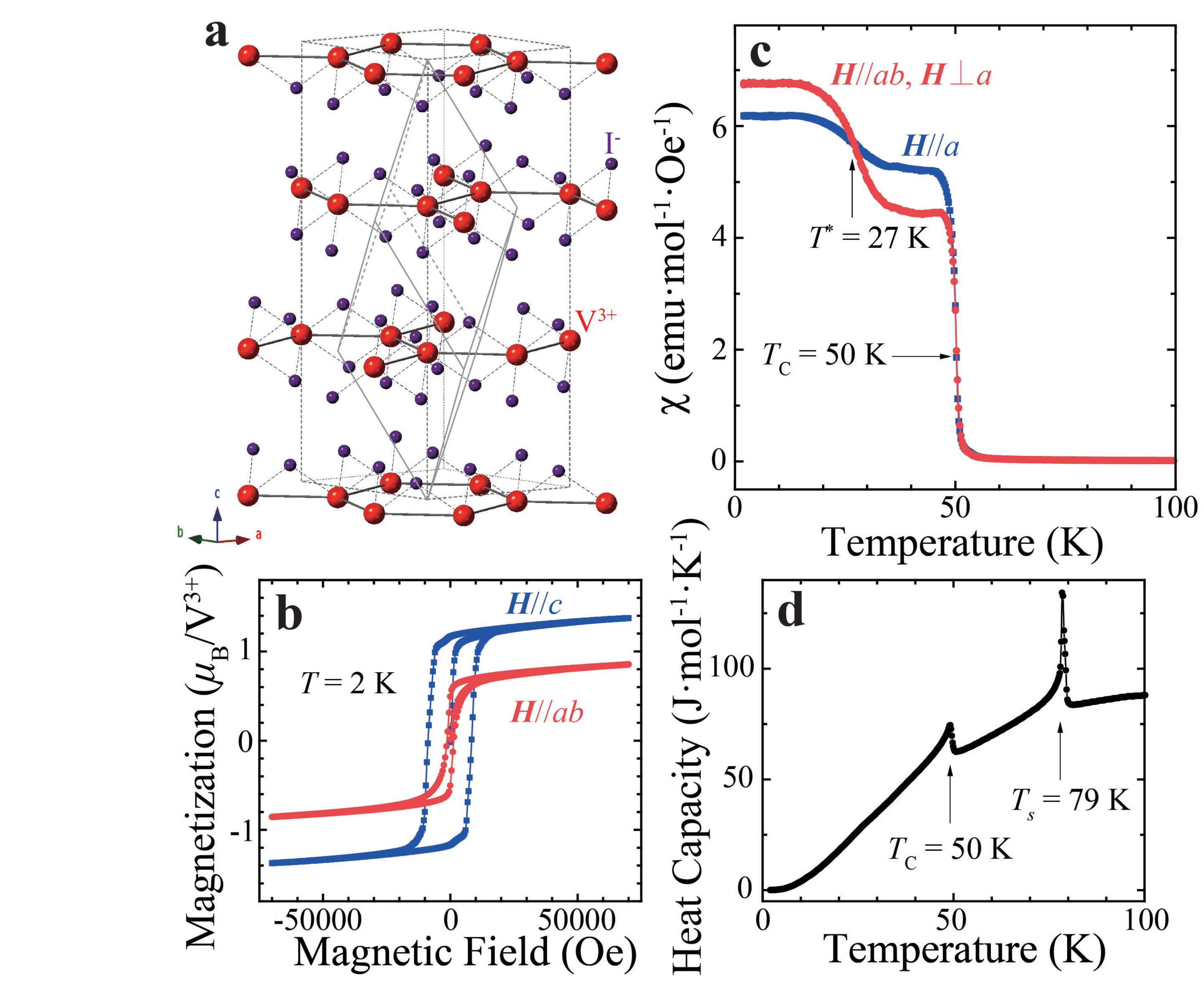
Spin reorientation in canted magnetic structures of the van der Waals ferromagnet VI_3

Yiqing Gu[1#], Yiqing Hao[1#], Yimeng Gu[1], Erxi Feng[2], Huibo Cao[2], Songxue Chi[2], Xiaoping Wang[2], Jun Zhao[1,3*]

