Intertwined dipolar and multipolar order in the triangular-lattice magnet $TmMgGaO_4$

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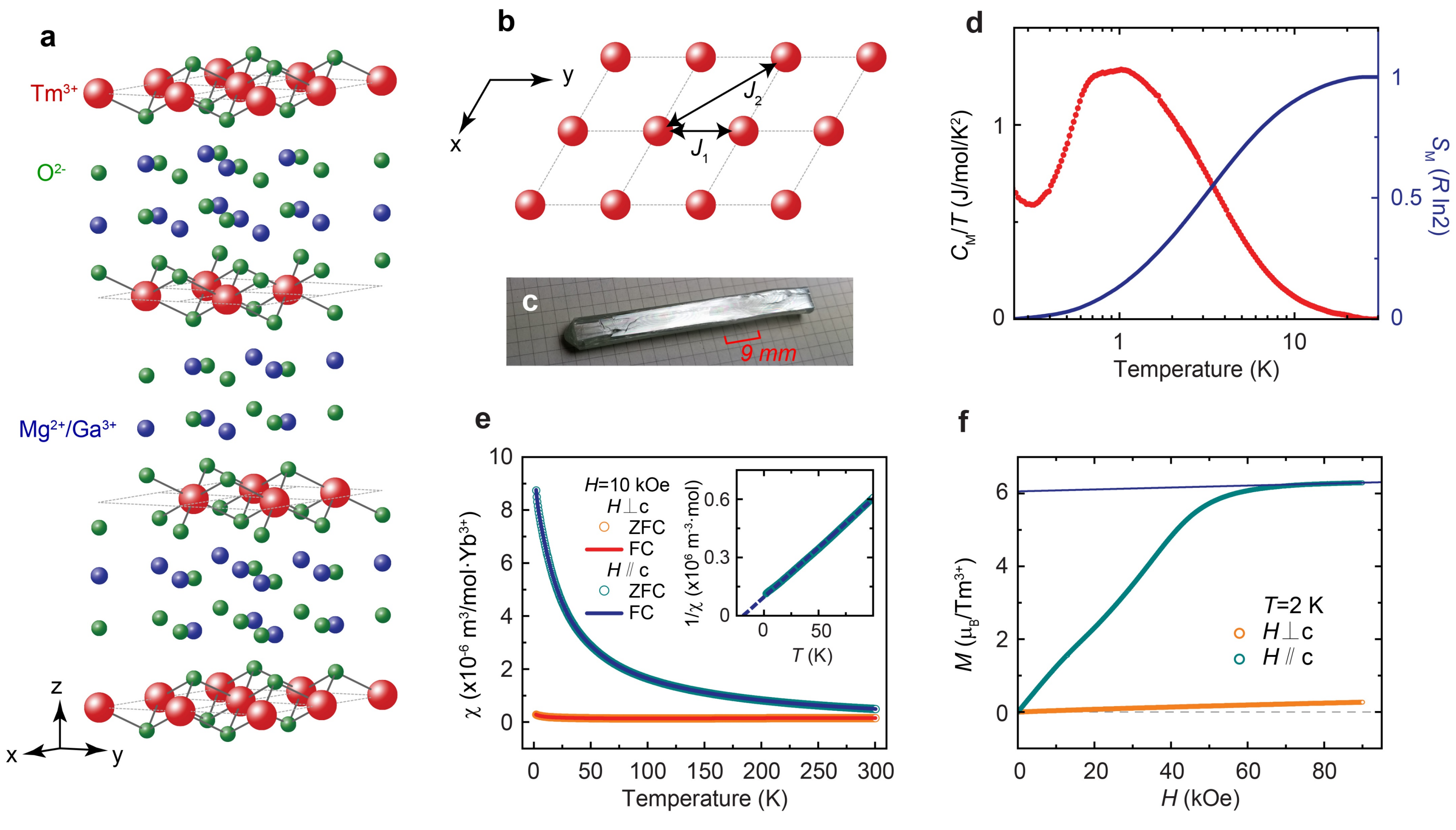
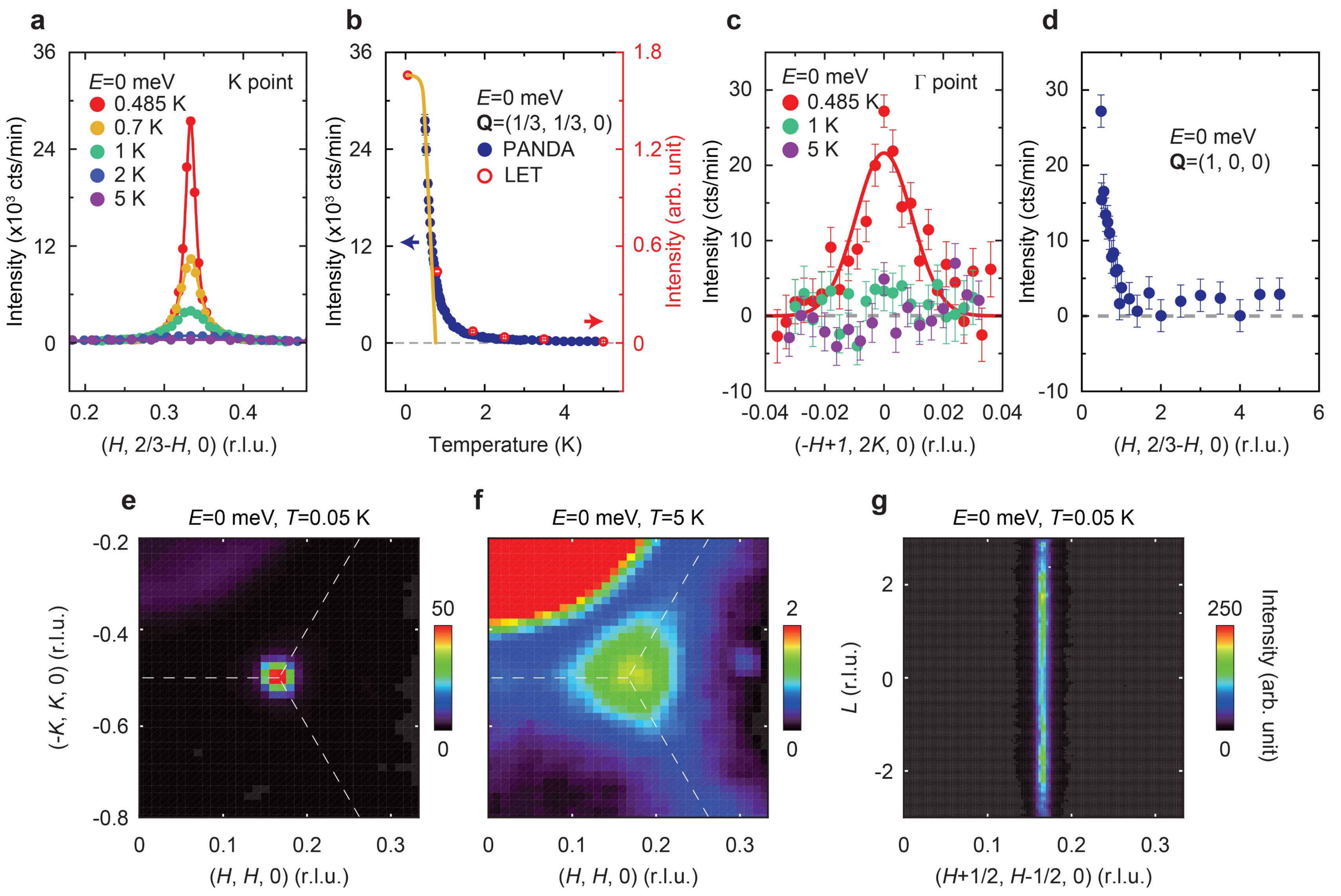
Abstract

