Far-Infrared Spectroscopy of Quantum Spin Chain: $PbCuSO_4(OH)_2$

E. Constable,* A.D. Squires,* K.C. Rule†* and R.A. Lewis*

*Institute for Superconducting and Electronic Materials, University of Wollongong, Wollongong NSW 2522, Australia

†Bragg Institute, ANSTO, Kirrawee DC NSW 2234, Australia

***Abstract*—This work presents far-infrared transmission spectroscopy on single crystal $PbCuSO_4(OH)_2$ using synchrotron radiation. The study covers the spectral region 150–400 cm^{-1} with electric field polarisation parallel to either the *a* or *b* crystal directions. The results reveal a number of anisotropic absorption features tentatively attributed to phonon modes.**

I. Introduction and Background

THE study of low dimensional spin systems is appealing because observable quantum mechanical effects are often present in the magnetic properties. The inherent low magnetic symmetry may lead to frustration between competing exchange interactions which can give rise to a number of intriguing magnetic phases, such as spin liquid states and non-collinear spin ordering. Such properties are uniquely linked to models that describe high temperature superconductivity and multiferroicity.

The naturally occurring copper-oxide, $PbCuSO_4(OH)_2$ (also known as linarite), is an example of a frustrated quasi-one-dimensional magnet. In linarite, the Cu^{2+} ions form spin S = 1/2 chains along the *b* crystal direction with competing nearest-neighbour ferromagnetic and next-nearest-neighbour antiferromagnetic interactions. The resulting magnetically frustrated topology has been shown to order through residual interchain coupling below $T_N = 2.8$ K [1]. The ordered ground state has been described as an elliptical helical spin structure featuring multiple field induced long-range ordered phases and possible short-range ordered regions. The helical magnetic structure incommensurate to the crystal lattice also supports a claim of multiferroicity [2].

The study of optical excitations in multiferroic materials can help to identify lattice coupling to the spin network. Careful monitoring of phonon frequency position and absorption strength with changing temperature and radiation orientation may reveal new magnetoelectric interactions in complex magnetic systems [3].

II. Results

The absorption spectra at 7.5 K for polarisations with $\bar{E} \parallel \bar{a}$ and $\bar{E} \parallel \bar{b}$ are shown in Fig. 1 (a) and (b). The spectral properties appear anisotropic with clear differences between the absorptions in each polarisation. For $\bar{E} \parallel \bar{a}$, the electric field is perpendicular to the spin chains. The spectrum features a broad absorption at lower frequencies. Above 216 cm^{-1} the transmissive properties improve with four distinct features observed between 240–300 cm^{-1}. For $\bar{E} \parallel \bar{b}$, the low-frequency absorption band occurs below 175 cm^{-1}. A sharp absorption is observed at 194 cm^{-1} with two broad absorptions at 290 cm^{-1} and above 348 cm^{-1}. A number of weaker features appear between 220 and 260 cm^{-1}. A shift in the onset of the transmissive region is observed as the polarisation is altered. With $\bar{E} \parallel \bar{a}$ the sample is seen to transmit in a window ranging

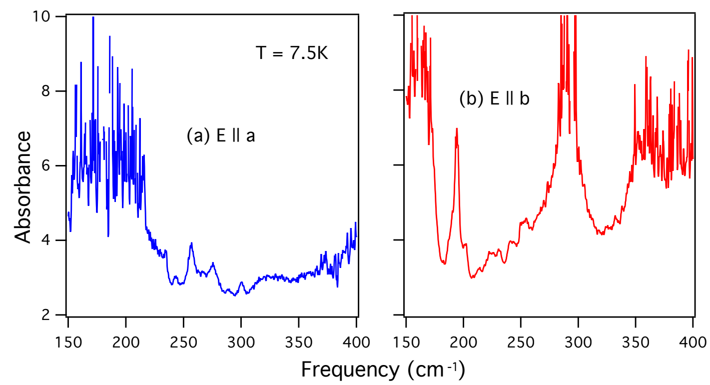

Fig. 1. Far-infrared absorption spectra of $PbCuSO_4(OH)_2$ by synchrotron radiation showing polarised light (a) $\bar{E} \parallel \bar{a}$ and (b) $\bar{E} \parallel \bar{b}$.

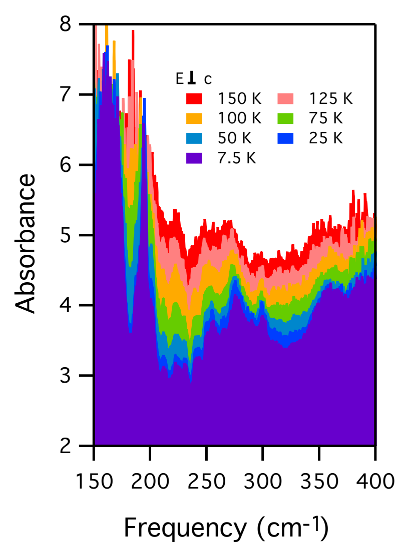

Fig. 2. Far-infrared absorption spectra of $PbCuSO_4(OH)_2$ by synchrotron radiation showing unpolarised temperature dependent measurements.

from 220 - 420 cm^{-1}. For $\bar{E} \parallel \bar{b}$ this window shifts to a region from 175 - 350 cm^{-1}.

Unpolarised temperature dependent spectra are shown in Fig. 2. In this geometry, the electric field is confined to the *ab* plane. The results indicate subtle hardening and strengthening of excitations with decreasing temperature. In particular, the sharp absorption at 194 cm^{-1} develops below 100 K. With all measurements performed well above the magnetic ordering temperature of 2.8 K, no magnetic coupling is expected. Based on the energy region and temperature properties, the observed features are most likely of phonon origin. Further analysis of these excitations at lower temperatures may reveal information regarding the proposed mutliferroic state and helical incommensurate spin structure.

III. Conclusion

The results present preliminary spectroscopic measurements of the novel quantum spin chain $PbCuSO_4(OH)_2$. A number of absorption features have been identified with temperature and polarisation dependence. The excitations are most likely phonons and their identification provides motivation for lower temperature studies. We would like to thank the Australian Synchrotron for travel funding assistance related to this work.

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