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Probing Signatures of Low-dimensional Magnetism in Layered Honeycomb Compound BaCo₂(PO₄)₂ Using High Frequency Electron Paramagnetic Resonance Spectroscopy

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Introduction

Layered honeycomb lattices are gaining interest due to the possibility of realizing Kitaev physics.¹ Of particular interest is BaCo₂(PO₄)₂ (γ -BCPO) which is a layered honeycomb lattice in which Co atoms form CoO₆ edge-sharing octahedra separated along the *c*-direction by non-magnetic layers of Ba and PO₄. The Co atoms in γ -BCPO form a quasi-2D honeycomb lattice. The strong crystalline electric field plus spin-orbit coupling of oxygen octahedra splits the energy levels of Co²⁺ in to a low-lying doublet thereby realizing a J_{eff} = 1/2. Hence, γ -BCPO is an ideal quantum magnet with effective spin 1/2 on a low dimensional honeycomb lattice. Theoretical work on frustrated *J*₁-*J*₂-*J*₃ exchange on a honeycomb lattice predicts a spin liquid phase.² We used EPR to probe the low dimensional magnetism in γ -BCPO and to estimate the *g*-values at low temperatures.

Experimental

The EPR experiment was motivated to probe the spin dynamics of Co^{2+} in γ -BCPO to obtain information about critical behaviour close to 4 K where the magnetic anomaly occurs, as a function of temperature and microwave frequency (90 – 400 GHz). Also to study the variation of *g*-value as a function of temperature and microwave frequency to infer the presence of spin-orbit coupling and Co^{2+} environment. Very high frequency (120 – 614 GHz) electron paramagnetic resonance (VHF-EPR) measurements have been performed from 1.5 – 80 K.

Results and Discussion

At any applied microwave frequency, we found no EPR signal from γ -BCPO above 20 K. For T < 20 K, a broad signal is visible at low fields, with the spectral features better defined. As the frequency changed from 120 to 336 GHz, the broad signal split into several narrow modes, and for frequency 406.6 GHz and 614.4 GHz, a broad, but complete signal appeared with no splitting. The effective *g*-value shifts from *g* = 14.62 to 11.18 as the frequency increases from 406.6 to 614.4 GHz, in line with a magnetic gap of 300 GHz. The EPR signal narrows and shifts to lower fields as the temperature is lowered from 20 to 1.5 K. The experimental EPR curves at different temperatures are presented in **Fig.1**.

Conclusions

The EPR features from our experiments indicate that the resonances are likely related to antiferromagnetic modes because of large internal fields in γ -BCPO.

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References

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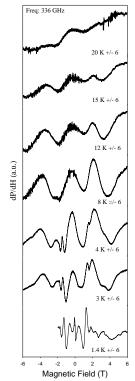


Fig.1 EPR spectra of γ -BCPO at different temperatures for 336 GHz.