Introduction

During this summer's REU program I worked in the Kapteyn/Murnane labs under Margaret Murnane. Professors Kapteyn and Murnane and their graduate students use the science behind ultrafast optics to probe realms that are unseen to the common eye. They employ a variety of techniques and ideas to probe tiny structures on thin films, observe the rotation and vibration of molecules, and generate laser-like beams of X-rays and ultraviolet light that can be manipulated at will. The driving force behind their experiments is the need to know what happens on a scale so small that conventional methods of observation can only provide marginal information about the system being observed. And they do it all with light.

Part 1: Ultrafast Optics

The ultrafast optics that the Kapteyn/Murnane group uses refers to the use of pulses of laser light which last for a time that is on the order of tens of femtoseconds or less [2, p. 165]. Several physical processes occur at these time scales, meaning that ultrafast optical pulses are required to resolve them in time. These processes include photosynthesis, human vision, protein folding, and molecular vibrations, just to name a few.

The group uses a Ti:sapphire laser design that was first thought up in the early 1990's by Henry Kapteyn. Such a laser works as follows: An Nd:YAG laser emitting at 1064 nm and frequency doubled to a wavelength of 532 nm is fired through an "optical Z" (an arrangement of two adjustable mirrors that allows for easy adjusting of the beam path) and into a short focal length positive lens. At the focal point of this lens lays the

Ti:sapphire crystal, a crystal of sapphire that has been lightly doped with titanium atoms. The crystal lies between two concave, partially reflecting mirrors. One of these mirrors allows the beam to enter the cavity from the lens and another partially transmits the beam into an absorber since it is not needed. See Fig. 1.
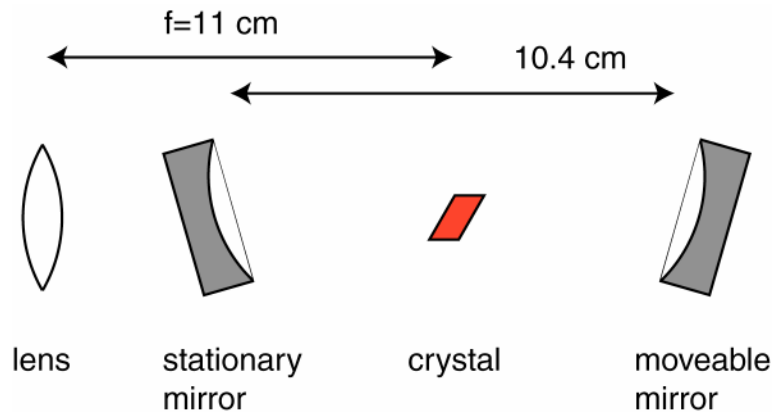

Fig. 1 A diagram of the cavity of a Ti:sapphire laser.

The purpose of the crystal is to produce the wide bandwidth laser light that can be harnessed to produce to ultrashort pulses. The titanium in the sapphire is needed to create an energy level of the crystal that is quantum mechanically suitable to emit laser light when stimulated by the pump laser. The light emitted from the crystal is reflected back and forth between the mirrors in the cavity, slowly amplifying the light until the crystal begins lasing. Two prisms and two other mirrors (one acting as the output coupler) are used to correct any distortions in the beam such as chirp. Chirp is a distortion of an ultrafast pulse where certain colors in the beam arrive before or after others [3, p. 2]

A useful aspect of this laser geometry is its ability to modelock. Modelocking is a method that is well-know to laser physicists for its ability to create short pulses in the laser's output. In a laser beam, there is often not just one frequency (or color) present, but

a band of frequencies that ranges between two values. The bandwidth of this output spectrum is determined by many things, but mostly by the type of laser used. All these frequencies are usually not matched in phase, meaning that a peak in the wavelength of one frequency usually doesn't coincide with a peak in another. Over a band of frequencies, these phase mis-matches result in an average output power that does not change in time but remains constant. Modelocking rearranges all of the phases in the colors of the beam so that they are closely lined up [4, p. 524]. By matching the phases of the colors, pulses in the power output of the light can be obtained. This phenomenon is similar to beating, in which the superposition of two waves of slightly differing frequencies results in a periodic modulation of the intensity of the resultant wave (in sound, these slightly differing frequencies sound like a beating pattern) [6, p. 196]. The peak power in a modelocked laser beam is usually more than the average power of a non-modelocked beam. Thus, modelocking is a good method for producing the high power, short duration pulses needed in ultrafast optics.

