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

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Abstract

Cuprates are a class of materials that have generated interest for their strongly correlated degrees of freedom, including spin and charge. High-temperature superconductivity is among one interesting quantum phenomenon that arises from this strong correlation. A subset of these materials, called 1D cuprates, have the characteristic Cu-O chain in one direction and provide an important avenue in better understandingthe electronic structure of these materials. We investigatedinvestigating the properties and dynamics of the 1D cuprate Sr₂CuO₃ in the low energy regime to search for nonequilibrium phases. To access this energy scale, we employed time-resolved time-domain terahertz spectroscopy. We observed polarization dependence in the material’s equilibrium state in the form of four-fold symmetry with increased absorption diagonal to the Cu-O chain direction, which we attribute to the birefringence of the material. We photoexcited the material with a laser pulse and saw various relaxation dynamics that may be attributed to several interesting quantum effects.

Introduction

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 not yet been observed experimentally. A class of materials called cuprates has been of particular interest in these experiments. As mentioned above, this correlationyields 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].

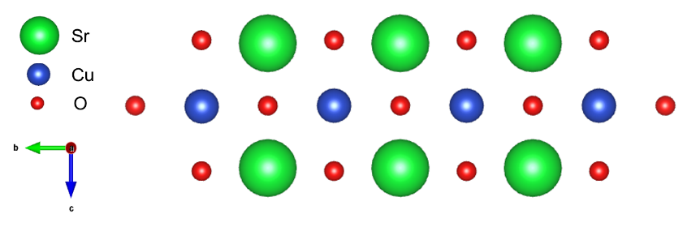


Figure 1: Crystal Structure of Sr₂CuO₃. Cu-O chain is represented by the alternating Cu and O atoms in the center row.

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. For instance, the Hsieh lab has studied iridates, which are a class of materials similar to cuprates in their Heisenberg exchange behavior, which is the interaction that causes the alignment or misalignment of magnetic spins, and their spin ½ ions. In strontium iridate, the Hsieh lab discovered the presence of bound states formed through magnetic interactions known as Hubbard excitons in the THz regime [3]. These excitons were the first of their kind as previously all discovered excitons were Coulombic excitons, bound through Coulomb’s force.



Sr

Cu

O

Methods and Resu

To look for such quantum phenomena, we first measured the terahertz response of the Sr₂CuO₃ sample in static transmission, meaning that the material is not excited by an initial laser pulse. Hence, we are measuring the material in a steady state; there are no photo-induced effects.

Terahertz light cannot be produced directly by a laser therefore we must generate it through other mechanisms. In the Hsieh lab, the terahertz light is generated through a nonlinear optical response of the original laser light with a ZnTe crystal. The signal can then be detected after passing through a second ZnTe crystal. The interaction of the terahertz light and the material provides information on any absorptions in the materials at these energies. These measurements are conducted to 1) explore the low energy properties intrinsic to the material and 2) distinguish the photo-induced effects from the material’s steady state when we later excite the material.

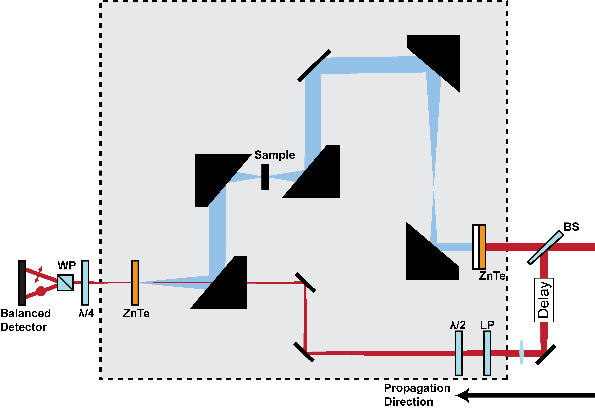


Figure 2: Static Transmission Layout [3]. The beam is separated into two paths by a beam splitter labeled BS. One beam reflects off several mirrors and is transmitted through the sample. The other beam detects the terahertz response of the material through the nonlinear interaction of the two beams with 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 are significant because the axes of the matter have different properties, therefore light’s interaction along various axes in the material can yield unique effects. For example, the axes have different atoms and hence different bond lengths and possibly electron interactions, which influence the axis’s response to light.

At the photodetector, we measure time traces of the signal to observe the material’s response to the terahertz light. Such time traces are depicted in Figure 3a. By Fourier transforming the pulses, we can analyze the data in the frequency space. Dips in the frequency spectrum, as seen at 1.4 THz in Figure 3b, signify energy absorption, which in turn signals that something is happening in the material at that frequency as some process in the material is using energy. The challenge then becomes determining what phenomenon is causing the absorptions.

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

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Figure 3: 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. 0 degrees represents the Cu-O chain direction of the material.

