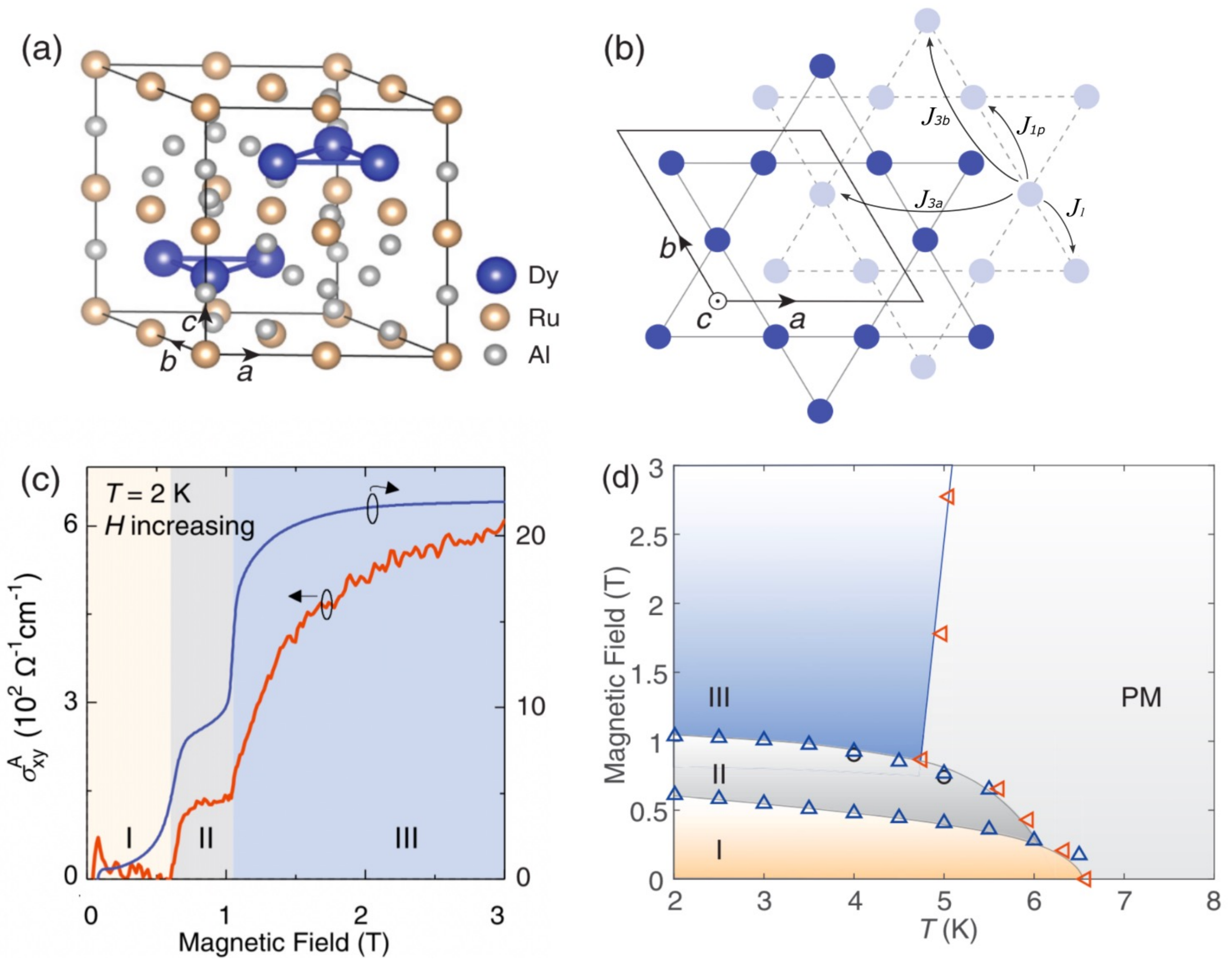
Spin dynamics in the trimer-host compound $Dy_3Ru_4Al_{12}$

Han Wang[1], Jiajun Mo[1], Akiko Kikkawa[2], Max Hirschberger[2,6], Fan Xiao[3], Taro Nakajima[2], Yixi Su[4], Barry Winn[5], Yasujiro Taguchi[2], Yoshinori Tokura[2,6], Taka-hisa Arima[2,7], Shang Gao[1,2]

