

# **Designing and Building an Imaging Spectrometer to Study Angular Dependence of Fluorescence of Aligned Gas Molecules**

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## **Abstract**

This paper outlines the design, construction, and alignment of an imaging grating spectrometer that studies the angular dependence of fluorescent light from aligned gas molecules. We are attempting to replicate and eventually expand on the experiment done by Jinping Yao, et al., described in their paper, "Alignment-dependent fluorescence emission induced by tunnel ionization of carbon dioxide from lower-lying orbitals".[1]

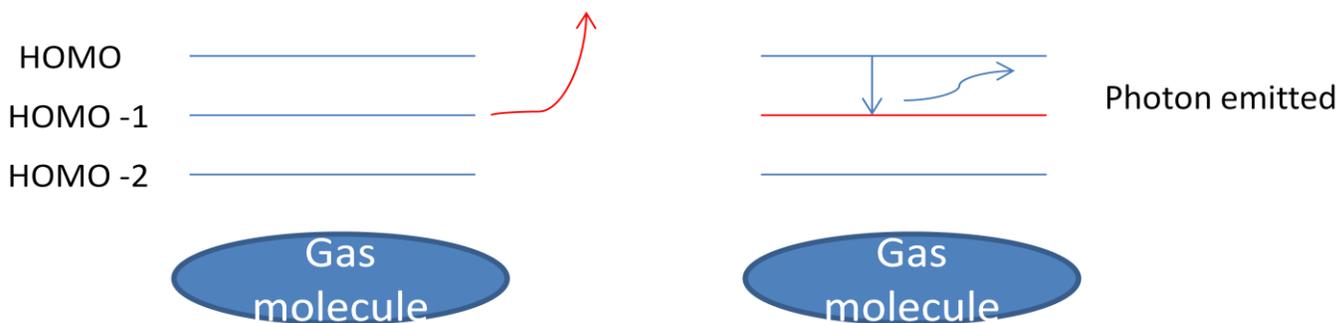
## **I. Introduction**

In order to begin designing the spectrometer, it is necessary to understand the light that is going to be measured and the mechanism that produces it. We begin with a Ti: sapphire laser that produces 30-40 femtosecond pulses centered at 800nm at a repetition rate of one kilohertz. This laser pulse is then divided in order to form a pump-probe setup. The pump pulse is then focused into the vacuum chamber where the gas molecules are emitted supersonically from a gas jet also running at 1 kHz. These now rotationally cold gas molecules are subjected to the strong electric field from the laser pulse and an electric dipole is induced when the negative electrons are pulled in the direction of the electric field. This electric dipole therefore feels a torque due to the electric field and the molecules begin to spin. Due to the quantization of angular momentum, these molecules will have revival structures where they will all align in the direction of polarization of the pump pulse after the laser pulse has left the area, creating what is called field free alignment. The much stronger probe pulse then enters the chamber at the time of the revival and coulomb explodes the gas molecules, i.e. ionizes electron which makes the gas molecules break their bonds along the molecular axis due to the repellent forces of the positively charged nuclei. These gas ions are then measured by a VMI (Velocity Map Imaging) setup where the ions are accelerated through a potential and collide with a phosphorescent screen. Based on the pattern that appears on the screen, the degree of alignment can then be back calculated. [2]