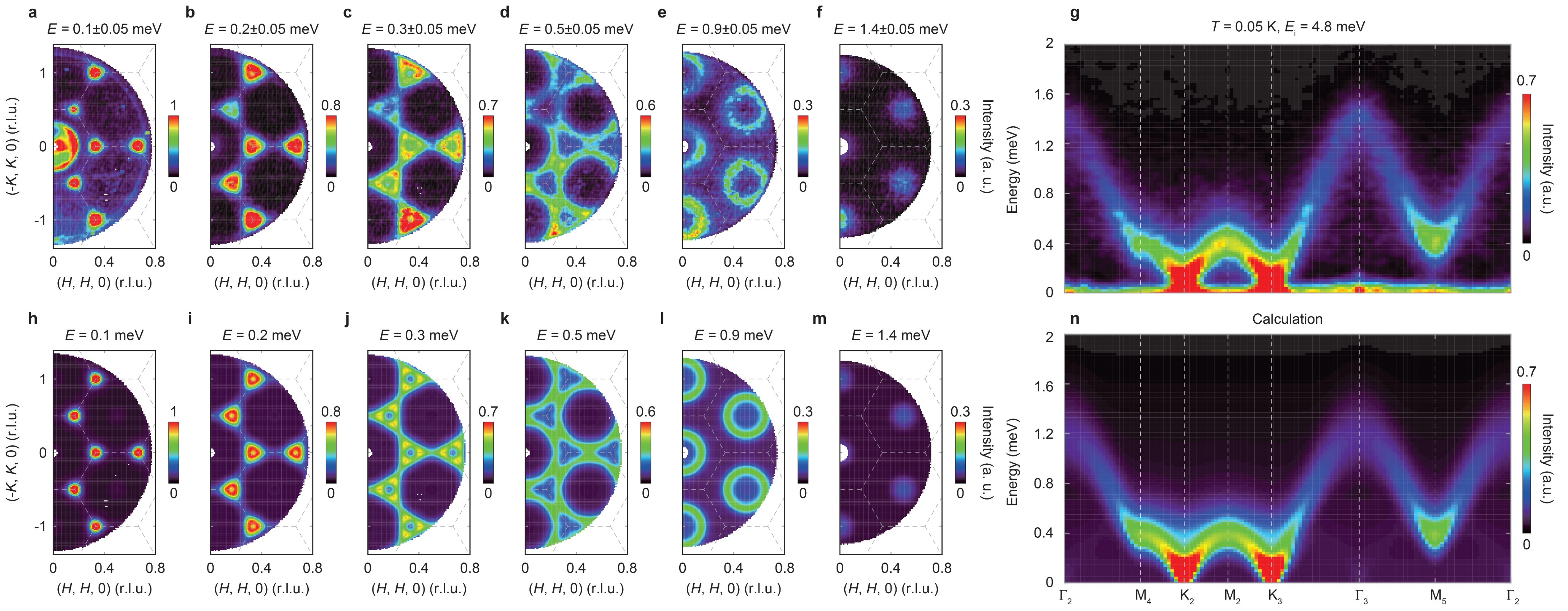
Certain magnetic materials exhibit exotic hidden-order phases, in which the order parameters are not directly accessible to conventional magnetic measurements. Here we study the rare-earth triangular-lattice magnet $TmMgGaO_4$. Clear magnetic Bragg peaks at K points are observed with sharp and highly dispersive spin excitations that cannot be explained by a magnetic dipolar order, but instead is the direct consequence of the underlying multipolar order that is "hidden" in the neutron diffraction experiments. We demonstrate that the observed unusual spin correlations and thermodynamics can be accurately described by a transverse field Ising model on the triangular lattice with an intertwined dipolar and multipolar order.



