Signatures of Kitaev interactions in van der Waals ferromagnet VI_3

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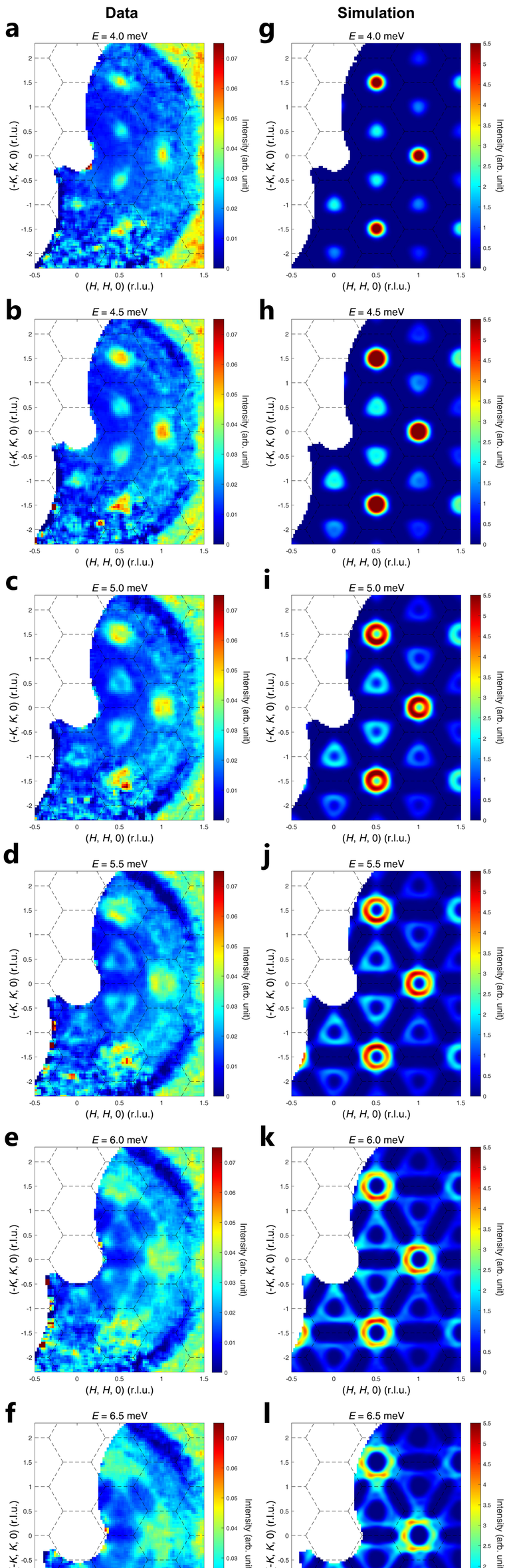
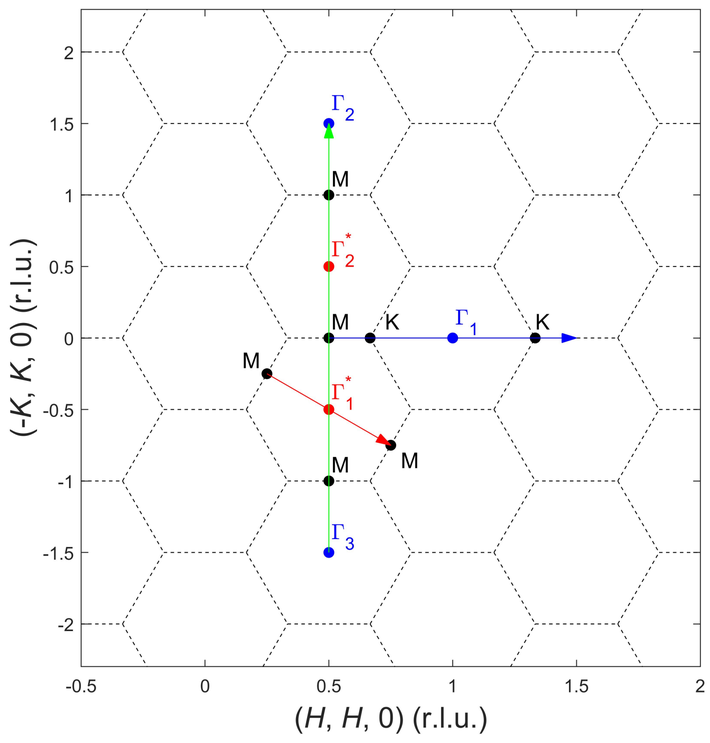
Abstract: **Materials manifesting the Kitaev model, characterized by bond-dependent interactions on a honeycomb lattice, can host exotic phenomena like quantum spin liquid states and topological magnetic excitations. However, finding such materials remains a formidable challenge. Here, we report high-resolution inelastic neutron scattering measurements performed on VI_3, a van der Waals ferromagnetic Mott insulator, covering a wide range of reciprocal space. Our measurements unveil highly anisotropic magnetic excitations in momentum space. Through a comprehensive comparative analysis of various models that incorporate diverse symmetry-allowed magnetic interactions, we find the observed excitations are well captured by a model with a large bond-dependent Kitaev interaction. These results not only help to understand the intriguing properties of VI_3, such as the pronounced anomalous thermal Hall effects and strong pressure/structure dependence of magnetism, but also open a new avenue for exploring Kitaev physics.**

