Stimulating Coherent Phonons in $CaFe_2As_2$ With Laser

by

Yongyang Huang

Department of Physics
University of Colorado Boulder

Defended March 31, 2016

Thesis Advisor:
Professor Daniel Dessau, Department of Physics

Defense Committee:
Professor Daniel Dessau, Department of Physics
Professor Jamie Nagle, Department of Physics
Professor William Wei, Department of History
This thesis entitled:
Stimulating Coherent Phonons in $CaFe_2As_2$ With Laser

by

Yongyang Huang

Committee

____________________________
Daniel Dessau

____________________________
Jamie Nagle

____________________________
William Wei
ABSTRACT

High-temperature superconductivity has attracted a lot of attention. Since 1986 when the first high-temperature superconductor was discovered by researchers of IBM, scientists never give up to increase the transition temperature so that superconductors can be applied widely in industries. However, it remains an enigma that what causes the superconductivity of the high-temperature superconductor. The study of conventional superconductors suggest that the possible cause of superconductivity is electron-phonon coupling. Vast research have been devoted into this direction. But generally stimulating phonons is not efficient enough to see how phonons of exact mode affect the properties of a certain material. In this work, we will investigate a method to resonantly stimulate phonons with an optical pulse train generator. By setting the position of each lens and mirror, we can match the frequency of the output pulses with the frequency of phonons and then stimulate them resonantly.

Then we will collect data with Angle Resolved Photoemission Spectroscopy. This allowed us to analyze the behaviors of electrons inside the sample after the excitation of phonons. We find that resonant stimulation indeed increase the intensity of vibration of phonons of corresponding frequency and decrease the intensity of phonons of other mode. This provides us a helpful tool to precisely stimulate phonons of certain mode and study the properties of material.
## Contents

Supervisory Committee ii

Abstract iii

Table of Contents iv

List of Figures v

Acknowledgements vii

1 Introduction 1

2 Introduction to Superconductor 2
  2.1 BCS Theory .................................................. 5
  2.2 High-Temperature Superconductor .............................. 6

3 Introduction to ARPES 8
  3.1 Photoemission Effect ........................................... 10

4 Experimental Consideration 11
  4.1 Pump-Probe Photoemission .................................... 11
  4.2 Stimulate Phonons Resonantly ................................ 12

5 Experimental Details 13
  5.1 Optical Pulse Train Generator ................................. 13
  5.2 Data Analysis .................................................. 17

6 Experimental Results 22

7 Conclusion 30
List of Figures

Figure 2.1 Reproduced by [10]. The non-superconductor materials always have certain resistance no matter what is the temperature which the superconductors will lose all the resistance once the temperature below the critical temperature. .............................. 3

Figure 2.2 Reproduced from Wikipedia [11]. The Meissner Effect. When the temperature is higher than transition temperature, the external magnetic field will permeate the material. When the temperature is lower than transition temperature, the external magnetic field is expelled by the surface current. ............................... 4

Figure 2.3 Reproduced by [12]. Once an electron scatters, the atoms or basis will be attracted by it and the lattice structure around it will be distorted and this region will be positively charged. Later, another electron with opposite spin will be attracted by this charged region. ................................................................. 6

Figure 2.4 Figure adapted from [12]. This figure shows the transition temperature vs. year of discovery of different superconductors. There is a large jump in 1986 when the high-temperature superconductors were discovered. ................................. 7

Figure 3.1 Reproduced from Wikipedia [9]. Firstly, a beam of photons with energy $h\nu$ hits the sample in a certain angle and some electrons get enough energy to escape to the vacuum. Then, the detector will collect these electrons and analyze their momentum and kinetic energy. Based on momentum and energy conservation, information about the initial momentum and energy of these electrons will be obtained ................................. 9
Figure 5.1 Cartoon of the optical pulse train generator. The blue lines means 8 mirrors. The orange small rectangles in the middle represents the 3 beam splitters. The two large rectangles are two movable stages controlled by computer. 16

Figure 5.2 How the optical pulse train looks like in reality. The direction of the light path is exactly the same with the cartoon. 16