[1]State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China [2]Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA [3]Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China

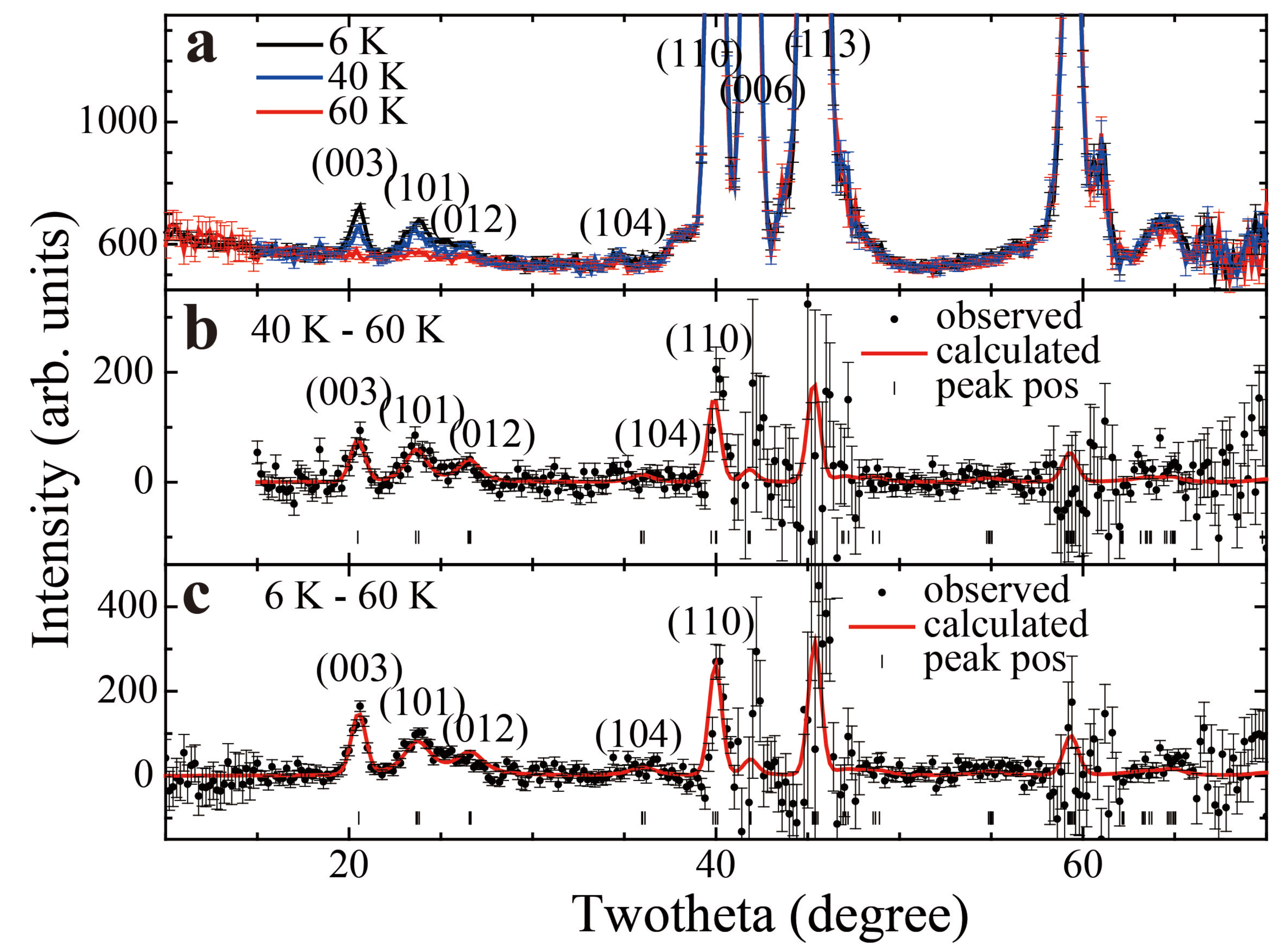
Abstract: Van der Waals ferromagnet VI_3 exhibits complex magnetic properties and great potentials for applications, but the microscopic information to its magnetic ordered states has yet been confirmed. Here, we reveal by neutron diffraction and susceptibility measurements that the magnetic ground state of VI_3 is a long-ranged ferromagnetic order. Despite the robustness of magnetic canting angle $\theta \sim 35°$, an in-plane spin reorientation is observed accompanied by structure distortion in honeycomb lattice. Such strong magnetic anisotropy and susceptible magnetoelastic effects are understood by strong spin-orbit coupling in VI_3. Our results elucidate the unneglectable role of orbital moments in VI_3 and provide a new paradigm in designing highly tunable spintronic devices.

