Helimagnons in a chiral ground state of the pyrochlore antiferromagnets

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Abstract – The Goldstone mode in a helical magnetic phase, also known as the helimagnon, is a propagating mode with a highly anisotropic dispersion relation. Here we study theoretically the magnetic excitations in a complex chiral ground state of pyrochlore antiferromagnets such as spinel CdCr₂O₄ and itinerant magnet YMn₂. We show that the effective theory of the soft modes in the helical state possesses a symmetry similar to that of smectic liquid crystals. An overall agreement is obtained between experiments and our dynamics simulations with realistic model parameters. By exactly diagonalizing the linearized Landau-Lifshitz equation in various commensurate limits of the spiral order, we find a low-energy dispersion relation characteristic of the helimagnons. Our calculation thus reveals the first example of helimagnon excitations in geometrically frustrated spin systems.

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Introduction. – The rich phenomenology associated with helical spin ordering has attracted considerable attention recently. Magnetic spirals have been shown to play a crucial role in inducing a spontaneous electric polarization in multiferroic materials [1]. The soft magnetic fluctuations in a helical spin-density-wave state of the itinerant magnet MnSi also holds the key to its non-Fermi liquid behavior [2]. Contrary to most well-known long-range spin orders such as ferromagnetism and antiferromagnetism, spin-wave excitations in a helical magnetic order are described by a theory similar to the elasticity of smectic liquid crystals [3,4]. In particular, despite the broken symmetry of the helical order is described by a O(3) order parameter, there only exists a single Goldstone mode similar to the smectic-like phonon mode which emerges on scales larger than the helical pitch [5,6]. These low-energy gapless excitations, dubbed helimagnons in ref. [5], exhibit a highly anisotropic dispersion relation.

The recent discovery of a novel chiral spin structure in spinel CdCr₂O₄ has generated much interest both theoretically and experimentally [7–17]. A very similar coplanar helical order has also been reported in the weak itinerant antiferromagnet YMn₂ [15,16]. The magnetic ions in these compounds form a three-dimensional network of corner-sharing tetrahedra (fig. 1), known as the pyrochlore lattice. The frustrated spin interactions due to the special connections of nearest-neighbor bonds in this lattice gives rise to an extensively degenerate ground state at the classical level [17,18]. As a result, magnetic ordering at low temperatures is determined by the dominant residual perturbations in the system.

The helical magnetic order in CdCr₂O₄ is stabilized by a combined effect of magnetoelastic coupling and relativistic spin-orbit interactions. Despite a rather high Curie-Weiss temperature |Θ_{CW}| ≈ 70 K, the spiral magnetic order sets in only at T_N = 7.8 K, indicating a high degree of frustration. The magnetic transition at T_N is accompanied by a structural distortion which lowers the crystal symmetry from cubic to tetragonal [8]. Recent infrared absorption measurements indicated a broken lattice inversion symmetry below T_N [12], consistent with the scenario predicted in ref. [7] that the magnetic frustration is relieved by the softening of a q = 0 optical phonon with odd parity. A long-range Néel order with ordering wave vector Q_0 = 2π(0, 0, 1) is stabilized by the lattice distortion. Moreover, the lack of inversion symmetry endows the crystal structure with a chirality. The collinear state is transformed into a magnetic spiral as the chirality is transferred to the spins through spin-orbit interaction. The shifted ordering vector Q = 2π(0, δ, 1) is consistent with the observed magnetic Bragg peaks.

In this paper we undertake a first theoretical calculation of the magnon spectrum in the helical ground state of
pyrochlore antiferromagnet. In particular, our microscopic calculation goes beyond the phenomenological approaches adopted in most studies of magnetic excitations in helimagnets. Based on a well-established microscopic model for the helical order, we exactly diagonalize the linearized Landau-Lifshitz-Gilbert (LLG) equation with a large unit cell; the size of the unit cell determines the ordering wave vector of the magnetic spiral in the commensurate limit. By comparing results obtained from different helical pitches, we find a spin-wave spectrum that is characteristic of helimagnons. Our work thus provides the first solid example of helimagnons in frustrated pyrochlore antiferromagnets.

We also performed dynamical LLG simulations on large finite lattices and found an overall agreement with the experimental data on CdCr$_2$O$_4$ at the high-energy regime. Along with the structural and Bragg scattering data, this justifies the validity of our microscopic Hamiltonian. However, the nature of the low-energy magnetic excitations remains unclear due to the lack of better experimental data. In fact, the observation that the incommensurate spiral might originate from the anisotropic DM interactions prompted the authors in ref. [8] to suggest a gapped magnon excitation ($\Delta \sim 0.6$ meV) in CdCr$_2$O$_4$. Based on analytical argument and exact numerical diagonalization, here we propose a different picture in which the low-energy elementary excitations of these chiral pyrochlore systems are emergent gapless helimagnons.

