Chiral spin-wave excitations of the spin-5/2 trimers in the langasite compound $Ba_3NbFe_3Si_2O_{14}$

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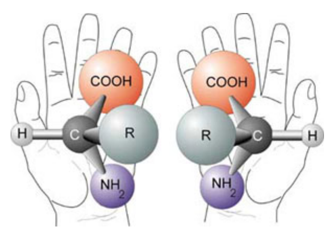
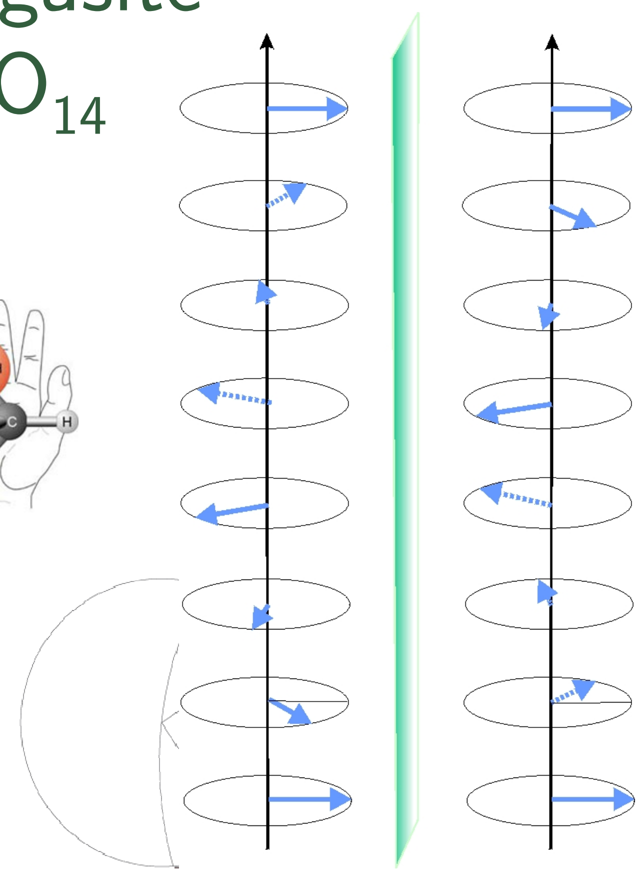




Theoretical and Experimental Magnetism Meeting
Rutherford-Appleton Laboratory
June 16-17, 2011

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, $\vec{Q} \simeq \frac{1}{7}\vec{c}^*$.
Polarized neutron diffraction indicated a single domain of helicity.

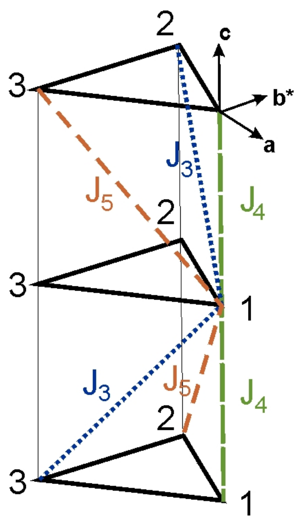
•C. Stock, L. C. Chapon, A. Schneidewind, Y. Su, P. G. Radaelli, D. F. McMorrow, A. Bombardi, N. Lee, and S.-W. Cheong:
Neutron scattering (elastic, polarized): diffuse scattering in paramagnetic phase.
Inelastic neutron scattering (unpolarized): Spin waves in the a*c* plane.

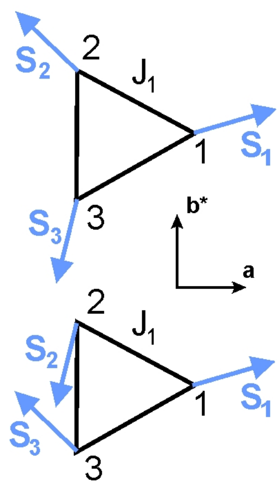
•M. Loire, V. Simonet, S. Petit, K. Marty, P. Bordet, P. Lejay, J. Ollivier, M. Enderle, P. Steffens, E. Ressouche, A. Zorko, and R. Ballou:
Inelastic neutron scattering (unpolarized and polarized): Spin waves in the b*c* plane.
Linear spin-wave theory: interpretation of the results.

