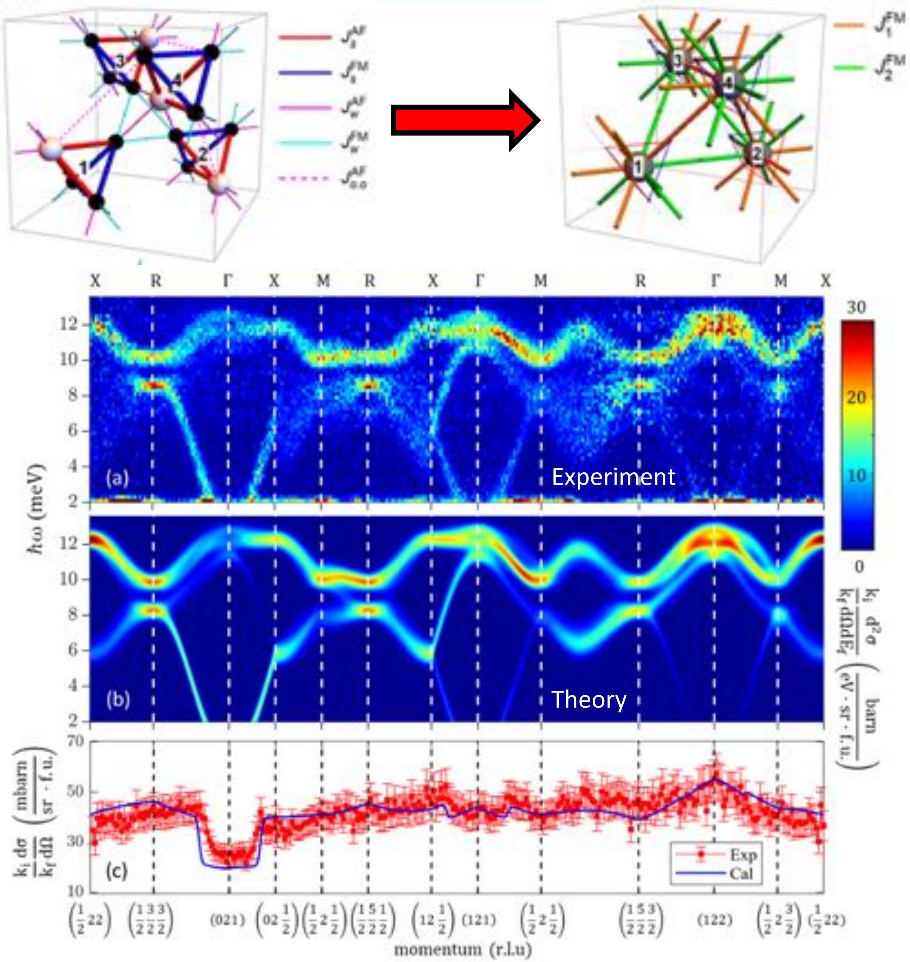
Magnons from molecular spins in chiral Cu_2OSeO_3



To describe the magnetism of the breathing pyrochlore Cu_2OSeO_3 (top) we treat each ferrimagnetic cluster as a single spin-1. This enables a focus on the simpler inter-cluster interactions. (a) and (c) show inelastic neutron scattering data along high symmetry trajectories that we describe in (b) and (c) with a small set of near neighbor interactions.

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Scientific Achievement

The asymmetric magnetic interactions that control the low energy excitations and the skyrmionic spin texture of Cu_2OSeO_3 were determined using inelastic neutron scattering and a coarse-grained description of this chiral breathing pyrochlore ferrimagnet.

Significance and Impact

Great simplification in describing the complex low energy magnetism of Cu_2OSeO_3 was achieved through a coarse-grained approach where four copper spins become one molecular spin-1 entity. Strongly selective X-point magnon decay into two long wavelength modes was discovered.