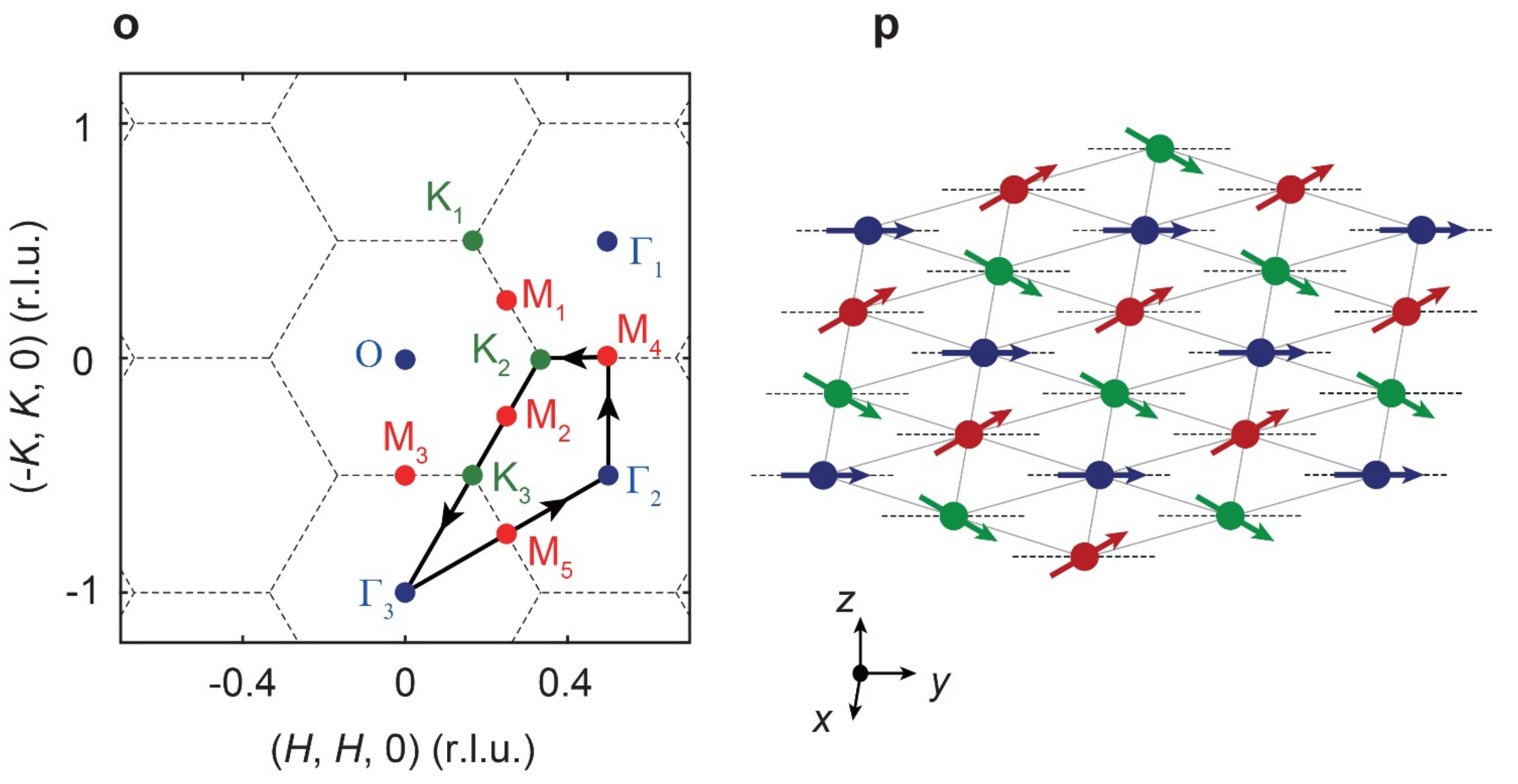
a, b, Schematic diagram of $TmMgGaO_4$ crystal structure. **c,** A photograph of a representative single crystal. **d,** Magnetic heat capacity and the corresponding magnetic entropy (0 T). **e,** Temperature dependence of the magnetic susceptibility. The inset shows the linear fitting of the inverse susceptibility with Curie-Weiss temperature of -19.1 K. **f,** Field dependence of the magnetization at 2 K. Linear fitting of the magnetization at high field gives Lande-*g* factor of 12.11(5) (solid blue line).



