Chiral spin-wave excitations of the spin-5/2 trimers in the langasite compound Ba$_3$NbFe$_3$Si$_2$O$_{14}$

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Background

• K. Marty, V. Simonet, E. Ressouche, R. Ballou, P. Lejay, and P. Bordet:
  X-ray scattering: spacegroup P321, single domain (enantiopure) crystal, chirality $\epsilon_T = -1$
  Neutron diffraction measurements: Fe moments helically ordered below 27 K, $\vec{Q} \approx \frac{1}{7} \vec{c}^*$. 
  Polarized neutron diffraction indicated a single domain of helicity.

• C. Stock, L. C. Chapon, A. Schneidewind, Y. Su, P. G. Radaelli, D. F. McMorrow,
  A. Bombardi, N. Lee, and S.-W. Cheong:
  Neutron scattering (elastic, polarized): diffuse scattering in paramagnetic phase.
  Inelastic neutron scattering (unpolarized): Spin waves in the a*c* plane.

• M. Loire, V. Simonet, S. Petit, K. Marty, P. Bordet, P. Lejay, J. Ollivier, M. Enderle,
  P. Steffens, E. Ressouche, A. Zorko, and R. Ballou:
  Inelastic neutron scattering (unpolarized and polarized): Spin waves in the b*c* plane.
  Linear spin-wave theory: interpretation of the results.
Ground state properties

Hund’s rules applied on Fe$^{3+}$ ion with five 3$d$ electrons \( \Rightarrow \ S = \frac{5}{2}, \ L = 0 \)

Trigonal space group \((P321)\). Different super-supersuperexchange paths for \(J_5\) and \(J_3\).
Structural chirality \(\epsilon_T = \pm 1\) (Marty et al., \(\epsilon_T = -1\)).

\[
T < T_N = 27 \text{ K}: \text{Helix with } \vec{Q} \simeq \frac{1}{7} \vec{c}^* \text{ or a turn angle } \phi = \epsilon_H \frac{2\pi}{7} \sim \text{helicity } \epsilon_H = \pm 1
\]

The angle between \(\langle \vec{S}_1 \rangle\) and \(\langle \vec{S}_2 \rangle\) is \(\gamma = \epsilon_\gamma \frac{2\pi}{3} \sim \text{spin triangle orientation}: \epsilon_\gamma = \pm 1\)

\[
\tan \phi = R \sin \gamma, \quad R = \frac{2(J_5 - J_3)}{2J_4 - J_3 - J_5} \quad \Rightarrow \quad \epsilon_H = \text{sign}(R) \epsilon_\gamma
\]
Cluster MF/RPA model

\[ \mathcal{H}_T = J_1 \left( \vec{S}_1 \cdot \vec{S}_2 + \vec{S}_2 \cdot \vec{S}_3 + \vec{S}_3 \cdot \vec{S}_1 \right) + D_c \left( \vec{S}_1 \times \vec{S}_2 + \vec{S}_2 \times \vec{S}_3 + \vec{S}_3 \times \vec{S}_1 \right) \cdot \hat{c} \]

\[ \mathcal{H} = \sum_i \mathcal{H}_T(i) + \frac{1}{2} \sum_{i, \xi} \sum_{j, \eta} J_{\xi \eta}(i j) \vec{S}_\xi(i) \cdot \vec{S}_\eta(j) \]

\[ J_1 = 1.25, \quad J_2 = 0.2, \quad J_3 = 0.1, \quad J_4 = 0.064, \quad J_5 = 0.29, \quad D_c = +0.0038 \]

(in units of meV).

Technicalities:

\[ \vec{S}_1 \otimes \vec{S}_2 \otimes \vec{S}_3 \Rightarrow 6^3 = 216 \text{ states/trimer} \]

MF-susceptibility (9 × 9 matrices): \( (\overline{\chi}^0)_{\xi \eta}(\omega) \)

7 trimer sublattices.

\[ I(\vec{q}, \omega) = \sum_{\alpha \beta} \frac{\delta_{\alpha \beta} - q_\alpha q_\beta / q^2}{3\pi \left(1 - e^{-\hbar \omega / k_B T}\right)} \]

\[ \times \sum_{\xi \eta} \text{Im} \left[ \chi_{\alpha \beta}(\vec{q}, \omega) e^{-i \vec{q} \cdot (\vec{R}_\xi - \vec{R}_\eta)} \right] \]