The downside to modelocking can be seen in the uncertainty principle for waves, $\Delta\omega\Delta t \geq \dfrac{1}{2}$. In order for a laser pulse to be short in duration, Δt, it must contain a large number of frequencies or colors, $\Delta\omega$ [1, p. 39]. Lasers such as helium neon, CO_2, or laser diodes do not have a high enough bandwidth (bandwidth means the number of frequencies in the beam) to produce pulses of a short enough time scale. Dye lasers have a wide bandwidth but Ti:sapphire lasers have one of the widest bandwidths (100 THz) of any lasing medium known [4, p. 480]. For this reason, Ti:sapphire is the lasing medium of choice for any ultrafast optical physicist.

High Harmonic Generation

Now that we know how to produce ultrashort, intense pulses, the next question is, what can we do with them? The Kapteyn/Murnane group has been working on the applications of ultrafast lasers for the past ten years and has pioneered many of the methods found in their labs.

The main physical process that the Kapteyn/Murnane group covers is high harmonic generation (HHG). HHG is a process that is very similar to second harmonic generation (SHG). SHG is also commonly known as frequency doubling, a process by which light is sent through a crystal in a manner in which two photons combine to form one photon of double the frequency. By Planck's relation for photon energies, $E = h\nu$, the new photon also has double the energy [3, p. 40].

Both HHG and SHG are nonlinear processes, which are processes that are dependent on the intensity of the light and can not be represented by a linear wave equation [6, p. 541]. The polarization of a material, or number of dipoles per unit volume [5, p. 166], is given by the equation

$$\vec{P} = \varepsilon_0 \chi \vec{E} \tag{1}$$

where ε_0 is the permittivity of free space, χ is the electric susceptibility, and $\vec{E}$ is the electric field applied to the medium [5, p. 179]. Equation (1) can be used in the optical wave equation,

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \tag{2}$$

to describe the flow of light through a linear material [3, p. 37]. In a nonlinear material where the intensity of the light is extremely large, the polarization depends on higher orders of the electric field,

$$P = \varepsilon_0 \left(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right) \qquad (3)$$

[3, p. 38]. Thus, the wave equation for the electric field becomes nonlinear and produces the effects found in SHG and HHG.

In HHG, photons are used to manipulate the atoms in a gas-filled capillary and produce photons that are integer multiples of the driving frequency (harmonics) of the incoming laser beam [2, p. 164]. HHG is usually used to produce extreme ultraviolet light (EUV) or x-rays. A gas must be used instead of frequency doubling crystals since EUV and x-ray light is absorbed by solids [1, p. 39]. The EUV light must also be allowed to pass out of the capillary and into vacuum since it is easily absorbed by air.

What actually happens on the molecular level of the gas goes like this: An incoming, intense pulse of laser light hits a gas molecule. The strong electric field of the pulse alters the potential energy of an electron enough to ionize the molecule and accelerate the electron away from the atom. Since the duration of the laser pulse is very short, the electric field will change directions quick enough before the electron gets away and accelerate it back towards the molecule. [1, p. 39] If the electron happens to recollide with the gas molecule, it will produce EUV or X-rays that have frequencies that are integer multiples of the driving pulse frequencies [2, p. 164]. Alternatively, as the HHG generated photons increase in frequency from the multiple electron collisions, their energies increase into the EUV and X-ray range.