Figure 5.3 One spectra of ARPES. The horizontal axis is kinetic energy and the vertical axis is emission angle in degrees which can be converted to momentum. 17

Figure 5.4 Integrate a spectra along y axis. This figure shows the delay time vs.kinetic energy. 18

Figure 5.5 Illustrate intensity vs. delay time. the highest peak shows the stimulation pulse. Along time, the vibration decays. 19

Figure 5.6 Subtract smoothed graph from the original graph to show the intensity of the vibrations. 20

Figure 5.7 Final FFT result for data set 3. These peaks represents some frequencies extract from last graph. 20

Figure 6.1 The first experiment data. There is only on pump pulse with average power 3.5 mW and shows no obvious lattice vibrations. 23

Figure 6.2 Pump with power 3.5 mW. Illustrate FFT amplitude vs. FFT frequency. This scan also shows no obvious vibrations. 24

Figure 6.3 FFT amplitude vs. FFT frequency with one single pulse which has a energy of 16.5 mW. This shows the main peak at the expected position. And we can also see the clear weaker peaks which are also in a reasonable position. 25

Figure 6.4 Twp-pulse pump stimulates phonons. The main peak is still in the right position while the other peaks are supressed. 26

Figure 6.5 4-pulse pump stimulation. Because this is not the natrual frequency of the material, the background noise is even stronger than before. Also, the main peak has been shifted with the change of the pump frequency. 27

Figure 6.6 This is a $2^4$ repetition rate enhancer with 8 optical arms. The longest arm is about 1 meters long. 28
ACKNOWLEDGEMENTS

I have to thank my advisor Professor Daniel Dessau. He gives me a lot of support and encouragement. As an excellent advisor, he taught me how to think out of the cage and always enlighten me to pay attention to somewhere I ignored. He is patient to help me to understand superconductivity, ARPES and anything confused me. His unique management of time and energy helps me to be an independent researcher.

Also, I would like to thank my mentor Dr. Stephen Parham. He is always the most helpful one. He helps me to design and build my first optical system. He is the one from whom I learned how to conduct experiments and what is a real researcher. He does not only teach me physics theories and technology skills, but also the right attitude toward science and experiments.

Conducting experiments is never an individual work, so I have to thank every one in the lab: Tom, Justin W., Justin G., Haoxiang, Xiaoqing, and Jason. They are always nice and willing to explain to me about everything. I am grateful for every lab member’s contribution to my project. I am glad to be one of this group.
Chapter 1

Introduction

In conventional superconductors, superconductivity is believed to be related to electron-phonon coupling[2]. Though, the exact mechanism of high-temperature superconductor is not found yet, there is a thinking that the iron pnictides is similar to cuprate superconductors. So that electron-phonon coupling may be also responsible for the superconductivity of this kind of high-temperature superconductor. To confirm this conjecture, we need to excite phonons and then analyze the behaviors of electrons to study the properties of the material. In the past, we usually stimulate phonons generally and ignore their mode. Therefore, the measurement is not precise enough and the oscillations are not clear enough. Now, we want to find a way to get stronger oscillations and selectively stimulate phonons.

In this thesis I will investigate the feasibility of resonantly stimulating coherent phonons in a sample using the optical pulse train generator. In the first several chapters, I will cover some necessary background in the topics of superconductivity, phonon-electron coupling and photoemission especially pump-probe photoemission. Since we use Angle Resolved Photoemission Spectroscopy (ARPES) to collect the escaping electrons which is powerful and helpful, I will devote one chapter to its basic operation and the advantages it has over other measuring instruments. Then I will talk about my experimental consideration and the real layout of my optical system and explain how it works. In the final chapter, I will show some of the data I got and investigate what we could learn from these data. To confirm that optical pulse train generator indeed stimulate phonons resonantly and it is helpful, I will compare my data with those from previous experiments.
Chapter 2

