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Lack of multiferroic behavior in $BaCuSi_2O_6$ is consistent with the frustrated magnetic scenario for this material

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Introduction

$BaCuSi_2O_6$ is a well-known quantum magnet that exhibits[1] a Bose-Einstein Condensation quantum phase transition[2] in applied magnetic fields. It contains Cu dimers that form singlets in zero magnetic field, and in applied fields as the singlet-triplet gap is suppressed a quantum phase transition occurs to canted XY antiferromagnetism between critical fields H_{c1} = 23 T and H_{c2} = 59 T. In addition, as the temperature is lowered, a rare frustration-induced dimensional reduction has been proposed from three to two dimensions.[3] Recently, however, a controversy has arisen about the details of the magnetic ordering due to the discovery[4] of a tetragonal to orthorhombic structural transition at 100 K with an incommensurate modulation along the b-axis. Multiple magnon modes were observed in neutron diffraction studies, while NMR found modulation of the spin structure along both the ab plane and the c-axis.[5] In this scenario the material is still a Bose-Einstein condensate system but the frustration is not perfect, calling into question the dimension reduction scenario. A recent study of $BaCuSi_2O_6$ combining inelastic neutron diffraction and density functional theory suggest that the material isn't even frustrated at all and that the spins are ordered ferromagnetically in the a-b plane and antiferromagnetically along the c-axis.[6] After a detailed symmetry analysis we have concluded that the magnetic scenario postulated by this most recent unfrustrated theory[6] will render $BaCuSi_2O_6$ a multiferroic between H_{c1} and H_{c2}, with electric polarization in easy axis of the a-b plane for magnetic fields along the c-axis via an inverse Dzyaloshinskii-Moriya mechanism. Electric polarization is a sensitive symmetry probe of magnetic order, since magnetic systems that break spatial inversion symmetry can induce an overall ferroelectricity in the crystalline lattice. In pulsed magnetic fields we can detect electric polarizations with unique sensitivity to sub-pC/m^2, which is orders of magnitude more sensitive than what can be detected in DC magnetic field.[7]

Experimental

Electric polarization of single crystals with silver paint contacts was measured in the above geometry in pulsed magnetic fields to 55 T down to 0.5 K, as well as for electric polarization along the c-axis.[7] The experiments were repeated for two different crystals in each of two orientations. Measurements were made in zero electric field, with electric field poling from high temperatures (though this is not expected to be helpful since magnetic order occurs only at high fields), and with electric fields applied during the measurement (100 V across a mm-sized crystal). The insulating properties of the crystal were confirmed by a loss tangent > 10^5.

Results and Discussion

No magnetic field-induced electric polarization change was resolved in any of these pulsed magnetic field measurements.

Conclusions

Our results are *not* consistent with the proposed unfrustrated scenario for $BaCuSi_2O_6$.

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