Counterroating Incommensurate Magnetic Order and Strong Quantum Fluctuations in Honeycomb NaNi$_2$BiO$_6$

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Introduction

The Kitaev model well-known for its spin liquid ground state, but it is just one of many bond dependent models on the honeycomb lattice. Here we present a first experimental realization of the bond-dependent magnetic 120° compass model exchange on a honeycomb lattice. The resulting incommensurate magnetism is characterized by strong quantum fluctuation.

NaNi$_2$BiO$_6$ is a magnetic Ni honeycomb compound with a mixture of 2/3 Ni$^{3+}$ ($S = 1/2$) and 1/3 Ni$^{2+}$ ($S = 1$) [1] shown in Fig. 1.

Neutron Scattering

Neutron diffraction shows the onset of magnetic order at low temperatures with an ordering wave vector $q = (\frac{1}{2}, \frac{1}{2}, 0.154 \pm 0.011)$, shown in Fig. 3. Magnetic refinements to this data (Fig. 4) and symmetry analysis reveal a two-step order whose spin order along the $c$ axis below $T_1$ and in the $ab$ plane below $T_2$.

In-plane Spin Structure

The $(\frac{1}{2}, \frac{1}{2})$ counterrotating in-plane order (Fig. 6) is not stabilized by isotropic interactions, and suggests anisotropic bond-dependent interactions. This particular structure matches the predicted ground state of the 120° compass model on the honeycomb lattice [2].

Interpretation: 120° compass exchange

Because of the unusual $(\frac{1}{2}, \frac{1}{2})$ order, the counterrotating spin structure, and the agreement with the theoretically predicted ground state, we attribute the magnetic structure in NaNi$_2$BiO$_6$ to the 120° compass interaction.

The origin of the 120° interaction in NaNi$_2$BiO$_6$ may be understood as arising from the anisotropy of the Ni-O-Ni bond (see Fig. 7). In this case, the anisotropy directions are 94.3° apart, which puts NaNi$_2$BiO$_6$ within the 120° phase but very close to the critical point of the Kitaev spin liquid phase ($87^\circ < \theta < 94^\circ$) [3].

Conclusion

This study constitutes the only experimental evidence we are aware of for magnetic 120° exchange on the honeycomb lattice, raising the possibility for discovering Kitaev-like spin liquid phases in 3d transition metal oxides.

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References


Heat Capacity

Heat capacity shows two second order transitions at $T_1 = 6.3$ K and $T_2 = 4.8$ K. Integrating $\Delta S = \int \frac{C}{T}$ reveals far less entropy than expected $R(2/3\ln(2) + 1/3\ln(3))$. This indicates either magnetic correlations above 20 K or residual entropy within the ordered phase.

Dynamic Magnetism

Sum rule analysis of the 1.8 K neutron spectrum (Fig. 5) indicates that 21(4)% of the magnetism is static within the ordered phase, which is only half of the maximum static moment $S_{max} = 38.9%$. This indicates strong magnetic fluctuations in the ground state.