pi/2$

for some critical angle $2\theta_c$. In this way, modes whose wavevectors are too close are excluded. That is, it enforces a “hard sphere” constraint $|\hat{k}_i \pm \hat{k}_j| \geq 2(1 - \cos 2\theta_c)$ on the wavevectors. The interaction between modes which satisfy this constraint is constant and reduces to the toy model. Therefore, if $V < U_s$, the ground state will include as many modes as geometrically possible by the “hard sphere” constraint. In the case $2\theta_c = \arccos(1/3)$, we obtain fcc, which has wavevectors at the vertices of a cube. In the case $2\theta_c = \pi/3$, we obtain bcc from the toy model. Therefore, if $V < U_s$, the ground state will include as many modes as geometrically possible by the “hard sphere” constraint. In the case $2\theta_c = \arccos(1/3)$, we obtain fcc, which has wavevectors at the vertices of a cube. In the case $2\theta_c = \pi/3$, we obtain bcc whose wavevectors are the vertices of a cuboctahedron. This can be seen as follows. A real-space bcc lattice corresponds to a fcc reciprocal lattice and the wavevectors $\pm k_j$ of the bcc spin crystals are the 12 nearest neighbors of the origin in the fcc reciprocal lattice. Because fcc is the cubic close packing of spheres, this represents the only arrangement of twelve vectors $|k_j| = Q$ which satisfies the constraint $|\hat{k}_i - \hat{k}_j| \geq 1$. The hexagonal close packing is not acceptable because it does not consist of pairs $\pm k_j$. In principle, this construction can be used to create models whose ground state contains an arbitrarily high number of modes.

We can now use the insight from these constructed models to understand certain features of the phase diagram of Figs. 2 and 3. In Fig. 4 we plot $V_\psi(\theta)$ as obtained from the expansion Eq. 15 at different places in the phase diagram of Fig. 2. In the bcc1 region of the phase diagram (curve C), $V_\psi(\theta)$ is similar to the one constructed above: close to constant and small for $\pi/3 \leq 2\theta \leq \pi/2$, big for $2\theta < \pi/3$. At the phase boundary between single-spiral and bcc1 (curve B), $V_\psi$ is still bigger than $U_s$, indicating that the quartet term $F_{nt}$ is essential to stabilize bcc1 close to the phase boundary. The phase boundaries between single-spiral and three-mode states (sc or $\Delta$) are exactly determined by the crossing of the minimum of $V_\psi(\theta)$ with $U_s$, as illustrated by curves D and E.

IV. STRUCTURE OF HELICAL SPIN CRYSTALS

A. Symmetry properties and topology of helical spin crystals.

Time-reversal symmetry $(T)$ reverses the magnetization direction $M \rightarrow -M$ and may be implemented in terms of the $\psi$-variables as $\psi_j \rightarrow -\psi_j$.

Under spatial translation by a vector $a$, the $\psi$-variables transform as $\psi'_j = \psi_j \exp(ik_j a)$, i.e., they experience a phase change.

If the set of vectors $k_j$ is linearly independent, every phase-change of modes simply amounts to a global translation. This is the case in helical spin crystals with up to three non-planar modes, e.g. in sc and $\Delta$. These states are periodic with Bravais vectors $a_1, a_2, a_3$, which are determined by $a_i \cdot k_j = 2\pi n_{ij}$. It follows that time-reversal $T$ is equivalent to a translation by $(a_1 + a_2 + a_3)/2$ for these states, which leads to a 2-fold symmetry inside the unit cell reminiscent of antiferromagnetism.

In contrast, helical spin crystals with more than three modes like bcc, fcc and $\square$ depend essentially on the relative phases between the helical modes. For these states, $T$ is not equivalent to any translation, since it is not possible that $\exp(ik_j a) = -1$ for all $k_j$. As a consequence, these states are doubly degenerate in addition to the translational degeneracy.

To obtain an understanding of the real-space picture of helical spin crystals, it is useful to study their symmetries under rotations (reflections change the chirality and therefore never appear in the symmetry group). For example, if the structure $M(\mathbf{r})$ has an $n$-fold rotation axis with direction $\hat{n}$, then $M \parallel \hat{n}$ along this axis. Therefore, $M_{\perp}$, which is the projection to the plane orthogonal to $\hat{n}$, has a node at the axis. The winding number $w_{\perp}$ (vorticity) of the node is restricted by symmetry to the values $1, 1 \pm n, 1 \pm 2n$, etc. The resulting pattern in the vicinity of the rotation axis, $M$ pointing along $\hat{n}$ in the center and $M_{\perp}$ winding around it, resembles that of a skyrmion or meron. Such patterns are currently being discussed in the context of helimagnets. Here, we observe that they naturally appear resulting from rotational symmetries. The simplest cases are winding numbers of $+1$ (for any $n$-fold axis) or $-1$ (only for $n = 2$).

Apart from proper rotations, the point group may contain anti-rotations, i.e., symmetry operations which are composed of a rotation followed by $T$. $n$-fold antirotation axes are only possible for even numbers $n$, since they imply $n/2$-fold rotation symmetry. In the special case $n = 2$, it merely follows that $M_{\perp} \parallel \hat{n}$ along the axis, where $\hat{n}$ is the axis direction. Higher anti-rotation symmetries with $n = 4, 6, \ldots$ imply $M = 0$ along the axis, i.e., they create a line-node in the magnetization. In the vicinity of this line-node, $M$ is approximately orthogonal to $\hat{n}$ (as can be seen by expanding $M(\mathbf{r})$ to linear order around a point on the axis). The winding number of $M_{\perp}$ around the node, is restricted by symmetry to the values
1 ± n/2, 1 ± 3n/2, 1 ± 5n/2, etc. The simplest case for a 4-fold anti-rotation axis is a winding number of −1 (anti-vortex line) and the simplest case for n = 6 is a winding number of −2.

