Normal Modes of a Spin Cycloid or Helix

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Although spin cycloids and helices are quite common, remarkably little is known about the normal modes of a spin cycloid or helix with finite length on a discrete lattice. Based on simple one-dimensional lattice models, we numerically evaluate the normal modes of a spin cycloid or helix produced by either Dzyaloshinskii-Moriya (DM) or competing exchange (CE) interactions. The normal modes depend on the type of interaction and on whether the nearest-neighbor exchange is antiferromagnetic (AF) or ferromagnetic (FM). In the DM case, there is only a single Goldstone mode; in the CE case, there are three. For FM exchange, the spin oscillations produced by non-Goldstone modes contain a mixture of tangential and transverse components. For the DM case, we compare our numerical results with analytic results in the continuum limit.

Spin cycloids and helices are ubiquitous in the field of magnetism. They appear in most multiferroics and in many other materials like rare earth intermetallics, and even superconductors. Cycloids with spins in the same plane as the ordering wavevector and helices (also known as spirals or proper screws) with spins perpendicular to partly satisfy neighboring exchange interactions and some competing energy like Dzyaloshinskii-Moriya (DM) or competing exchange (CE) interactions. Cycloids and helices have attracted great attention not only for their response to competing energies but also for applications based on their control with electric or magnetic fields.

The excitation spectrum of a cycloid or helix provides a dynamical “fingerprint” of the microscopic interactions and anisotropies responsible for its formation. Yet remarkably little is known about the spectrum of spin-wave (SW) modes for a cycloid or helix, especially one with a finite period on a discrete lattice. This paper studies simple one-dimensional lattice Hamiltonians for DM and CE cycloids or helices with either antiferromagnetic (AF) or ferromagnetic (FM) nearest-neighbor exchange. Our work seeks to answer several questions. Are the mode spectra and SW amplitudes different for the four cases (AF/DM, FM/DM, AF/CE, and FM/CE) considered? Which SW modes are observable by inelastic neutron scattering (INS) and which by optical spectroscopy? How is the continuum limit approached in these four cases?

A simple one-dimensional lattice Hamiltonian for a DM cycloid is

\[ H_{\text{DM}} = -J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} - D \sum_i \mathbf{y} \cdot (\mathbf{S}_{i+1} \times \mathbf{S}_i), \]

where neighboring sites and are separated by lattice constant along the z axis. The DM interaction along \( \mathbf{y} \) constrains the spins to lie in the xz plane. A helix with spins in the yz plane would be produced by a DM interaction along \( \mathbf{x} \). A one-dimensional lattice Hamiltonian for a CE cycloid or helix is

\[ H_{\text{CE}} = -J_1 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} - J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2}, \]

where \( J_2 \) is the next-nearest-neighbor exchange coupling between sites \( i \) and \( i + 2 \). For either sign of \( J_1 \), the DM interaction or AF exchange \( J_2 < 0 \) frustrates simple AF or FM order to produce a cycloid or helix.

Using classical spins, it is easy to show that a cycloid or helix of period \( M \) is produced by the DM interaction \( D \) in \( H_{\text{DM}} \) when

\[ D = J_1 \tan(2\pi \delta). \]

For AF \( J_1 < 0 \), \( \delta = p/2M \) where \( p \) is the number of \( 2\pi \) rotations (not counting the AF oscillations) in distance \( 2Ma \) and the ordering wavevector is \( \mathbf{Q} = (2\pi/a)(0.5 + \delta) \mathbf{x} \). The cycloid or helix is periodic in distance \( Ma \) if \( p \) is odd (even) and \( M \) is odd (even). For FM \( J_1 > 0 \), \( \delta = p/M \) where \( p \) is the number of \( 2\pi \) rotations in distance \( Ma \) and \( \mathbf{Q} = (2\pi/a) \delta \mathbf{x} \).