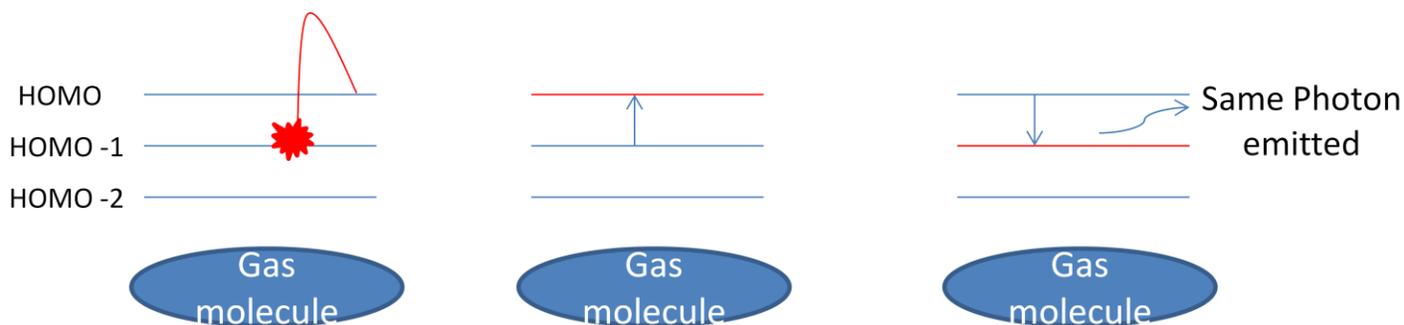
The phenomenon that my spectrometer will study occurs when the probe pulse enters the vacuum chamber in the aligned molecules which then fluoresce. There are two possibilities as to how this excitement process can occur: the first is for the HOMO (highest occupied molecular orbital) to decay into a spot where the HOMO-1 or HOMO-2 has tunnel ionized. The second possibility is that the HOMO is ionized and accelerated away in the laser electric field then comes back and collides with the molecule when the electric field switches direction. Unlike in high harmonic generation when the electron will recombine with the molecule, here the electron merely collides with the molecule which excites either the HOMO-1 or the HOMO-2 to the now vacant HOMO. The HOMO then decays again

into the same orbital it came from, emitting the same photon as if the HOMO-1 or HOMO-2 were tunnel ionized. (This possibility was not discussed in the paper that we are attempting to replicate.) Both of these processes will have angular dependence, but whether they have the same angular dependence is unknown. On a more technical note, the reason why more normal molecular excitement is not a large factor is because the Keldysh parameter is  $\ll 1$ . When this parameter, which is inversely proportional to the intensity of the laser focus, is less than one, then tunnel ionization is favored over multiphoton excitement.[3] The figures below show in a pictorial form the photon that gets emitted in the two different processes. Both of these processes after happen for HOMO-2 as well (not pictured). Why this process is angular dependent can be seen by looking at the molecular ADK theory.[4] What this means for building the spectrometer is that the energy level difference between the HOMO and the HOMO-1 or HOMO-2 is in the UV range, so the spectrometer will be built to measure UV light.

Tunnel ionization and decay



HOMO ionization, collision, and decay



## II. Design and Equipment

There are two major parts of the spectrometer design: the apparatus that will get the light from the laser focus to the spectrometer, and the actual spectrometer itself (the whole setup is illustrated in the figure below). The process begins with a Ti:sapphire laser pulse about 30-40fs in duration with a repetition rate of 1kHz being focused into the vacuum chamber. A collection lens then sits

perpendicular to the laser focus with a 45 degree mirror sitting behind the collection lens in order to send the light back out of the vacuum chamber through a window that is on the same flange as the window for the incoming laser pulse. The collection lens must be sitting so that the laser focus is right at the focal length in order to have collimated light coming out of the vacuum chamber. The collimated light then becomes raised in a periscope and into another lens so that an image of the laser focus will appear vertical (due to the periscope having mirrors pointed 90 degrees from each other) and the image will appear at the location of a small vertical slit, marking the beginning of the spectrometer. The spectrometer will take a picture of the slit for each wavelength present in the incoming beam. After passing through the slit, which now acts as a point source, there is a collimating lens that will send a collimating beam into the grating. Following the grating is another focusing lens that will create the images of the slit at the position of the camera.

The diffraction grating was the central piece in the setup and all other pieces were bought around it. The grating was chosen to have a Littrow angle of 26.7 degrees for 500nm (i.e. when the angle between the normal line of the grating and the incoming beam is 26.7 degrees, 500nm light goes back in the same direction as the incoming beam) in order to have good efficiency in the UV region, and 1800 grooves/mm which produced an angular separation between 250nm and 500nm to also be 27 degrees by using the diffraction grating equation:

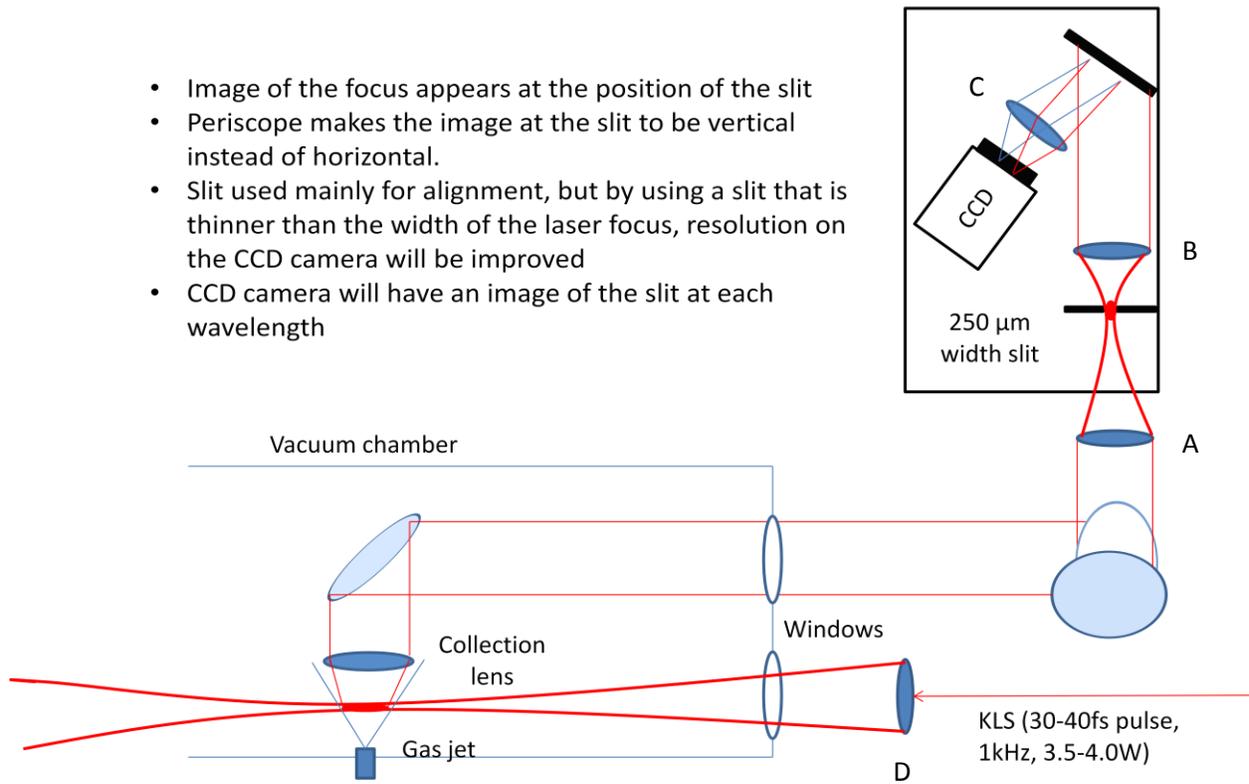
$$\sin(\theta_{reflected}) - \sin(\theta_{Littrow}) = \frac{m\lambda}{d}$$

where  $\lambda$  is the wavelength and  $d$  is the distance between grooves. (We are also of course looking at first order diffraction, so  $m=1$ .) With an angular separation of 27 degrees, the lens after the grating would need to have an  $f/\#$  of 2.0 (meaning the focal length is twice the diameter) in order to gather that range of light if the lens were to be placed at the focal distance from the grating; thus a one inch lens with a focal length of 5cm was used in this spot. In order to have the magnification of the slit image on the camera to be one, another one inch lens with a focal length of 5cm was used to collimate the light from the slit.

Because a very small amount of light is expected to be emitted from the aligned molecules, a collection lens inside the chamber with the lowest  $f/\#$  would be desirable, but the lens also has to sit far enough away from the gas jet so that it does not cause back reflections of the gas to interfere with the experiment. Also, in order to get the beam through the window of the vacuum chamber, the lens could not be more than an inch in diameter. All of these constrictions resulted in buying a lens that was the exact same as the ones in the spectrometer: one inch diameter with a focal length of 5cm. Since there is a one inch beam coming out of the vacuum chamber, two inch diameter mirrors had to be purchased in order to fit the beam on the surface at tilts of 45 degrees. The final piece of the puzzle is the lens that will focus the light onto the slit. A one inch lens with a focal length of 10cm was chosen in order to get a total magnification of two in the system to fill as much of the camera detector as possible. All mirrors bought were metallic and designed to have maximum reflectivity in the 250-600nm region, and the lenses were all uncoated, UV fused silica, plano-convex lenses. All optics were purchased from Newport

with model numbers: SPX016 (collection lens and lenses A and C), SPX022 (lens B), 20D20AL.2 ( 3 mirrors), 10RG1800-500-1 (grating).