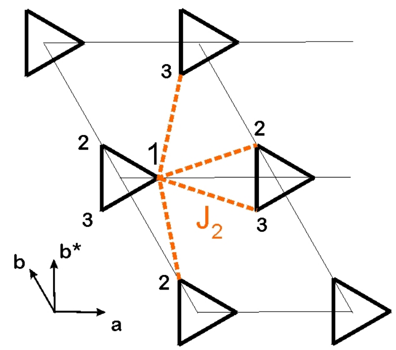
Ground state properties

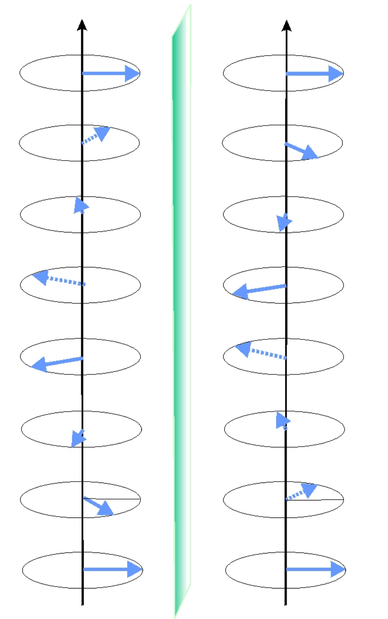
Hund's rules applied on Fe^{3+} ion with five $3d$ electrons $\quad\Rightarrow\quad S = \frac{5}{2},\ L = 0$

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









$\epsilon_H = -1 \qquad \epsilon_H = +1$

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

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

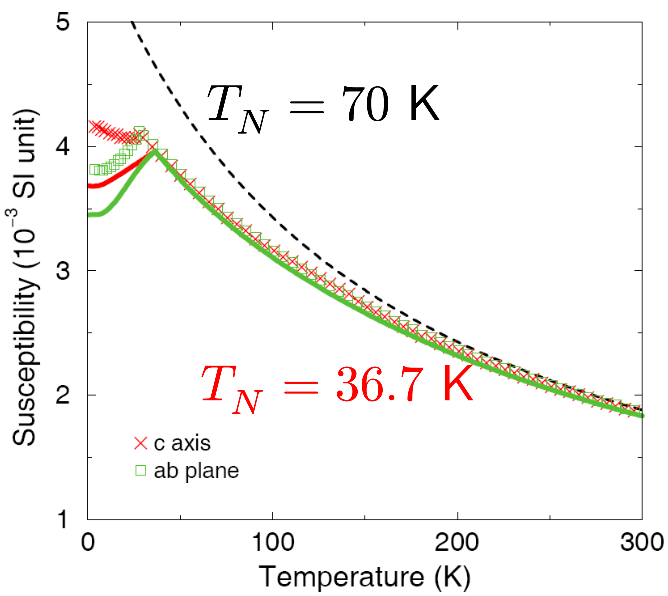
$$\tan\phi = R\sin\gamma, \quad R = \frac{2(J_5 - J_3)}{2J_4 - J_3 - J_5} \quad \Rightarrow \quad \epsilon_H = \mathrm{sign}(R)\,\epsilon_\gamma$$

Cluster MF/RPA model

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

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

$J_1 = 1.25,\ J_2 = 0.2,\ J_3 = 0.1,\ J_4 = 0.064,\ J_5 = 0.29,\ D_c = +0.0038$
(in units of meV).

