Comparative Study of Field-Dependent Electronic Excitations in Closely Related Ni^{2+} Compounds

V.C. Long and L.A. LaViolet (Colby College, Physics); C.P. Landee (Clark University, Physics), R.D. Willett (Washington State U., Chemistry), S.A. McGill (NHMFL, Visible Optics)

Introduction

A quartet of nearly isostructural Ni^{2+} spin chains have recently presented a puzzle because three are Haldane systems, characterized by a spin gap and corresponding critical field H_C in the magnetization, whereas the fourth exhibits spin glass behavior [1]. The four are NENP, NENB, NINO, and NTNB ($Ni(en)_2NO_2ClO_4$, $Ni(en)_2NO_2BF_4$, $Ni(tn)_2NO_2ClO_4$, $Ni(en)_2NO_2BF_4$; with en = $C_2N_2H_8$ and tn = $C_2N_2H_{10}$);NTNB is the spin glass. Previous magneto-optical studies revealed that H_C also appears in field-dependent absorption intensities of NENB and NENP [2].We have investigated NTNB and NINO by the same method in order to shed more light on the unexpected magnetic behavior of NTNB.

Experimental

We measured the low temperature polarized absorbance of NINO and NTNB single crystals at Colby College and the field (H) dependent absorbance at NHMFL using a 30 T DC resistive magnet. We calculated the absorbance difference (AD) to highlight small changes in the absorption bands with applied field, where AD = A(H) – A(H=0T).

Results and Discussion

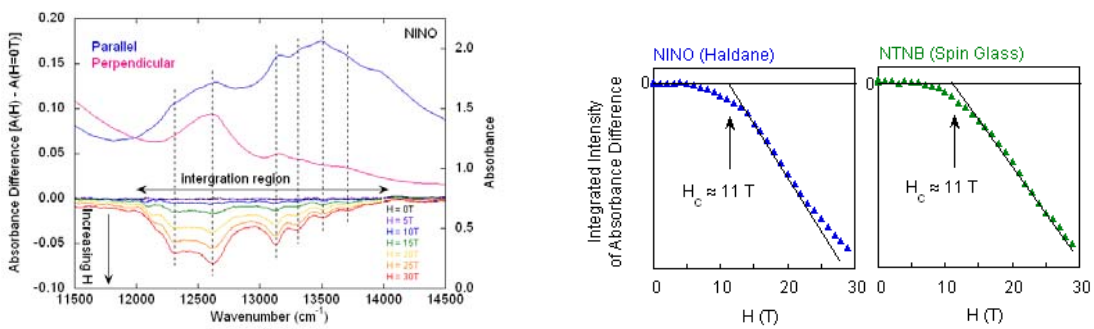

Fig. 1 On the left (upper region of plot) is plotted the low temperature zero-field spin forbidden bands of NINO and (lower region of plot) the field sensitivity of those bands, displayed as the absorbance difference. NTNB has nearly identical spectra. On the right is plotted the integrated intensity of the absorbance difference (area under the AD curves) versus field for the integration region shown by a double arrow on the left, for both NINO and NTNB.

In Fig. 1 left, the absorbance difference spectra reveal the field-dependent spin-forbidden Ni^{2+} *d-d* absorption bands with several vibrational replicas spaced by ~200 cm^{-1}. Similar to the related Haldane compounds NENB and NENP [2], the SF bands of NINO and NTNB are suppressed by applied field. Surprisingly, both NINO and NTNB show a critical field in the integrated intensity versus field (Fig. 1, right), consistent with a Haldane state in NTNB, in contrast to earlier reports [1].

Conclusions

Our results suggest either 1) an interesting coexistence of Haldane and spin glass phases in NTNB or else 2) sample variability due to difference defect concentrations or to self annealing behavior of NTNB samples over time.

Acknowledgements

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References

[1] Willett, R.D., *et al*., EMRS2007, Strasbourg, France (2007).
[2] Long,V.C., *et al*., Phys. Rev. B **76**, 024439 (2007).