Incoherent Shaped Radiation for Control of Atomic Excitation and Energy Up-Conversion

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1. Background

a. Quantum control

The field of quantum control is rapidly evolving with many successful applications. The commonly applied paradigm is to shape a coherent pulse of light to form a "photonic reagent", which is employed to manipulate atomic and molecular dynamics phenomena. The shaping of the pulse is often accomplished using a genetic machine learning algorithm. Several arbitrarily shaped pulses are generated and tested for their ability to perform a desired task. The pulses which are more effective are then used to generate a new generation of pulses. This process continues until successive generations cease to be more effective. This method allows quantum control to find solutions to problems that could not be predicted which in turn provides valuable insight into the underlying dynamics of a system. However, this process requires the manipulated system's underlying dynamics to be non-linear. Otherwise, the optimizations and the attempts to manipulate the system will be unsuccessful.

Additionally, thus far control experiments have exclusively been performed with coherent sources which are expensive. Therefore, as of now, large scale control applications are cost prohibitive. However, recent simulations suggest that substantial control can be obtained using spectral intensity shaped incoherent light sources. A suitable incoherent light source would be much cheaper than fs laser systems, and even a modest level of control would be a major advance. Thus, one of the major goals of the project I worked on this summer is to experimentally confirm that control can be accomplished with incoherent sources. For the purposes of this project, we are defining incoherent radiation as radiation with a wide spectral range that lacks an overall phase.

b. Energy Conversion

Specifically, the systems that we control are crystals doped with lanthanide ions that undergo energy up-conversion. Energy up-conversion is a process by which light of one frequency is converted into light of a higher frequency. Energy up-conversion generally occurs

via one of two processes: Energy Transfer Up-Conversion and Excited State Absorption. In both cases, the process begins by light exciting the sample. In Energy Transfer Up-Conversion, two excited ions (or molecules depending on the system involved) then interact and transfer energy resulting in one ion going to a higher energy level and the other ion going to a lower energy level. The ion in the higher energy state then decays radiatively resulting in the emission of high frequency light. In Excited State Absorption, an excited ion absorbs additional photons and then decays radiatively resulting in the emission of high frequency light.

These systems are excellent candidates for quantum control for numerous reasons. First, in order for a system to be a potential candidate for quantum control, it needs to have non-linear dynamics. Energy up-converting systems are inherently non-linear because they are dependent on the absorption of multiple photons. Second, it is straight-forward to measure up-conversion. Since fluorescence intensity is proportional to population, excited state populations can be measured without having to account for light coming from excitation sources. Third, there is implicit value in studying these materials. Energy up-conversion currently has numerous uses including bio-imaging, alternative energy, and authentication. However, the advent of up-converting materials is still relatively recent and additional research could enable novel uses of these materials as well as enhance current uses.

2. Goals

The primary goal of this project is to tailor the intensity of an incoherent light source over its spectral range in order to control a system of interacting lanthanide ions. The tailored spectrum of the radiation constitutes the photonic reagent, and optimization of its spectral structure is performed using a genetic machine learning algorithm. If quantum control can be successfully performed using incoherent light, the utility of the field of quantum control will be greatly increased. A secondary goal is to analyze the implications of the system's response to understand the underlying dynamics occurring in an up-converting material. This could increase the existing applications for energy converting materials.

3. Summary of Work

a. Optimizing the Fluorescence of Rhodamine 6G

A large portion of the work accomplished this summer went into constructing an apparatus that would be capable of performing these control experiments. Our initial attempts

went into shaping a wide spectrum of visible light from a flash-lamp. To accomplish this goal, we used a Spatial Light Modulator (SLM) which was capable of generating an arbitrary non-thermal spectrum of light. In order to ensure that the SLM would be able to successfully shape the light, we ran a simple calibration experiment. The objective was to optimize the fluorescence of a well-known fluorescence standard—Rhodamine 6G. If we were able to correctly deduce the absorption spectrum of Rhodamine 6G using our experimental set-up, we could reasonably apply this method to deduce how various wavelengths interacted with various up-conversion processes.