Model Hamiltonian. – Our starting point is a classical Heisenberg model on the pyrochlore lattice described by the Hamiltonian

$$\mathcal{H} = \sum_{(ij)} (J + K_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{(ij)} D_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + J_2 \sum_{(i)} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{(iiij)} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here $J$, $J_2$ and $J_3$ are nearest-neighbor (NN), second- and third-neighbor exchange interactions, respectively, $K_{ij}$ is the exchange anisotropy, and $D_{ij}$ is the Dzyaloshinskii-Moriya (DM) interaction [19,20] between NN spins. The isotropic NN exchange is the dominant term with an antiferromagnetic sign $J > 0$. It can be recast into $(J/2) \sum_r |\mathbf{M}(\mathbf{r})|^2$ up to a constant, where $\mathbf{M}(\mathbf{r})$ denotes the vector sum of spins on a tetrahedron at $\mathbf{r}$. Minimization of this term requires the vanishing of total magnetic moment on every tetrahedron but leaves an extensively degenerate manifold [17,18].

The anisotropy $K_{ij}$ in the nearest-neighbor exchange coupling constants is due to the combined effect of the lattice distortion and magnetoelastic coupling. In CdCr$_2$O$_4$, the lattice symmetry is lowered from cubic ($Fd\bar{3}m$) to tetragonal ($I4_1/amd$) structure [21]. The fact that the lattice translational symmetry is preserved by the tetragonal distortion indicates that the structural transition at $T_N$ is triggered by the softening $q = 0$ optical phonons. In particular, a softened phonon with odd parity breaks the inversion symmetry and stabilizes a collinear spin order with wave vector $Q_0 = 2\pi(0,0,1)$ [22,23].

The antisymmetric exchange, or DM interaction, is allowed on the pyrochlore lattice, where the bonds are not centrosymmetric as required by the so-called Moriya rules [20]. Moreover, the high symmetry of the pyrochlore lattice completely determines the orientations of the DM vectors up to a multiplicative factor. The explicit form of the vectors $\mathbf{D}_{ij}$ can be found in refs. [24–26].

Finally, we also include second- and third-neighbor spin couplings $J_2$ and $J_3$ in the model Hamiltonian. These further-neighbor exchanges are important for the modeling of various Cr spinels, as indicated by the $ab$ initio calculations [27]. In the limit dominated by a large NN exchange, the ground-state properties actually depend only on the difference between $J_2$ and $J_3$. The relative shift in energy for any pair of ground states due to a small $J_3$ is identical to the effect of a $J_2$ of the same magnitude with an opposite sign [28].

Coplanar spirals. – To understand how the coplanar helical order is stabilized in the large-$J$ limit, we briefly review the continuum theory based on a gradient expansion of the order parameters [7]. We also make a crucial generalization of the continuum theory by uncovering a larger class of continuously degenerate helical states. An important consequence of this additional degeneracy is the existence of gapless helimagnons. The antiferromagnetic order on the pyrochlore lattice is characterized by three staggered order parameters $L_i$ ($i = 1, 2, 3$). For example, $L_1 = (S_0 + S_1 - S_2 - S_3)/4$ describes the staggered spins on two opposite [011] and [011] bonds, here the subscripts 0, 1, 2, 3 denote the four sublattices of the pyrochlore lattice (see fig. 1). The order parameter for the collinear order is $L_1(\mathbf{r}) = \mathbf{n}_1 \exp(iQ_0 \cdot \mathbf{r})$, and $L_2 = L_3 = 0$, where $Q_0 = 2\pi(0,0,1)$, and $\mathbf{n}_1$ is an arbitrary unit vector. Note that the phase factor $e^{iQ_0 \cdot \mathbf{r}} = \pm 1$ changes sign between consecutive layers of tetrahedra along the $z$-direction (cf. the two layers of green tetrahedra in fig. 1).

