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Magnetic excitations and heat capacity of fayalite, Fe_2SiO_4

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Abstract

We have used inelastic neutron-scattering measurements to study the magnetic excitations in the antiferromagnetic and paramagnetic phases of polycrystalline fayalite, Fe_2SiO_4. Sharp, nondispersing excitations are found in the ordered state, at 3.3, 5.4, 5.9, and 11.4 meV, and are interpreted as arising from the spin-orbit manifold of the Fe^{2+} ions. These excitations are increasingly damped with increasing temperature, merging into a quasielastic continuum near the 65 K Neel temperature, although their energy does not vary with temperature. We have calculated the contribution of the heat capacity arising from these magnetic excitations and found that it compares favorably with the magnetic heat capacity deduced experimentally. Our analysis indicates that the M1 and M2 sites behave distinctly. The M1 site behaves quasi-locally and appears in the heat capacity as a Schottky anomaly that explains the shoulder in the heat capacity curve near 20 K, while the M2 site contributes predominantly to the critical lambda anomaly. The behavior of fayalite illuminates the nature of magnetic states in several related minerals, including others that also show shoulders and lambda anomalies in the heat capacity (tephroite), those that show only lambda anomalies (cobalt olivine and liebenbergite), and those that show only non-lambda anomalies (bronzite, anthophyllite, and almandine). We find no evidence to support the recent claim that some transition metal silicate and germanate olivines exhibit strong geometric frustration.

Keywords: Fayalite, neutron diffraction, calorimetry, phase transition, magnetic properties