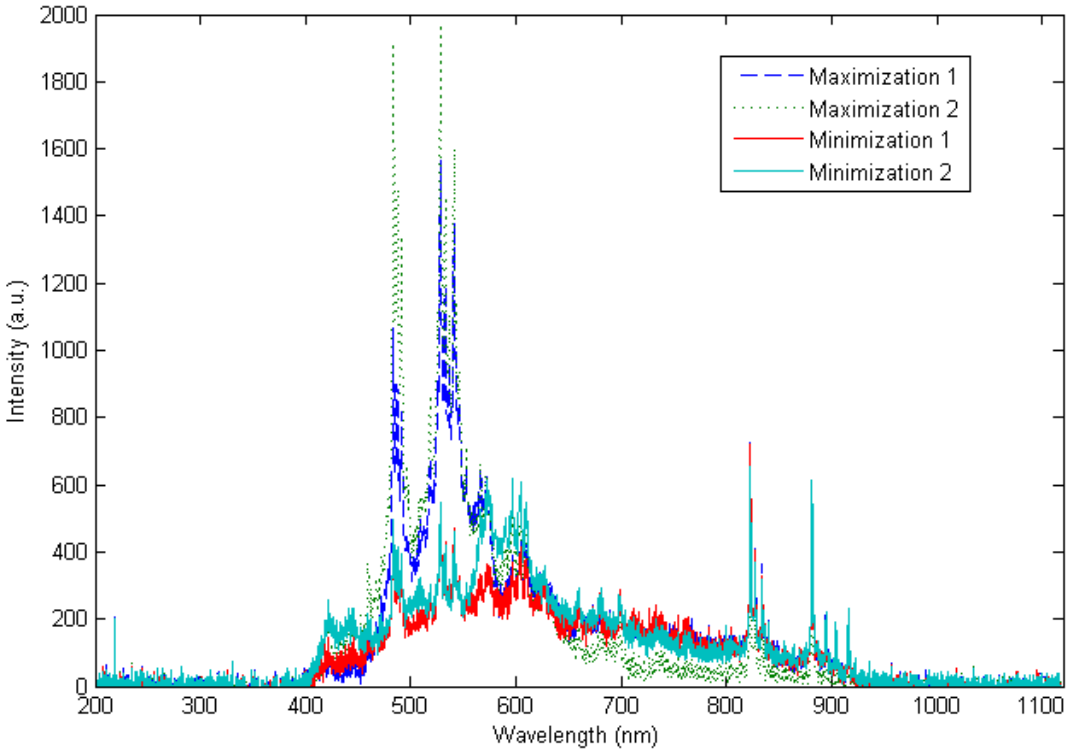

Figure 1: The excitation spectrum of our optimization trials.

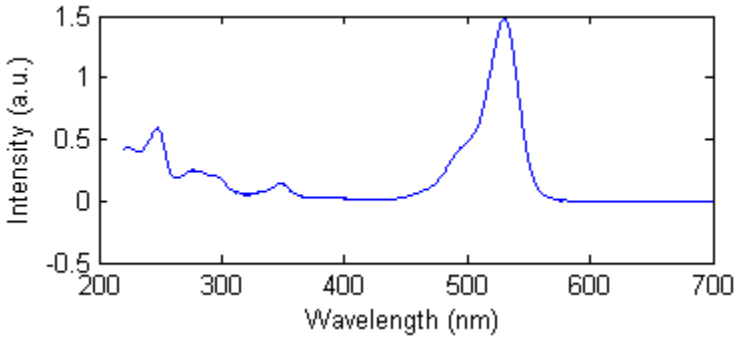

Figure 2: The absorption spectrum of Rhodamine 6G.[1]

These attempts were successful: we were able to successfully control the fluorescence of a solution of Rhodamine 6G using a genetic algorithm. Our methodology involved spectrally shaping light from the flash-lamp with the SLM and then refocusing that shaped light onto a solution of Rhodamine 6G. The solution's fluorescence was focused, sent through an interferometric filter that corresponded to Rhodamine 6G's fluorescence, and then recorded by a PMT. Our genetic algorithm shaped 80 genes which corresponded to various segments of the visible radiation emitted by the flash-lamps. The success of these optimization demonstrated that our setup was capable of maximizing the fluorescence of a sample.