- Image of the focus appears at the position of the slit
- Periscope makes the image at the slit to be vertical instead of horizontal.
- Slit used mainly for alignment, but by using a slit that is thinner than the width of the laser focus, resolution on the CCD camera will be improved
- CCD camera will have an image of the slit at each wavelength



### III. Alignment

Alignment begins with first fixing the beam entering the chamber and placing lens D at the correct position so that the focus is at the position of the gas jet. Since seeing the plasma with the eye is not possible through the setup, a wire was placed at the brightest part of the plasma in order to scatter light bright enough to be able to see quite clearly with fluorescent paper. A BBO was also added in the incoming beam so that 400nm wire would be scattered off the wire and followed through after the grating. Once the wire was fixed in position, the collection lens is placed at the correct distance from the wire so that the light coming through the lens was collimated. This was checked easily by making sure the lens focusing onto the slit, lens A, was filled completely with the light from the collection lens. The mirror inside the vacuum chamber should be at a 45 degree angle and should bring the beam out of the vacuum chamber without clipping. Following that, the periscope is placed so that the bottom mirror is facing the vacuum chamber, but sits on a 45 degree upward mount such that the beam after hitting that mirror was going straight up. The top mirror in the periscope, which is sitting on a 45 degree downward mount, should now be sending the beam parallel with the optical table and perpendicular to the incoming beam direction. The outgoing beam after the periscope will be the line that the rest of the spectrometer optics up to the grating will be on.

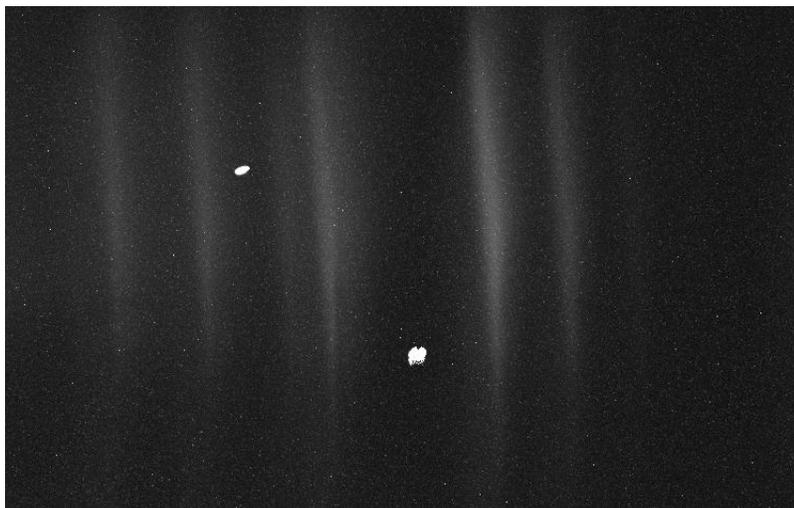
After the periscope, the position of lens A is not all that critical other than that it must be capturing the whole beam from the collection lens and focusing to a point where the slit will have room to be placed. Before inserting the slit, it is useful to align lens B such that the beam is once again collimated, meaning that the distance between the lens A and lens B is the sum of the two focal lengths. Another good thing to note is that since lens A has a focal length of 10cm and lens B has a focal length of only 5cm, the collimated beam after lens B will only be half of the original diameter. After the lenses have been fixed, the slit should be inserted at the focus position of lens B such that the beam can still be seen after the slit. When using the 250 micron slit, the whole image passes through the slit, thus meaning that the slit is not increasing the resolution of the system, but it will still to decrease background and mark the position of the beam path in case anything gets knocked or has to be replaced.

Following the placement of lens A, lens B, and the slit, a suitable position for the grating should be chosen. This position is not very critical since the light coming into the grating is collimated. Lens C should then be placed as close to the beam headed into the grating as possible so that the grating angle is as efficient as possible. Lens C should also be placed at the focal length from the grating if catching the 250-500nm range is desired, but moving the lens farther in will just increase the range of the spectrum while farther away from the grating will catch less. After lens C is fixed, the grating can just be rotated to send the 400nm light scattered from the wire into lens C at the desired location, followed by the CCD which will just sit at the focus of lens C to see an image of the slit at each wavelength. Once a good image of the wire appears on the camera, the wire can be removed and the plasma can then be imaged with at higher exposure time than the wire. Optics that can be played with to try to get a sharper and brighter plasma image is the position of the CCD which is on a translational stage, the collection lens and lens A which are on screw mounts to very slowly change the position, and the lens D which sits on a 3-D translational stage.

