

Bond-dependent interactions and ill-ordered state in the honeycomb cobaltate $\text{BaCo}_2(\text{AsO}_4)_2$

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Following the proposed materialization of Kitaev-bond-dependent spin liquid physics in honeycomb lattices of heavy transition metals with 4d or 5d electrons [1], it has been proposed that this can be extended to 3d transition metals, in particular Co^{2+} [2]. A first step in validating the prospect of finding a quantum spin liquid is to demonstrate the presence of these anisotropic bond-dependent interactions in such materials. These could promote new types of behavior or provide insight into certain materials not elucidated to date. This is the case of $\text{BaCo}_2(\text{AsO}_4)_2$, a honeycomb cobaltate whose ground state and Hamiltonian have been debated for decades [3]. We have investigated the magnetic properties of a $\text{BaCo}_2(\text{AsO}_4)_2$ single-crystal through neutron diffraction and inelastic scattering, as well as by very-low temperature magnetization and AC susceptibility measurements. The latter measurements, which reveal slow dynamics and non-equilibrium responses, are consistent with an original ill-ordered magnetic compound with intrinsic defects as proposed previously [4]: collinear zig-zag ferromagnetic chains in a up-up-down-down arrangement interspersed with additional chains to agree with the propagation vector of 0.27 imposed by competing interactions. To interpret these results, we propose an exchange model with bond-dependent anisotropic interactions on the first neighbors and Heisenberg interactions up to the fourth neighbors. Monte Carlo calculations show that our model successfully reproduces key experimental observations, namely spin-wave dispersions (figure), magnetization curves with a 1/3 magnetization plateau, and the faulty collinear spin configuration, leading to a coherent picture that had not been achieved to date [5]. This highlights the potential of including these new ingredients (anisotropic Kitaev and off-diagonal interactions) in understanding long-standing puzzling behaviors and discovering exotic physics.

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