RPA-susceptibility:

\[ \overline{\chi}(i j, \omega) = \overline{\chi}_i^0(\omega) \left\{ \delta_{i j} + \sum_{j'} \overline{\delta}(i j') \overline{\chi}(j' j, \omega) \right\} \]
Spin waves in a simple helix

The spin-wave mode shown is the one, where all spins are precessing in phase, \( q_{\text{rel}} \approx 0 \).

\[ \omega t = 0: \text{the } ab \text{ component of } \Delta \vec{S} \text{ (the green arrow) has the same wave vector and the same sense of rotation (helicity) as the helix.} \]

\[ \omega t = \pi / 2: \text{the } c \text{ component of } \Delta \vec{S} \text{ (the orange arrow) has zero wave vector and is independent of the sense of rotation.} \]
Spin waves in the triangular helix

The spins along a line parallel to the $c$ axis precess in the same way as in the case of a simple helix, but the basis of three spins in the triangles implies the presence of three different polarizations of the spin-wave excitations:

- $\vec{q} \parallel \vec{c}$
- $\vec{S}_{1\perp} + \vec{S}_{2\perp} + \vec{S}_{3\perp} = \vec{0}$
- $S_{1c} + S_{2c} + S_{3c} = 0$

Detected by the $c$ component

Detected by an $ab$ component
Unpolarized neutron scattering

Polarized neutron scattering I


\[
\frac{d\sigma^{\pm/\mp}}{d\Omega} = \sum_{ij} e^{i\vec{k} \cdot \vec{r}_{ij}} p_i p_j^* \left[ \vec{S}_{\perp i} \cdot \vec{S}_{\perp j} \mp i\hat{z} \cdot (\vec{S}_{\perp i} \times \vec{S}_{\perp j}) \right]
\]

\[
S(\vec{k}, \omega) = \frac{I^\pm(\vec{k}, \omega) + I^\mp(\vec{k}, \omega)}{2}, \quad C(\vec{k}, \omega) = \frac{I^\pm(\vec{k}, \omega) - I^\mp(\vec{k}, \omega)}{2}, \quad \hat{z} = \hat{k}
\]

Simple helix with helicity $\epsilon_H$:

Static:  \[
\frac{C(\vec{k})}{S(\vec{k})} = \frac{2 \cos \theta}{1 + \cos^2 \theta} \left[ \delta(\vec{G} - \vec{Q} - \vec{k}) - \delta(\vec{G} + \vec{Q} - \vec{k}) \right] \epsilon_H
\]

Dynamic:  \[
\frac{C(\vec{k}, \omega)}{S(\vec{k}, \omega)} = 0 \quad \text{or} \quad \frac{C(\vec{k}, \omega)}{S(\vec{k}, \omega)} = \pm \frac{2 \cos \theta}{1 + \cos^2 \theta} \epsilon_H
\]

where $\cos \theta = \vec{k} \cdot \vec{Q} / ||\vec{k}|| ||\vec{Q}||$ and $\vec{G}$ is a reciprocal lattice vector.
Polarized neutron scattering II

X-ray exp. (Marty et al.) \(\Rightarrow\) \(\epsilon_T = -1\).

Unpolarized neutron exp. (Loire et al.) \(\Rightarrow\)
\[R = \frac{2(J_5 - J_3)}{2J_4 - J_3 - J_5} < 0, \text{ or } \text{sign}(R) = \epsilon_T\]

or \(\epsilon_H = \epsilon_T \epsilon_T\)

\(\epsilon_H = +1\) when \(\epsilon_T = -1\)

\(\Rightarrow\) \(D_c > 0\)
Conclusion

• Dynamic chiral effects have been observed before – in MnSi (critical fluctuations) and in CsMnBr$_3$ (in an applied field) – but not as clearly exposed as here in Ba$_3$NbFe$_3$Si$_2$O$_{14}$.

• M. Loire, V. Simonet, S. Petit, K. Marty, P. Bordet, P. Lejay, J. Ollivier, M. Enderle, P. Steffens, E. Ressouche, A. Zorko, and R. Ballou performed the first polarized neutron experiments showing the unique properties of the spin waves in Ba$_3$NbFe$_3$Si$_2$O$_{14}$.

• The main conclusions of Loire et al. based on linear spin-wave theory were the same as presented here.

• The spin triangles are relatively strongly frustrated, and the cluster-MF/RPA calculations show that the single-spin MF approximation is unreliable.

• The spin dynamics derived from a boson representation (Holstein-Primakoff) works “surprisingly” well – the results derived from the cluster-MF/RPA calculations are nearly the same as predicted by linear spin-wave theory (in the zero temperature limit).