Research Details

- 50 Cu_2OSeO_3 single crystals (m=5.1 g) were synthesized and assembled for inelastic neutron scattering on SEQUOIA (ORNL) and MACS (NIST).
- High-quality neutron data were accounted for in detail by theoretical modelling based on a molecular magnetic form factor
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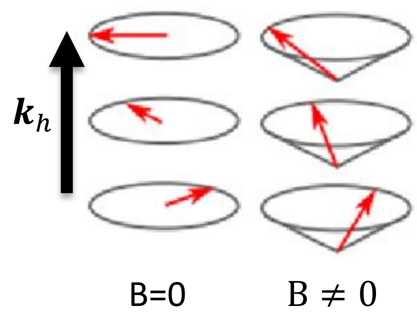
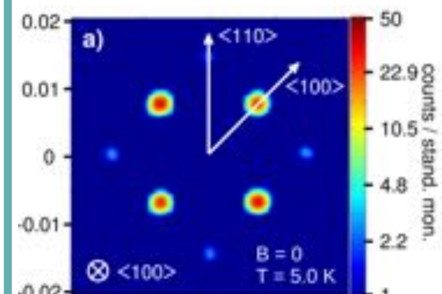
Phase diagram and mesoscopic spin texture in Cu_2OSeO_3

$$F[\mathbf{M}(\mathbf{r})] = \int d^3r[\underbrace{A(\nabla\mathbf{M})^2}_{\text{FM}} + \underbrace{aM^2 + cM^4}_{\text{Determine } |M|} + \underbrace{D\mathbf{M}\cdot(\nabla\times\mathbf{M})}_{\text{DM}}]$$

Low temperature

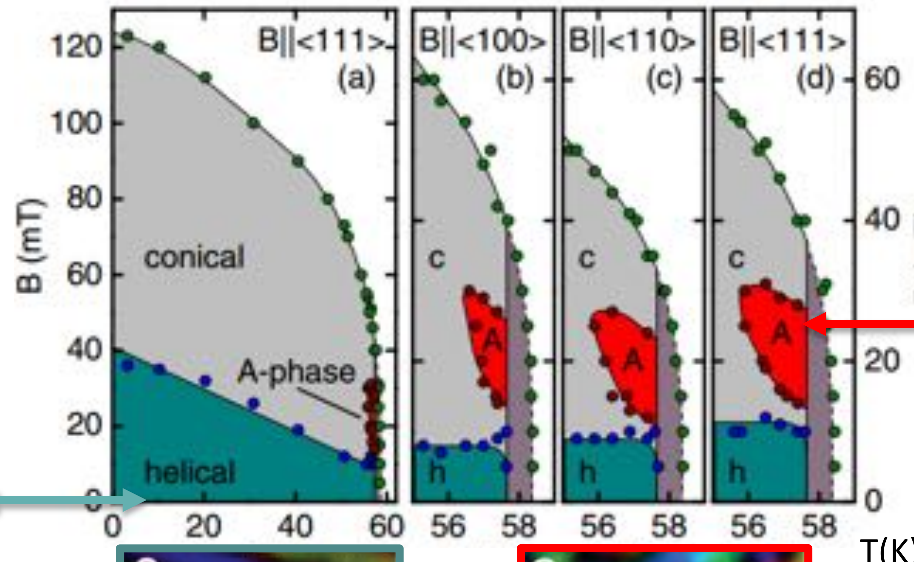


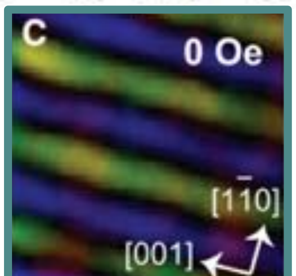


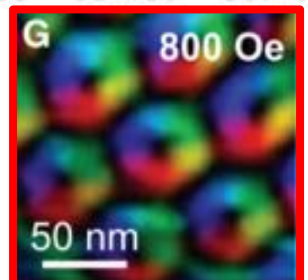
$k_h = 0.0145(11)$ r.l.u

$k_h = \frac{D}{2A}$ theory

Polarized SANS resolves the chirality to be the same as the crystal chirality

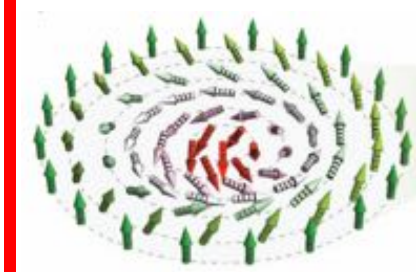


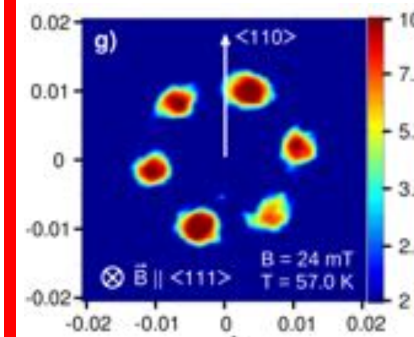




(TEM) Seki et al. Science 336 (2012): 198-201.
(SANS) Adams, T., et al. PRL 108 (2012): 237204.
(Polarized SANS) Dyadkin, V., et al. PRB 89 (2014): 140409.
(Theory) Petrova and Tchernyshyov. PRB 84. (2011): 214433.