Assuming a slow variation of staggered magnetizations in the helical order, we parameterize the order parameters as: $L_i(\mathbf{r}) = e^{iQ_0 \cdot \mathbf{r}} \phi_3(\mathbf{r}) \hat{n}_i(\mathbf{r})$, where $\mathbf{n}_i$ are three orthogonal unit vectors, $\phi_1 \approx 1 - \frac{1}{2}(\phi_2^2 + \phi_3^2)$, and $\phi_2, \phi_3 \ll \phi_1$ [7]. In the $J \to \infty$ limit, vanishing of total magnetization on tetrahedra requires $\mathbf{n}_3 \parallel \partial_y \mathbf{n}_1$, and the $\phi$ fields can be analytically expressed in terms of the director fields $\mathbf{n}_i$. The effective energy functional in the gradient approximation can be solely expressed in terms of the director fields $\mathbf{n}_1$ and $\mathbf{n}_2 \parallel \partial_y \mathbf{n}_1 \times \mathbf{n}_1$ [7]:

$$\mathcal{F} = D \int d\mathbf{r} \left[ \hat{n}_1 \cdot (\hat{\mathbf{x}} \times \partial_x \hat{n}_1 + \hat{\mathbf{y}} \times \partial_y \hat{n}_1 - \hat{\mathbf{z}} \times \partial_z \hat{n}_1) \right] + K_u \int d\mathbf{r} \left[ (\partial_x \hat{n}_1)^2 + (\partial_y \hat{n}_1)^2 + 2(\partial_z \hat{n}_1)^2 - (\hat{n}_2 \cdot \partial_z \hat{n}_1)^2 \right]. \quad (2)$$

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The magnetic order of the DM terms suggests spiral solutions with \( \hat{\theta}(\mathbf{r}) = \mathbf{Q} \cdot \mathbf{r} + \varphi_0 + \hat{\psi}(\mathbf{Q} \cdot \mathbf{r} + \varphi_0) \), where \( \mu = 0, 1, 2, 3 \) is the sublattice index, \( \varphi_0 = \psi_1 = \pi\delta + \psi, \quad \varphi_2 = \pi + \psi, \quad \) and \( \varphi_3 = \pi(1 + \delta) + \psi \), with \( \psi \) an arbitrary constant related to the \( U(1) \) symmetry of the coplanar spiral, i.e., \( \theta(\mathbf{r}) = \mathbf{Q} \cdot \mathbf{r} + \psi \).

**Helimagnons in the continuum limit.** – We next consider a phenomenological approach for the low-energy magnetic excitations in the helical ground state. For a spiral with a fixed axis, the obvious soft modes are the \( U(1) \) phase fluctuations \( \psi \) associated with the rotation angle \( \theta(\mathbf{r}) \). A naive guess of the effective energy functional of the soft mode would be \( \mathcal{F} \propto \int d\mathbf{r} |\nabla \psi|^2 \), similar to that of planar magnets. A linear phase variation along the direction perpendicular to the helical axis, \( \psi = \alpha \zeta \), costs an energy \( \propto K_u \alpha^2 \) according to this functional. However, such a phase variation corresponds to a rotation of the helical axis in the \( xy \) plane and does not cost energy on account of the \( O(2) \) invariance discussed above. Consequently, there cannot be any \( (\partial_\theta \psi)^2 \) term in the effective energy density. The correct energy functional for the soft modes is [3–5]

\[
\mathcal{F} = \frac{1}{2} \int d\mathbf{r} \left[ c_3 (\partial_\theta \psi)^2 + c_{1\perp} (\partial_\psi \psi)^2 + \frac{c_2}{Q^2} (\partial_\theta \psi)^2 + rm^2 \right],
\]

where \( c_3, c_{1\perp} \sim K_u, c_2 \sim J_3 \) are elastic constants, and the true soft mode is a generalized phase accompanied by a rotation of the director field. We have also included a zero wave vector ferromagnetic mode \( m \) which is soft due to spin conservation. It is worth noting that the first term in eq. (5) is absent in the conventional helical order discussed in ref. [5]. As discussed above, it originates from a rather large \( J_3 \) that gives a penalty to variations along the \( z \)-direction. Schematically the coarse-grained sublattice magnetizations are

\[
\mathbf{S}_\mu(\mathbf{r}) \sim \mathbf{S}_0(\mathbf{r}) + m(\mathbf{r}) \hat{\mathbf{y}} + \psi(\mathbf{r}) \left[ \cos(\mathbf{Q} \cdot \mathbf{r} + \varphi_\mu) \hat{\mathbf{x}} - \sin(\mathbf{Q} \cdot \mathbf{r} + \varphi_\mu) \hat{\mathbf{z}} \right],
\]