[1]Department of Physics, University of Science and Technology of China, China
[2]RIKEN Center for Emergent Matter Science CEMS, Japan
[3]Laboratory for Neutron Scattering, Paul Scherrer Institut, Switzerland
[4]Jülich Center for Neutron Sciences at MLZ, Germany
[5]Neutron Scattering Division, Oak Ridge National Laboratory ORNL, USA
[6]Department of Applied Physics, University of Tokyo, Japan
[7]Department of Advanced Materials Science, University of Tokyo, Japan

1. Crystal structure and phase diagram

- In $Dy_3Ru_4Al_{12}$ (space group $P6_3/mmc$), the Dy^{3+} ions form breathing kagomé layers that are composed of two different sizes of corner-sharing triangles [1,2].
- Below T_N = 6.5 K, $Dy_3Ru_4Al_{12}$ enters an antiferromagetically ordered phase with propagation vector $\boldsymbol{q}$ = (1/2, 0, 1/2) [1].
- In a magnetic field along the *c* axis, two additional phases were observed, including a 1/3 magnetization plateau phase (II) and a nearly saturated phase (III).
- The existence of spin trimers with a nonzero scalar spin chirality has been proposed as the origin of an unconventional geometrical Hall effect [3].



2. Experimental and theoretical methods

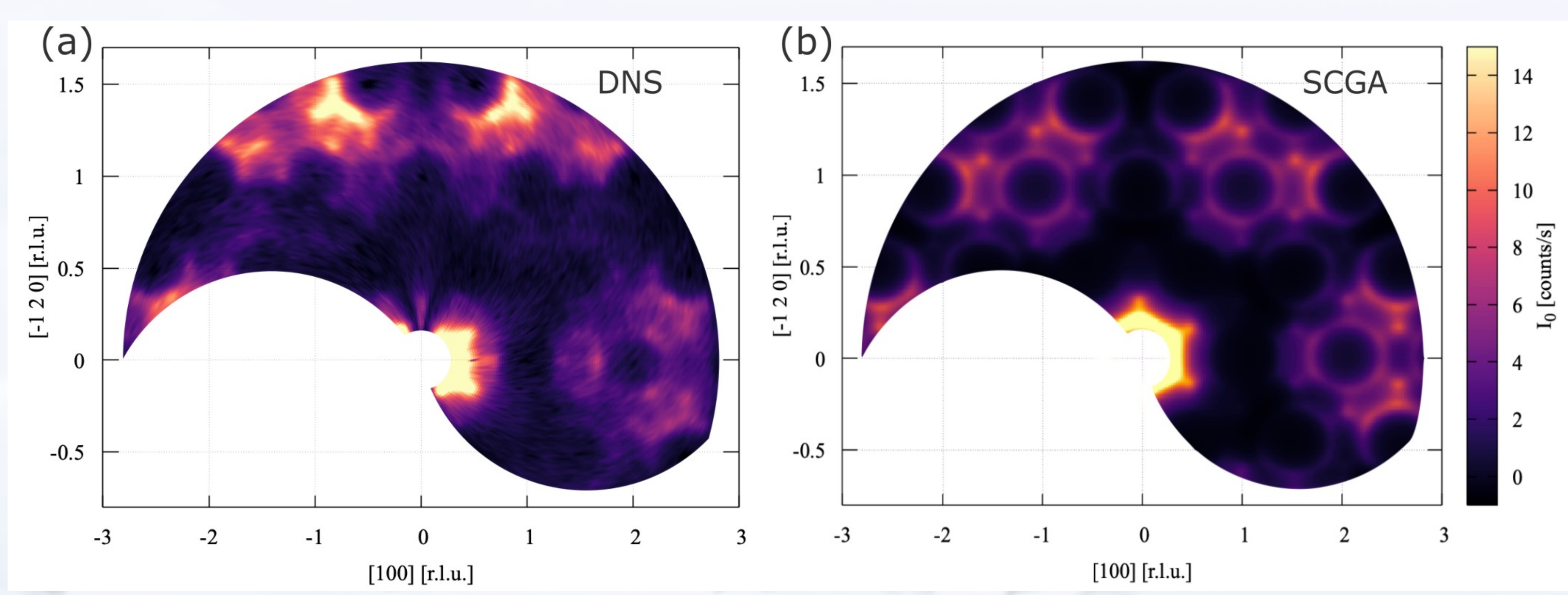
- Crystals of $Dy_3Ru_4Al_{12}$ were grown using the Czochralski technique at the RIKEN CEMS to study the spin correlations below and above T_N.
- Polarized neutron diffuse scattering experiments were performed on the DNS diffractometer at the MLZ. Crystals were polished to thin slices of dimensions of 3.0×0.8 ×0.15 mm^3 to reduce neutron absorption caused by the Dy element [4].
- Inelastic neutron scattering (INS) experiments were performed on the HYSPEC spectrometer at the ORNL to study the spin excitations in $Dy_3Ru_4Al_{12}$ below T_N.
- The self-consistent Gaussian approximations *(SCGA)* method, as implemented in the JuliaSCGA program, is employed to analyze the short-range quasi-elastic spin correlations. In this method, the spin correlations under the Gaussian approximation can be expressed as [5]:

$$\langle S^{\alpha}_{\mu}(-\boldsymbol{q})S^{\beta}_{\nu}(\boldsymbol{q})\rangle = \delta^{\alpha\beta}[\lambda\mathbb{1} + \beta\mathcal{J}(\boldsymbol{q})]^{-1}_{\mu\nu},$$

- The Molecular Dynamics Simulation *(MD)* method, as implemented in the JuliaMD program, is employed to simulate and study the motion and interaction of localized spin systems [6]. In this method, The motion of a particle under the action of an average field can be expressed as:

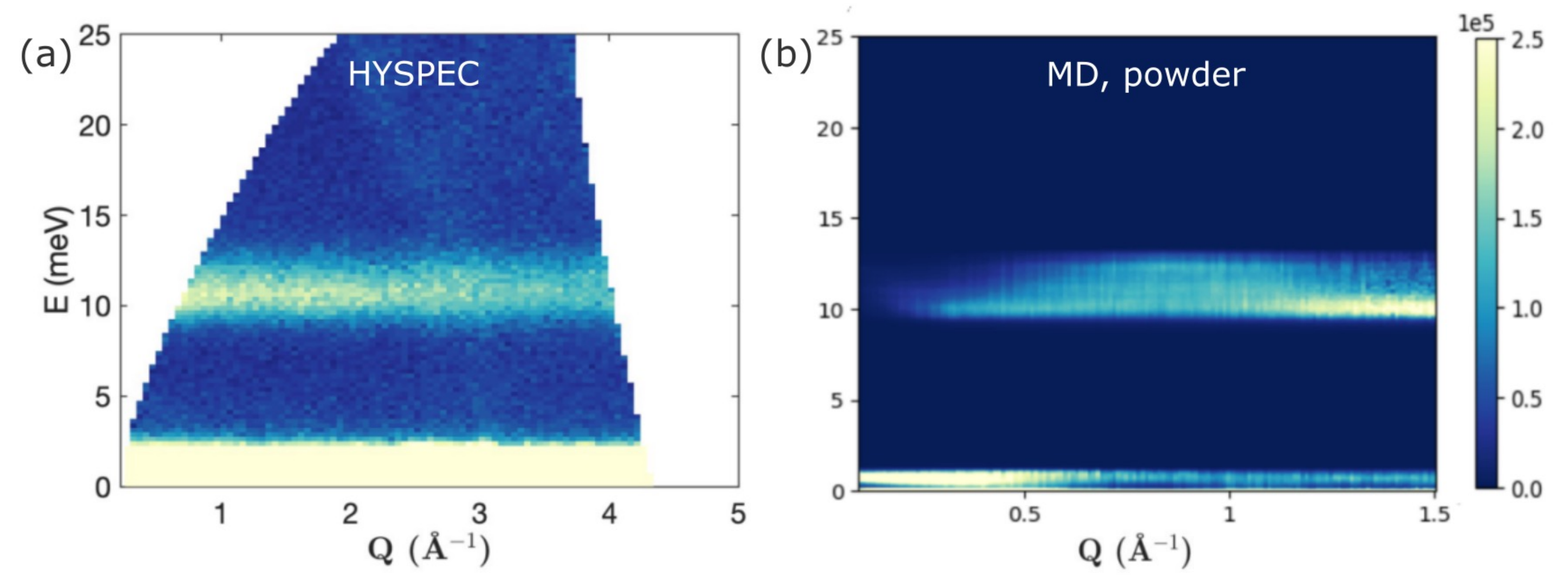
$$\frac{d\mathbf{M}}{dt} = -\gamma'\mathbf{M}\times\mathbf{H}_{\text{eff}} - \lambda\mathbf{M}\times(\mathbf{M}\times\mathbf{H}_{\text{eff}})$$