A cycloid or helix with period \( M \) is also produced by the next-nearest-neighbor exchange \( J_2 \) in \( H_{\text{CE}} \) when

\[ J_2 = -\frac{|J_1|}{4} \sec(2\pi \delta). \]

For \( J_1 < 0 \), \( \delta = p/2M \) and for \( J_1 > 0 \), \( \delta = p/M \) as above. The ordering wavevectors \( \mathbf{Q} \) are the same as for DM interactions. Only collinear AF or FM order is possible when \( |J_2| < |J_1|/4 \).
For specificity, we shall consider a cycloid with spins in the \(xz\) plane containing \(Q\). While this condition is guaranteed by the DM interaction along \(y\) in \(H_{DM}\), the spin plane is not determined by the CE interactions in \(H_{CE}\). The spins can then be constrained to the \(xz\) plane by adding a small (infinitesimal) easy-plane anisotropy \(K\) along \(y\). On the other hand, a helix with spins in the \(yz\) plane perpendicular to \(Q\) would be created by a DM interaction or easy-plane anisotropy along \(x\). All the numerical results below also hold for a spin helix with the appropriate rotation of the spin reference frame. Our new continuum-limit results for the FM/DM case are also valid for both cycloids and helices.

A classical cycloid with spins in the \(xz\) plane is given by

\[
S_r = S(\sin(Qr), 0, \cos(Qr)),
\]

where \(S\) is the spin and \(R = ra\) is the position of site \(r\). Since \(S_{1+M} = S_r\), the magnetic unit cell contains \(M\) spins with \(1 \leq r \leq M\). For AF or FM interactions, cycloids with \(\delta = 1/10\) are sketched in Fig.1. Notice that the tangent

\[
t_r = (\cos(2\pi r), 0, -\sin(2\pi r))
\]

does not alternate sign with the AF modulation in Fig.1(a).

We solve for the SW modes of these two models by performing a 1/S expansion about the classical limit and then diagonalizing a \(2M \times 2M\) equation-of-motion matrix. We always take \(S = 5/2\). The predicted INS intensities \(S(q, \omega)\) are plotted in Fig.2 for all four cases with \(\delta = 1/10\). Clear signatures are exhibited by the spectra produced by DM and CE cycloids and by AF and FM interactions. For CE cycloids, the SW modes always fall within the first structural Brillouin zone between \(H = 0\) and 1, as can be seen from the SW frequencies plotted as dashed curves. For DM cycloids, the SW branches extend beyond the first Brillouin zone. For example, three SW branches arise from \(H = \pm 1/10\) and \(H = 0\) for the DM/FM case in Fig.2(b).

The normal modes evaluated at wavevector \(H = m\delta\) (integer \(m\)) may appear in optical measurements since zone folding maps those wavevectors onto \(q = 0\). To understand the different mode spectra in our four cases, we plot the SW dispersions versus wavevector \(q\) in Fig.3. Any normal mode crossed by two SW branches is doubly degenerate.
For AF interactions, we obtain two classes of modes labeled $\Phi_{\pm n}$ and $\Psi_{\pm n}$ (doubly degenerate for $n > 0$). In the AF/DM case, the single Goldstone mode $\Phi_0$ corresponds to a uniform spin rotation about $y$. The mode spectrum in Fig.3(a) is close to the spectrum predicted by de Sousa and Moore\cite{10} in the continuum limit with $h\omega_i(\Phi_{\pm n}) = 2S|D|n$ and $h\omega_i(\Psi_{\pm n}) = 2S|D|\sqrt{1+n^2}$. In the AF/CE case, the three Goldstone modes are $\Phi_0$ and $\Psi_{\pm 1}$. Their three-fold splitting away from $H = 0$ is plotted in the inset to Fig.3(c) and can also be seen in Fig.2(c). Goldstone modes $\Psi_{\pm 1}$ are associated with rotations out of the $xz$ plane, assuming that the easy-plane anisotropy $K$ vanishes. Of course, this rotation costs energy in the AF/DM case.