Introduction to Superconductor

Superconductors are one kind of materials that conduct electricity with zero resistance when below a certain temperature called $T_c$. Besides this widely accept fact, superconductors have another property called Meissner Effect[1], which was found in 1933. Meissner Effect is a mechanism for diamagnetism which means that in weak field, superconductors almost exclude all the magnetic flux. It is reached by generating current on the surface of the superconductor. The surface current will generate a magnetic field which will cancel the external magnetic field and result in a 0 magnetic field inside the superconductor. Figure 2.1 and Figure 2.2 shows the properties of a superconductor. Superconductivity was firstly found in 1911 by Heike Kamerlingh Onnes on solid mercury. In that moment, he used liquid helium as a refrigerant and kept the temperature at 4.2K. Later, in 1913, lead was found to express superconductivity at the temperature 7K, and in 1941 niobium nitride was found to show superconductivity at the temperature 16 K. Evermore, scientists take a large of time and energy to study superconductors, they try to explain this phenomenon and increase the transition temperature $T_c$. 
Figure 2.1: Reproduced by [10]. The non-superconductor materials always have certain resistance no matter what is the temperature which the superconductors will lose all the resistance once the temperature below the critical temperature.
Figure 2.2: Reproduced from Wikipedia [11]. The Meissner Effect. When the temperature is higher than transition temperature, the external magnetic field will permeate the material. When the temperature is lower than transition temperature, the external magnetic field is expelled by the surface current.
2.1 BCS Theory

It was until 1911 that the first microscopic theory of superconductivity was established by Bardeen, Cooper and Schrieffer. In 1957, Bardeen, Cooper and Schrieffer formally published their theory of superconductivity. According to the inventors' names, this theory is named BCS theory. The core of the BCS theory is that the vibration of the lattice of crystal will cause the interaction between two electrons when the energy difference between the electrons states involved is less than the phonon energy [7]. BCS theory won Bardeen, Cooper and Schrieffer a Nobel Prize in 1972. Cooper showed the existence of this two-electron pair, so it is called Cooper pair. Figure 2.3 illustrates how an electron distort the charge equilibrium of the lattice. First, an electron scatters from one state to another. Because of the movement of this electron, the local balance of charge is broken and the lattice is locally positive charged. Later, this positive charged region will attract another scattering electron with opposite spin. In general, these two electrons are paired and make a Cooper pair. To be more specific, two electrons are paired in the way of virtual exchange of phonons [7]. In another word, we could also say that this is an electron-phonon coupling. This effect also provides a powerful means to coherently control the electronic structure of materials[4].
2.2 High-Temperature Superconductor

After the emergence of BCS theory, most of the superconductivity phenomena can be explained. However, in 1986, Bednorz and Müller announced that they found a new superconductor named Lanthanum Barium Copper Oxide which has a transition temperature at 35K. This discovery violated the prediction of BCS theory which said that the maximum $T_c$ based on electron-phonon coupling is about 32K [1]. Later, superconductors with $T_c$ at 41K, 55K, even 90K were founded continually. Figure 2.4 shows a timeline of the discovery year of superconductors with different transition temperature.
Figure 2.4: Figure adapted from [12]. This figure shows the transition temperature vs. year of discovery of different superconductors. There is a large jump in 1986 when the high-temperature superconductors were discovered.

It seems like BCS theory cannot explain superconductivity for high-\(T_c\) superconductors. However, recently, some scientists suspect that the mechanism of high-\(T_c\) superconductor is still related to electron-phonon coupling[2]. Till now, it is not clear how closely related Fe-based and Cu-based superconductors are, or that they should have the same mechanism of pairing. One class could be phonon-based and one class could be something else. My project is working on the Fe-based superconductor, but there are some people like Reznik[8] also think that Cu-based ones may be electron-phonon.
Chapter 3