Crystal structure and thermodynamic property of VI_3



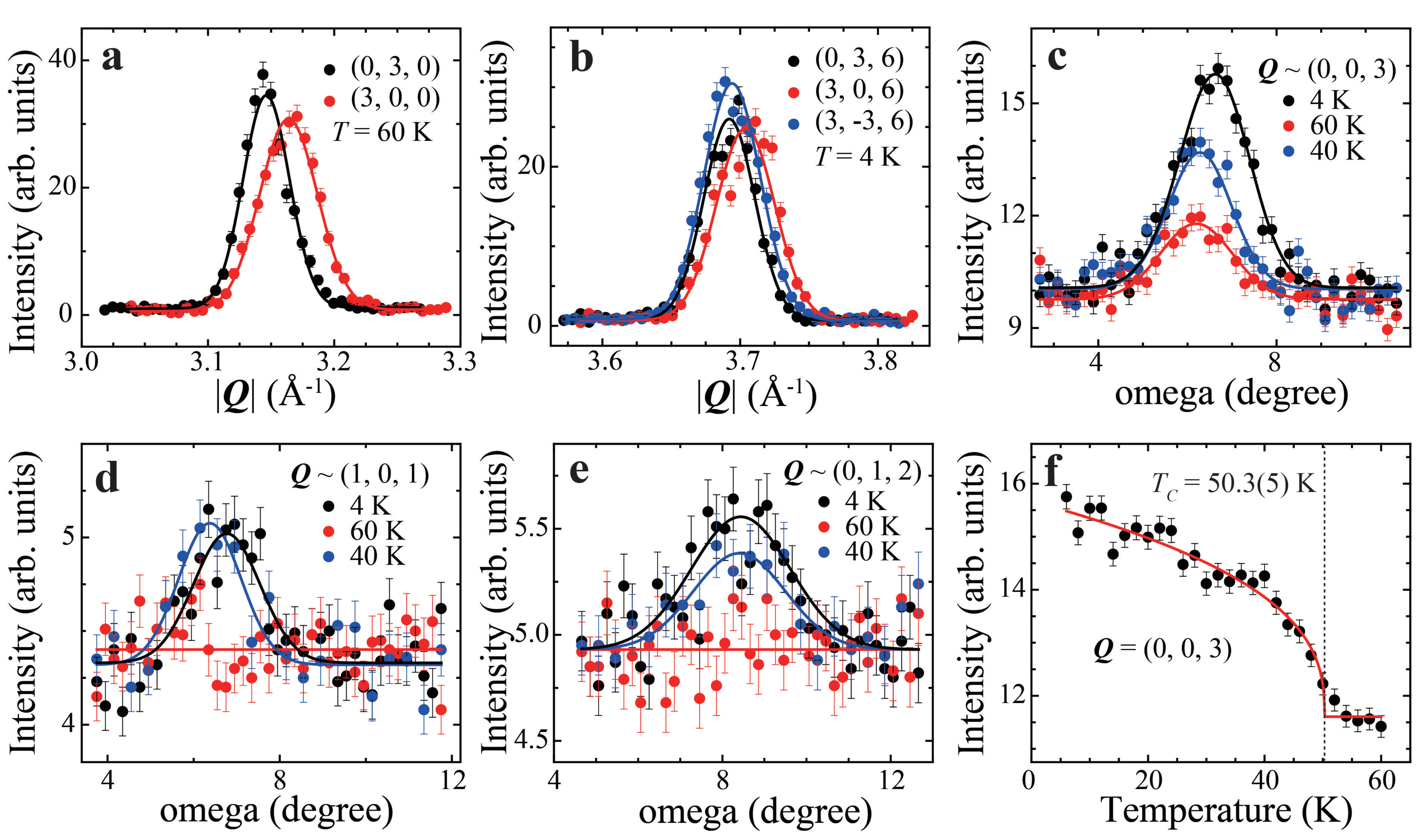
a, Crystal structure of VI_3. **b**, Field-dependent magnetization in *c*-axis (blue) and *ab*-plane (red) at 2 K, showing clear ferromagnetic hysteresis loops. **c**, Field-cooled magnetic susceptibilities. 200 Oe field was applied along hexagonal *a*-axis (blue) and its orthogonal direction within *ab*-plane (red). T^* represents the proposed transition temperature of spin reorientation. **d**, Temperature-dependent specific heat of VI_3, two peaks are found at T_C = 50 K and T_s = 79 K.