a, b, Constant energy cuts across the magnetic dipolar Bragg peak, the K point, and the temperature dependence of its intensity. **c, d,** Constant energy cuts and temperature dependence around the magnetic multipolar Bragg peak, the Γ point. **e,f,** Momentum dependence of the magnetic Bragg peak at the K point. **g,** *L* dependence of the elastic signals at K point.



a-g, Measured spin wave excitations at *T*=0.05 K. **h-n,** Calculated spin wave dispersion by linear spin wave theory ($J_1^{zz}=0.54\,meV, J_2^{zz}=0.026\,meV,\ \ h=0.62\,meV$). Due to the multipolar nature, only S^{zz} are considered. **o,** Sketch of the reciprocal space. **p,** Schematic of the three-sublattice magnetic structure. The multipolar components (S^x and S^y) form a ferro-multipolar order while the dipolar one (S^z) leads to the ordering at K points.



MODEL: The low energy degrees of freedom of Tm^{3+} ions is a pair of nearly degenerated singlets (denoted as $|\Psi^{\pm}\rangle$) that are separated by an energy gap *h* induced by the crystalline electric field. The effective spin-1/2 operators can be defined as:

$$S_i^x = i/2(|\Psi_i^-\rangle\langle\Psi_i^+| - |\Psi_i^+\rangle\langle\Psi_i^-|),\ \ S_i^y = 1/2(|\Psi_i^+\rangle\langle\Psi_i^+| - |\Psi_i^-\rangle\langle\Psi_i^-|),\ \ S_i^z = 1/2(|\Psi_i^+\rangle\langle\Psi_i^-| + |\Psi_i^-\rangle\langle\Psi_i^+|)$$

The transverse components S^x and S^y are time-reversal even and transform as multipolar moments under space group operations that do not couple to neutrons and external field directly while the S^z component are odd under time reversal and transform as dipoles. The effective model is the transverse field Ising model that takes the form:

$$H = \sum_{\langle ij\rangle} J_1^{zz} S_i^z S_j^z + \sum_{\langle\langle ij\rangle\rangle} J_2^{zz} S_i^z S_j^z - h \sum_{\langle ij\rangle} S_i^y$$

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