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Magnetic field dependence of spin-forbidden electronic excitations reflects the Haldane or paramagnetic ground state V.C. LONG, J.R. MONTAGUE, A.C. KOZEN, Colby College, Waterville ME, X. WEI, National High Magnetic Field Laboratory, B.R. LANDRY, K.R. PEARSON, M.M. TURNBULL, C.P. LANDEE, Clark University, Worcester MA — We compare the zero-field and magnetic field-dependent optical spectra of the Haldane chain compound NENB ($Ni[en]_2NO_2BF_4$; en = $C_2N_2H_8$) and the paramagnetic compound, $Ni(en)_3(ClO_4)_2$,H_2O. Due to similar electronic coordination of Ni^{2+}, the two materials show similar zero-field $d-d$ electronic transitions, including a spin-forbidden (SF) transition at 1.58 eV, overlapping a broad spin-allowed band at 1.45 eV. The relatively greater intensity of the SF band in the Haldane compound suggests activation by a spin exchange mechanism, whereas a spin-orbit coupling origin is likely in the paramagnet. A second narrower SF spin flip transition appears in NENB at 1.66 eV. In both compounds, the SF excitations are sensitive to applied field H. In NENB, the SF intensity is suppressed by H, consistent with behavior of spin exchange-activated bands. In $Ni(en)_3(ClO_4)-2,H_2O$, the SF field sensitivity appears to combine an energy shift and intensity decrease. Details of the H dependence reflect the magnetic ground state of the material: the field sensitivity commences only above $H_C \approx 10$ T, in the Haldane compound, whereas the field-induced modifications begin immediately at $H = 0$ T in the paramagnet.

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