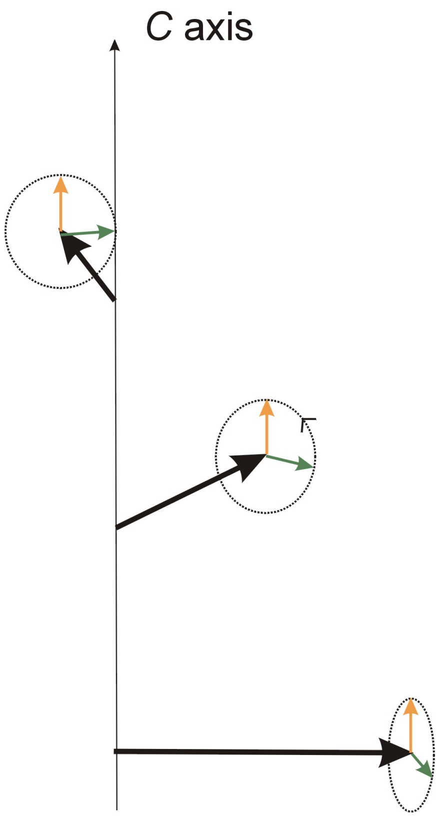


Technicalities:
$\vec{S}_1\otimes\vec{S}_2\otimes\vec{S}_3 \quad\Rightarrow\quad 6^3 = 216$ states/trimer
MF-susceptibility (9×9 matrices): $\left(\overline{\overline{\chi}}^{\,0}\right)^{\xi\eta}(\omega)$
7 trimer sublattices.

$$I(\vec{q},\omega) = \sum_{\alpha\beta}\frac{\delta_{\alpha\beta}-q_\alpha q_\beta/q^2}{3\pi\left(1-e^{-\hbar\omega/k_BT}\right)} \times \sum_{\xi\eta}\mathrm{Im}\left[\chi^{\xi\eta}_{\alpha\beta}(\vec{q},\omega)e^{-i\,\vec{q}\cdot(\vec{R}_\xi-\vec{R}_\eta)}\right]$$

RPA-susceptibility: $\overline{\overline{\chi}}(ij,\omega) = \overline{\overline{\chi}}{}_i^0(\omega)\left\{\delta_{ij}+\sum_{j'}\overline{\overline{\mathcal{J}}}(ij')\overline{\overline{\chi}}(j'j,\omega)\right\}$

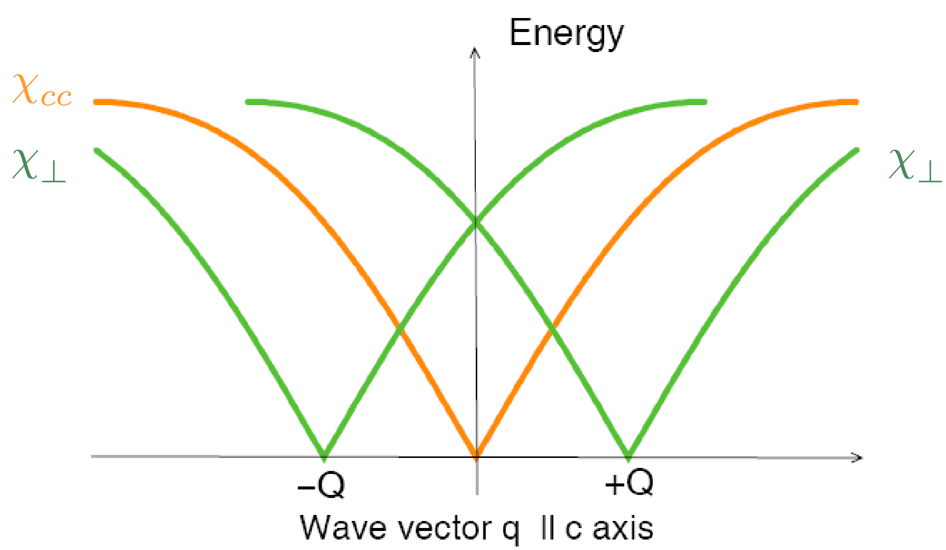
Spin waves in a simple helix



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

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

$\omega t = \pi/2$: the c component of $\Delta\vec{S}$ (the orange arrow) has zero wave vector and is independent of the sense of rotation.