Introduction to ARPES

Angle resolved photoemission spectroscopy (ARPES) is a powerful tool to investigate the electronic structure of a solid system[1]. Instead of integrating over momentum, ARPES gives information on the direction, speed and scattering process of valence electrons in the sample which means that it provides information on both energy and momentum and makes it possible to study band structure and Fermi surface. Figure 3.1 is a simple cartoon illustrates how ARPES analyzes electrons. The operation of ARPES is based on photoemission. Hence, it is necessary to introduce photoemission. Compared with normal ARPES, trARPES extends and compliment the conventional ARPES by adding femtosecond time-resolution. Instead of conducting normal photoemission, trARPES sends a femtosecond infrared laser pulse to excite electrons first. After certain delay time, the probe pulse will be send to finish the photoemission and the detector will collect the escaping electrons. This process will give a time revolution for several hundred femtoseconds.
Firstly, a beam of photons with energy $h \nu$ hits the sample in a certain angle and some electrons get enough energy to escape to the vacuum. Then, the detector will collect these electrons and analyze their momentum and kinetic energy. Based on momentum and energy conservation, information about the initial momentum and energy of these electrons will be obtained.
3.1 Photoemission Effect

Photoemission effect was firstly observed by Heinrich Hertz in 1887. Later, Philipp von Lenard discovered the important laws in experiments that the number of emitting electrons is proportional to the intensity of incident light. But these electrons’ kinetic energy does not depend on the intensity of incident light but the wavelength of the incident light. Until 1905, Einstein explained this phenomenon in theory. According to Einstein, if a photon with enough energy is absorbed by an electron in a solid, this electron will escape to vacuum. This theory won Einstein a Nobel Prize in 1921 and laid a foundation of quantum physics. The final energy of the escaping electron is given by:

\[ E_f = E_i + h\nu - \Phi \]

Where \( E_f \) and \( E_i \) represent the final and initial energy, \( h\nu \) represents the energy of one photon and \( \Phi \) is the work function. Since most materials have a work function around 4 – 5 eV[1], once we control the photon energy and measure the final energy, the initial energy of the electron is obvious. In ARPES, we additionally use momentum conservation. Therefore, we could extract the momentum distribution of electrons based on their photoemission angles [1].
Chapter 4

Experimental Consideration

The properties and collective phenomena of materials are determined by the electrons with lowest binding energy. Therefore, the electron-phonon coupling is strongly related to materials’ properties. Our goal is to stimulate phonons in certain mode and study the electrons. If we could control the phonons, then we will get stronger vibrations which gives us clearer ringing and is helpful for our study.

4.1 Pump-Probe Photoemission

Normal photoemission can also help us to study the interaction between electrons and phonons in a different way. However, what we want is to stimulate phonons first and then, after certain delay time, shot another probe photons with UV laser to let some electrons escape into vacuum and analyze the momentum and energy of these electrons. Hence, we need pump-probe photoemission. Instead of simply hitting the sample with a beam of photons with enough energy, we hit the sample with some lower-energy photons first, which are usually infrared photons. These photons cannot provide energy to electrons to escape into vacuum. But, they are strong enough to cause the vibrations of the lattice, that is, stimulate phonons. After photoexcitation, hot electrons will be generated, populating states above the Fermi level, which then decay through allowed electron-electron and electron-phonon channels[5]. After certain delay time we want, we will hit the sample with another beam of photons with higher energy, which are usually ultraviolet photons. These photons will provide enough energy to electrons to enable them to leave the sample. Finally, the detector will collect and analyze these electrons. As a result, we could
learn about the band structure and electron distribution in the sample.

4.2 Stimulate Phonons Resonantly

To make the vibrations of lattice to be as obvious as possible, we need to stimulate phonons as intensive as we can. The most efficient way is stimulating it resonantly. Just like we play with swings. If the frequency of the pushing equals to the frequency of the swing itself, then we can use less energy to push the swing to a very high position. Now, we want to do the same thing to the phonons.
Chapter 5

Experimental Details

The most important part of the pump-probe photoemission is the pump. Therefore, we need to design and build a system to create pumps in a certain frequency. One way to achieve this goal is to design and build an optical pulse train generator which will produce a bunch of pulses to stimulate phonons. Because coherent phonons are lattice vibrations having the same frequency, using multiple pulses to control them is possible [6]. Also, according to previous experiments, the determined FFT (Fourier transformation) frequencies do not vary with the excitation density and are temperature independent [2], we do not need to specifically control the temperature of the environment around our sample.