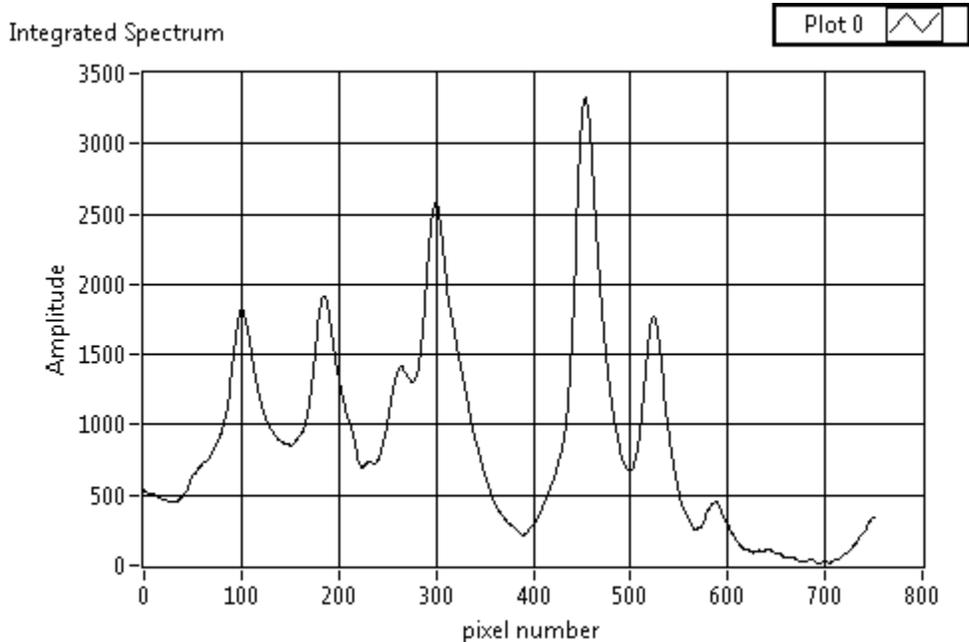
#### IV. Results and Future Work

The REU project finished with images of plasma generated in air.

This is the CCD image from a plasma generated from 3.4W from the 1kHz, ~40fs, Ti:sapphire laser and the exposure time on the camera is 5.587 seconds, meaning that very little light is being emitted from the plasma in the direction of the collection lens. (The two bright white spots are damaged areas on the camera.)



This next image is the graph of the background subtracted image that is summing the Y values for each X value. The peaks are quite well defined, and there the peak at pixel number 300 is believed to be at 391nm due to a strong emission of nitrogen at that wavelength (the 400nm wire spot was a little to the right of the 391nm peak). Wavelength increases from left to right.



Since the plasma data looks good, the next step is to calibrate the spectrum (which can easily be done by sticking a known source as the position of the laser focus), close the vacuum chamber, and insert the gas jet. With the gas jet inserted and giving signal to the camera. The pump and probe setup can then be aligned and sent into the chamber to begin the measurement of alignment dependent fluorescence. A more long term goal of the project is to measure both HHG (High Harmonic Generation) and the alignment dependent fluorescence to hopefully distinguish between the contributions of the two different fluorescent processes described in the introduction.

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## References

- [1] Jinping Yao, Guihua Li, Xinyan Jia, et.al, "Alignment-dependent fluorescence emission induced by tunnel ionization of carbon dioxide from lower-lying orbitals", arXiv:1302.3400v2, (2013)
- [2] F. Rosca-Pruna, M. J. J. Vrakking, Phys. Rev. Lett. 87, 153902 (2001)
- [3] S. Petretti, Y. V. Vanne, A. Saenz, A. Castro, and P. Decleva, Phys. Rev. Lett. 104, 223001 (2010)
- [4] S.-F. Zhao, C. Jin, A.-T. Le, T. F. Jiang, and C. D. Lin, Phys. Rev. A 81,033423 (2010)