Some Problems with HHG

The efficiency of HHG at the moment is limited to between 10^{-4} and 10^{-5} [1, p. 41]. Most of the losses in HHG occur in the plasma formed by the gas being ionized [7, p. 51].Despite this limitation, useful photon flux levels are still obtained (around the order of 10^{12} photons/sec) [1, p. 41]. One of the major ongoing studies in the lab is trying to find ways to increase the efficiency of HHG to higher flux levels to improve the quality of the experiments being done.

Another big problem in the area of HHG is the problem of phase matching between the X-rays that are produced. Typically, an X-ray will be produced at a certain point on the waveform of the driving laser pulse. (See Fig. 2). In any medium, the index of refraction (and thus the velocity of the light wave) will change as a function of the frequency of the light [6, p. 119]. Because of this, the X-rays travel at a different velocity than the driving laser light. The X-rays will interfere with each other after being created at the certain points of the driving laser waveform, causing the sum of the all these X-ray's phases to cause destructive interference and thus also to limit the efficiency of HHG. This can be somewhat corrected by adjusting the pressure of the gas in the capillary to balance the dispersions of the driving light and the higher harmonics so that the driving light interferes constructively and boosts the higher harmonics' intensities [1, p. 41]. Much theoretical work is being done by the group in the area of phase matching.
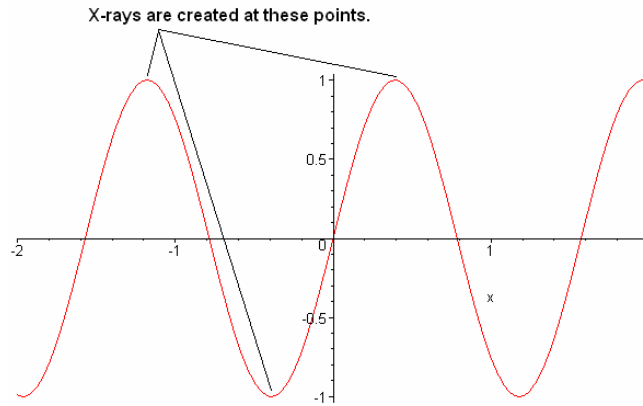
Fig. 2 Driving laser waveform and the points where the X-rays are created.

Part 2: Tools and Techniques Used in Ultrafast Optics

Working in ultrafast optics requires several tools and techniques that one usually doesn't typically use when working with optics in general. For example, to be able to use the X-rays that are generated through HHG, one must have a vacuum chamber setup in which they can travel since they are so easily absorbed by air. As another example, most electronic sensors have response times that are several orders of magnitude above the duration of an ultrashort laser pulsewidth. This means that traditional semiconductor electronics (such as an oscilloscope) can not be used to measure the characteristics of the laser pulse and an alternative method must be used. This part deals with several of the tools and projects I worked on that helped with the diagnostics and data taking in the lab.

Spectrometers

The first tool that I worked on and built was a red spectrometer. It is called a red spectrometer because it was primarily used to get intensity information in the red part of the optical spectrum. A spectrometer is simply a collection of lenses and a diffraction

grating that splits incoming light into its component colors. The setup of my spectrometer is seen in figure 3.
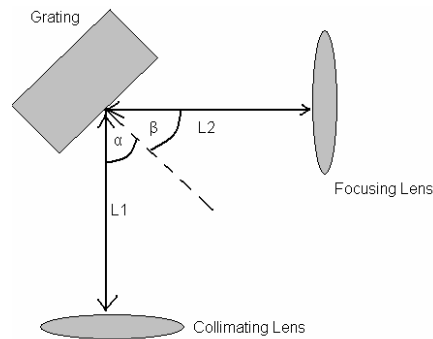

Fig. 3 A rough schematic of the layout of my spectrometer.

The spectrometer was built on a small optical bench that I obtained from the lab. A collimating 5 cm focal length lens was used to take the incoming light source and expand it to a diameter that was large enough to easily work with. The light emerging from a lens that comes from a source placed at the lens's focal point is parallel to the axis of the lens, according to the thin lens equation $\frac{1}{s} + \frac{1}{s'} = \frac{1}{f}$, where s is the distance of the light source from the lens, s' is the distance of the image from the lens, and f is the focal point [6, p. 51]. If s = f, then s' becomes infinity and hence parallel rays are observed.