Under finite external field and temperature

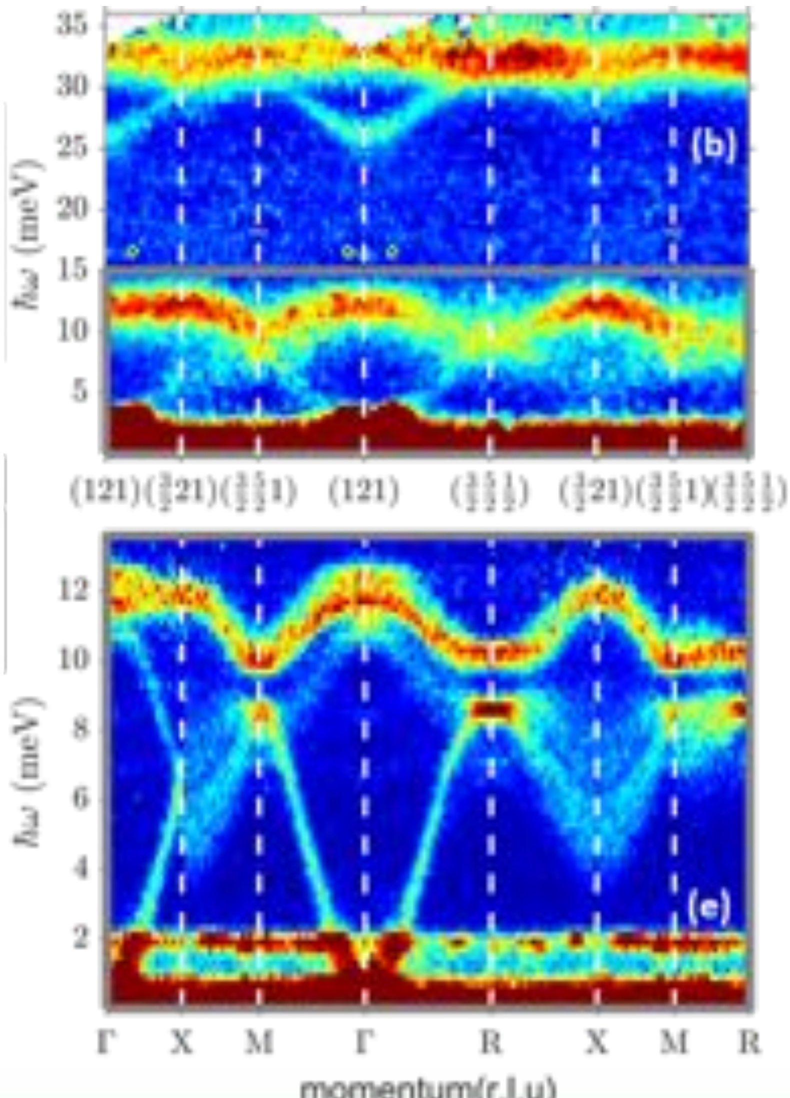


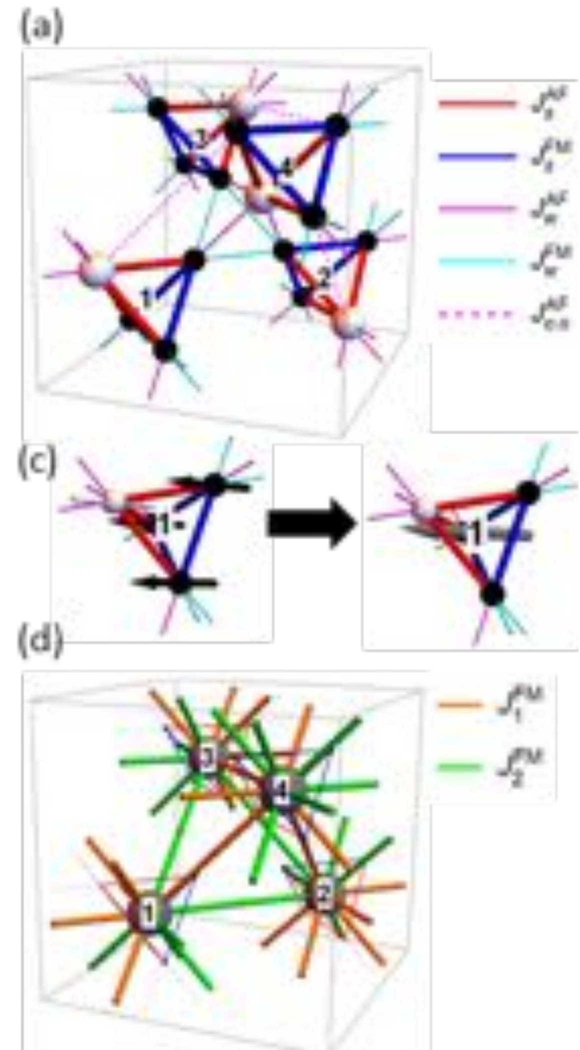


Superposition of three spin helices with wavevector $\boldsymbol{q}$ forming 120° configuration

