The Millikelvin Temperature Dependence of the Magnetization of an *S* = 1 Linear-Chain Antiferromagnet with *D*/*J* ≈ 1

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Introduction

Linear-chain, integer-spin antiferromagnets, commonly known as Haldane systems, remain the focus of considerable interest for numerous fundamental and technological reasons. After Haldane identified the gap in these systems, the boundary between the Haldane and Large-*D* phases was identified at $D/J \approx 1$ [1], where *D* and *J* are the zero-field splitting and nearest-neighbor interaction parameters, respectively. Although no condensed matter system has been localized near this critical boundary, theoretical and numerical interest continues due to the possibility of identifying the transition in quantum gases. In fact, recent numerical work suggests that the critical ratio is $[D/J]_C = 0.97$ [2,3], while the ground state properties have been explored numerically [4,5].

After $[Ni(HF_2)(3\text{-}Clpy)_4]BF_4$ was identified as an $S = 1$ linear-chain antiferromagnet with $D/J \approx 1$ [6], our work was initiated to explore this system at temperatures below the previous lower limit of nominally 0.5 K [7]. The crystal structure of $[Ni(HF_2)(3\text{-}Clpy)_4]BF_4$ consists of one-dimensional zig-zag chains, whereby NiN_4F_2 octahedra are linked only by HF_2^- ligands. Fitting of the low-field, high-temperature $\chi(T)$ data suggested $J = 4.86$ K, whereas modeling of UV-Vis data indicated $D \approx 4.3$ K [6]. Since $D/J \approx 0.88$ and an unexpected magnetic signal was detected at 0.5 K [6], the present work was preformed to measure the temperature dependence of the magnetization in fields up to 10 T.

Experimental

Low frequency (53 Hz) magnetic susceptibility studies were performed in the Williamson Hall Annex of the NHMFL High-B/T Facilities located at UF. The experimental approach has been described elsewhere [8], and for the present work, isothermal data were collected at 50 mK and then nominally at 100 mK intervals from 100 mK to 1 K while sweeping the magnetic field up to 10 T. Due to the solvent used for the sample, a sample holder was constructed with Kel-F® by *3M* (now known as *Daikin* Neoflon® PCTFE).

Results and Discussion

Although the analysis of the data is continuing, the presence of an additional magnetic signal below 500 mK has been detected [7]. The temperature dependence of this feature, along with its magnetic field dependence, is being addressed by the on-going analysis, which is complicated by the presence of noise in the data from nominally 0.5 T to 2 T. These noisy features are thought to arise from flux jumps while varying the magnetic field.

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