Figure 1 shows the spectra of light shaped by the SLM after four optimization experiments. These spectra describe the shaped light as it hit our sample and do not reflect any interactions between the shaped light and the Rhodamine 6G solution. The major difference between the maximization and minimization optimizations is the spectral intensity around 500-600 nm. In the maximization trials, this spectral range's intensity was greatly increased while in the minimization this regime's intensity was greatly reduced. It can then be inferred that Rhodamine 6G absorbs in this region. Looking at the literature, this absorption spectrum is confirmed: as you can see in Figure 2, Rhodamine 6G's absorption peak is around 500-550 nm. So, this method could theoretically be used to maximize the fluorescence of a sample.

Ultimately, as we moved forward we went away from this form of controlling light. This change occurred because most up-converting phosphors absorb in the NIR and the flash-lamp emits mainly in the visible. As a result, in order to see energy up-conversion we needed to get a new light source. We transitioned to using a series of NIR laser diodes. Specifically, we use three laser diodes: a 808 nm laser diode with an output power of 200 mW, a 980 nm laser diode with an output power of 100 mW, and a 1550 nm laser diode with an output power of 100 mW. For the purposes of our project, these laser diodes are incoherent because we combine them resulting in a wide spectral range and a lack of an overall phase. Laser diodes can be modulated directly and do not require the usage of an external machine like an SLM to be shaped. For simplicity, we are shaping the laser diodes directly. However, it is possible that we will move back to using the SLM at some point if we start evaluating samples that absorb significantly in the visible range.

b. Sample Analysis

The other major problem in setting up our experiment was acquiring samples that would undergo up-conversion. We spent a substantial amount of time and effort trying to generate our own such samples with limited success. As a result of these difficulties, we began collaborating with a local company—Intelligent Material Solutions—to aid in the fabrication of such materials. We were able to obtain several different types of samples. First, we received a series of powder samples. They were Yttrium Oxysulfide doped with Erbium, Ytterbium, and Holmium; Yttrium Oxysulfide doped with Erbium, Ytterbium, and Thulium; Yttrium Oxysulfide doped with Erbium and Ytterbium; and Yttrium Oxide doped with Erbium and Ytterbium. Next, we received a series of nano-crystals suspended in colloids. They were $LiYF_4$ doped with Ytterbium and Thulium; $LiYF_4$ doped with Ytterbium and Holmium; $LiYF_4$ doped with Ytterbium, Erbium, and Thulium; and $NaYF_4$ doped with Ytterbium and Erbium. However, as of now, we have only really analyzed the powder samples.

Every powder sample exhibited up-conversion fluorescence that was visible to the eye. Their up-conversion luminescence is displayed in Figures 3-6. Each line corresponds to the up-conversion luminescence of a sample when pumped by a single laser diode centered around a single wavelength. These plots do not subtract out the pump intensities, so the peaks seen at 980/808 nm when the excitation source is of the same wavelength should be considered mainly as a measurement of the intensity of the excitation source rather than the fluorescence itself. Also note that in some of the samples the spectrometer got saturated resulting in an artificial plateau appearing on the top of a fluorescence peak.