We attribute part of the four-fold symmetry to birefringence. Birefringence is when the light’s path through the sample is delayed more in one direction than in the perpendicular direction. Therefore, the refractive indices of the material, which are a measure of this delay, are different. Our calculation of the refractive index along the chain and perpendicular to the chain plotted in Figure 4 supports this claim. We see that the refractive index along the Cu-O chain is in the 3-3.1 range whereas the refractive index perpendicular to the Cu-O chain is in the 2.65-2.9 range. Hence, light is more delayed in the Cu-O chain direction than in the perpendicular direction.

A graph of a line graph

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Figure 4: Real refractive index along the Cu-O chain direction (red) and perpendicular to the chain direction (green).

This birefringence, however, does not entirely explain the dip at 1.4 THz. To further understand this phenomenon, we plan to analyze the optical conductivity of the material at this frequency from the data we took to better understand the electronic behavior of the system. In addition, we plan to ensure that the absorption is not simply an artifact of our terahertz spectroscopy set-up.

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

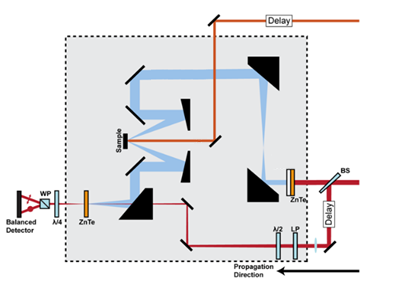


Figure 5: Pump-probe reflectivity set up [3]. The beam is separated into two paths just as in the transmission set-up. In this case, the first beam is reflected off the sample. In addition, there is a third beam path at the top. This beam is the pulse that excites the sample, while the two other beams 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 of the reflected light as a function of time. Figure 6a is a fitting of a pump-probe scan along the Cu-O chain direction. We fit the data with two exponentials and a constant background to study the decay of the system. Second, we took transient electro-optic sampling (TEOS) scans, shown in Figure 6b, where we measured the terahertz pulse output at fixed time delays between the pump and the probe. The TEOS scans are similar to the measurements we took in the static transmission case yet we measured the delay at various times after the pump pulse to understand how the material’s dynamics are changing with time due to the added elections.

a.A graph of a function

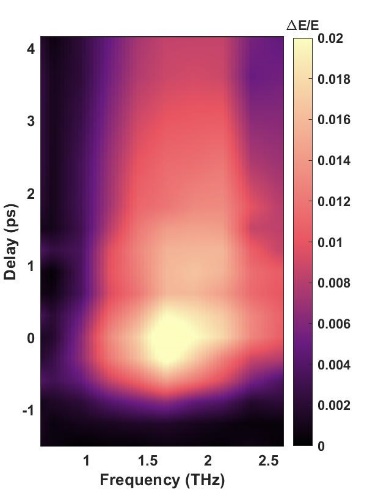
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Figure 6: a) Exponential fitting of pump-probe scan along the Cu-O chain direction. Two exponential functions are fitted, one with a time decay of 0.8 ps and the other with a time decay of 7 ps. The dashed line C represents a constant added background. 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.

Discussion

We can understand Figure 6a as being a vertical slice of Figure 6b if Figure 6b was in the time domain and not yet Fourier transformed to the frequency domain. Both results depict a fast decay time and a slower decay time, shown as the two exponential decays plotted in 6a and the rate of change in intensity of the plotted change in electric field in Figure 6b. When electrons, and hence energy, are added to a system in the form of an ultrafast laser pulse, often this additional energy will cause a response in the material. Then, the material will relax, releasing this energy, and return to its initial state. The time of this relaxation can signify what sort of response has occurred. Therefore, the two decay times we observe signal that there may be two phenomena occurring in the material when electrons are added to the system with the pump pulse. In some cases, the faster decay signals an electronic response whereas the slower decay may be due to heating. The two decays could also be due to some recombination effects where the fast decay is the relaxation of the phenomena that are formed immediately after the pulse whereas the slow decay is from the same phenomena though they form and relax some time after the pump due to the remaining excess energy in the system. One theory for the observed phenomena is that we are seeing Coulombic excitons, which as previously mentioned, are bound states between an electron and electron hole. However, further measurements are required to investigate this theory.

We plan to conduct fluence dependence measurements, where we increase or decrease the power of the laser pump. If we are observing excitons, higher fluence should yield a greater response as more excitons are produced from the higher number of added electrons. In addition, we plan to cool down the sample to investigate whether we see additional absorptions and/or different time decays in the material. These low temperature measurements may be interesting because the optical conductivity of Sr₂CuO₃ increases at lower temperatures in the terahertz regime. This increase means that more current is induced in the material due to the laser pulse, which could affect the response of the material and/or produce a stronger output signal.

Much of the phenomena we observed raises questions and requires 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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