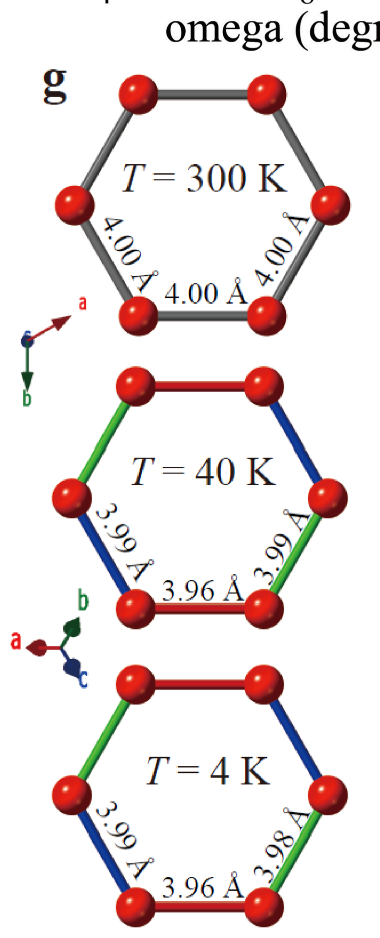
Neutron powder diffraction (NPD) measurements



a, NPD intensities of VI_3. The wavelength used is 2.359 Å. Black, blue, and red lines represent diffraction intensities measured at 6 K, 40 K and 60 K, respectively. The strong magnetic peaks and main nuclear Bragg peaks are indexed using hexagonal notation. **b-c**, Rietveld refinement of the magnetic peaks at 40 K and 6 K using canted ferromagnetic structure. For both 40 K and 6 K, magnetic moments are tilted 35(2)° from the normal to VI_3 layers, showing the robustness of magnetic canting angle to the monoclinic-to-triclinic transition at 32 K.