For FM interactions, we obtain only one class of modes labeled $\Theta_{\pm n}$ (doubly degenerate for $n > 0$). In the FM/DM case, the single Goldstone mode $\Theta_0$ again corresponds to a uniform spin rotation about $y$. In the continuum limit of the FM/DM case, we find that $h\omega_i(\Theta_{\pm n}) = S(D^2/J)n\sqrt{1+n^2}$. In the FM/CE case, the three Goldstone modes are $\Theta_0$ and $\Theta_{\pm 1}$ with the three-fold splitting plotted in the inset to Fig.3(d). As in the AF/CE, the extra Goldstone modes are associated with rotations of the spin state out of the $xz$ plane. In all four cases, the Goldstone modes are “massless,” which means that the dispersion is linear near $H = 0$.

The spin oscillation $\Delta S_r^{(n)}(q, t)$ at site $r$ produced by SW mode $n$ with wavevector $q$ is generally given by\cite{11,12,14,17}

$$\Delta S_r^{(n)}(q, t) = 2\sqrt{N} \text{Re} \left\{ e^{-i\omega_n t - i\xi_n} \delta S_r(n, q) \right\}, \quad (7)$$

$$\delta S_r(n, q) = (0|S_r|n, q), \quad (8)$$

where $|0\rangle$ is the ground state, $|n, q\rangle$ is an excited state containing a single SW with energy $\omega_n(q)$ at wavevector $q$, and $S_r$ is the quantum spin operator at site $r$. Like the SW frequency $\omega_n(q)$, the SW amplitude $\delta S_r(n, q)$ is the same at wavevectors $q = 0$ and $q = Q$.

A close examination of the SW amplitudes for DM and CE cycloids with AF or FM interactions reveals that

$$\delta S_r(\Phi_{\pm n}) = \left\{ \xi_1^{(n)} t_r - i\xi_2^{(n)} y \right\} e^{\pm 2\pi in\delta r} \quad (AF), \quad (9)$$

$$\delta S_r(\Psi_{\pm n}) = \left\{ \rho_1^{(n)} y - i\rho_2^{(n)} t_r \right\} e^{\pm 2\pi in\delta r} \quad (AF), \quad (10)$$

$$\delta S_r(\Theta_{\pm n}) = \left\{ \gamma_1^{(n)} t_r - i\gamma_2^{(n)} y \right\} e^{\pm 2\pi in\delta r} \quad (FM), \quad (11)$$

for either $q = 0$ or $q = Q$. In each case, the real and positive coefficients are the same for the degenerate $\pm n$ modes and are normalized by $\xi_1^{(n)2} + \xi_2^{(n)2} = 1$, $\rho_1^{(n)2} + \rho_2^{(n)2} = 1$, and $\gamma_1^{(n)2} + \gamma_2^{(n)2} = 1$. The complex factors in the brackets imply that the tangential and transverse spin oscillations are out of phase. These factors switch sign when the helicity of the cycloid is reversed.

The SW amplitudes for the Goldstone modes are purely transverse (out of the cycloidal plane) or tangential (in the cycloidal plane). For AF interactions, $\delta S_r(\Phi_0) = t_r(-1)^r$ in both the DM and CE cases. In the AF/CE case, $\delta S_r(\Psi_{\pm 1}) = \exp(\pm 2\pi i\delta r)(-1)^r y$. For FM interactions, $\delta S_r(\Theta_0) = t_r$ in both the DM and CE cases. In the FM/CE case, $\delta S_r(\Theta_{\pm 1}) = \exp(\pm 2\pi i\delta r)y$. Although not a Goldstone mode, the SW amplitude $\delta S_r(\Psi_0) = (-1)^r y$ of the AF/CE mode $\Psi_0$ is purely transverse but out of phase with the cycloid.