We have thus demonstrated the emergence of topological objects like merons and antivortices, which are not expected to be stable in the present context of a vectorial order parameter in three dimensions but which are stabilized by symmetry. Thus, rotation axes are meron lines and 4-fold anti-rotation axes are anti-vortex node-lines.

1. Symmetry and real-space picture of bcc1

The bcc helical spin crystals consist of six helical modes with wavevectors $\mathbf{k}_1 = [110]$, $\mathbf{k}_2 = [110]$, $\mathbf{k}_3 = [011]$, $\mathbf{k}_4 = [011]$, $\mathbf{k}_5 = [101]$ and $\mathbf{k}_6 = [101]$, as shown in Fig. 5 and has the periodicity of a body centered cubic lattice. We chose the convention [see Eq. (4)]

$$e^{j'}_{j} = \frac{\mathbf{z} \times \mathbf{k}_j}{|\mathbf{z} \times \mathbf{k}_j|} \tag{19}$$

for j = 1,..., 6 with $\mathbf{z} = [001]$, and we consider negative chirality $[\text{i.e., } \mathbf{M} \cdot ((\nabla \times \mathbf{M}) < 0]$, such that $e^{j'}_{j} = \mathbf{z} \times e^{j}_{j}$.

The geometry of the wavevectors allows for three quartet terms in the free energy produced by $T_x = \psi_1^* \psi_2 \psi_3 \psi_6$, $T_y = \psi_1^* \psi_3 \psi_4 \psi_6$ and $T_z = -\psi_3 \psi_4 \psi_6^*$. The sign in the definition of $T_z$ has been introduced for convenience.

The transformation properties of the $\psi$-variables and the three quartic terms under rotations are shown in Table I. From this, it can be deduced that the (rotation-invariant) quartet contribution to the free energy is

$$F_{\text{nt}} = 2\lambda_{\text{nt}} \text{Re}(T_x + T_y + T_z). \tag{20}$$

The value of the parameter $\lambda_{\text{nt}}$ is not determined by symmetry. Direct calculation yields

$$\lambda_{\text{nt}} = \frac{\pi}{4} U/\left[\frac{\pi}{3} - 2 \text{arccos}\left(\frac{1}{3}\right)\right] = \frac{1}{2} U/\left(\frac{\pi}{4}, \pi\right). \tag{21}$$

There are thus two bcc states, depending on the sign of $\lambda_{\text{nt}}$. In bcc1 (bcc2), which corresponds to $\lambda_{\text{nt}} > 0$ (< 0), the phases of the $\psi$’s are such that $T_x$, $T_y$, $T_z$ are all negative (positive). Three out of the six phases are arbitrary due to global translation symmetry. This means that the magnetic pattern of bcc1 and bcc2 is uniquely determined up to translational and time-reversal degeneracy.

The solution for $\lambda_{\text{nt}} > 0$, bcc1, turns out to be the bcc structure with the highest point group symmetry. By selecting the coordinate origin conveniently, we obtain $-\psi_1 = \psi_2 = -i \psi_3 = i \psi_4 = \psi_5 = -i \psi_6 = SM_0$ for bcc1, where $S$ = ±1 is the time-reversal symmetry label and $M_0 > 0$ is the amplitude. From Table I we deduce that $\mathbf{M}(r)$ changes sign under a $\pi/2$ rotation about the $x$, $y$ or $z$ axis. That is, the magnetic point group is $O(T)$ (international notation $4\overline{2}d$) with 4-fold anti-rotation axes at $\langle 100 \rangle$, 3-fold rotation axes at $\langle 111 \rangle$ and 2-fold anti-rotation axes at $\langle 110 \rangle$.

The real-space representation of the bcc1 state is

$$\mathbf{M}(r) = SM_0 \left( \frac{\sqrt{3}}{2} s_x (c_y - c_z) - 2 s_y s_z \right) \left( \frac{\sqrt{3}}{2} s_y (c_z - c_x) - 2 s_z s_x \right), \tag{22}$$

where $s_x = \text{sin}(Qx/\sqrt{2})$, $c_x = \text{cos}(Qx/\sqrt{2})$, etc. The resulting pattern was shown in Fig. 2 of our earlier paper. In Fig. 5 we show the symmetry axes. As discussed above, the magnetization must vanish along the 4-fold anti-rotation axes, which are anti-vortices with winding number −1. The $x$, $y$, $z$ axes, and their translations according to the bcc periodicity, form two interpenetrating cubic lattices of such line-nodes. The cubic space diagonals are 3-fold and the red arrowed lines of Fig. 6 are 2-fold rotation axes. In the vicinity of these lines, the magnetization field is skyrmion-like (i.e., $\mathbf{M}_\perp$ has a winding number of +1).

The fact that the bcc1 state breaks $\mathcal{T}$ in a non-trivial way and cannot be restored by any translation is manifest in the occurrence of the $\mathcal{T}$-breaking order parameter $\langle M_x M_y M_z \rangle = SM_0^3/2 \neq 0$, which is a magnetic octupole. This curious property may lead to distinctive anomalous effects, e.g. in the magnetotransport. Octupolar magnetic ordering has recently been discussed in different contexts.

| $R_z$ | $\psi_1$ | $\psi_2$ | $\psi_3$ | $\psi_4$ | $\psi_5$ | $\psi_6$ | $T_x$ | $T_y$ | $T_z$ |
|-------|--------|--------|--------|--------|--------|--------|------|------|------|
| $R_x$ | $i\psi_6$ | $-i\psi_6$ | $-\psi_3$ | $\psi_4$ | $i\psi_2$ | $i\psi_1$ | $T_x^*$ | $T_y^*$ | $T_z^*$ |

TABLE I: Transformation properties of the $\psi$-variables and three quartic terms (defined in Section IV A 1) of the bcc spin crystals under rotations. $R_z$ and $R_x$, respectively, are $\pi/2$ rotations around the $z$- and $x$-axis. These two rotations generate the cubic point group $O$ and therefore, the behavior under any rotation which maps the 12 wavevectors onto each other may be obtained by combining these two operations.
netization. The red (dark gray) lines are 2-fold rotation axes with 4-fold anti-rotation symmetry and vanishing magnetization. The structure has 3-fold rotation symmetry about all cubic space diagonals.