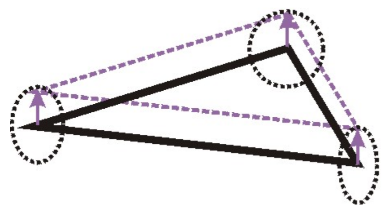
Righthanded helix.
chirality/helicity: $\epsilon_H = +1$

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:

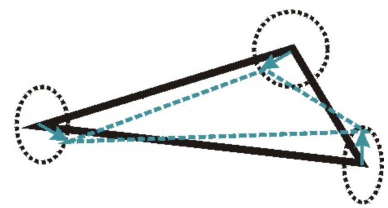
c mode

$\vec{q} \parallel \vec{c}$

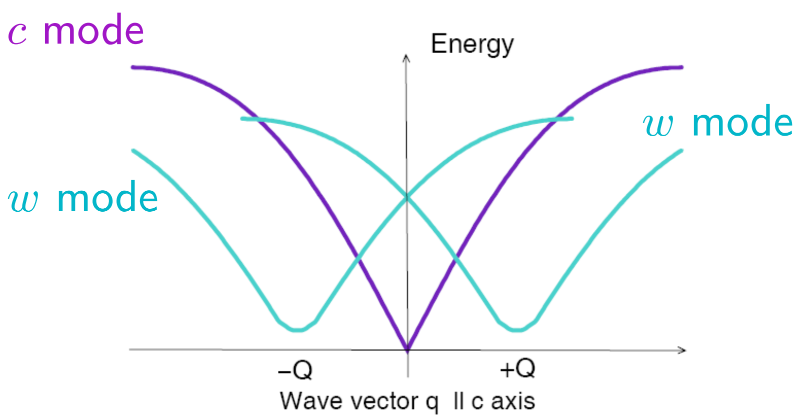


$$\vec{S}_{1\perp} + \vec{S}_{2\perp} + \vec{S}_{3\perp} = \vec{0}$$

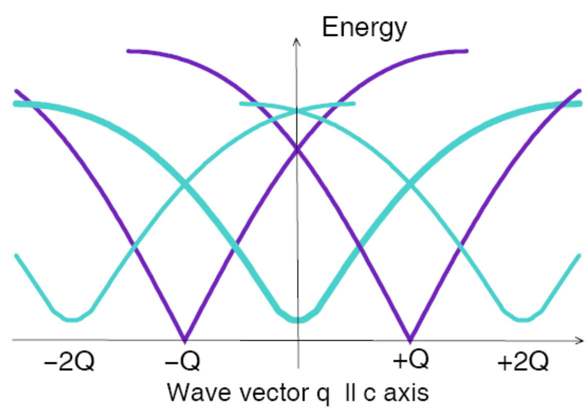
$2 \times w$ modes



$$S_{1c} + S_{2c} + S_{3c} = 0$$



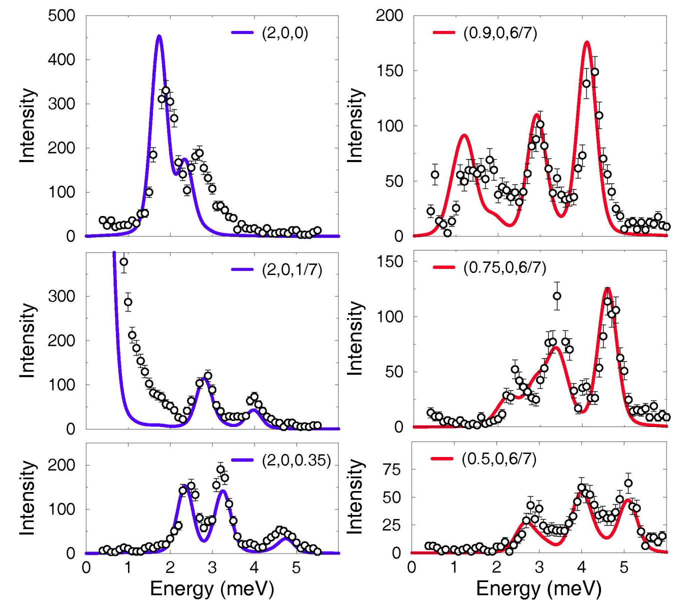
Detected by the c component

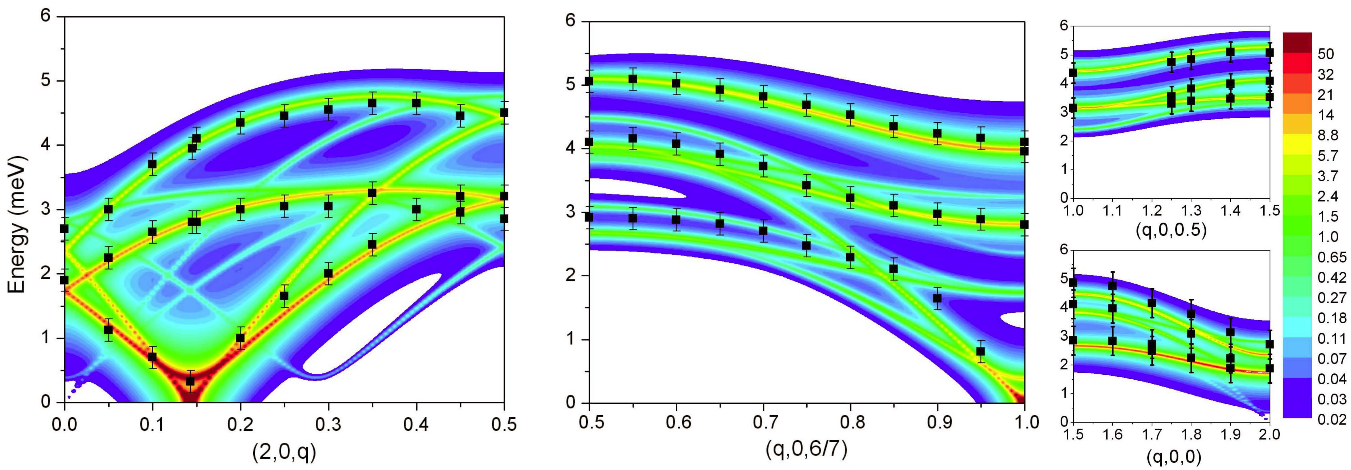


Detected by an ab component

Unpolarized neutron scattering