5.1 Optical Pulse Train Generator

An optical pulse train generator is an optical system which will split one laser pulse into a bunch of pulses and set their repetition rate. To build an optical pulse train generator, we only need beam splitters and mirrors. To make \(2^n\) pulses, \(n\) beam splitters and \((4n + 2)\) mirrors are needed [3]. Beam splitters are used to divide one beam into equally two beams, and mirrors are used to change the direction of beams and set the light path for each pulse in order to set the frequency of the pulse train. Figure 5.1 and Figure 5.2 show how the optical pulse train generator looks like.

What we build is a \(2^3\) optical pulse train generator with 3 beam splitters and 8 mirrors which means, in theory, we will get 8 pulses in the end. There are 4 pairs of mirrors, and each pair set an arm of the system. Each of these 8 pulses will travel a
different combination of arms. So the length of each arm will set the length of the light path of each beam, thus set the repetition rate of the pulse train. In fact, it is unnecessary to use this many pulses to stimulate phonons. Hence, we just use half of them which will come out parallel to the incident laser pulse. But we still want 3 beam splitters. Because the intensity of our incident laser is usually about 16 mW. If there are only 2 beam splitters, the energy of each output pulse will be too high for stimulating goal. As a result, there will be multi-photon photoemission which adds a background signal of count. Hence we have to keep the pump power low enough that this background doesn’t ruin the data.

The main difficulties I encountered is aligning these optical elements. To ensure that the light paths of each pulse are exact the length I designed and the beams are in a horizontal plane, I have to make sure that all the mirrors and beam splitters are vertically aligned and their reflecting angle could change the direction of incident lights and keep the light in the right path. What I did is firstly covering all the other arms and only left one shortest light path. I used ruler to measure the height of beam in different positions and adjust mirrors and beam splitters to let the beam in the same height everywhere. In the end, I let the beam to pass two small holes in the same height and in a straight line. These two small holes could be treated as two points to set a straight. After these procedures, I set one pulse in the right light path and settled the output path for all the pulses. Then, I uncovered other arms one by one. After adding each arm, I will adjust the mirrors that set this arm to make the output light from this arm perfectly overlapped with the one I just aligned. To confirm that all the pulses are indeed overlapped, I will let the output light to travel a long distance and still be overlapped.

To control the movable stages precisely, I use Labview to set up a program to control these arms. In my program, we just need to type in the delay between pump pulse and probe pulse, the program will automatically calculate the distance that the mirrors need to move to satisfy the delay time and move them to the right positions. There are two modes in the program. The first one is to set two arms separately. In this mode, we do not need the 4 pulses to be evenly spaced and we could set different spacing time between different pulses by setting the arms separately. The another mode is to set the two arms together. In this mode, we want these 4 pulses are evenly spaced. Therefore, once we input a delay time, the
program will regard this as the spacing time between any neighbor pulses and calculate the distances that both arms should travel and move them one by one.

Previous experiments tell us that the natural frequency of Fe-based superconductor is 5.6 THz[2][4]. In addition, resonant stimulation requires the accuracy in frequency of our pulse train. Hence, two of the arms are built on movable stages which are controlled by computer and can move backward and forward. Once we input the ideal frequency, computer will calculate the corresponding spacing time according to the equation:

$$T = f^{-1}$$

Where T is the spacing time and f is the frequency. Then according to

$$S = cT$$

where S is the distance and c is the speed of light, the length of the arms will be obtained. Then, the movable arms will be set to the corresponding position to achieve the desired frequency.