\[ R_{\pm} = R_{\pm}^{(3)} R_{\pm}^{(3)} R_{\pm}^{(3)} \]

| \( R_{\pm} \) | \( R_{\pm} \) | \( R_{\pm} \) | \( R_{\pm}^{(3)} \) | \( R_{\pm}^{(3)} \) |
|---|---|---|---|---|
| \( \psi_1 \) | \( \psi_2 \) | \( \psi_3 \) | \( \psi_4 \) | \( \psi_5 \) |
| \( \psi_2 \) | \( \psi_3 \) | \( \psi_4 \) | \( \psi_5 \) | \( \psi_1 \) |
| \( \psi_3 \) | \( \psi_4 \) | \( \psi_5 \) | \( \psi_1 \) | \( \psi_2 \) |

TABLE II: Transformation for the \( \psi \)-variables of sc under rotations. \( R_x \) and \( R_z \) are \( \pi/2 \)-rotations, \( R_{\pm}^{(3)} \) is a \( 2\pi/3 \)-rotation and \( R_{\pm}^{(3)} \) a \( \pi \)-rotation around the indicated axis.

2. Symmetry and real-space picture of sc

The simple cubic (sc) helical spin crystal consists of three modes with \( \mathbf{k}_1 = [100], \mathbf{k}_2 = [010] \) and \( \mathbf{k}_3 = [001] \). It forms a periodic structure with a cubic unit cell and the lattice constant is \( \lambda = 2\pi/Q \). The convention for \( \mathcal{C}_j \) is given by Eq. (11), where the unit vector \( \mathbf{z} \) is replaced by [111].

The transformation properties of the \( \psi \)-variables under rotations are given in Table II. By choosing the center of coordinates corresponding to \( \psi_1 = \psi_2 = \psi_3 = iM_0 \), we obtain from Table II that the point group symmetry is \( D_3(D_3) \) (international notation 32). That is, the chosen origin has a 3-fold rotation axis along \([111]\) and three two-fold axes along \([110], [10\bar{1}] \) and \([0\bar{1}1]\). Obviously, \( \mathbf{M} \) must vanish at a point of such high symmetry. Hence there is a point node at the origin.

Symmetry operations consisting of a rotation followed by an appropriate translation yield similar point nodes at \( \frac{1}{3} \mathbf{k}_1 \) (with 3-fold axis along \([111]\)), \( \frac{1}{3} \mathbf{k}_2 \) (3-fold axis \([111]\)) and \( \frac{1}{3} \mathbf{k}_3 \) (3-fold axis \([111]\)). Finally, each of these nodes is doubled inside one unit cell because a translation by \( \frac{1}{3} \mathbf{k}_i \) amounts to \( \mathbf{M} \rightarrow -\mathbf{M} \). The 2- and 3-fold rotation axes form a complex array of skyrmion-like lines, all with winding numbers of +1. The real-space representation is

\[ \mathbf{M}(\mathbf{r}) = SM_0 \begin{pmatrix} \mathbf{c}_y - s_z \\ \mathbf{c}_z - s_x \\ \mathbf{c}_x - s_y \end{pmatrix}, \]  

where \( s_z = \sin[Q(x + \lambda/8)], \mathbf{c}_x = \cos[Q(x + \lambda/8)] \), etc.

B. Higher harmonics Fourier modes

As briefly mentioned in Section 4.3 magnetic ordering in wavevectors \( \pm \mathbf{k}_j \) generally induces higher harmonics in the magnetic structure. In the presence of magnetic order \( \mathbf{m}_{k_j} \neq 0 \), the Landau free energy for the modes \( \mathbf{m}_q \), which do not belong to the set \( \mathbf{m}_{k_j} \), is (to quartic order)

\[ \Delta F = \sum_q \tilde{\mathbf{h}}_\psi(q)|\mathbf{m}_q|^2 - \mathbf{h}_\psi(q) \cdot \mathbf{m}_q^* - \mathbf{h}_\psi^*(q) \cdot \mathbf{m}_q, \tag{24} \]

with \( \tilde{\mathbf{h}}_\psi(q) = \mathbf{h}(q) = O(1) \) and

\[ \mathbf{h}_\psi(q) = -4 \sum_{q_1,q_2,q_3} U(q_1,q_2,q_3) \langle \mathbf{m}_{q_1} \cdot \mathbf{m}_{q_2} \rangle \mathbf{m}_{q_3}, \tag{25} \]

where the sum is restricted to \( q_1,q_2,q_3 \in \{ \pm \mathbf{k}_j \} \) such that \( q_1 + q_2 + q_3 = \mathbf{q} \). The origin of the exchange field \( \mathbf{h}_\psi \) is the coupling term Eq. (5). In the following, we assume that \( \tilde{\mathbf{h}}(q) > 0 \). Obviously, Eq. (24) then leads to induced modes

\[ \mathbf{m}_q = \frac{\mathbf{h}_\psi(q)}{\tilde{\mathbf{h}}(q)} \tag{26} \]

at momenta \( \mathbf{q} = \pm \mathbf{k}_j \pm \mathbf{k}_j \). These modes modify the detailed magnetic structure, but they do not change its symmetry, since the field \( \mathbf{h}_\psi \) respects all the symmetries of the spin crystal.

We now briefly discuss the consequences for the three helical magnetic structures under consideration.

A single spin-density wave involving wavevectors \( \pm \mathbf{k} \) might create higher harmonics at \( \pm 3\mathbf{k} \) via Eq. (26). However, in the case of spin spirals, \( \mathbf{m}_q^2 = 0 \) and therefore \( \mathbf{h}_{\psi,3\mathbf{k}} = 0 \). Thus, there are no higher harmonics created by a single spin spiral.