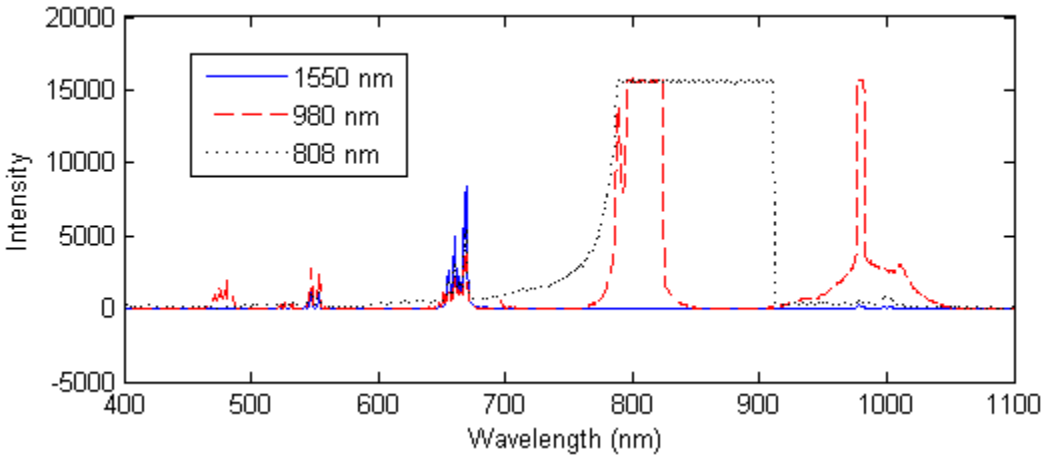

Figure 3: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium, Ytterbium, and Thulium

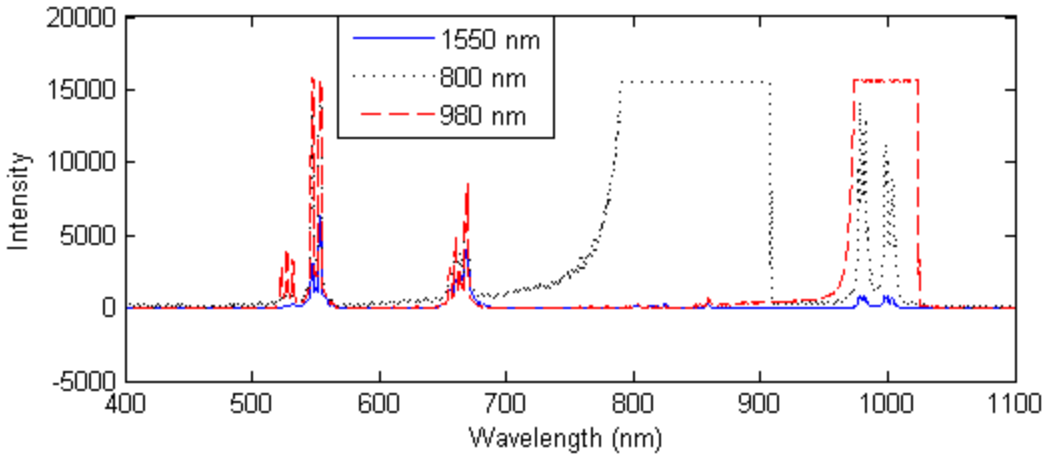

Figure 4: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium, Ytterbium, and Holmium