While even and odd $M$ were handled differently for AF interactions, physical results only depend on the wavevector parameter $\delta$. The amplitude coefficients are plotted versus $\delta$ in Fig.4. In either the AF/DM or AF/CE case, $\xi_1^{(n)}$ and $\rho_1^{(n)}$ approach 1 for all $n$ in the continuum limit $\delta \to 0$. All the SW amplitudes become purely tangential or transverse as $\delta \to 0$ but coefficients with larger $n$ converge much more slowly than for smaller $n$. Figures 4(a) and (c) plot $\xi_1^{(n)}$ and $\rho_1^{(n)}$ as closed and open circles, respectively. For larger $n$, $\xi_1^{(n)}$ and $\rho_1^{(n)}$ are quite close, but deviations can be seen for smaller $n$ away from $\delta = 0$.

For FM interactions, the behavior of the coefficients is more complex. While $\gamma_1^{(n)} \to 1/\sqrt{2}$ as $\delta \to 0$ and $n \to \infty$ in both the FM/DM and FM/CE cases, $\gamma_1^{(n)}$ have higher (FM/DM) or lower (FM/CE) limits for smaller $n > 0$. Recall that $\gamma_1^{(1)} = 0$ for the FM/CE case while $\gamma_1^{(0)} = 1$ for both FM cases. In the continuum limit of the FM/DM case, we have proven that $\gamma_1^{(n)} \to \sqrt{(1+n^2)/(1+2n^2)}$. Although we lack a rigorous proof, we numerically find that $\gamma_1^{(n)} \to |n^2 - 1|/\sqrt{2n^4 + 2n^2 + 1}$ in the continuum limit of the FM/CE case. So non-Goldstone modes always mix tangential and transverse components for FM interactions.
Another way to look at these results is by plotting the coefficients versus mode index for a fixed δ = 1/M in Fig. 5. For AF interactions, the coefficients quickly fall off from their asymptotic δ → 0 limits of ξ(n) = 1 and ρ(n) = 1 with increasing n. As in Fig. 4, the results for ξ(n) (closed circles) and ρ(n) (open circles) are very close. Figures 4 and 5 suggest that for the maximum n = M/4, ξ(n) and ρ(n) approach 1/√2 as M increases. In the FM/DM case, γ(n) falls off monotonically with n for all M and analytic results in the continuum limit are indistinguishable from numerical results for M = 80. In the FM/CE case, γ(n) increases with n starting with γ(1) = 0. In both FM cases, γ(n) remains fairly constant as a function of n beyond n = 10 or so and approaches 1/√2 for large M.

What do these results imply about the observability of the SW modes? The contribution of mode n to the spectral weight S_{αα}(Q, ω) is proportional to

\[ \omega_n \left| \sum_{r=1}^{M} e^{-iQ \cdot r} \delta S_{αα}(n) \right|^2. \tag{12} \]

Using Eqs. (4) and (11), it is straightforward to show that the three modes Ψ1 (α = x or z), Ψ2 (α = y), and Ψ3 (α = x or y) contribute for AF interactions while the three modes Θ1 (α = x or z), Θ2 (α = y), and Θ3 (α = x or y) contribute for FM interactions. These modes are responsible for the INS intensity \[ S(q, ω) = S_{yy}(q, ω) + S_{zz}(q, ω) \] plotted in Fig. 1.

The purely magnetic contribution of mode n to the optical absorption is proportional to

\[ \omega_n \left| \sum_{r=1}^{M} \mathbf{h} \cdot \delta \mathbf{S}_r(n) \right|^2 = \frac{\omega_n}{4\mu_B^2} |\langle 0 | \mathbf{h} \cdot \mathbf{M} | n, q = 0 \rangle|^2, \tag{13} \]

where \( \mathbf{h} \) is the magnetic polarization of light and \( \mathbf{M} = 2\mu_B \sum_{r=1}^{M} \mathbf{S}_r \) is the magnetization per unit cell. This is nonzero for \( \Psi_{\pm \pm} \) in the AF/DM case and for \( \Theta_{\pm \pm} \) in the FM/DM case, both when \( \mathbf{h} = x \) or \( z \). So for nonzero \( \delta \), optical spectroscopy will detect two modes (\( \Psi_{\pm \pm} \)) in the AF/DM case, two (\( \Theta_{\pm \pm} \)) in the FM/DM case, and none in the CE cases. Only the FM/DM \( \Theta_{\pm \pm} \) modes remain optically active as \( \delta \to 0 \).