The sc spin structure with principal ordering wavevectors \( \langle 001 \rangle \) with \( |\mathbf{k}_j| = Q \) generates higher harmonics along \( \langle 111 \rangle \) (with \( |\mathbf{q}| = \sqrt{3}Q \)) and along \( \langle 012 \rangle \) (with \( |\mathbf{q}| = \sqrt{5}Q \)). Note that throughout the current and last sections, all crystal directions refer to the magnetic crystals. The orientation of a magnetic crystal with respect to the atomic crystal depends on the anisotropy term \( F_a \), which will be considered in Section 5.4.

In contrast to the former cases, bcc structures couple linearly to the \( \mathbf{q} = 0 \) mode (i.e., the uniform magnetization), since some triples of ordering vectors add to zero. This coupling will be further investigated in Section 5.5. Here, we only notice that for the bcc1 and bcc2 states, \( \mathbf{h}_{\psi,\mathbf{q}=0} = 0 \). This results can be understood in terms of the symmetry of these states. In the case of bcc1, the point group symmetry is too high to support a non-zero axial vector \( \mathbf{h}_{\psi,\mathbf{q}=0} \). Therefore, bcc1 and bcc2 do not create a spontaneous net magnetization. The next set of
wavevectors which can be reached by adding three ordering vectors are along (001) (with $|q| = \sqrt{2}Q$). However for bcc1 and bcc2, direct calculation shows $h_{\psi} = 0$ for these modes. As before, this can be understood in terms of symmetry. Higher harmonics along (001) would have the structure of a sc spin crystal. We have seen in Section [4] that the point group symmetry of sc is lower than that of bcc1. Therefore, bcc1 can not create such an exchange field $h_{\psi}$. We conclude that bcc1 creates no secondary Bragg peaks at (0, 0, $\sqrt{2}Q$), etc. The same is true for bcc2. The shortest wavevectors which are created by bcc1 or bcc2 as higher harmonics are along (112) (with $|q| = \sqrt{3}Q$). Others are at (110) ($|q| = 2Q$), (013) ($|q| = \sqrt{5}Q$), (111) ($|q| = \sqrt{6}Q$) and (123) ($|q| = \sqrt{7}Q$).

V. RESPONSE TO CRYSTAL ANISOTROPY, MAGNETIC FIELD AND DISORDER.

A. Effect of crystal anisotropy

So far, our free energy has been completely rotation invariant. In the magnetically ordered states, full rotation symmetry is spontaneously broken, but any global rotation of the spin structure leaves the energy invariant. This degeneracy is lifted by an additional anisotropy term $F_a$, which couples the magnetic crystal to the underlying atomic lattice. The crystal anisotropy energy is small and may be treated as a perturbation which merely selects the directional orientation, but does not otherwise affect the magnetic state.

1. Single-spiral state

In the case of a single-spiral state, crystal anisotropy is a function $F_a(\mathbf{k})$, where $\mathbf{k}$ is the spiral direction. The function $F_a(\mathbf{k})$ may depend on various parameters, it should be symmetric under the point group of the (atomic) crystal lattice and satisfy $F_a(\mathbf{k}) = F_a(-\mathbf{k})$. For concreteness, we assume the cubic point group $T$, relevant for the B20 crystal structure. We further assume that $F_a(\mathbf{k})$ is a slowly varying function, since a singular or rapidly oscillating function in reciprocal space would translate into a (non-local) interaction between magnetic moments and the atomic crystal. Such a function $F_a(\mathbf{k})$ generally has its minimum at either (100) or (111), which can be shown in two different ways.

The first argument is based on combining symmetry with Morse’s theory of critical points. Morse theory implies that

$$\text{maxima} - \text{saddles} + \text{minima} = 2 \quad (27)$$

for a function on the unit sphere. Symmetry requires that $F_a(\mathbf{k})$ has stationary points (points with vanishing first derivative, i.e., maxima, minima or saddles) at (100) (6 directions), (111) (8 directions) and (110) (12 directions). If $F_a(\mathbf{k})$ is slowly varying, we suspect that these are the only stationary points, since adding more maxima, minima and saddles means that the function is more rapidly oscillating. Under this hypothesis, it follows from Eq. (27), that the (110) directions are saddle points and that the extrema are at (111) and (100). For (110) to be minima, $F_a(\mathbf{k})$ needs to have additional stationary points (e.g. saddles) at non-symmetric, parameter-dependent locations. We conclude that an anisotropy which favors (110) would need to be more rapidly oscillating than required by symmetry.

The second argument is based on an expansion of $F_a(\mathbf{k})$ in powers of the directions cosines $\hat{k}_x, \hat{k}_y, \hat{k}_z$:

$$F_a(\mathbf{k}) = \alpha (\hat{k}_x^4 + \hat{k}_y^4 + \hat{k}_z^4) + \alpha' \hat{k}_x^2 \hat{k}_y^2 \hat{k}_z^2 + \ldots, \quad (28)$$

where we retained the first two terms allowed by cubic symmetry. Because of the smallness of the wave-vector sphere $Q$, one typically expects $|\alpha'| \ll |\alpha|$ and subsequent terms even smaller. It is easily checked that for most values of the parameters $\alpha, \alpha'$, Eq. (28) has its global minima at either (100) (for $\alpha < \min(0, \alpha'/18)$) or (100) (for $\alpha > \max(2\alpha'/9, \alpha'/18)$). Only in the narrow parameter regime $0 < \alpha < 2\alpha'/9$, the minima are indeed at (110). We conclude that crystal anisotropy which favors (110) may only appear in a narrow regime between two phases which favor (111) and (100), respectively.

Accordingly, (111) or (100) are the selected spiral directions in all cubic helimagnets known so far. The preferred direction in MnSi at low pressure is (111) and in Fe$_x$Co$_{1-x}$Si, it is (100). In FeGe, there is a phase transition between these two directions but no intermediate phase with (110) spiral orientation has been reported. However, the neutron scattering data in the partially ordered phase of MnSi clearly show a maximum signal along the (110) crystal directions. While it is initially tempting to interpret the partially ordered state of MnSi as a single-spiral state that has lost its orientational long-range order by some mechanism, one would still expect a maximal scattering intensity in the energetically preferred lattice direction. Theories of the partially ordered state in terms of disordered helical spin-density waves thus depend on a crystal anisotropy that prefers spiral directions along (110). As we have shown, this seems very unlikely.

