Spectroscopic study of magnetoelastic CrOCl

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Introduction

Multiferroics defines a class of materials that exhibit more than one primary ferroic orders simultaneously, or more generally, for materials which show strong couplings between the orders. Using the high magnetic field facility at NHMFL, Musfeldt *et al* have discovered a strong correlation between the magnetic order and ferroic orders as evidenced by the correlation between the magnetic order and the optical phonon modes (IR- and Raman-active) in several multiferroic materials. [1] Among multiferroics, CrOCl is a magnetoelastic material that the antiferromagnetic order (T_N=13.5K) is related to a lattice structural distortion leading to a induced strain in the crystal. [2] Another phase transition at 27K is identified by the specific heat measurement and an applied magnetic field is known to induce a spin-flip transition at around 4T; whereas the characters of these new phases and the correlation between the magnetic and lattice orders are not yet studied and understood.

Experimental

Greenish leaves-like crystals were synthesized and grown by H. Zhou at NHMFL. The thin CrOCl flakes are carefully laid upon Scoth tape to create a film-like sample of appreciable cross area for optical measurements. The optical properties of CrOCl flakes on tape are investigated using IR transmittance and reflectance spectroscopy at 4K using superconducting and resistive magnet in SCM3 and cell 8, respectively. Efforts have been focused on identifying phonon modes in CrOCl and how are these modes changes with increasing magnetic field.

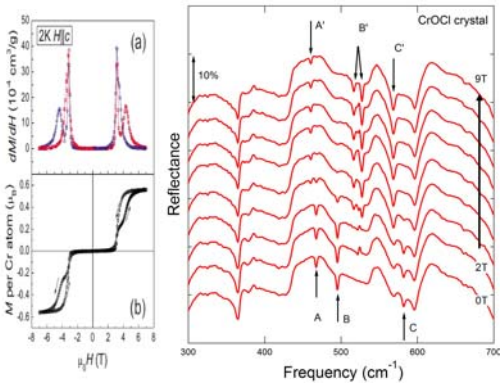

Fig.1 Left: A magnetic measurement result taken from Ref. [2]. A spin-flip transition occurs at around 4T, where the sample turns from AFM to FM order. Right: Reflectance spectra of CrOCl flakes at selective fields are displayed. The spectra are vertically shifted for clarity. Several absorptions due to IR-active phonon modes are found. In particular, mode A, B and C demonstrate a strong correlation to magnetic order, while mode B splits into two modes in the high-magnetic-field phase, which suggests a change in symmetry in the crystal

Results and Discussion

A set of reflectance spectra at the magnetic fields around the spin flip transitions (4T) is plotted in Fig. 1. Several sharp optical absorptions have been observed and they can be attributed to the optical phonon modes in CrOCl lattice. The spectroscopic symbols and the corresponding atomic vibrations for these optical phonon modes remain to be identified, but it can be clearly seen that several of them (as indicated by the arrows) changes as the sample goes through the spin-flip transition. Moreover, one of the modes splits into two at the spin-flip transition and it suggests that not only were the bonding between the atoms changes with the magnetic order (as evidenced by a change in optical phonon frequency and line width.), the lattice symmetry also changes at the transition. The evolution of these modes agrees with the earlier magnetic measurements about the spin-flip transition including the hysteresis of the magnetic transition. [2] To the best of my knowledge, never a multiferroic material exhibits such strong correlation between the magnetic order and optical phonon modes, while with a change in the lattice symmetry of the crystal.

Conclusions

We have investigated the IR transmittance and reflectance of CrOCl flakes at 4K up to 35T. Several optical phonon modes have been identified, while a strong correlation between lattice order and magnetic order is discovered. Further works are required to reveal the properties of these IR-active phonon modes and a future request for magnet time to identify the Raman-active optical phonon modes in this crystal.

References

[1] "Magnetoelastic Coupling through the Antiferromagnet-to-Ferromagnet Transition of Quasi-Two-Dimensional $[Cu(HF_2)(pyz)_2]BF_4$ Using Infrared Spectroscopy", J. L. Musfeldt *et al*, Phys. Rev. Lett. **103**, 157401(2009)

[2] "Observation of strong magnetoelestic coupling in a first-order phase transition of CrOCl", J. Angelkort *et al.*, Phys. Rev. B **80**, 144416 (2009)