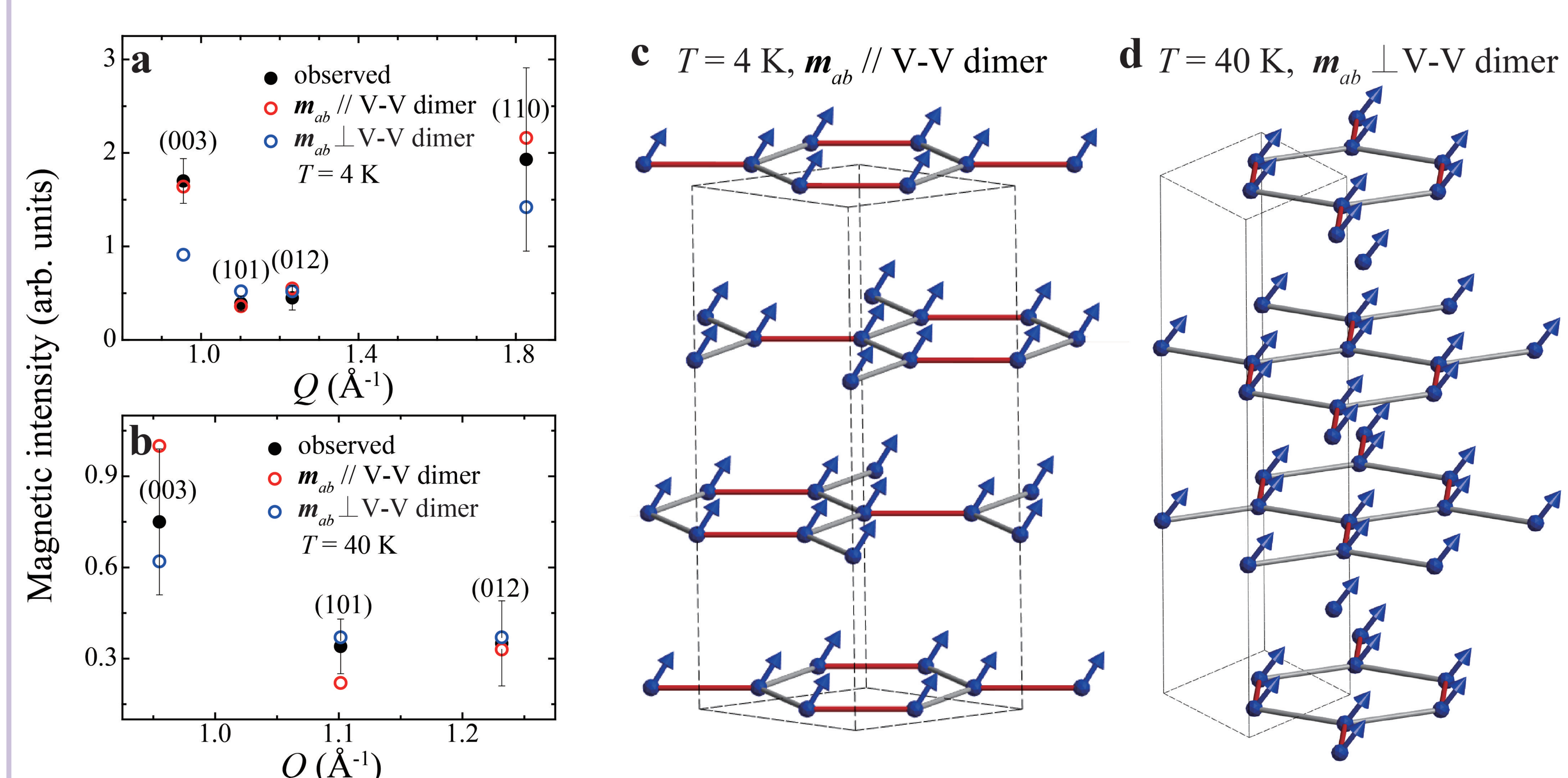
Neutron diffraction on single-crystalline VI_3





Nuclear (**a-b**) and magnetic (**c-e**) Bragg peaks measured in single-crystalline VI_3. The wavelength used is 1.542 Å. **f**, Temperature dependence of $(003)_h$ magnetic Bragg peak. The red line shows fitting using critical exponential function, yielding T_C = 50.3(5) K and critical exponent β = 0.19(2). **g**, V-V bond lengths at different temperature. Red, blue and green color indicates three types of symmetric inequivalent V-V bonds below T_s.

Magnetic structure refinement of VI_3



a-b, Magnetic structure refinement of VI_3 at 4 K and 40 K. Black dots represent the observed intensities, red and blue circles represent calculated intensities assuming the in-plane moment of V^{3+} parallel and perpendicular to V-V dimers, respectively. **c-d,** Magnetic structures of VI_3 at 4 K and 40 K. Blue arrows represent moment directions and red bonds represent V-V dimers. The refined moments are $\boldsymbol{m}_{//\mathrm{dimer}}$ = 0.84(5) μ_B, $\boldsymbol{m}_c$ = 1.08(8) μ_B at 4 K and $\boldsymbol{m}_{\perp\mathrm{dimer}}$= 0.51(5) μ_B, $\boldsymbol{m}_c$ = 0.65(6) μ_B at 40 K. The robust magnetic canting angle is confirmed by both powder and single-crystal neutron diffraction measurements.

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