2. Helical spin crystals

For multi-mode spin crystals, $F_a$ is no longer determined by a single direction $\mathbf{k}$, so the arguments of Section [4] do not apply. Rather, the anisotropy energy depends on three Euler angles, which rotate the full three-dimensional magnetic structure relatively to the atomic crystal. In other words, $F_a$ is a function of the rotation group $SO(3)$. Relative to some standard orientation $\mathbf{k}_j$ of the mode directions, the leading-order anisotropy term
where $R$ is a rotation operator and $g(\hat{k}) = \hat{k}_x^4 + \hat{k}_y^4 + \hat{k}_z^4$. As before, we have assumed a cubic point group symmetry. In the case $a > 0$, the modes of the bcc spin crystals get locked to the $⟨110⟩$ directions. The orientation of sc is four times degenerate if $a > 0$. The four minima of $F_a$ are obtained from the standard orientation along $(100)$ through a $π/3$-rotation around any of the four space diagonals, such that the three spiral modes point along $(122)$. In the opposite case $(a < 0)$, sc is oriented along $(100)$. This time, it is the bcc spin crystals that get rotated by $π/3$ around any $(111)$ axis to reach one of four stable orientations. Under such $π/3$-rotations, three of the six modes remain along $(110)$ and three move to $(114)$. Each individual $(114)$ direction appears only in one of the four solutions but each $(110)$ direction appears in two of four solutions.

If there is more than one degenerate orientation, the sample typically breaks up into domains such that full cubic symmetry is restored in the neutron scattering signal. Table III lists the directions of magnetic Bragg peaks for the different cases. Out of the three prominent phases in our phase diagram, the bcc1 spin crystal is the only one that can explain the neutron-scattering peaks along $(110)$ in the ’partial order’ phase of MnSi. It does so most naturally for the case $a > 0$, which is the known sign of the anisotropy in MnSi at low pressure.

In order to compare the energy scale of $F_a$ (i.e., the locking energy) for the different magnetic states, we note the following. At the phase boundary between two equal-amplitude-spiral-crystal phases (one phase with amplitudes $|ψ_1| = \ldots = |ψ_N|$ and the second phase with $|ψ_1| = \ldots = |ψ_N|$) the amplitudes of the two neighboring phases are related by

$$N|ψ_j|^2 = \tilde{N}|ψ_j|^2.$$

(30)

In the vicinity of the phase boundary, the anisotropy term is therefore proportional to the mode-average $1/N \sum_j g(R^j_k)$. Using this result, we find that the effective anisotropy energy is smaller for the bcc spin crystals than for the single-spiral state by a factor of $4 - 4.5$.49 For the sc state, the locking energy is anisotropic. Certain rotations are equally costly in energy as for the single-spiral case while some small rotations about the minimum for $a > 0$ are softer than for the single-spiral by a factor of 4.5.

**B. Effect of magnetic field**

A uniform external magnetic field $\mathbf{H}$ couples to the $q = 0$ mode of the magnetization, $\mathbf{m} = ⟨\mathbf{M}⟩$, via Zeeman coupling. The uniform magnetization, in turn, couples to the helical modes $ψ_j$ through

$$F = F|_{m=0} + r_0 \mathbf{m}^2 + U(0,0,0) \mathbf{m}^4 - h_0(0) \mathbf{m} + 2 \sum_j (U(k_j,−k_j,0) \mathbf{m}^2 + U(k_j,0,0) \mathbf{m}_{1−j}^2)|ψ_j|^2,$$

(31)

where $\mathbf{m}_{1−j} = \mathbf{m} − (\mathbf{m} \cdot \hat{k}_j) \hat{k}_j$ and we have used Eqs. (5),(25). Plumer and Walker argued that $U(0,0,0) ≈ U(k_j − k_j,0) ≈ U_s$, which we will use in the following for simplicity.

1. Response of the single-spiral state

The behavior of the single-mode spiral state under a magnetic field has been studied both experimentally7,8,9,10,12 and theoretically37,38. It is characterized by a strongly anisotropic susceptibility, induced by the last term in Eq. (31). For a fixed spiral direction $\mathbf{k}$, the susceptibilities parallel and orthogonal to the spiral direction are given by

$$χ_∥ ≈ Δ^{-1},$$

$$χ_⊥ ≈ δ + \frac{2 U′}{U_s}|r(Q)|^{-1},$$

(32)

where $Δ = 2[r_0 − r(Q)] = 2JQ^2$ and $U′ = U(k,0,0)$. Well below the critical ordering temperature, $Δ ≪ 2|r(Q)|$ and therefore $χ_∥ \gg χ_⊥$. This strong anisotropy leads to a spin reorientation transition at $H = H_{sr}$, where the spiral axis gets oriented along the field direction. The value of $H_{sr}$ depends on the anisotropy [Eq. (25)] and the field direction. For $α > 0$ and $\mathbf{H}∥ ⟨100⟩$, Plumer and Walker obtained

$$H_{sr}^2 = \frac{4α}{χ_∥ − χ_⊥} \geq 40αΔ,$$

(33)

where we have used $χ_∥ ≫ χ_⊥ > 0$. Once the spiral is oriented, the susceptibility is large (equal to $χ_∥$).

The spiral amplitude decreases as a function of the external field and vanishes at

$$H_c = |ψ_0|Δ,$$

(34)

where $|ψ_0|^2 = |r(Q)|/(2U_s)$. Above $H_c$, the magnetization is uniform.

| $a > 0$ | spiral No. | bcc No. | sc No. |
|--------|----------|--------|--------|
| $a < 0$ | ⟨111⟩   | 4      | (110)  | 1      | (122)  | 4      | (100)  | 1      |

*When averaged over domains, Bragg peaks along ⟨110⟩ are twice as intense as peaks along ⟨114⟩.