In this project, our pump pulse is infrared laser and the probe pulse is ultraviolet laser. The average power of our infrared laser is about 16 mW, the repetition rate is 20 KHz and the spot of the light has a diameter of 180 micrometers. From these information, we could calculate the pump fluences $F_{abs}$.

$$F_{abs} = 16 \frac{mJ}{sec} \frac{1sec}{20000 pulses} \frac{1 pulse}{\pi (90 \mu m)^2} \left( \frac{10^4 \mu m}{cm} \right)^2 \frac{J}{2 \times 10^4 \times \pi \times 8100 (cm)^2} = 3 \frac{mJ}{(cm)^2}$$

Our sample is a piece of $CaFe_2As_2$ which is a kind of Fe-based superconductor. This sample is cleaved in vacuum and all the procedures are conducted in vacuum. According to the data from Wolf [2], the rings of the sample in independent of temperature. Therefore, we do not pay much attention on the environment temperature and the typical temperature of the environment is about 200 K.
Figure 5.1: Cartoon of the optical pulse train generator. The blue lines means 8 mirrors. The orange small rectangles in the middle represents the 3 beam splitters. The two large rectangles are two movable stages controlled by computer.

Figure 5.2: How the optical pulse train looks like in reality. The direction of the light path is exactly the same with the cartoon.
5.2 Data Analysis

Our data collected from ARPES is a spectra and we need to figure out the frequency of the vibration from it. Now I will take one of the data sets to show the process of analyze a spectra.

First, our spectra from ARPES looks like Figure 5.3. Each point in the spectra means electrons with corresponding kinetic energy and momentum.

![Figure 5.3: One spectra of ARPES. The horizontal axis is kinetic energy and the vertical axis is emission angle in degrees which can be converted to momentum.](image)

This graph is a concatenation of spectra from different delay time. So it shows momentum vs. kinetic energy vs. delay time. We can tell from the graph that the Fermi level is about 2.05 ev. Then we integrate the area in the blue rectangle which means integrating by momentum along y axis and get the graph as shown in Figure 5.4 which illustrates delay time vs. kinetic energy.
Figure 5.4: Integrate a spectra along $y$ axis. This figure shows the delay time vs. kinetic energy.

Now, we integrate the area in the blue rectangle which means integrating by kinetic energy along $x$ axis. In this graph, the Fermi level is about 2.04 THz, so our integrating area is just above the Fermi level and we can count the hot electrons that escape from the material. When the delay time is 0 ps, the pump pulse hits the sample and excites some electrons and the probe pulse hit the sample at the same time. As time goes by, the hot electrons decay and back to the stable states. Finally, we get a graph shows the intensity vs. delay time as showed in Figure 5.5.
Figure 5.5: Illustrate intensity vs. delay time. The highest peak shows the stimulation pulse. Along time, the vibration decays.

This figure shows the intensity of hot electrons vs. delay time. The delay time means the spacing time between the pump pulse and probe pulse. When the delay time is negative, it means that the probe pulse is before the pump one, so there is hardly existing hot electrons. When delay time is 0, it means the pump pulse and probe pulse are perfectly overlapped and we can see the intensity of the hot electrons reach the largest number. After the pump pulse, the hot electrons will decay along time. As a result we can tell from the figure that the intensity decays along time. But because we stimulated phonons, and phonons and electrons are coupled, there are still some ringings on the top of the decay graph. These ringings are what attract us and they show the electron-phonon coupling in the material. To get rid of the background noise, we smooth the graph with boxcar smoothing of 5 points. Then we subtract the smoothed graph from the original graph. Figure 5.7 show the results.
Figure 5.6: Subtract smoothed graph from the original graph to show the intensity of the vibrations.

We pay our attention on the region after stimulation which means after the highest point and apply the fast Fourier transformation (FFT) to the region after the highest point which means the 0 delay time to exclude the noise bump before tzero. The background noise is an artifact of smoothing because the real data rises faster than the boxcar width. The result is showed in Figure 5.8.

Figure 5.7: Final FFT result for data set 3. These peaks represents some frequencies extract from last graph.
There are many interesting physics hiding in the FFT graph. We will see more FFT graphs for different stimulating mode in next chapter and analyze them in detail to see what we could learn about the sample and our optical pulse train generator.
Chapter 6

Experimental Results

In this chapter, I will show some results of the pump-probe photoemission. When materials receive a stimulation, the atoms receive impulse force and they initiate to keep pace with the neighbors. These atomic motions are called coherent phonons. Because we pump the phonons with infrared laser, what we excite are optical phonons which means the out-of-phase movements of the atoms in the lattice. We pump the coherent phonons in different ways to see if our optical pulse train generator could really control the phonons and how great the influences are that the pump process has on the lattice vibrations mode.

