Exotic Magnetic Excitations and Magnon-phonon coupling in Honeycomb Ferromagnet VI_3

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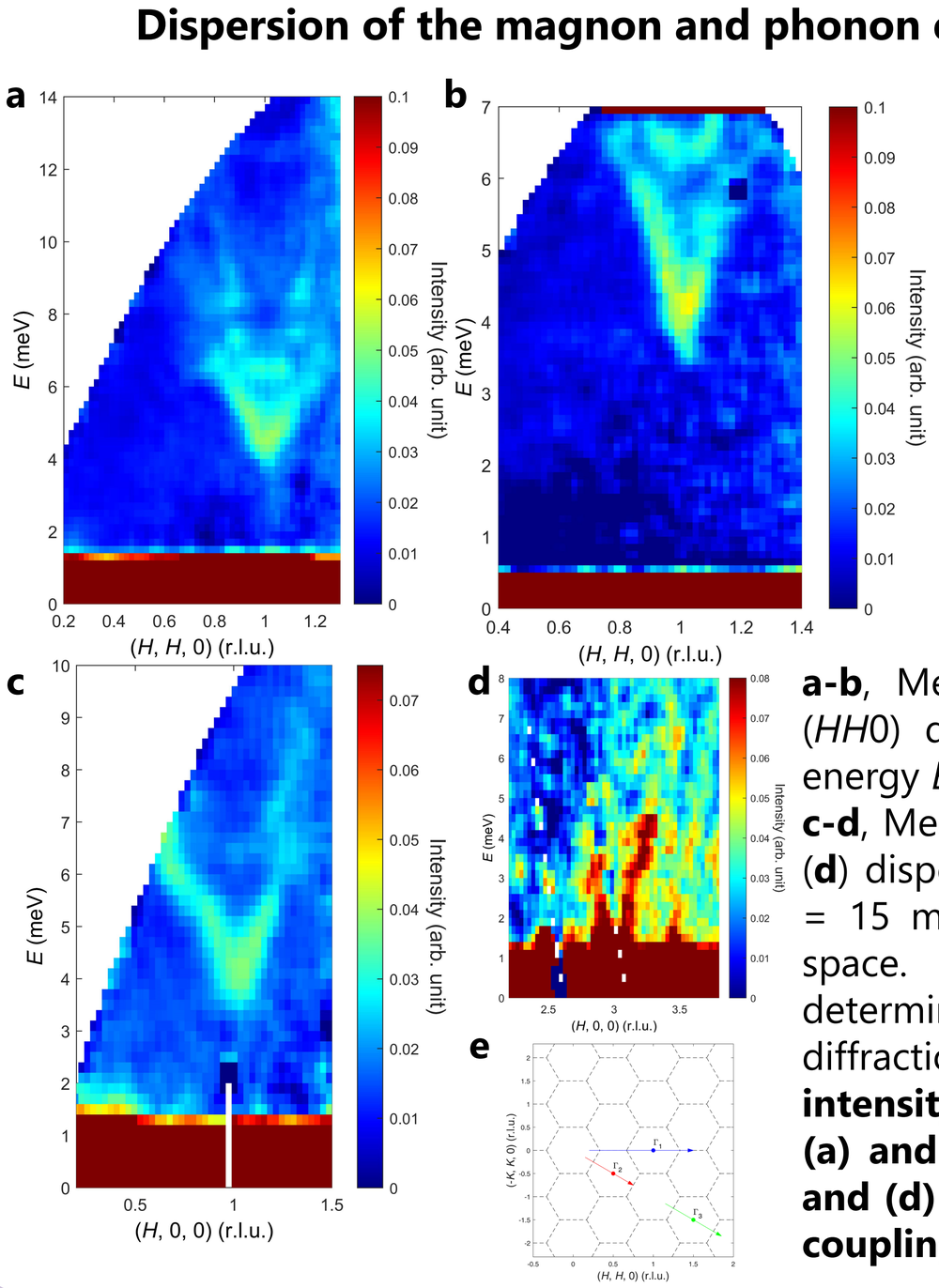
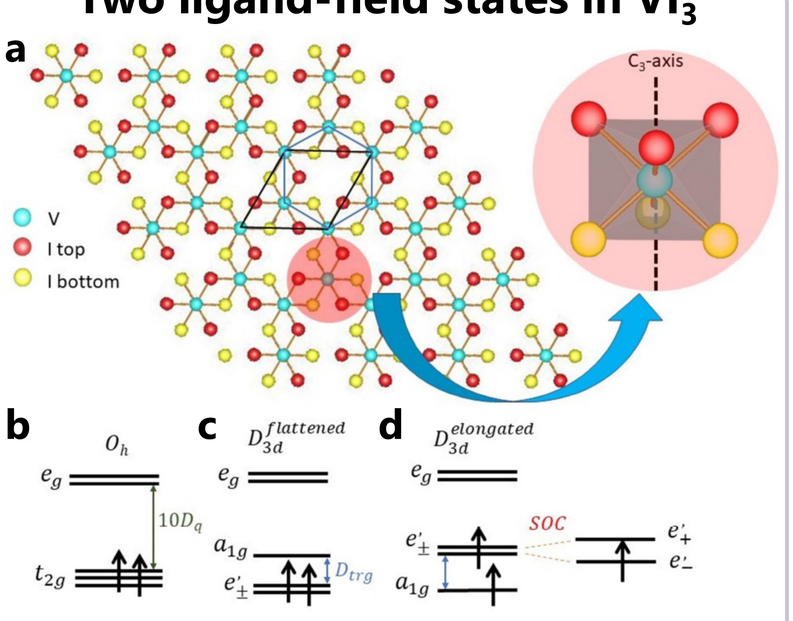
Abstract: **Van der Waals magnet VI_3 exhibits intriguing magnetic ordering properties and great potentials for applications. Here, we present inelastic neutron scattering measurements in single crystalline VI_3. Our experiments reveal two acoustic magnon branches with distinct dispersions: (i) The low-energy branch sharply disperses from Γ points, showing highly anisotropic momentum dependence, which is damped significantly at high energies; (ii) The higher-energy branch disperses more gently from Γ points. These exotic phenomena could be possibly understood by two ligand-field states possessing large and quenched orbital moment with a small energy gap. Furthermore, indication of magnon-phonon coupling was also observed, suggesting an intricate interplay between spin, orbital and lattice degrees of freedom in this system.**

