Coherent Phonons in $CrGeTe_3$

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1 Introduction

Light can interact with a crystal in various ways, such as transfer heat, excite electrons and more. When the pulses are shorter than the period of a phonon thay can excite coherent lattice dynamics i.e. phonons. In a previous work [1] it was shown that light can also enhance magnetic interactions in systems where the magnetic interaction is based on the superexchange mechanism. It was tested on $CrSiTe_3$ where the angle between the metal and ligand ions is $\theta < 90°$. This resulted in a force which opposes the pure electrostatic force between the ions. The abrupt change in exchange interaction results in an effective force which acts on the magnetic ions thus exciting coherent phonons. In this research we want to test the hypothesis shown in the paper by working on a closely related compound $CrGeTe_3$ where $\theta > 90°$. In this geometry the electrostatic and exchange induced forces are predicted to act in the same direction which will dramatically affect the lattice dynamics following the optical excitation.

2 Theory

$CrSiTe_3$ is a layered ferromagnetic insulator where in each layer a Cr ion is surrounded by an octahedra of 6 Te ions. The angle formed between neighbouring Cr ions and their connecting Te ligand is $\theta < 90°$.

When excited with an ultrafast laser pulse a charge transfer (CT) excitation can occur from the Te ions to the Cr ion. This CT excitation has two effects, reduction of the electrostatic attraction between the Cr and Te ions which exerts a coulomb force $\vec{F}_{\mathrm{C}}$, and enhancement of the spin exchange energy which exerts an exchange force $\vec{F}_{\mathrm{ex}}$.

The two forces launch coherent phonon oscillations with A_g symmetry which can be observed using the pump probe technique (section (3)). In contrast to $\vec{F}_{\mathrm{C}}$ which is almost independent of temperature $\vec{F}_{\mathrm{ex}}$ depends strongly on temperature since it is nullified in the absence of magnetic correlations which are absent at $T \gg T_{\mathrm{Curie}}$. As temperature is lowered $\vec{F}_{\mathrm{ex}}$ gets stronger with the increasing strength of magnetic correlations until at a certain temperature $\tilde{T}$, $\left|\vec{F}_{\mathrm{C}}\right| = \left|\vec{F}_{\mathrm{ex}}\right|$.

At temperatures greater than $\tilde{T}$ CT excitations will launch an A_g phonon with a phase $\phi = 0$ relative to the arrival of the laser pulse. As temperature is lowered below $\tilde{T}$ the phase will shift to $\phi = \pi$ due to the total force now acting in the opposite direction.

In the closely related compound $CrGeTe_3$, $\theta > 90°$, in contrast to $CrSiTe_3$ where $\theta < 90°$. This means that the two forces, $\vec{F}_C$ and $\vec{F}_{ex}$, will act in the same direction no matter the temperature, so we suspect that as we cool the sample the amplitude of the phonon will increase as $\left|\vec{F}_{ex}\right|$ grows in size.

3 Experiment

A pump-probe setup consists of a short pulses laser which is split into two different beams. The first, called the pump, excites the sample and the second called the probe is used to optically study its physical properties. The length of each beam can be controlled to create the desired time delay between the pump and probe pulses. This delay enables us to measure (with the probe beam) the dynamics of optical properties as a function of the time delay between the pulses on femtosecond timescales.

Over the summer I have built such a system, below is a preliminary dataset taken on $CrGeTe_3$ at $T = 5$K.

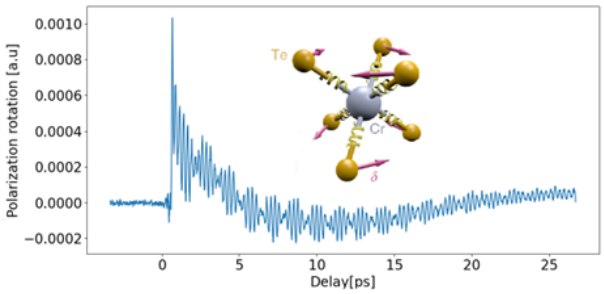

Figure 1: Measurement of the magneto optic kerr effect (MOKE) of $CrGeTe_3$ using a pump-probe setup. An abrupt increase in the signal follows the arrival of the pump pulse at Delay=0. The bi-exponential decay is overlayed with oscillations at multiple frequencies, one of which is of the sought after A_g phonon.

In this project we propose to systematically measure the transient optical properties of $CrGeTe_3$ as a function of temperature and beam parameters e.g energy, power to understand the interplay between lattice electrons and their spins in $CrGeTe_3$.

References

[1] A. Ron, S. Chaudhary, G. Zhang, H. Ning, E. Zoghlin, S.D. Wilson, R.D. Averitt, G. Refael and D. Hsieh, "Ultrafast Enhancement of Ferromagnetic Spin Exchange Induced by Ligand-to-Metal Charge Transfer", *Phys. Rev. Lett.*, vol. 125, no. 197203, 2020.