3. Short-range spin correlations



- An exotic diffuse scattering pattern in the L = 0 plane was observed at T = 8.5 K.
- Due to unperfect neutron absorption correction, the diffuse pattern is not exactly symmetric as expected for a hexagonal system.
- Using the SCGA method, the scattering pattern can be described by a minimal J_1-J_{1p}-J_{3ab}-J_{c12}-D_2 model, where J_1, J_{1p}, J_{3a}, and J_{3b} are intralayer couplings, J_{c1} and J_{c2} are the shortest interlayer couplings. The fitted coupling strengths are J_1 = -1.4 (FM), J_{1p} = -0.26(1), J_{3ab} = 0.11(1) (AFM) , J_{c12} = -0.013(2), D_2 = 0.08(1) meV.

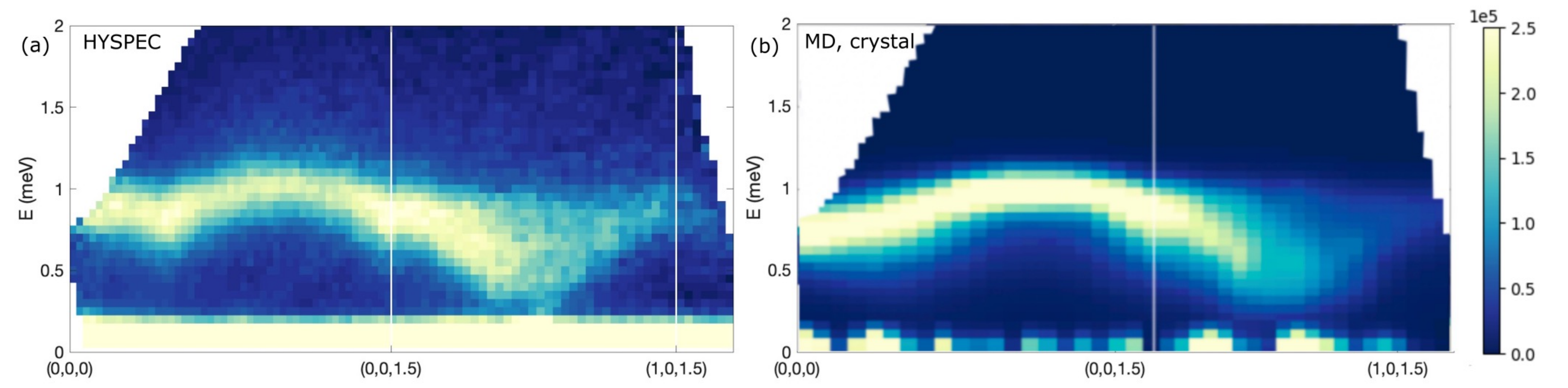
4. Spin dynamics at high energies



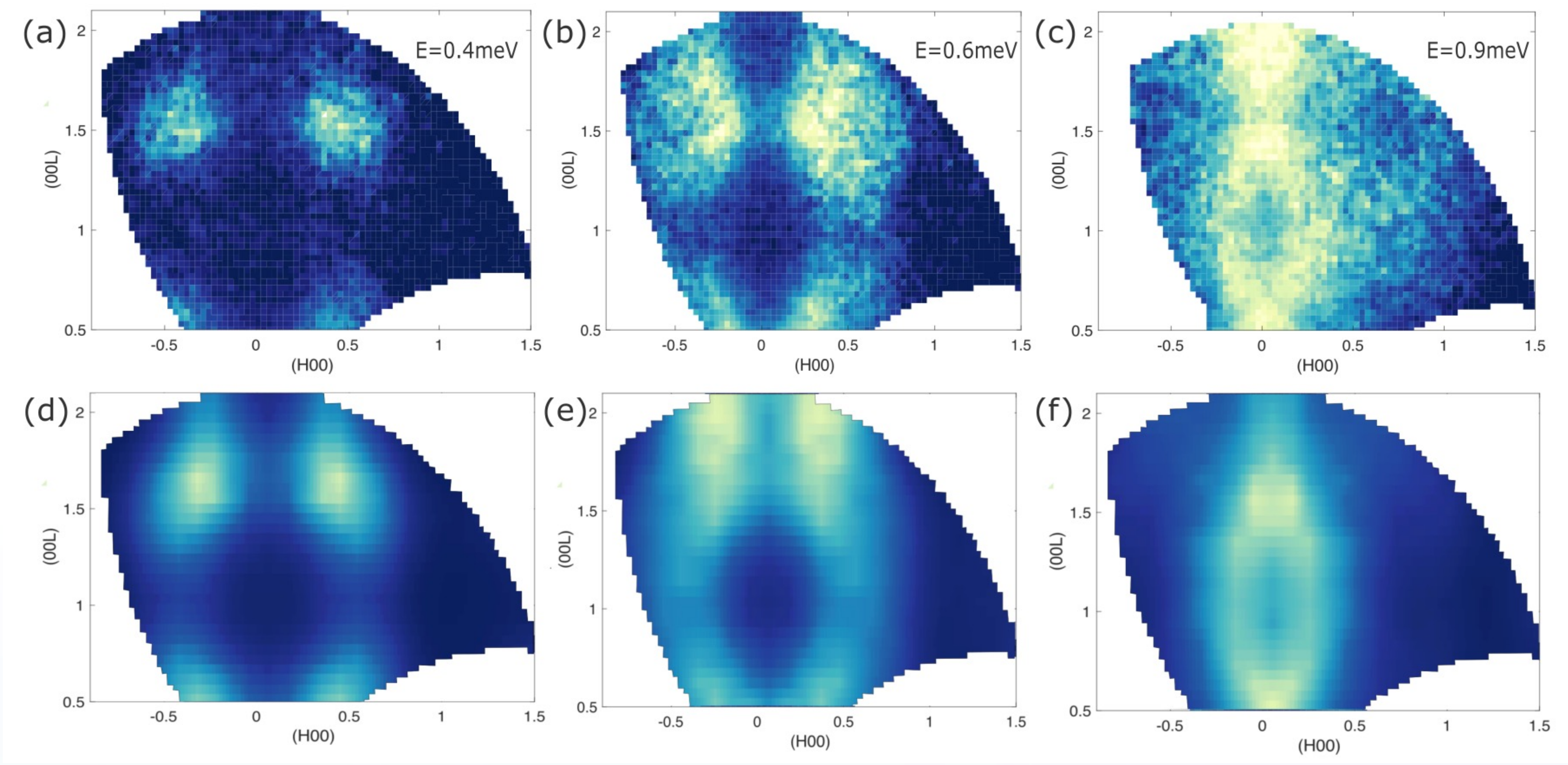
- Inelastic neutron scattering experiments performed at T = 2 K on a powder sample reveal the existence of flat magnetic excitations at energy E ~ 11 meV.
- The flat magnon band may be attributed to the intra-trimer excitations and relatively weak inter-trimer couplings.

5. Spin dynamics at low energies

- A low-energy magnon band with a bandwidth of ~ 0.7 meV is observed at E < 1 meV.
- The dispersion of the low-energy band is not perfectly captured by the minimal model. Therefore, a refined J_1-J_{1p}-J_{3ab}-J_{c12}-D_2-J_{n12} model is considered, where J_{n12} are the two shortest couplings between the second-neighbor layers. The strengths of the added interactions are J_{n1} = -0.02meV, J_{n2} = 0.03meV.



- Comparisons of the experimental and calculated constant-E slices using the refined spin model:



6. Conclusions & Acknowledgements

- Combined diffuse and inelastic neutron scattering experiments were performed on the trimer-host compound $Dy_3Ru_4Al_{12}$ to study its spin correlations above and below T_N.
- Through molecular dynamics and self-consistent Gaussian approximation calculations, we determine the spin Hamiltonian underlying the breathing kagomé lattice formed by the Dy^{3+} spins, which paves the way for the understanding of the ordering phenomena of spin trimers in $Dy_3Ru_4Al_{12}$.

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Email: wanghanyi@mail.ustc.edu.cn