Dispersion of the magnon and phonon excitations in VI_3 at 5 K



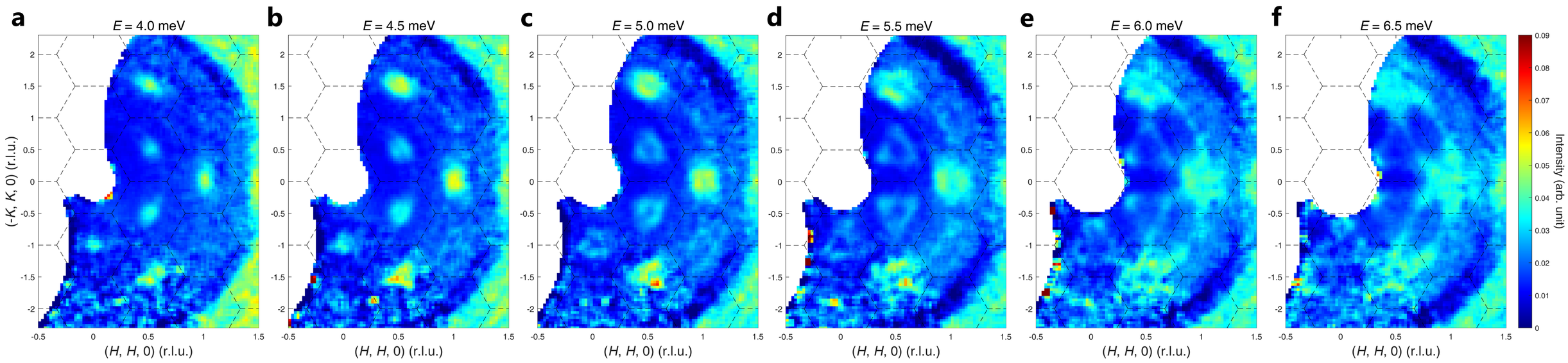
a-b, Measured magnon dispersion in (*HH*0) direction with incident neutron energy E_i = 15 meV (**a**) and 7.7 meV (**b**). **c-d**, Measured magnon (**c**) and phonon (**d**) dispersion in (*H*00) direction with E_i = 15 meV. **e**, Sketch of the reciprocal space. **f**, The magnetic structures determined from our previous neutron diffraction experiments [1]. **The intensity reduction near 7 & 8 meV in (a) and similar kink anomalies in (c) and (d) indicate the magnon-phonon coupling in VI_3.**

Two ligand-field states in VI_3



The observed two acoustic magnon branches can be understood by two ligand-field states with ~ 5.6 meV energy difference calculated by ref. [2] : **1.** The low-energy mode is excited from large-orbital-moment state (**d**, Ground state). Above ~ 6 meV, due to considerable spectral weight is transferred to the transition from (**d**) to (**c**) state, this mode is weakened and damped significantly. **2.** The higher energy mode is excited from quenched-orbital-moment state (**c**), which needs additional excitation energy.

Momentum dependence of the low-energy magnetic excitations in VI_3



The momentum dependence of magnetic excitations in VI_3 measured at T = 5 K for the indicated energies E = 4.0 meV (**a**), 4.5 meV (**b**), 5.0 meV (**c**), 5.5 meV (**d**), 6.0 meV (**e**), and 6.5 meV (**f**). The low-energy magnetic excitations show highly anisotropic momentum dependence dispersing from the Γ points, which cannot be simply described by the Heisenberg model with single-ion anisotropy. This indicates anisotropic two-ion magnetic interactions need to be considered for the large-orbital-moment state of VI_3.

Conclusion

- The magnetic excitation spectra of VI_3 display two acoustic magnon branches, which could be understood by two ligand-field states possessing large and quenched orbital moment with a small energy difference.
- Evidence for magnon and phonon coupling was also revealed, indicating the complex interplay between spin, orbital and lattice degrees of freedom in this system.

Acknowledgments

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References

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[2] D. Hovancik, C. Piamonteze, J. Pospisil, K. Carva, V. Sechovsky. arXiv: 2210.11278 (2022).

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