Experiments at 2.5 K by
C. Stock, L. C. Chapon, A. Schneidewind, Y. Su,
P. G. Radaelli, D. F. McMorrow, A. Bombardi,
N. Lee, and S.-W. Cheong,
Phys. Rev. B **83**, 104426 (2011).





Polarized neutron scattering I

Experiments at 1.5 K by 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, Phys. Rev. Lett. **106**, 207201 (2011).

R. M. Moon, T. Riste, and W. C. Koehler, Phys. Rev. **181**, 920 (1969)

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

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

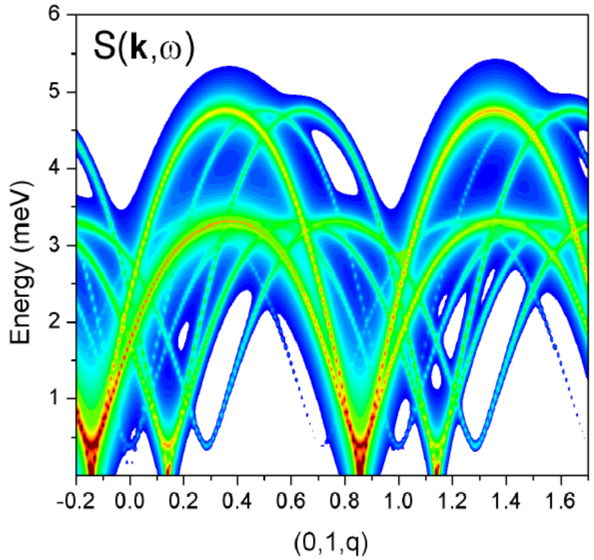
Simple helix with helicity ϵ_H:

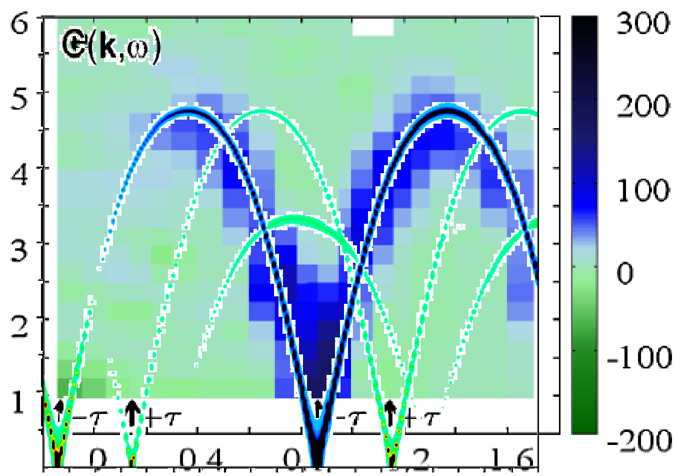
Static: $$\frac{C(\mathbf{k})}{S(\mathbf{k})} = \frac{2\cos\theta}{1+\cos^2\theta}\left[\delta(\mathbf{G}-\mathbf{Q}-\mathbf{k})-\delta(\mathbf{G}+\mathbf{Q}-\mathbf{k})\right]\epsilon_H$$

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

where $\cos\theta = \mathbf{k}\cdot\mathbf{Q}/|\mathbf{k}||\mathbf{Q}|$ and $\mathbf{G}$ is a reciprocal lattice vector.

Polarized neutron scattering II



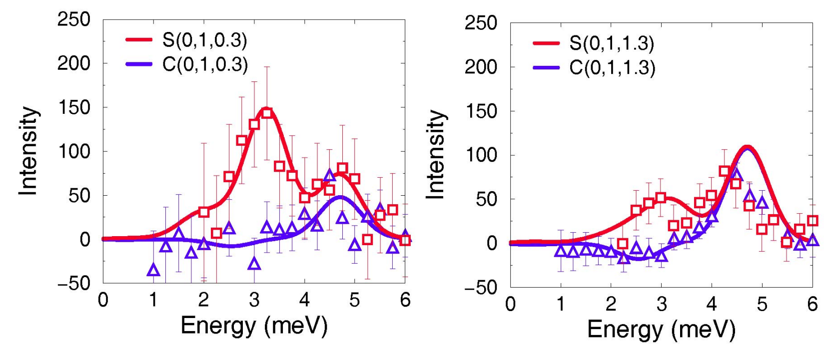


X-ray exp. (Marty et al.) $\Rightarrow \quad \epsilon_T = -1.$

Unpolarized neutron exp. (Loire et al.) $\Rightarrow$

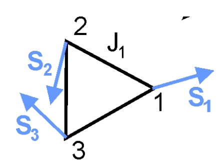
$R = \dfrac{2(J_5 - J_3)}{2J_4 - J_3 - J_5} < 0$, or $\mathrm{sign}(R) = \epsilon_T$

or $\epsilon_H = \epsilon_\gamma \, \epsilon_T$



$\epsilon_H = +1$ when $\epsilon_T = -1$

$\Rightarrow \quad D_c > 0$



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 $Ba_3NbFe_3Si_2O_{14}$.

•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 $Ba_3NbFe_3Si_2O_{14}$.

•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).