According to previous experiments, there should be at least three sharp intense peak in the FFT frequency graphs[2]. The main peak should be at $\omega_1 = 5.6$ THz. Also, there should be another two weaker peaks at $\omega_2 = 3.3$ THz and $\omega_3 = 2.6$ THz. These features are likely to be fingerprints of coherent lattice vibrations and are observed in ARPES due to the coupling between the electronic system and the coherent phonons [2]. Hence, once we could cause the lattice vibrations, we are supposed to see these peaks.

To test our optical pulse train generator, we firstly only use one red pulse to stimulate phonons. We did this experiment twice with different intensities. Figure 6.1 and Figure 6.2 show the results of two different sets of data.
Figure 6.1: The first experiment data. There is only one pump pulse with average power 3.5 mW and shows no obvious lattice vibrations.

In this scan, our pump energy is only 3.5 mW which is too low, so that it is hard to tell the ringing from the background noise. Therefore, the graph shows no typical peaks at the supposed positions. Also, there are a bunch of peaks after 8 THz which are not reasonable, because they are above our Nyquist Frequency. The Nyquist Frequency comes from the Nyquist theorem which means that we must have at least two sample points per period of a sinusoidal wave in order to figure out its frequency. The temporal frequency of our project is 40 fs which means that the oscillation we are looking at will be sample every 40 fs. According to

$$f = \frac{1}{T} = \frac{1}{40(\text{fs})} = 25 THz$$

But we need to sample points, we can only find the accurate frequency of waves at half of the frequency we get above which is 12.25 THz. As a result, all the peaks above 12.25 are not reasonable. However, as we get closer and closer to 12.25 THz, the samples get less and less and the aliasing effects occurs which means the data will not be accurate enough. So we usually do not pay attention to any frequencies above 8 THz.

To confirm the result, we do the same scan again. The result of data set 2 is showed in Figure 6.2.
Figure 6.2: Pump with power 3.5 mW. Illustrate FFT amplitude vs. FFT frequency. This scan also shows no obvious vibrations.

From the FFT graph of the data set 2, we still do not see these fingerprints peak which means that this pump power is indeed not high enough to stimulate phonons to give obvious ringing and it cannot help us to study the electron-phonon couplings. So, we improve the power of the infrared laser to about 16.4 mW and collect data set 3. Figure 6.3 shows the result of data set 3.
Figure 6.3: FFT amplitude vs. FFT frequency with one single pulse which has a energy of 16.5 mW. This shows the main peak at the expected position. And we can also see the clear weaker peaks which are also in a reasonable position. 

This is a very clear graph of lattice vibrations with the fingerprints peaks we expect. The main peak is at $\omega_1 = 5.6$ THz compared with the frequency obtained by Wolf 5.6 THz [2]. Besides, we find a weaker peak at $\omega_3 = 2.8$ THz. In the paper, the peak is at around 2.6 THz. Though this weaker peak shifts a little bit, it still in a acceptable range. So we could say it is consistent with the previous data. But, our second strong peak is found at $\omega_2 = 3.8$ THz instead of the 3.3 THz. Our data is consistent with the theoretical Raman active mode because the Raman active mode was found at 16 meV corresponding to 3.8 THz[2] which is exactly where our peak is. As a result, data set 3 shows that we successfully stimulate phonons in the $CaFe_2As_2$ and our optical pulse train generator successfully stimulated coherent phonons and we could try to use it to control these coherent phonons with the pulse train.
With the success in single pulse stimulation, we try to improve the number of pump pulses into 2 and stimulate phonons resonantly. Figure 6.5 shows the result of a two-pump-pulse stimulation.

Figure 6.4: Two-pulse pump stimulates phonons. The main peak is still in the right position while the other peaks are suppressed.