**TABLE III**: Crystal directions of magnetic Bragg peaks and number of degenerate orientations for three magnetic structures, sc, bcc and single-spiral, with crystal anisotropy given by Eq. (29).
2. Response of the bcc1 spin crystal

In the bcc spin crystal states, the linear response is isotropic, because their symmetry group does not allow for an anisotropic susceptibility tensor. As a consequence, there is no orientation of the bcc state towards the magnetic field at the level of linear response (i.e., from energies up to order \( H^2 \)). However, there is a sub-leading contribution to the energy \( \propto (M_x M_y M_z) H_x H_y H_z \). This contribution splits the degeneracy between the \( S = 1 \) and \( S = -1 \) states and it may lead to a reorientation of the bcc crystal towards the field.

In terms of the six \( \psi \)-variables of bcc, the exchange field \( \mathbf{h}_\psi(0) \), which enters Eq. \( (31) \), amounts to

\[
\mathbf{h}_\psi(0) = -\mu \text{Re} \left[ (5 - i \sqrt{2}) \tilde{\mathbf{h}}_\psi \right],
\]

where \( \mu = U(\mathbf{k}_{j_1}, \mathbf{k}_{j_2}, \mathbf{k}_{j_3}) \) for \( \mathbf{k}_{j_1} + \mathbf{k}_{j_2} + \mathbf{k}_{j_3} = 0 \) and

\[
\tilde{\mathbf{h}}_\psi = \psi_1 \psi_3^* \psi_6^* \begin{pmatrix} 1 & 1 & -1 \\ 1 & -1 & 0 \\ -1 & -1 & -1 \end{pmatrix} + \psi_1^* \psi_4 \psi_5 \begin{pmatrix} 1 & 1 & -1 \\ 1 & -1 & 0 \\ -1 & -1 & -1 \end{pmatrix}.
\]

The (isotropic) inverse spin susceptibility (see Appendix) in the bcc1 state is composed of three contributions

\[
\chi^{-1}_{\text{bcc1}} = \chi^{-1}_{\text{bare}} + \chi^{-1}_{\text{phase}} + \chi^{-1}_{\text{amp}}.
\]

The first term

\[
\chi^{-1}_{\text{bare}} = 2 \left( r_0 + \frac{U_{\text{bcc1}}}{4 U_{\text{bcc}}^2} |r(Q)| \right),
\]

where \( U_{\text{bcc1}} = 1/[U_{\text{bcc}} + 4 V_{r}(\pi/4) + 4 V_{s}(\pi/6) - \lambda_{nt}] \), can be derived in analogy to the single-spiral case. In fact, \( \chi_{\text{bare}} \) is a “mixture” of \( \chi_{s} \) and \( \chi_{nt} \), determined geometrically by the angles between the mode directions \( \mathbf{k}_j \) and the magnetic field. It follows that \( \chi_{\text{bare}} \ll \chi_{s} \), provided \( U_{\text{bcc1}} \sim U_{\text{bcc}} \) (the two couplings are equal at the phase boundary between single-spiral and bcc1). The remaining terms in Eq. \( (37) \)

\[
\begin{align*}
\chi^{-1}_{\text{phase}} &= \frac{\mu^2 |r(Q)|}{3 U_{\text{bcc1}} \lambda_{nt}}, \\
\chi^{-1}_{\text{amp}} &= -\frac{25 \mu^2 |r(Q)|}{12 U_{\text{bcc1}} [U_{\text{bcc}} - 4 V_{r}(\pi/4) + \lambda_{nt}]},
\end{align*}
\]

stem from the response of the bcc magnetic structure to the field. That is, they originate from the adjustments of relative phases and amplitudes, respectively, of the helical modes as a result of the term \( -\mathbf{h}_\psi(0) \cdot \mathbf{m} \) in Eq. \( (33) \). The effect of \( \chi_{\text{phase}} \) and \( \chi_{\text{amp}} \), which are necessarily negative, is to increase the susceptibility of the bcc1 state.

The change in the relative amplitudes and phases of the six interfering spirals as a function of the magnetic field may be calculated (see Appendix). For example, the linear response of the amplitudes of bcc1 is

\[
\begin{pmatrix}
\delta |\psi_1| \\
\delta |\psi_2| \\
\delta |\psi_3| \\
\delta |\psi_4| \\
\delta |\psi_5| \\
\delta |\psi_6|
\end{pmatrix} = \frac{5 \mu S}{4 [U_{\text{s}} - V_{r}(\pi/4) + \lambda_{nt}]} \begin{pmatrix}
-m_z \\
m_z \\
-m_x \\
m_x \\
-m_y \\
m_y
\end{pmatrix}.
\]

This response should be observable by neutron scattering, if it is possible to prepare the sample in a single-domain state (i.e. without mixture of the two time-reversal partners). For example, a field in \( \hat{z} \) direction affects \( |\psi_1| \) and \( |\psi_2| \), the amplitudes of the modes propagating orthogonally to \( \hat{z} \) (Fig. 5), which get enhanced and suppressed by the magnetic field, respectively.

The expected effects of external magnetic field on the resistivity of bcc spin crystal are presented elsewhere.

VI. EFFECT OF IMPURITIES: A POSSIBLE ROUTE TO 'PARTIAL ORDER'.

While the helical spin crystal states are expected to show Bragg spots at particular wavevectors, a variety of effects such as thermal or quantum fluctuations or disorder can destroy the long range order while preserving the helical spin crystal structure at shorter scales. Here, we investigate in more detail the effect of non-magnetic disorder on helical spin crystal structures.