The spin dynamics is governed by a generalized Landau-Lifshitz equation [4]:

\[
\frac{\partial \mathbf{S}_\mu(\mathbf{r})}{\partial t} = -\gamma \mathbf{S}_\mu(\mathbf{r}) \times \frac{\delta \mathcal{F}}{\delta \mathbf{S}_\mu(\mathbf{r})},
\]
energy is measured in units of $JS$. The diagonalization of a large unit cell reveals the results are fitted to the phenomenological dispersion $\varepsilon_k = \sqrt{\alpha k^2 + \beta k^4}$. The magnon dispersion in this commensurate limit, a highly accurate description of the ground state is required. To this end, we start from the coplanar spiral solution, eq. (4), which is exact in the infinite-$J$ limit, and numerically integrate the LLG equation [29]

$$\frac{\partial \mathbf{S}_i}{\partial t} = \mathbf{S}_i \times \frac{\partial \mathbf{H}_i}{\partial \mathbf{S}_i} + \frac{\alpha_G}{\gamma S} \mathbf{S}_i \times \frac{\partial \mathbf{S}_i}{\partial t},$$  \hspace{1cm} (9)

with a large damping constant $\alpha_G$ to relax the spins to its true ground state.

We then introduce small deviations to spins in the obtained ground state: $\mathbf{S}_i = \mathbf{S}_i^0 + \mathbf{\sigma}_i$, where $\mathbf{\sigma}_i \perp \mathbf{S}_i^0$ denotes transverse deviations. Note that because of this constraint, there are only two degrees of freedom associated with $\mathbf{\sigma}_i$ at each site. The LLG equation is linearized with respect to the small deviations $\mathbf{\sigma}_i$:

$$\frac{\partial \mathbf{\sigma}_i}{\partial t} = \mathbf{S}_i^0 \times \sum_j (J_{ij} \mathbf{\sigma}_j - \mathbf{D}_{ij} \times \mathbf{\sigma}_j) - \mathbf{H}_i^0 \times \mathbf{\sigma}_i,$$  \hspace{1cm} (10)

where $J_{ij}$ includes $J$, $K_{ij}$ and further-neighbor exchanges, $\mathbf{H}_i^0 = \sum_j (J_{ij} \mathbf{S}_j^0 - \mathbf{D}_{ij} \times \mathbf{S}_j^0)$ is the equilibrium local exchange field, and we have set the damping constant $\alpha_G$ to zero. The eigenmodes of the LLG equation has the form $\mathbf{\sigma}_i = \mathbf{\sigma}_i \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t)$, where the site index $i = (r, s)$, where $r$ denotes the position of the extended unit cell containing $\Lambda$ cubes and $s$ labels spins within this block. Note that the above eigenmode $\{\mathbf{\sigma}_s\}$ has a momentum $\mathbf{Q} + \mathbf{k}$ due to the underlying structure of the supercell. Arranging $\mathbf{\sigma}_s = (\mathbf{\sigma}_s^x, \mathbf{\sigma}_s^y)$ into a column vector $\mathbf{\sigma}_s = (\mathbf{\sigma}_s^x, \mathbf{\sigma}_s^y, \cdots, \mathbf{\sigma}_s^x, \mathbf{\sigma}_s^y)$ of dimension $n = 2 \times 4\Lambda \times 4$, the
linearized LLG equation is recast into an eigenvalue problem $T({\mathbf k}) \cdot \mathbf u = -i\omega_{k\alpha} \mathbf u$. Numerical diagonalization of the $n \times n$ matrix $T({\mathbf k})$ gives the dispersion of the low-energy magnons in the vicinity of ordering wave vector $Q$.

The resulting magnon dispersions along the three principal directions are shown in fig. 2(a)–(c). The number of spins in the unit cell is defined by the value of the spiral pitch $\delta$. We have considered $\delta = 0.2, 0.1, 0.05$, which gives $N_s = 80, 160, 320$, respectively. The numerical spectra are fitted to a phenomenological dispersion: $\varepsilon_k = \sqrt{\alpha k^2 + \beta k^4}$. The dispersion parameters $\alpha$ and $\beta$ obtained from the fitting are shown in fig. 2(d)–(f) as a function of the helical wave number $\delta$. First, we note that the dispersion along the $y$ (helical direction) and $z$ (staggering direction) axes can be well approximated by a linear relation, $\varepsilon_k \sim vk$, with $v = \sqrt{\beta}$ weakly dependent on $\delta$, see fig. 2(d). The large spin-wave velocity along the $z$-axis can be attributed to a large $J_3$, consistent with the analysis of gradient expansion [7].

