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Giant shifts of crystal field excitations with temperature as a consequence of internal magnetic exchange interactions

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Crystal field theory, invented in the 1930s by Hans Bethe, provides an explanation of the crystal field excitations (CFE) observed in inelastic neutron scattering (INS) spectra of rare-earth compounds [1]. However, some long withstanding problems remain. Our inelastic neutron scattering experiments on vanadates CeVO3 and TbVO3 did reveal an unexpected large shift of the energies of the crystal field excitations as a function of temperature. Thus far, only few publications on INS experiments mention shifts in crystal field excitation (CFE) energy in spectra above and below magnetic phase transition temperatures [2,3,4]. Recent IR transmission measurements also identified a CFE energy shift in hexagonal DyMnO3 with temperature and upon the application of an external magnetic field [5]. However, no studies report a detailed microscopic theory and to the best of our knowledge does not exist in literature.

The vanadates CeVO3 and TbVO3 share the same orthorhombic Pbnm crystallographic structure featuring tilted, corner-sharing octahedra and possess a Cz-type antiferromagnetic structure below Néel temperatures 124 K and 110 K, respectively [6-9]. In both vanadates the CFE energies shift strongly below the magnetic phase transitions.

We have used quantum-mechanical point-charge calculations to determine the energies of observed CFEs to model their large shift as a function of temperature. Two mechanisms have been simulated: (i) distortions of the crystallographic lattice due to magnetostriction, or (ii) internal magnetic exchange interactions with CF levels at the onset of the magnetic order. The effect of lattice distortions measured by neutron diffraction [7,8] produces a negligibly small shift of CFE energy, therefore cannot drive the shift. Results accounting for internal magnetic exchange fields arising from the ordered V3+ spins reveal a shift which agrees excellently with neutron data. The CFE energy shift is well reproduced with the same shift in CFE energy and intensity. Therefore, the unexpected large shift of CFE energies with temperature has been confirmed by point-charge model theoretical calculations and can be attributed to an internal magnetic exchange interaction.

In addition to the CFEs, spin-wave excitations (magnons) are present in both vanadate materials below the magnetic phase transition. In TbVO3 there appears to be an anticrossing-like behaviour between magnon and CFE at 14 meV. Such an anticrossing has been reported in far-IR transmission investigations in Tb3Fe5O12 garnet [12]. In order to investigate this observation in TbVO3, magnon dispersion calculations have been performed to clarify the exact nature of the interaction.

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