The collimated beam next hits a reflective diffraction grating and splits into a spectrum of the colors in the beam. The grating had a line density of 600 lines/mm and was one inch wide. Several things I had to consider when choosing this grating were the free spectral range, the grating dispersion, and the resolution of the spectrometer. The free spectral range is the range of overlap between the different orders of the spectrum [Predrotti, p. 351]. Since the spectrum I was to be working with from the laser only ran from 700 nm to 900 nm, there was no worry about any overlap from different orders of the spectrum. The grating dispersion is the angular spread that the light in any specific

order will cover on a screen [6, p. 352-353]. The linear dispersion is just the linear

distance covered by an order of the spectrum and is given by $f \dfrac{d\theta_m}{d\lambda}$, where f is the focal

length of the focusing lens used and $\dfrac{d\theta_m}{d\lambda}$ is the angular spread per unit of wavelength in

the spectrum. The units of the linear dispersion are millimeters per nanometer. Finally,

the resolution is a quantification of how well a spectrometer can display spectral lines

that are separated by a small distance and is given by $\Re = mN$, where m is the order and

N is the number of illuminated grooves on the grating [6, p. 353-354]. Collimating is now

seen to be important since an expanded beam will hit more grooves and offer increased

resolution. All three of these characteristics had to be tweaked to attain a spectrometer

that was good enough to image a spectrum and obtain useful information from it.

A USB camera with the lens and infrared filter removed was placed at the focus

of the focusing lens. I wrote a LabVIEW program that was capable of interacting with the

camera and taking a color image of it (see figure 4 for the spectrum of a white light

source obtained from my spectrometer). The color image was converted to grayscale and

the intensity of each column of pixels was integrated to obtain a line profile of the power

vs. wavelength. The point of the spectrometer was to have a simple tool to get quick

diagnostics on a Ti:sapphire laser beam without having to resort to expensive,

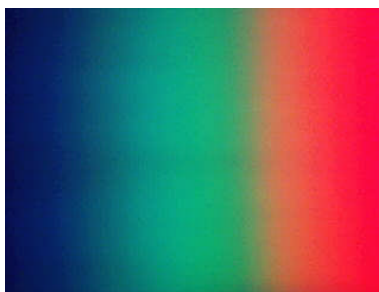commercial products that did more than what the group needed.

Fig. 4 White light spectrum obtained from my spectrometer.

Vacuum Chambers

The second thing that I worked on this summer was a small vacuum chamber. I built the chamber to test whether or not a CCD camera would work when placed in a vacuum. Since this was a "does it work or not" project, there wasn't any data to report. Nevertheless, I still had fun building it and taking apart a CCD camera.

A vacuum chamber is simply a vessel that is shut off from the atmosphere and has the gas in it removed to a certain extent. No true vacuum really exists. For that reason, there are several "degrees" of vacuum. Rough vacuum ranges from atmospheric pressure to about 10^{-3} torr; high vacuum goes from 10^{-3} to 10^{-8} torr; ultrahigh vacuum ranges from 10^{-8} to about 10^{-12} torr. The method used to reduce the pressure in the chamber is known as pumping. Several different pumps exist, including roughing pumps, turbopumps, and vapor diffusion pumps. Roughing pumps are pumps that are primarily used to reduce a system down to the lower range of rough vacuum. A rotary vane pump which uses a vane attached to a rotor to move air out of the system is an example of a roughing pump. Turbopumps are pumps that consist of many rotating rotors and stationary stators. As the rotors rotate, they impart momentum to the gas molecules and send them moving towards the exhaust. Turbopumps are high vacuum pumps. They are very inefficient at reducing a

chamber from atmosphere, but are very good at moving air in the high vacuum range. Vapor diffusion pumps are high vacuum pumps that use jets of oil to impart momentum to the gas molecules and move them out the exhaust.