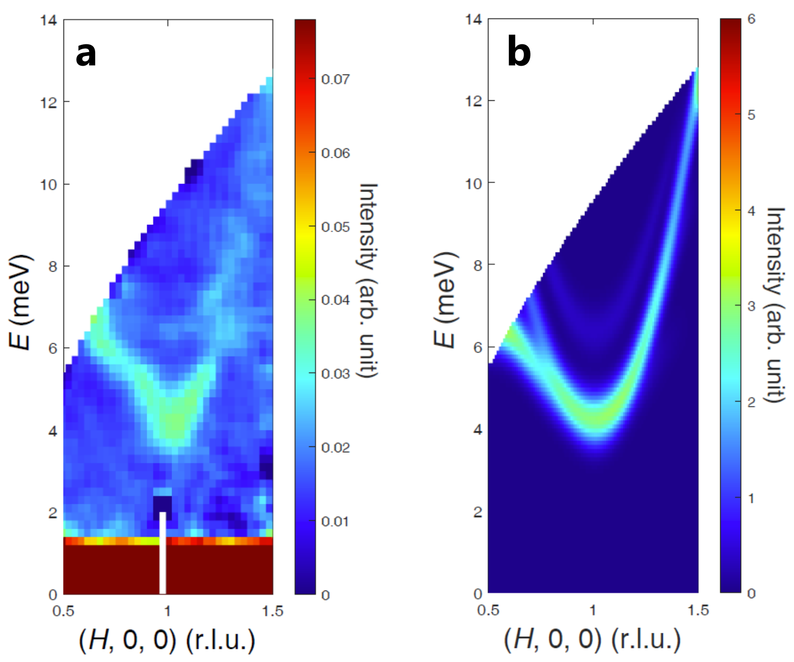
The low-energy magnetic excitations in VI_3



← Constant energy slices of magnetic excitations in VI_3 within the (H, K) plane at T = 5 K. (**a**)–(**f**) Measured constant energy slices with energy transfer E = 4.0, 4.5, 5.0, 5.5, 6.0, 6.5 meV which are integrated over $E \pm 0.5$ meV and $-3 \le L \le 3$. The incident neutron energy is E_i = 15.1 meV. (**g**)–(**l**) Constant energy slices at the specified energies generated through simulations using the *J-K-Γ-Γ′-A* model.

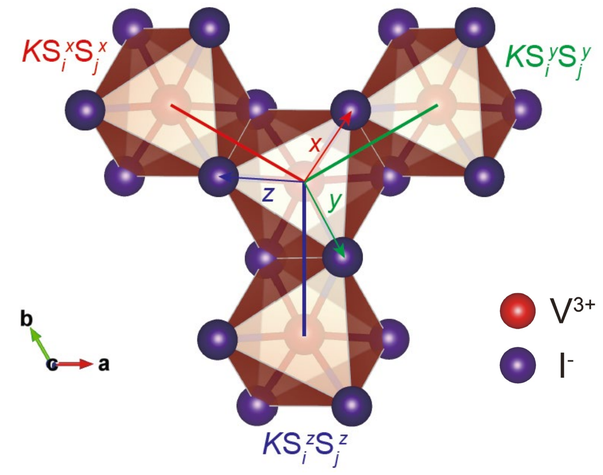


↑ Reciprocal lattice and high symmetry directions



↑ Dispersion of magnetic excitation spectra of VI_3 at 5 K. (**a**) The momentum-dependent magnetic excitations along the M-Γ_1^*-M path at 5 K. **A distinct bend anomaly is observed at ~ 5.5 meV in the low-energy branch.** (**b**) Simulated magnetic excitations along the M-Γ_1^*-M path.

Kitaev interactions



Bond-dependent Kitaev interactions in the local coordinates of VI_3. In materials featuring a honeycomb lattice and edge-shared ligand octahedra, the Kitaev interaction manifests as three kinds of bonds, each associated with bond-dependent Ising axes that are orthogonal to one another.

J-K-Γ-Γ′-A model

$$H = H_{J-A} + H_{\mathrm{K}-\Gamma} + H_{\Gamma'}$$

$$H_{J-A} = \sum_{\langle i,j \rangle} J\boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_j A\left(\boldsymbol{S}_j \cdot \boldsymbol{n}\right)^2$$

$$H_{\mathrm{K}-\Gamma} = \sum_{\langle i,j \rangle \in \alpha\beta(\gamma)} \left[KS_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right)\right]$$

$$H_{\Gamma'} = \sum_{\langle i,j \rangle \in \alpha\beta(\gamma)} \Gamma'\left(S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta\right)$$

Best fitted parameters:

J = -1.04 meV, K = -7.8 meV, Γ = 0.3 meV, Γ' = -1.2 meV, A = -0.6 meV

Conclusion

- Inelastic neutron scattering measurements on VI_3 uncover distinct anisotropic magnetic excitations that are well captured by a model with large bond-dependent Kitaev interactions
- This model suggests that VI_3 resides near the S = 1 Kitaev spin liquid phase despite having a ferromagnetic ground state
- Vanadium magnets could be a new platform for the exploration of Kitaev spin liquid physics

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References

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