On the other hand, the spectrum along the $x$-axis exhibits a predominant quadratic behavior at small $k$, similar to that of a ferromagnet. Although the continuum theory predicts an exact quadratic dispersion along the $x$-direction, eq. (8), careful fitting shows a small linear component $\alpha$, which can be attributed to the discrete cubic symmetry of the underlying lattice model. Dimensional analysis indicates that $\alpha$ should scale as $Q^2$ [5], while the coefficient $\beta \sim 1/Q^2$ according to the helimagnon dispersion, eq. (8). These scaling relations are indeed confirmed by our exact diagonalization as demonstrated in figs. 2(e) and (f).

**Magnons in CdCr$_2$O$_4$.** — Although the exact diagonalization method provides a rather accurate magnon spectrum, the complicated band-folding due to a large unit cell makes it difficult to compare the results with experiment. Instead, here we employ a less accurate but more direct approach based on simulating the linearized LLG equation (10) in a large finite lattice [30]. The simulation is initiated by a short pulse localized at $r_1 = 0$. Since the system is driven by a white source with flat spectrum, we expect magnetic excitations of various energy and momentum are generated in our simulations. The spin-wave spectrum is then obtained via the Fourier transform of the simulation data. Figure 3(a) shows the numerical spin-wave spectrum in a collinear $Q_0 = 2\pi(0,0,1)$ Néel order, which is in perfect agreement with that from analytical calculation.

We next apply the method to obtain the magnon spectrum of the spiral order with $Q = 2\pi(0,\delta, 1)$, and compare the results with the experimental result measured using inelastic neutron scattering [8]. In our calculation, we use the measured helical pitch $\delta \approx 0.1$ to fix the ratio of the DM interaction to the exchange anisotropy $K_{su}$. The absolute value of $K_{su}$ is estimated by the magnon energies at the zone center. As for further-neighbor exchanges, *ab initio* calculations find a negligible $J_2$ and a large $J_3$ with antiferromagnetic sign [7,27]. Using the following set of parameters: $J = 1.35$ meV, $K_u = 0.21$ meV, $D = 0.14$ meV, and $J_3 = 0.28$ meV, we obtained good agreement with the experimentally measured spectrum in the high-energy regime ($\omega \gtrsim 1$ meV), as shown in fig. 3(b).

**Concluding discussions.** — We have studied the spin-wave excitations in a complex helical magnetic order on the pyrochlore lattice. Our exact diagonalization calculation of the low-energy dispersion in the commensurate limit of the long-period spiral clearly demonstrates the existence of helimagnon excitations in the frustrated pyrochlore antiferromagnet. This novel magnetic excitation can be observed in the chiral ground state of spinel CdCr$_2$O$_4$ and the itinerant magnet YMn$_2$. We have also performed dynamical LLG simulations with parameters inferred from structural data and *ab initio* calculations of CdCr$_2$O$_4$. The calculated spectra in the higher-energy regime agrees well with the experimental data. As our calculation is based on a spin Hamiltonian that is consistent with the structural distortion and magnetic order, the agreement on the dynamics part further corroborates the microscopic model in which the magnetic spiral arises from the interplay between distortion-induced exchange anisotropy and DM interactions.
Although helimagnons in unfrustrated magnets such as MnSi [31] and Ba$_2$CuGeO$_7$ [32] have been extensively investigated experimentally, there has been very few such studies in geometrically frustrated systems. The spiral magnetic order in YMn$_2$ and CdCr$_2$O$_4$ provides a unique opportunity to study these novel excitations in the frustrated pyrochlore lattice. However, the current experimental data on CdCr$_2$O$_4$ cannot resolve the issue of whether the low-energy magnon spectrum is gapless [8]. In fact, it was proposed that a spectral gap is opened at the incommensurate ordering wave vector due to the anisotropic DM interaction [8]. Such an DM-induced spectral gap is indeed reported in the magnetic spiral state of Cs$_2$CuCl$_4$ [33]. On the other hand, a prerequisite for helimagnons is the emergence of a continuous rotational symmetry for the helical axis, which we clearly demonstrate by generalizing the continuum theory for spiral orders in pyrochlore. Moreover, our numerical calculation based on microscopic spin Hamiltonian unambiguously demonstrates the existence of gapless helimagnons in the helical magnetic order on pyrochlore lattice. In addition to inelastic neutron scattering, such gapless spin excitations could be detected by other experimental probes such as Raman scattering. We hope that our work could stimulate further experimental investigations on helimagnons in geometrically frustrated magnets.

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