UNUSUAL MAGNETIC AND PRESSURE RESPONSE OF THE S = 1 ANTI-FERROMAGNETIC LINEAR-CHAIN MATERIAL [Ni(HF$_2$)(3-Clpy)$_4$]BF$_4$

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An S = 1 antiferromagnetic polymeric chain, [Ni(HF$_2$)(3-Clpy)$_4$]BF$_4$ (py = pyridine), has been identified to have nearest-neighbor antiferromagnetic interaction $J/k_B = 4.86$ K and single-ion anisotropy $D/k_B = 4.3$ K, while avoiding long-range order down to 25 mK [1]. With $D/J = 0.88$, this system is close to the $D/J \approx 1$ gapless quantum critical point between the topologically distinct Haldane and Large-$D$ phases. The magnetization was studied over a range of temperatures, 50 mK $\leq T \leq 1$ K, and magnetic fields, $B \leq 10$ T [2]. The results allow an upper bound of the critical field, $B_C$, which closes the Haldane gap, to be estimated. Specifically, $B_C \leq (35 \pm 10)$ mT, which is close to the predicted 46 mT [3], when using the reported values of $J$, $D$, and $g$ [1]. In low fields, the magnetic signal increases with decreasing temperature for 400 mK $< T < 800$ mK but is independent of temperature for 50 mK $\leq T \leq 400$ mK. This observation is consistent with a significant increase in the specific heat arising from the accumulation of entropy in the vicinity of the quantum critical point.

Using a commercial magnetometer equipped with home-made pressure cells [4], the low-field (0.1 T), high-temperature ($T \geq 2$ K) magnetic susceptibility was studied as function of pressure up to 1.47 GPa. Preliminary analysis of these data suggest the response observed at ambient pressure [1] changes between 0.24 GPa and 0.35 GPa, and the unusually strong Curie-like response intensifies. In other words, externally applied pressures might be tuning the material through the $D/J \approx 1$ gapless quantum critical point. To our knowledge, theoretical predictions of the thermodynamic quantities in the critical region have not appeared in the literature.

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