Magnetic Order and Magnetic Exchange Interactions in the Quasi-Two-Dimensional Magnets $[Cu(pyz)_2(HF_2)]X$ with $X = BF_4$ and PF_6

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We report on combined experimental and theoretical efforts to elucidate the magnetic properties of the organic magnets $[Cu(HF_2)(pyz)_2]X$ with $X = BF_4$ and PF_6. By use of high-resolution specific-heat measurements on high-quality single crystals we were able to resolve small lambda-type anomalies indicative for a phase transition into a long-range ordered state. First-principles density-functional calculations allow to estimate the exchange couplings and confirm a three-dimensional antiferromagnetic order between adjacent Cu centers with a stronger coupling within the layers of pyz-bridged Cu and a much weaker coupling across the HF_2 linker units. Specific-heat data in magnetic field, B, reveal a slightly anisotropic nonmonotonic dependence of the Néel temperature T_N with B. The increase of T_N at low fields can be understood by the reduction of phase fluctuations that are prominent in these highly anisotropic quasi-two-dimensional magnets. Towards higher fields, B suppresses the amplitude of the antiferromagnetic order parameter, and correspondingly T_N, by aligning the spins parallel to B. This nonmonotonic behavior of T_N, as well as the magnetic part of the specific heat, cannot be described by a mean field theory that neglects the crucial role played by phase fluctuations. In contrast, we show that both properties are very well reproduced by Quantum Monte Carlo simulations. Pulsed-field magnetization data corroborate the strong anisotropy of the exchange interactions. The ratio of the inter- to intralayer exchange interaction is estimated as 0.0025 (for $X = BF_4$) and 0.009 ($X = PF_6$). High-field electron spin resonance measurements reveal a magnetic easy-plane anisotropy.

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