Counterrotating Incommensurate Magnetic Order and Strong Quantum Fluctuations in Honeycomb $NaNi_2BiO_6$

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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-depdent magnetic 120° compass model exchange on a honeycomb lattice. The resulting incommensurate magnetism is characterized by strong quantum fluctuation.

$NaNi_2BiO_{6-\delta}$ 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.

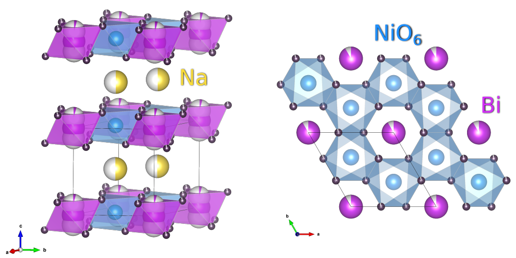

Figure 1: Crystal structure of $NaNi_2BiO_6$.

Heat Capacity

Heat capacity shows two second order transitions at $T_{c1} = 6.3\,\mathrm{K}$ and $T_{c2} = 4.8\,\mathrm{K}$. Integrating $\Delta S = \int \frac{C}{T} dT$ 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.

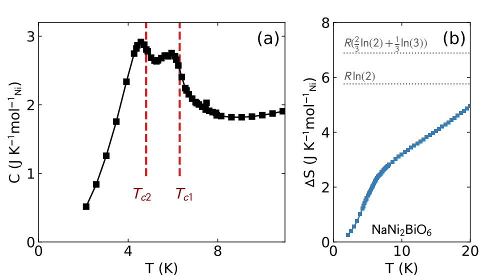

Figure 2: (a) heat capacity and (b) entropy of $NaNi_2BiO_6$.

Neutron Scattering

Neutron diffraction shows the onset of magnetic order at low temperatures with an ordering wave vector $\mathbf{q} = (\frac{1}{3}, \frac{1}{3}, 0.154 \pm 0.11)$, shown in Fig. 3. Magnetic refinements to this data (Fig. 4) and symmetry analysis reveal a two-step order where spins order along the c axis below T_{c1} and in the ab plane below T_{c2}.

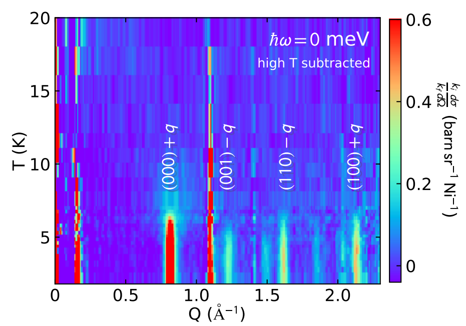

Figure 3: Temperature dependent elastic neutron diffraction of $NaNi_2BiO_6$ showing the onset of magnetic order.

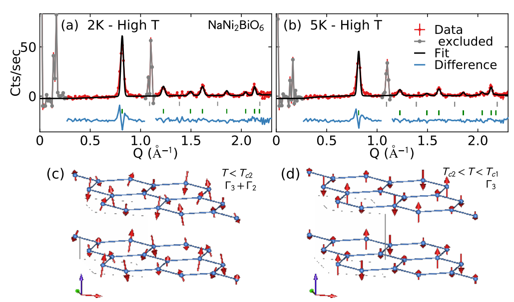

Figure 4: Refined magnetic structures of $NaNi_2BiO_6$ at 5 K and 2 K.

In-plane Spin Structure

The $(\frac{1}{3}, \frac{1}{3})$ 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].

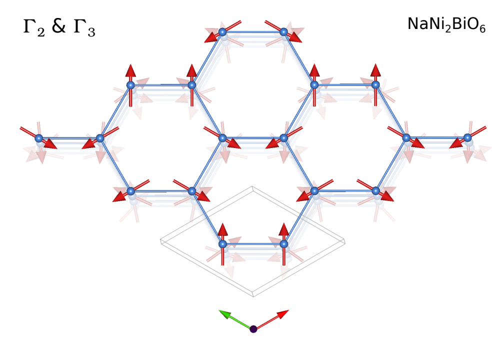

Figure 6: In-plane magnetic structure of $NaNi_2BiO_6$.

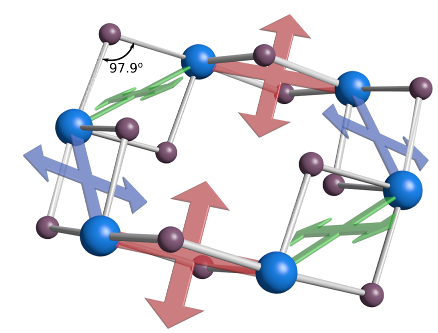

Figure 7: Ni-O-Ni exchange pathways of $NaNi_2BiO_6$, with anisotropy directions indicated by green, blue, and red arrows.

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 $\frac{S^2}{S(S+1)} = 38.9\%$. This indicates strong magnetic fluctuations in the ground state.

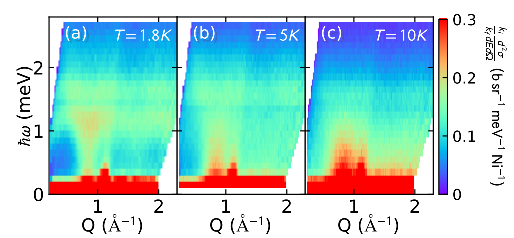

Figure 5: Inelastic neutron scattering of $NaNi_2BiO_6$.

Interpretation: 120° compass exchange

Because of the unusual $(\frac{1}{3}, \frac{1}{3})$ order, the counterrotating spin structure, and the agreement with the theoretically predicted ground state, we attribute the magnetic structure in $NaNi_2BiO_6$ to the 120° compass interaction.

The origin of the 120° interaction in $NaNi_2BiO_6$ may be understood as arising from the anisotropy from the Ni-O-Ni bond (see Fig. 7). In this case, the anisotropy directions are 94.3° apart, which puts $NaNi_2BiO_6$ within the 120° phase but very close to the critical point of the Kitaev spin liquid phase ($87° < \theta < 94°$) [3].

Conclusion

This study constitutes the only experimenal 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.

Acknowledgments

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References

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