My vacuum chamber was a small, hollow stainless steel cube with a hole in each face. Figure 5 is a picture of the completed chamber. Six blank NW 40 plates were used as is or modified to close off the chamber. A pressure sensor is seen on the left. Copper tubing attached to a venting valve and the roughing pump is on the right. A CCD camera is soldered into the back plate and viewable through the front window. During the main test, the pump ran for four and a half hours, maintaining a pressure of about 1 torr. The camera continued to work throughout the whole test, proving that CCD cameras will work in vacuum and image correctly.
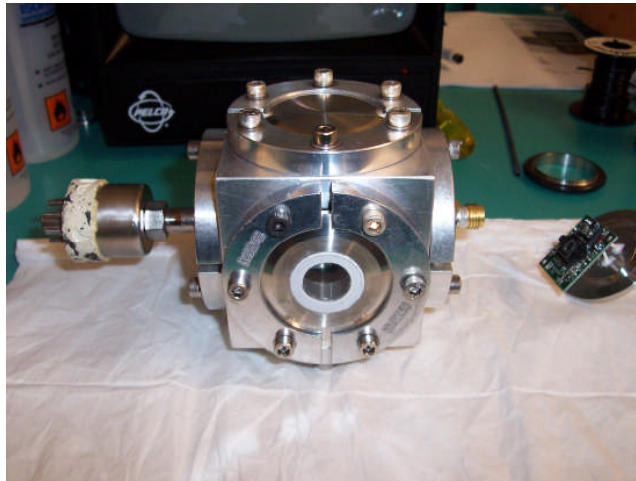

Fig. 5 The vacuum chamber.

Vibration Sensor

I just wanted to include this since it was a cool little machining project I worked on. I cut out a groove in a brass plate so that a small glass cube could sit in it firmly. The glass cube had two open ends. The end touching the plate was caulked in place. In one

corner of the brass plate I drilled and counterbore a hole for a screw for an optical post. On the post sat a mount that I found around the lab. Into the mount screwed a custom aluminum piece I designed and machined to hold a laser pointer. The glass cube was then filled halfway with water and the laser pointer aimed so that the beam reflected off the water's surface.

When the surface that the brass plate sat on vibrated, the surface of the water jiggled about and caused the laser pointer's reflection to dance about on the wall. The more the reflection danced about, the more the surface was vibrating. While this was not a method that could be easily quantified, it was a quick and easy test to see how much a surface was vibrating.
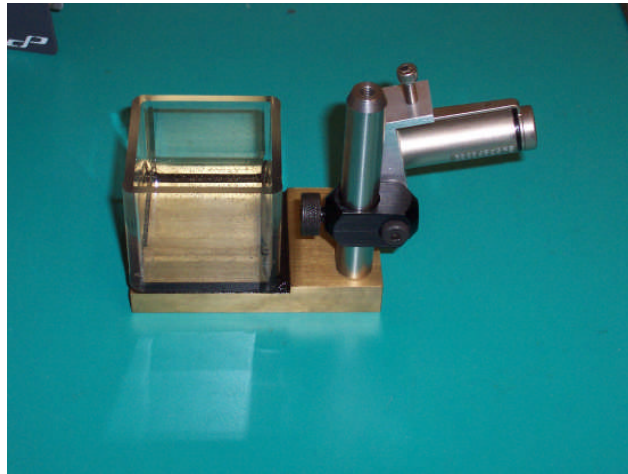

Fig. 6 My vibration sensor.

Conclusion

This REU experience was a very good one. I learned a lot about the hands on practical things that graduate students need to know, like taking initiative and machining. I also learned about the physics that is carried on by K/M labs at JILA and ultrafast

optics. I think the lessons learned will be invaluable when I get to graduate school. I now know how research works, so the learning curve will not be as great as if had I not attended the REU. Plus, it was fun!

References

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