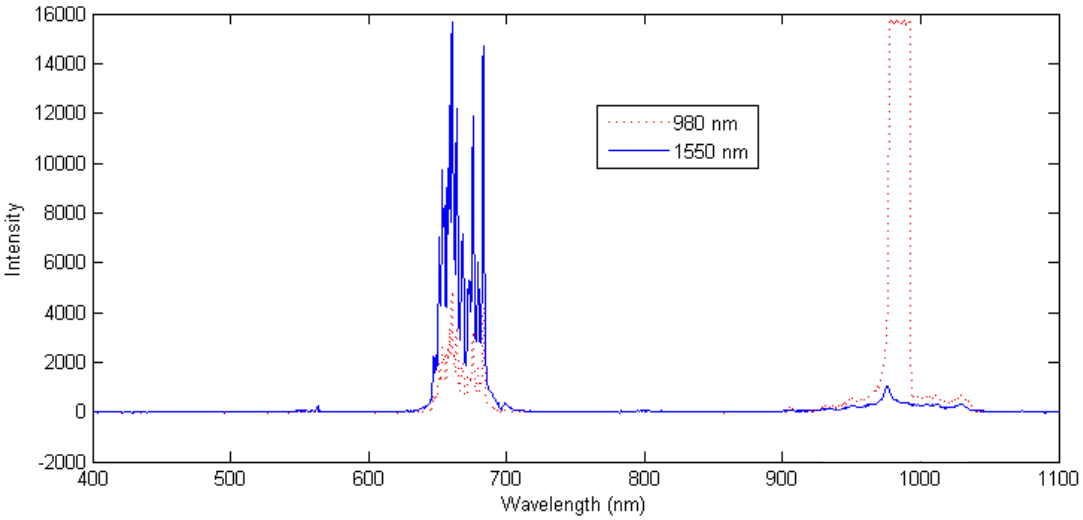

Figure 5: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium and Ytterbium

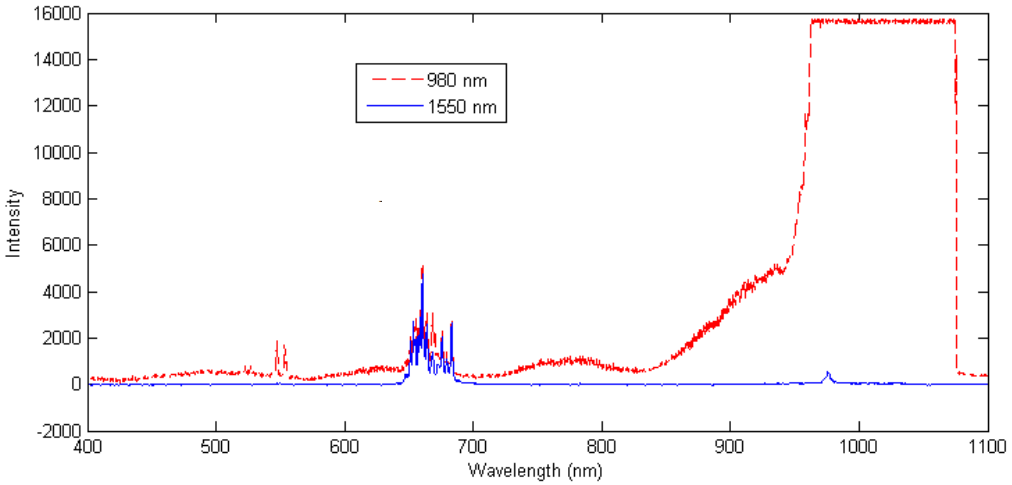

Figure 6: Up-conversion luminescence of Yttrium Oxide doped with Erbium and Ytterbium

The other substantial conclusion reached this summer was that these various up-conversion processes were related. For our project to work, the various energy up-conversion processes that occur in our samples need to interact. Otherwise, our experiments could be reduced to a series of one-dimensional optimizations which are not control experiments. Fortunately, we've seen and proven the existence of these interactions. Our method was to excite our samples with different laser diodes and then compare the samples' responses to being simultaneously pumped with several different laser diodes to the superposition of their responses to each individual laser diode. These excitation sources appear to act cooperatively to increase up-conversion florescence as the up-conversion from simultaneous excitation is greater than the superposition of the separate excitations. Figures 7-10 show this trend is consistent throughout all of our samples.