The spacing time between these two pump pulses are 180 fs which is the natural period of the $A_{1g}$ mode of coherent phonons. From this we could get the frequency of this mode.

$$f = 1/T = 1/180(fs) = 5.6THz$$

Hence, we expected to stimulate phonons resonantly. And the result is promising. The main peak is at $\omega_1 = 5.5$ THz and there is a weaker peak at $\omega_2 = 3.8$ THz which are consistent to the theoretical value. Compared with the graph of data set 3, the two weaker peaks are relatively much lower than the main peak which means that the lattice vibrations in these frequencies are effectively suppressed and we indeed control the coherent phonons. This result also confirms that resonant stimulation of phonons will give us a better recognition of the phonons in the natural frequency of the material.
Finally, we want to apply all the four pulses to stimulate phonons and test if we could drive the phonons and force them to respond to a different frequency. Our result is showed in Figure 6.6. This time, we do not try to stimulate the coherent phonons resonantly. Instead, we set the spacing time between two close pump pulses to be 200 fs which is slight larger than the resonant spacing time 180 fs. According to equation

\[ f = \frac{1}{T} \]

The stimulated frequency of coherent phonons should be 5.0 THz. In our graph, the highest peak is at the position \( \omega_1 = 5.1 \) THz, which is exactly consistent with our driving frequency. This result shows that the optical pulse train generator can not only stimulate phonons resonantly, but also drive the phonons in a mode that we want and achieve the goal of controlling the phonons.

![Figure 6.5: 4-pulse pump stimulation. Because this is not the natural frequency of the material, the background noise is even stronger than before. Also, the main peak has been shifted with the change of the pump frequency.](image)
Take all the data sets in total, compared with the results from Wolf [2], it is obvious that our data have much stronger background noise which can be improved by increasing the repetition rate. So my next project is to design and build a repetition rate enhancer. The repetition rate enhancer is a similar optical system with the optical pulse train generator as showed in Figure 6.7. I just finished building this repetition rate enhancer and aligning these mirrors and beam splitters. Ideally, this repetition enhancer will split each beam of light into 16 pulses which means that our repetition rate could be improved from 20 KHz to $20 \times 16 = 320$ KHz. Such a great improvement is supposed to give us better data quality. Compared with the precisely spaced pulses produced by the optical pulse train generator, the pulse train produced by the repetition rate enhancer does not have to be perfectly equally spaced. But it does not matter. Because the repetition rate means the number of pulses per second. Our largest delay time is 4 ps. The spacing time corresponding to 320 KHz is $\frac{1}{320(KHZ)} = 3\mu second$ which is much longer than the 4 ps. Therefore, even the pulses are distributed slightly uneven, they won’t affect the result. Besides, the error of the length of each arm is less than 1mm which means
the spacing time error is less than \( \frac{1 \text{ (mm)}}{3 \times 10^8 \text{ (m/s)}} = 3 \text{ (ps)} \). However, the repetition rate enhancer still needs to be tested before applying to real application.
Chapter 7

Conclusion

Femtosecond time- and angle- resolved photoemission spectroscopy is determined to be a powerful tool to investigate the response of electron structure around the Fermi level. In this study, we try to quantum control the coherent phonons in $CaFe_2As_2$ sample to study the electron-phonon coupling. To achieve this goal, we apply the pump-probe photoemission to stimulate phonons and analyze the response of the lattice structure. The optical pulse train generator is designed to complete this mission. In our study, we stimulate phonons with different fluence and various number of pump pulses. Also, we try to drive the coherent phonons. The results shows that the optical pulse train generator is effective to control the coherent phonons and will help us to study the electron-phonon coupling and electron structure in a material.

Future works are still necessary. For example, when we applied a window to crop the graphs, it is equal to time the original graph by a square wave with sharp edge. This process will cause the edge effect and create some ripples in our FFT frequency graphs and affect the accuracy of our data. Therefore, in the future, we need to figure out a way to reduce this edge effect. For example, we might multiply a square wave with smooth edges instead of sharp ones.

Also, our background noise is still very strong. My next project is the repetition rate enhancer. We need to test it and then try to apply it to the optical pulse train generator project to see if it can improve the quality of our data.
Bibliography