Although the experimentally studied hemimagnets are very clean from the electrical resistivity point of view, the helical magnetic structures are sensitive to disorder at a much longer length scale. In addition, the low energy scales required to distort them means that one needs to consider disorder effects. An observation that can immediately be made is that for the physically relevant case of non-magnetic disorder (\( V_{\text{dis}}(\mathbf{r}) \)), the single-spiral state and the spin crystal states respond very differently. By symmetry, the coupling of disorder to the magnetic structure is given by \( F_{\text{dis}} = \langle V_{\text{dis}}(\mathbf{r}) | \mathbf{M}(\mathbf{r}) \rangle^2 \). Hence, single-spiral states which are unique in having a spatially uniform magnitude of magnetization \( | \mathbf{M}(\mathbf{r}) | = \text{constant} \) are unaffected by this coupling; in contrast the spin-crystal states necessarily have a modulated magnitude and hence are affected by non-magnetic disorder. Therefore the neutron scattering signal of the spin-crystal state is expected to have more diffuse scattering than the single mode state. This is consistent with the experimental observation that the high pressure phase has diffuse scattering peaked about \( (110) \) while the low pressure phase has sharper spots, consistent with identifying the two as spin-crystal and single-spiral states respectively.

The effect of disorder on the spin-crystal state is closely related to the problem of the ordering of an XY model in the presence of a random external field. The phase rotation symmetry of the XY model captures the translational invariance of the spin-crystal in the clean state.
Disorder destroys this invariance and behaves like a random field applied to the XY system. Using the insights from the study of that problem in three dimensions, one expects that for weak disorder a Bragg glass will result, where although true long range order is destroyed, power law divergent peaks at the Bragg wavevectors remain, and the elastic constants remain finite. For stronger disorder one expects this algebraic phase to also be destroyed, and recover a short range correlated phase without elasticity. Nevertheless, for the case of the bcc1 and bcc2 crystals, due to time reversal symmetry (T) breaking in these states, the disordered states also spontaneously break time reversal symmetry, and hence a phase transition is expected on cooling despite the absence of long range order. It is difficult to predict which of these two scenarios (Bragg glass or only T breaking) is more appropriate for MnSi. In the latter case one may estimate the spreading of the Bragg spots due to disorder by considering the energetic cost to deform the spin-crystal state in different ways.

Ignoring elastic contributions, there are two distinct types of deformations—one that involves a change in the magnitude of the ordering wavevectors $\delta q_\parallel$ and others that do not change the wavevector magnitude but rotate the structure from its preferred orientation: $\delta q_\perp$. The second is expected to be low in energy because rotations of the structure are locked by the crystal anisotropy term, which is weak. From Eq. (28) we obtain the energy cost to shift the ordering vector by $\delta q_\perp$ along the sphere $|q| = Q$

$$\delta F_\perp = \frac{4\alpha}{3\kappa} \left( \frac{\delta q_\perp}{Q} \right)^2,$$

where $\kappa = 1$ for the single-spiral. For multi-mode spin crystals, the energy cost of rotation is reduced, as explained in Section V A 2. Thus, $\kappa \approx 4$ for the bcc spin crystals. In contrast, deformations that change the magnitude of the ordering wavevectors, must contend with the elastic anisotropy term, and hence pay a higher energy penalty

$$\delta F_\parallel = \frac{1}{2} \Delta \cdot |\psi_0|^2 \cdot \left( \frac{\delta q_\parallel}{Q} \right)^2.$$

Assuming the disorder couples to these deformations equally, we can estimate the ratio of their amplitudes in the limit of weak deformations, by equating Eqs. (41) and (42). It follows

$$\left( \frac{\delta q_\perp}{\delta q_\parallel} \right)^2 = \frac{3\kappa\Delta |\psi_0|^2}{8\alpha}.$$

Using Eqs. (33) and (42), we can relate this ratio to the experimentally known ratio between the critical magnetic fields for, respectively, reorienting and polarizing the single-spiral state

$$\frac{\delta q_\perp}{\delta q_\parallel} \gtrsim \sqrt{\frac{3\kappa}{2} \frac{H_c}{H_{sr}}}.$$

We can now apply these results to the case of MnSi and test the hypothesis that the ‘partial order’ state is in fact a disordered bcc spin crystal. Setting in $\kappa = 4$ and the experimentally measured critical fields for MnSi, $H_c = 0.6$ Tesla and $H_{sr} = 0.1$ Tesla, one obtains $\delta q_\perp/\delta q_\parallel \gtrsim 15$. Neutron scattering experiments do indeed find that the transverse broadening is larger than the longitudinal broadening, but since the latter is resolution limited, this only gives us an upper bound that is consistent with the estimate above: $[\delta q_\perp/\delta q_\parallel]_{\text{expt.}} > 2.3$. Nevertheless, the trend that the width of the spot is greater along the equal magnitude sphere than transverse to it is clearly seen in the experimental data.

Thus, weak non-magnetic disorder of the atomic crystal is expected to destroy magnetic long range order in multi-mode helical spin crystal states and lead to a neutron scattering signal compatible with the observations in the ‘partial order’ phase of MnSi. However in the case of bcc spin crystals, time-reversal symmetry breaking is expected to persist even in the presence of disorder. The scenario of interpreting ‘partial order’ in MnSi as a bcc1 state disordered by impurities thus predicts quasi-static local magnetic moments and implies a finite temperature phase transition on cooling into this phase.

**VII. CONCLUSION**

We have analyzed the magnetic properties of non-centrosymmetric weak ferromagnets subject to DM spin-orbit coupling. This problem falls into the general class of systems where the low energy excitations live on a surface in reciprocal space rather than on discrete points. The addition of DM interactions to a ferromagnetic state produces a large degeneracy of magnetic states characterized by arbitrary superpositions of spin helices of a fixed helicity and fixed wavevector magnitude. This enormous degeneracy is broken by interactions between modes, and the single-spiral state is realized for slowly varying interactions, by virtue of its unique property of having a spatially uniform magnitude of magnetization.

For more general interactions, multi-mode helical spin crystal states are obtained. We show that for the model interactions considered, the phase diagram is largely determined just by considering the interactions between pairs of modes. The phase that is eventually realized may be readily deduced from the range of angles in which this interaction drops below a critical value. In particular, the bcc structure is stabilized by virtue of the fact that its reciprocal lattice, fcc, is a close packed structure. These results may also be relevant in other physical situations where crystallization occurs, such as the Larkin-Ovchinnikov-Fulde-Ferrel instability in spin-imbalanced superconductors, which may potentially be realized in solid state systems, cold atomic gases and dense nuclear matter.