A distinct low energy regime for magnetism in Cu_2OSeO_3

50 co-aligned crystals m= 5.1 g
mosaic < 2°





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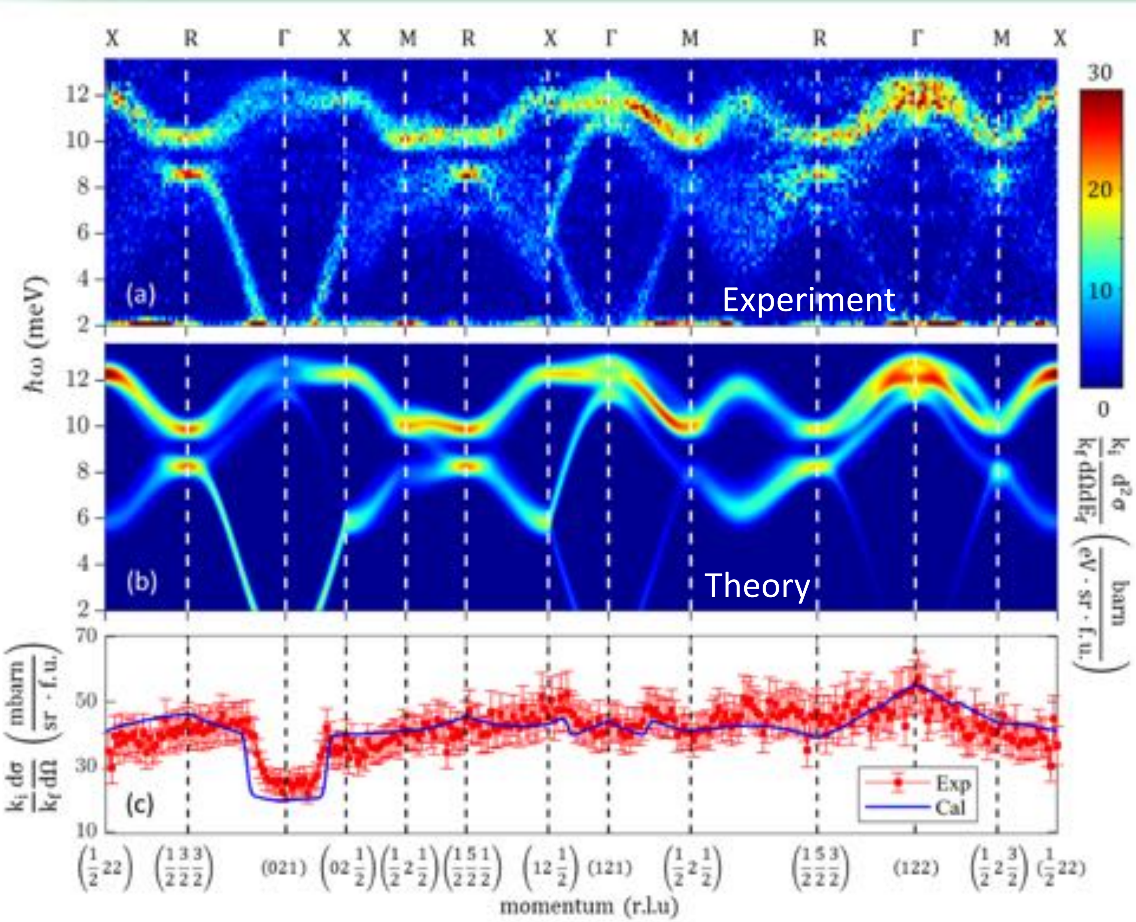
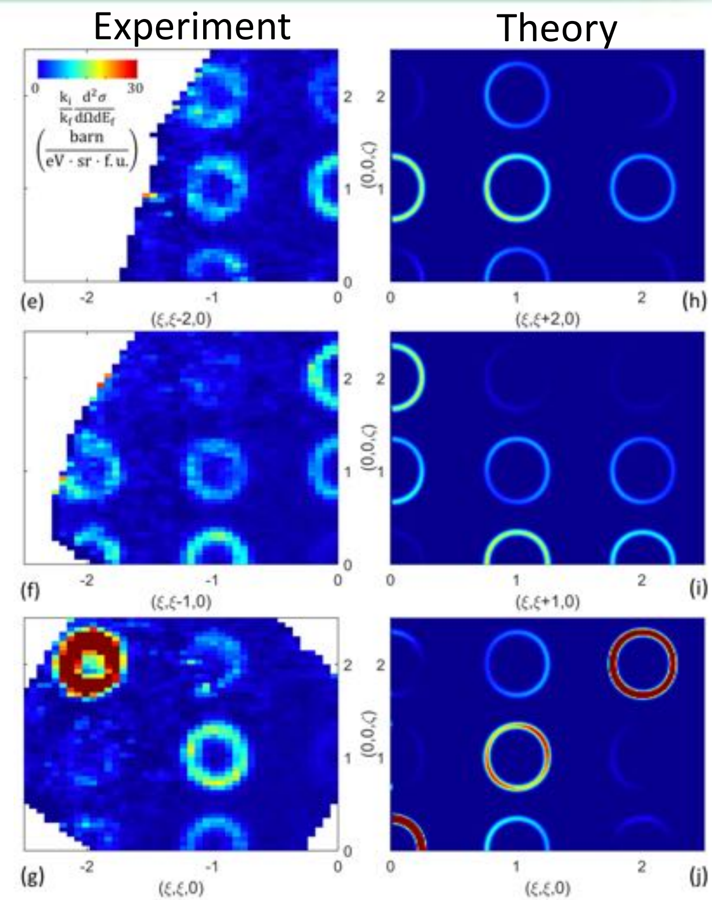
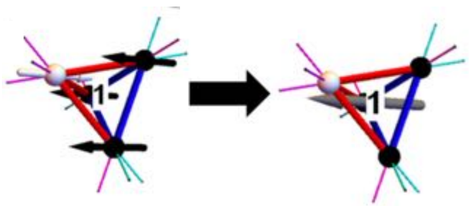
Modeling the low energy sector: molecular form factor





