HIGH-FIELD ELECTRON SPIN RESONANCE STUDIES OF A NEW HALDANE SPIN SYSTEM NENB

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The investigation of low-dimensional quantum magnets in strong magnetic fields is currently a very attractive area of research. Among others, $S=1$ Heisenberg antiferromagnets are particularly interesting objects for investigation. In accordance with Haldane’s conjecture [1], the ground state of an isotropic $S=1$ chain is a spin singlet, separated from the first excited state by an energy gap $\Delta \sim 0.4 J$ (where $J$ is the intrachain spin interaction). Although the Haldane conjecture is widely accepted, there are many open questions regarding high-field properties of Haldane materials. Here we report on a synthesis and ESR characterization of a new $S=1$ spin chain material $\text{Ni}((\text{C}_2\text{H}_8\text{N}_2)\text{NO}_2\text{Cl})_4$ (hereafter NENB).

The compound was synthesized in similar fashion to NENP [2]. $\text{Ni}((\text{BF}_4)_2\cdot6\text{H}_2\text{O}$ and 3 equivalents of 1,2-ethanedia mide (en) were dissolved in water and the solutions combined. The resulting purple solution grew large crystals of $\text{[Ni(en)\text{3}]}(\text{BF}_4)_2$ over the course of a few days. The product was isolated and air-dried to give a greater than 65% yield of purple crystals. NENB and the Zn-doped material were then prepared as follows:

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2 \text{[Ni(en)\text{3}]}(\text{BF}_4)_2 + (1-x) \text{Ni}((\text{BF}_4)_2\cdot6\text{H}_2\text{O} + x \text{Zn}((\text{BF}_4)_2\cdot6\text{H}_2\text{O} + 3 \text{NaNO}_2 \rightarrow \text{[Ni}_{1-y}\text{Zn}_y\text{(en)2(NO}_2\text{)](BF}_4)
\]

The reagents were combined in aqueous solution and the water allowed to evaporate slowly over the course of several weeks, resulting in the formation of large (>50 mg) single crystals of NENB. Crystals were harvested at low yields (typically 20-40%) to reduced changes in composition of the doped crystals, and the identity of the product confirmed by infrared spectroscopy and powder X-ray diffraction.

The ESR measurements were done in the frequency range of 140-700 GHz and in fields up to 25 T, using a tunable-frequency BWO (Backward Wave Oscillator) spectrometer [2]. A complete frequency-field diagram of magnetic excitations in NENB is shown in Fig. 1. To the best of our knowledge this is the most complete frequency-field diagram of magnetic excitations in Haldane spin chain compounds, obtained using high-frequency ESR. Several modes have been observed. The behavior of the modes A, B, C, D, E and G is well agreed with a frequency-field dependence of magnetic excitations found in NENP and NINO [3 and ref. herein]. In addition, two modes (F and H), whose origin is not clear at the moment, were observed in the ESR spectra. The interpretation of the excitation spectrum of NENB, particularly its high-field mode E is in progress. As pointed out by Huang and Affleck [4], including the Dzyaloshinskii-Moriya term in the analysis is very essential.

References