Notice that different parts of \( \delta \mathbf{S}_r(n) \) contribute to the INS intensity and to the optical absorption. For the AF/DM \( \Psi_{\pm \pm} \) and FM/DM \( \Theta_{\pm \pm} \) modes, the tangential parts of \( \delta \mathbf{S}_r(n) \) contribute to the optical absorption while the transverse parts contribute to the INS intensity.

Although few modes are optically active for the simple Hamiltonians \( \mathcal{H}_{\text{DM}} \) and \( \mathcal{H}_{\text{CE}} \), several physical perturbations can activate other modes. \( \Psi_2 \) and \( \Psi_3 \) become optically active for \( \mathbf{h} = y \). Hybridization with \( \Phi_0 \) then activates \( \Phi_{\pm 2} \), also for \( \mathbf{h} = y \). The alternating tilt of the cycloid on neighboring hexagonal planes mixes transverse and tangential components, thereby activating \( \Phi_1 \) and \( \Phi_{\pm 1} \).

How do other well-known materials with cycloidal or helical states fall into the four cases considered here? With helical AF2 and cycloidal AF5 states created by long-range competing AF interaction, \( \mathcal{H}_{\text{DM}} \) and \( \mathcal{H}_{\text{CE}} \), \( \mu_W \) falls into the AF/CE class. Although itinerant MnSi is a member of the FM/DM family and its inelastic neutron-scattering spectra agrees with Fig. 2(b).

Of the three observed modes in MnSi, only the central \( \Theta_1 \) mode is predicted to be optically active. A rare member of the FM/CE class, \( \text{Sr}_3\text{Fe}_2\text{O}_7 \) has a helical state produced by the competition between FM nearest-neighbor double exchange and AF next-nearest neighbor exchange.

To summarize, we have evaluated the normal modes of a spin cycloid or helix produced by either DM or CE interactions and for either AF or FM nearest-neighbor exchange coupling. In the continuum limit for AF exchange, the SW amplitudes for all modes are either purely tangential or transverse. But for FM exchange, the SW amplitudes for all modes except the Goldstone modes contain both tangential and transverse components, even in the continuum limit. Whereas the mode spectrum for DM interactions contains only one Goldstone mode, the mode spectrum for CE interactions contains three Goldstone modes. Our results explain why
only a subset of these modes are observable using neutron scattering or optical absorption.

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12 For a helix (also known as a spiral or proper screw) propagating along x, the spins would have values

\[ S_r = S(0, \sin(Qra), \cos(Qra)) \]  

(14)

with the tangent

\[ t_r = (0, \cos(2\pi \delta_r), -\sin(2\pi \delta_r)). \]  

(15)

The transverse direction in Eqs.[9][11] would then be x instead of y.

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18 Inelastic neutron scattering measures

\[ S(q, \omega) = \sum_{\alpha, \beta} \{ \delta_{\alpha \beta} - q_{\alpha} q_{\beta}/q^2 \} S_{\alpha \beta}(q, \omega) \]

\[ = S_{yy}(q, \omega) + S_{zz}(q, \omega) \]

for q along x.

19 For odd M and AF exchange, modes \( \Phi_{\pm(2n+1)} \) and \( \Psi_{\pm 2n} \) are missing from the spectrum because \( t_M = -x \) differs from \( t_0 = x \) and \( S_{M} \neq S_0 \). Consequently, those modes only become observable under some perturbation that breaks the invariance of the system when translated by \( M a \) with odd \( M \). One possibility is the dimerization of the lattice with period 2a. Another is the AF coupling between adjacent hexagonal planes but FM coupling within each plane, as for BiFeO3[20]. With FM exchange, \( t_M = x \) for either even or odd \( M \) so the complete spectrum of modes \( \Theta_{\pm a} \), always appears.

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