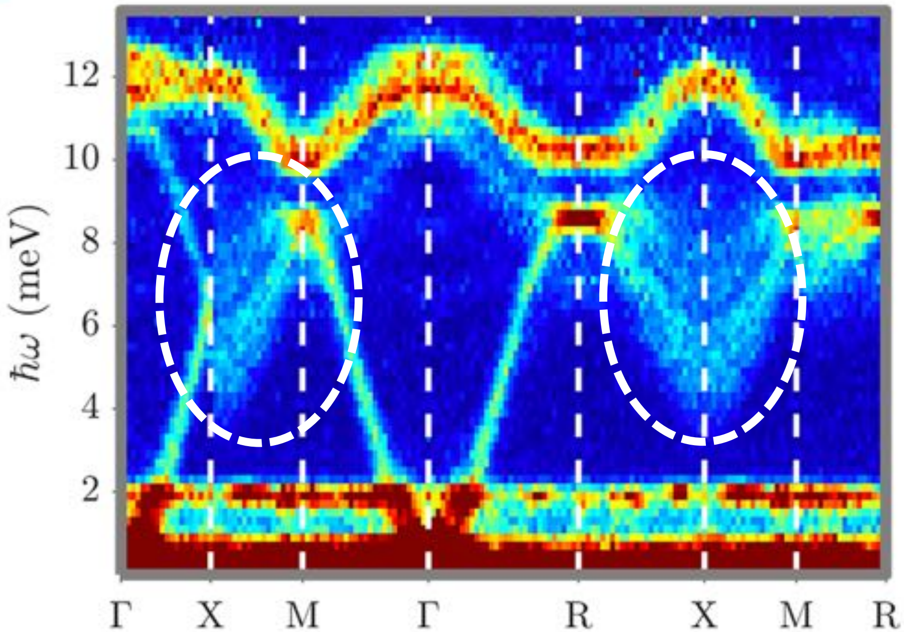


$$\langle\lambda'|\, s^{\beta}_{\mathrm{ldn}}\,|\lambda\rangle = \begin{cases} -\frac{1}{4}\,\langle\lambda'|\, S^{\beta}_{\mathrm{ld}}\,|\lambda\rangle & n=1\\ \frac{5}{12}\,\langle\lambda'|\, S^{\beta}_{\mathrm{ld}}\,|\lambda\rangle & n=2,3,4\end{cases}$$

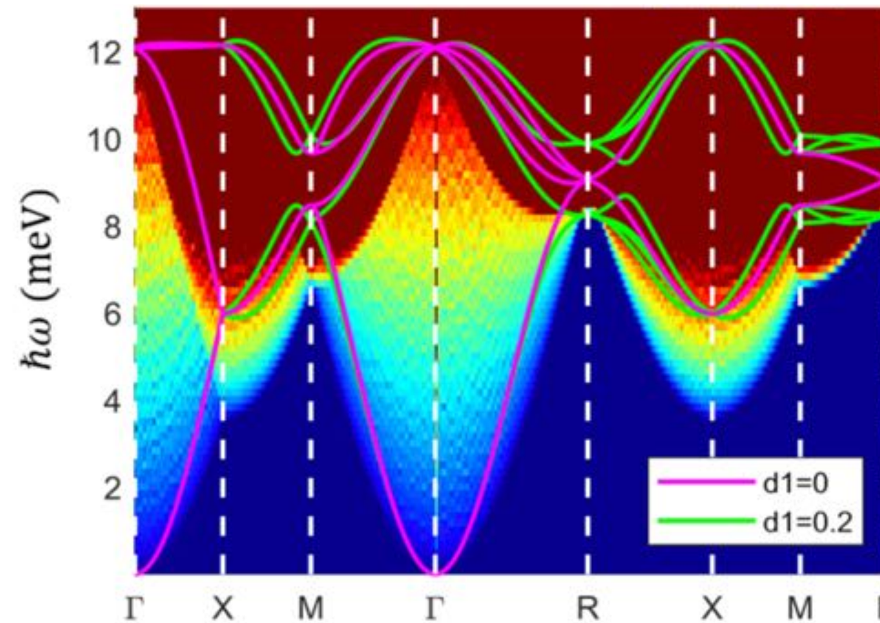
$$\tilde{F}_d(\mathbf{Q}) = \left(-\frac{1}{4}e^{i\mathbf{Q}\cdot(\mathbf{r}_{\mathrm{ld}1}-\mathbf{r}_{\mathrm{ld}})} + \frac{5}{12}\sum_{i=2}^{4} e^{i\mathbf{Q}\cdot(\mathbf{r}_{\mathrm{ld}i}-\mathbf{r}_{\mathrm{ld}})}\right)F(\mathbf{Q})$$

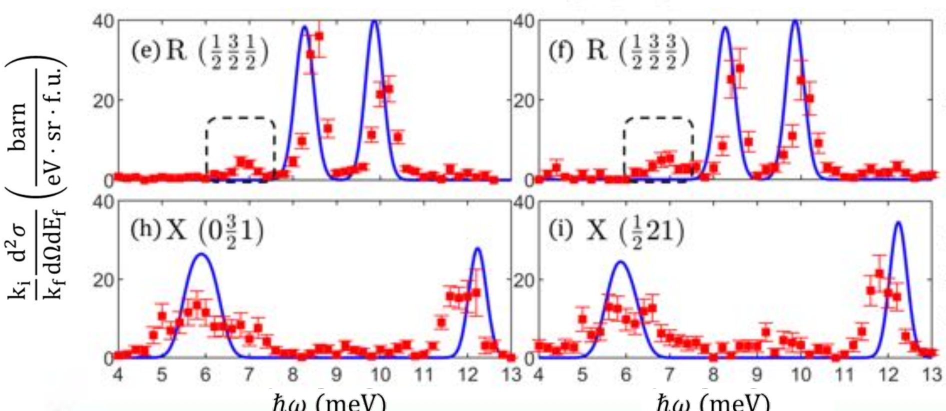
Selective x-point decay to long wavelength magnons