Helical spin crystals typically give rise to complicated real space magnetic structures which we discussed in this
paper. In particular, topological textures like merons and anti-vortices can be seen about special axes in particular realizations, although these are not expected to be stable given the order parameter and spatial dimensionality of the system. We show here that such topological structures exist as a consequence of symmetry, which also dictates the absence of certain higher Bragg reflections, which a naive analysis would predict.

The response of helical spin crystals to crystalline anisotropy and applied magnetic field are considered with a special emphasis on the bcc structures which are contrasted against the response of the single helix state. An unusual transfer of spectral intensity in the presence of an applied magnetic field, which is strongly dependent on the direction of applied field is noted for the bcc structures. This is a consequence of broken time reversal symmetry in the absence of a net magnetization (which is symmetry forbidden). The unusual magnetotransport in such a state, a linear in field magnetoresistance and quadratic Hall effect, has been discussed briefly in Ref. [23] and was elaborated upon in Ref. [24].

Helical spin crystals exhibit Bragg peaks at specific wave-vectors, and hence are not directly consistent with the experimental observation of 'partial order'. The point of view taken in our earlier work [22] is that the short distance and short time properties are captured by the appropriate helical spin crystal structure. Studying the properties of helical spin crystals with long range order is a theoretically well defined task with direct consequences for a proximate disordered phase with similar correlations up to some intermediate scale. The mechanism that leads to the destruction of long range helical spin crystal order is unclear; in Ref. [25], this was assumed to be the coupling to non-magnetic disorder. Then, as elaborated in this paper, beginning with a bcc helical spin crystal a neutron scattering signature consistent with that of 'partial-order' may be obtained. However, within the simplest version of this scenario, one also expects a finite temperature phase transition where time reversal symmetry breaking develops, and static magnetic order which may be seen in nuclear magnetic resonance or muon spin rotation experiments. Other mechanism for the destruction of long range order of the bcc spin crystal state, such as thermal or quantum fluctuations may also be considered, but are left for future work.

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APPENDIX: LINEAR RESPONSE

Let the free energy $F$ depend on real internal variables $\mathbf{x} = (x_1, \ldots, x_n)$ (related to various order parameters) and on $\mathbf{m} = (m_1, m_2, m_3)$, which couples linearly to the external field $\mathbf{H}$. The internal variables are chosen such that for $\mathbf{m} = 0$, the minimum energy shall be at $\mathbf{x} = 0$. Expanding $F$ to second order in $\mathbf{x}$ and $\mathbf{m}$ yields

$$ F = \frac{1}{2} \mathbf{x}^T \mathbf{A} \mathbf{x} + \mathbf{m}^T \mathbf{B} \mathbf{x} + \frac{1}{2} \mathbf{m}^T \mathbf{C} \mathbf{m}, $$

where $\mathbf{A}$, $\mathbf{B}$ and $\mathbf{C}$ are matrices, $\mathbf{A} = \mathbf{A}^T$, $\mathbf{C} = \mathbf{C}^T$. All eigenvalues of $\mathbf{A}$ are positive. Eq. (A.1) can be written as

$$ F = \frac{1}{2} (\mathbf{x} - \mathbf{x}_m)^T \mathbf{A} (\mathbf{x} - \mathbf{x}_m) + \frac{1}{2} \mathbf{m}^T \chi^{-1} \mathbf{m}. $$

where $\mathbf{x}_m = -\mathbf{A}^{-1} \mathbf{B}^T \mathbf{m}$ and $\chi^{-1} = C - \mathbf{B} \mathbf{A}^{-1} \mathbf{B}^T$. It follows that under the influence of an external field $\mathbf{H}$, the equilibrium internal variables get shifted to $\mathbf{x} = \mathbf{x}_m$ and the linear response is given by $\mathbf{m} = \chi \mathbf{H}$. Thus, there are two contributions to the inverse susceptibility: $\chi_{\text{bare}}^{-1} = C$ and $\chi_{\text{ext}}^{-1} = -\mathbf{B} \mathbf{A}^{-1} \mathbf{B}^T$. The latter comes from the internal response of the $\mathbf{x}$-variables to the magnetic field.

These general results are applied in Section [VB2] where the internal variables $x_1, \ldots, x_9$ are the deviations from their equilibrium value at $\mathbf{m} = 0$ of six amplitudes $|\psi_j|$ and three phases (holding the other three phases fixed). In this case, $\chi_x$ leads to both the phase and amplitude related terms in Eq. (59).
In non-magnetic crystals, the coupling term is cubic and the shift to directions $\mathbf{F}$ would still beat the neighboring phases in the fcc atomic lattice, since crystal anisotropy is neglected. To obtain the phase diagram of Figs. 2 and 3, we have used the approximation $1 + x^2 + (1 - x)^2 \approx 2 + (4\sqrt{2} - 2)x^2$ for $x = \cos \theta$ in the evaluation of $V_\phi(\theta)$. The error is smaller than 0.7%.

The shift to directions $e^{i\phi}$ is very small with $\phi \sim 0.02$ and even without the deformation, fcc would still beat the neighboring phases in the fcc"-region.

In non-magnetic crystals, the coupling term is cubic and therefore higher harmonics are generated at all sums of two momenta $\mathbf{k}_1 \pm \mathbf{k}_2$. In magnetic structures, higher-harmonics wavevectors are restricted to sums of three ordering momenta. In this regime, all 26 high-symmetry points are exchanged. See for example B. Binz, D. Baeriswyl and B. Douçot, Eur. Phys. J. B 25, 69 (2002).

Without having a rigorous proof, we suspect that in the context of Section III.C “very big” just means “bigger that $U_a$”. This bound has proven sufficient in all cases that we have tested.