Studying Low-Energy Symmetry Properties of 1D Cuprates with Terahertz Spectroscopy

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In the past decade, scientists have discovered that there are exciting properties and phases in materials at very low energies. This discovery has created a new area of study in condensed matter physics. Such low energy dynamics, accessed using ultrafast lasers, could be hiding quantum phenomena that have never been observed experimentally. A class of materials called cuprates has been of particular interest in these experiments. Cuprates are known for their strongly correlated degrees of freedom, including spin and charge. This correlation yields high-temperature superconductivity and other interesting quantum features. Some believe that by better understanding cuprates’ quantum properties we may learn more about the origins of high-temperature superconductivity.

The low energy range in question is referred to as the terahertz (THz) frequency regime and spans from 1 to 200 meV. Studying the THz regime requires the use of a femtosecond (10⁻¹² s) laser in a technique known as time-resolved time-domain THz spectroscopy. The laser sends pulses of 35 fs in width every millisecond. This mechanism allows us to first photo-excite, or pump, the system with a pulse and then probe the system with another pulse at various time delays to measure how the material changes due to the electron doping from the pump pulse [2].

The Hsieh lab used THz spectroscopy to study the 1D cuprate Sr₂CuO₃ to search for nonequilibrium phases. 1D cuprates are materials with the CuO₂ structure that is characteristic of cuprates but in one direction [1]. Sr₂CuO₃ has not yet been studied in the THz regime though we believe there is interesting physics at this level due to prior experiments in the THz regime.

To look for such quantum phenomena, we first measured the Sr₂CuO₃ sample in static THz transmission. This set-up measures the material in a steady-state; there are no photo-induced effects. In addition, we detect the pulse after it passes through the sample, compared to detecting the pulse reflected off the sample as we measured later. These measurements are conducted to 1) explore the low energy properties intrinsic to the material and 2) later distinguish the photo-induced effects from the material’s properties.

Diagram of a diagram of a liquid nitrogen flow

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Figure 1: Static Transmission Layout [3]. The beam is separated into two paths by a beam splitter. One beam reflects off several mirrors and is transmitted through the sample. The other beam detects the changes imposed by the material on the first beam through the nonlinear interaction of the two beams with the ZnTe crystal before the detector.

We measured our Sr₂CuO₃ samples in static transmission using a technique we developed called Rotational Anisotropy Terahertz (RA-THz) spectroscopy. We placed the sample in a rotation mount and measured the THz transmission at different sample orientations. This rotation allowed us to study the polarization dependence of phenomena in the sample. The changes in the polarization matter because the light’s interaction along different axes in the material can yield unique effects. These differences are depicted by the light pulses that are measured at the photodetector. By Fourier transforming the pulses, we can analyze the data in the frequency space. Dips in the frequency spectrum, as seen in the figure below, signify that something interesting is happening in the material at these frequencies because more energy is being absorbed. The challenge then becomes determining what phenomenon is causing this feature.

From our static transmission measurements of Sr₂CuO₃, we see a four-fold symmetry in the frequency spectrum. This four-fold symmetry persists throughout our frequency bandwidth. Additionally, around 1.4 THz, we see a dip in the signal 45 degrees from the Cu-O chain direction.

a)A graph of a waveform

Description automatically generated with medium confidenceb)A graph of a graph of a graph

Description automatically generated with low confidencec)A circular graph with a cross

Description automatically generated with medium confidence

Figure 2: a) Time domain THz transmission pulse at intervals of 15 deg from 0 to 345 deg. b) Fourier transform of THz transmission pulses. c) Slice of THz transmission at 1.4 THz as a function of sample orientation. The polar plot has been rotated so that the 45 degree label corresponds to 45 degrees from the chain direction of the material.

We attribute part of the four-fold symmetry to birefringence, meaning that the light’s path through the sample is delayed more in the y axis than in the x axis, or vice versa. Our calculation of the refractive index along the chain and perpendicular to the chain displayed below supports this difference in refractive index. This birefringence, however, does not entirely explain the dip at 1.4 THz. The additional feature causing this dip remains an open question.

A graph of a line graph

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Figure 3: Real refractive index along the Cu-O chain direction (red) and perpendicular to the chain direction (green).

After the static transmission measurements, we conducted pump probe reflectivity terahertz spectroscopy. This configuration differs from the previous measurements in that we measured the terahertz light that reflected off the sample and photo-excited the sample with a pump pulse.

Diagram of a diagram of a device

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Figure 4: Pump-probe reflectivity set up [3]. The beam is separated into two paths just as in the transmission set-up. In this case, the first beam is reflected off the sample. In addition, there is a third beam path at the top. This beam is the pulse that excites the sample, while the two other beams probe the state of the sample.

In this reflection geometry, we took two types of measurements. First, we took pump-probe scans where we measured the change in electric field as a function of time. Figure 5a is a fitting of a pump-probe scan along the Cu-O chain direction. We fit the data with two exponentials to study the decay of the system. Second, we took transient electro-optic sampling (TEOS) scans, shown in Figure 5b, where we measured the terahertz pulse output at different delay times between the pump and the probe. The TEOS scans are similar to the data we measured in the static transmission case yet at various times after the pump pulse to understand how the material’s dynamics are changing due to the added elections.

a.A graph of a function

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Figure 5: a) Exponential fitting of pump-probe scan along the Cu-O chain direction. Two exponential functions are fitted, one with a time decay of 0.8 ps and the other with a time decay of 7 ps. b) Change in electric field, hence the induced signal, as a function of time and frequency. The color bar represents the change in electric field over the total electric field.

In Figures 5a and 5b, we see a fast decay time and a slower decay time, depicted as two different rates of change in the change in intensity of the plotted change in electric field in Figure 5b. These two decays suggest that there are two phenomena occurring due to the photo-excitation.

We plan to repeat these measurements with a different sample to check the reproducibility of the results. In addition, we plan to cool down the sample to investigate whether we see additional features in the material. Much of the phenomena we observed remain an open question and require more investigation. Nevertheless, the abundance of interesting physics in the terahertz regime is clear. Such experiments reveal the exciting aspects of strongly correlated systems. We hope to persuade scientists of the importance of low energy dynamics.

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