Inelastic magnetic neutron scattering



Two-magnon density of states





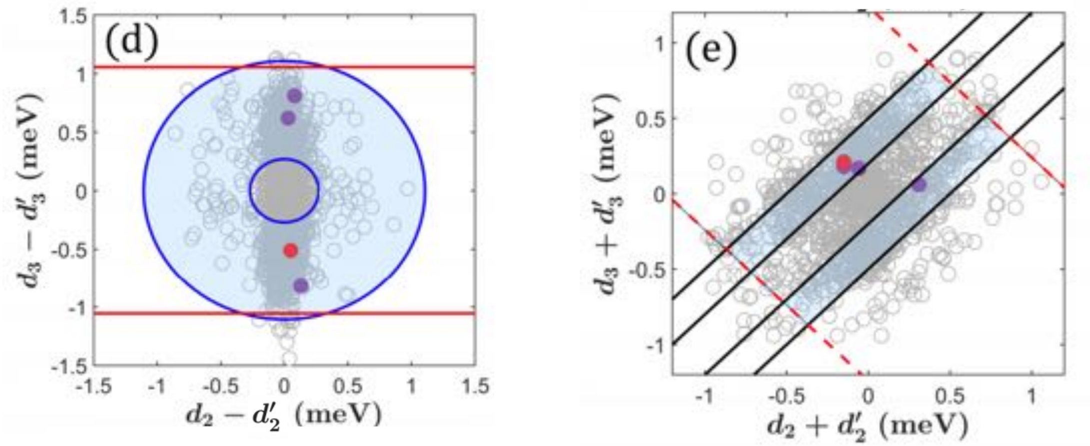
$$\mathbf{Q} \quad = \mathbf{q}_1 + \mathbf{q}_2$$
$$\hbar\omega = \epsilon(\mathbf{q}_1) + \epsilon(\mathbf{q}_2)$$

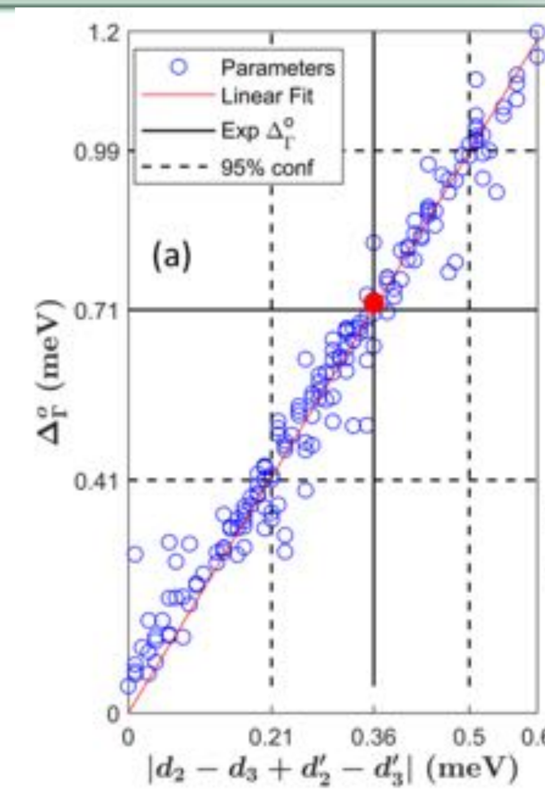
Magnons decay is pronounced at the X-point where the decay is to two long wavelength Γ-point magnons

A low energy molecular spin Hamiltonian for Cu_2OSeO_3

$$\mathcal{H}_J = \sum_{\langle ij \rangle} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle\langle ij \rangle\rangle} J_2 \mathbf{S}_i \cdot \mathbf{S}_j$$

$$\mathcal{H}_D = \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \sum_{\langle\langle ij \rangle\rangle} \mathbf{D}'_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$





Parameter Sectors	Parameter(meV)								Calculated Result	
	J_1	J_2	d_1	d_2	d_3	d'_1	d'_2	d'_3	k_h(r.l.u)	χ^2
$\|J_1\| < \|J_2\|, d_1 > 0$	$-0.58^{+0.08}_{-0.03}$	$-0.93^{+0.03}_{-0.05}$	$0.24^{+0.01}_{-0.03}$	-0.05	-0.15	$-0.16^{+0.01}_{-0.03}$	-0.10	0.36	0.0143	13.26
$\|J_1\| < \|J_2\|, d_1 < 0$	$-0.56^{+0.06}_{-0.04}$	$-0.95^{+0.09}_{-0.05}$	$-0.16^{+0.02}_{-0.03}$	-0.06	0.40	$0.24^{+0.02}_{-0.03}$	-0.09	-0.22	0.0129	13.54
$\|J_1\| > \|J_2\|, d_1 > 0$	$-0.96^{+0.07}_{-0.03}$	$-0.54^{+0.03}_{-0.05}$	$0.22^{+0.04}_{-0.01}$	-0.08	-0.36	$-0.18^{+0.04}_{-0.01}$	-0.14	0.42	0.0162	16.47
$\|J_1\| < \|J_2\|, d_1 < 0$	$-0.94^{+0.07}_{-0.02}$	$-0.55^{+0.03}_{-0.05}$	$-0.15^{+0.01}_{-0.03}$	0.22	-0.38	$0.25^{+0.01}_{-0.03}$	0.09	0.44	0.0151	15.04

D□ D'□

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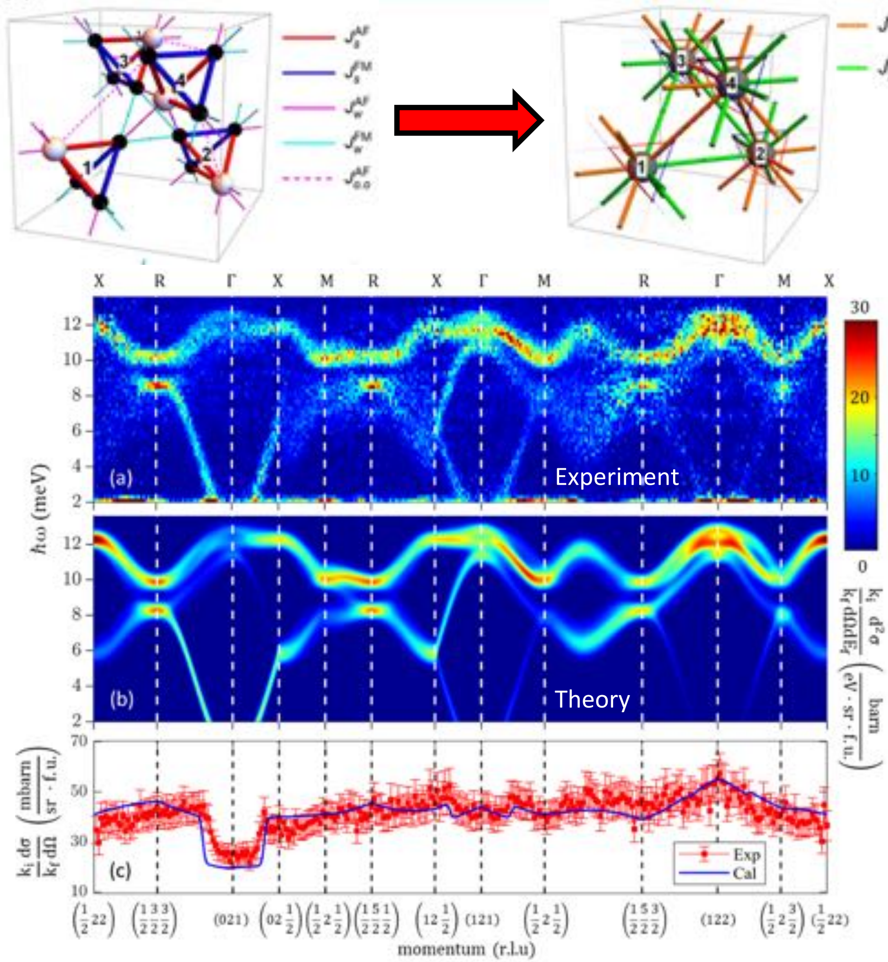

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