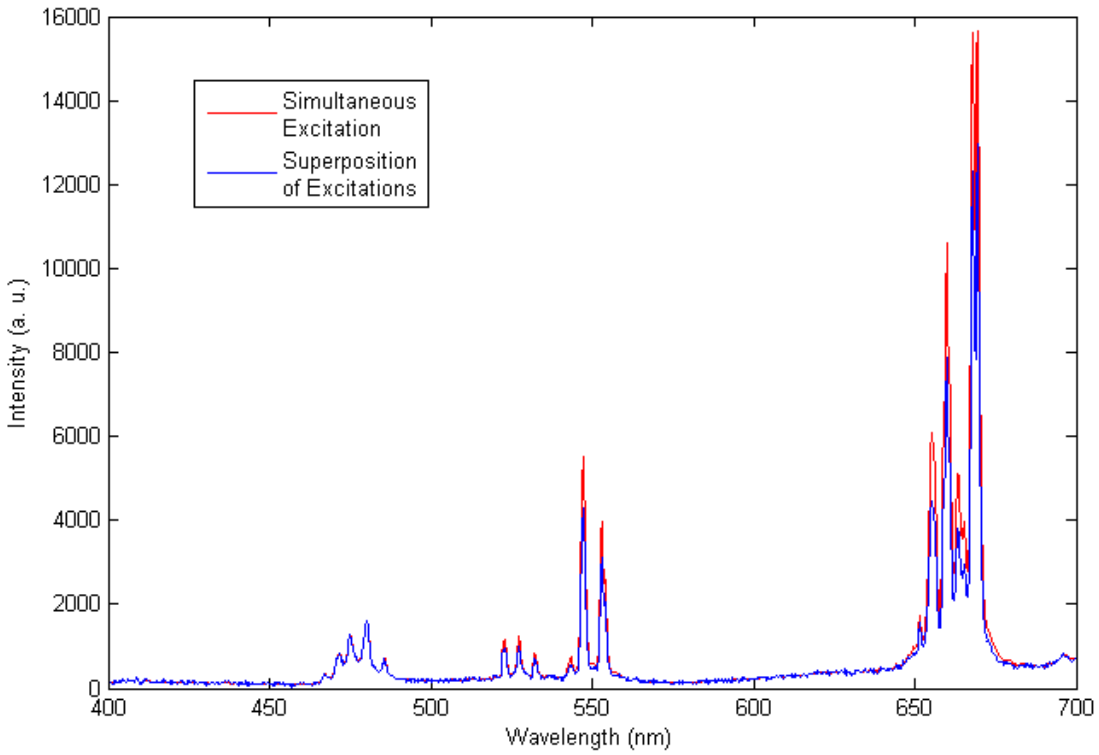

Figure 7: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium, Ytterbium, and Thulium when excited by our laser diodes separately compared to the superposition of its response to individual laser diode

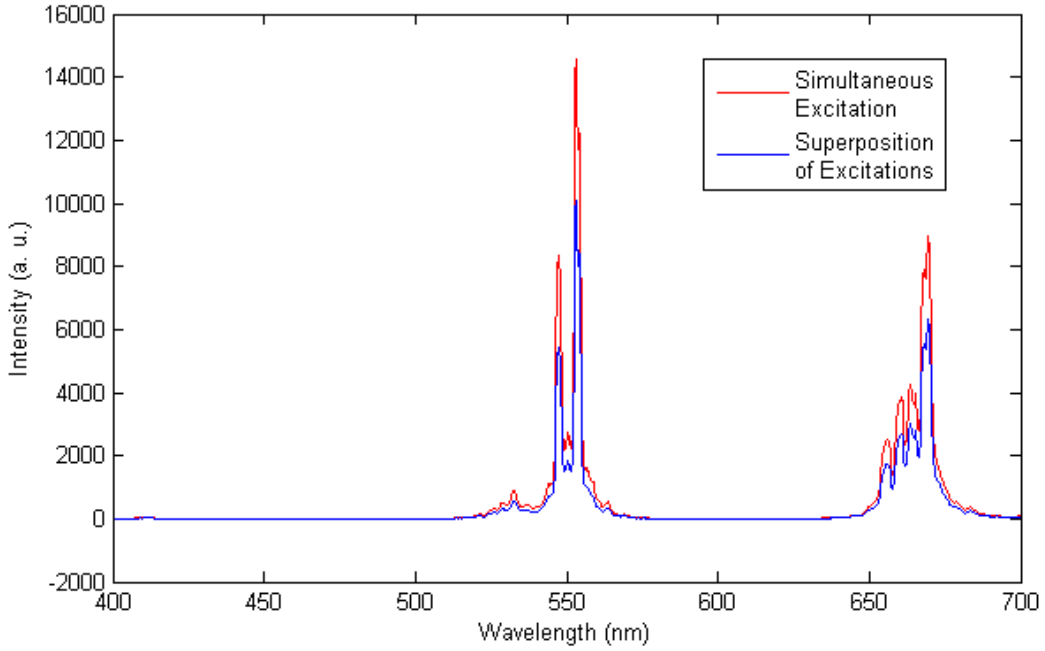

Figure 8: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium, Ytterbium, and Holmium when excited by our laser diodes separately compared to the superposition of its response to individual laser diode

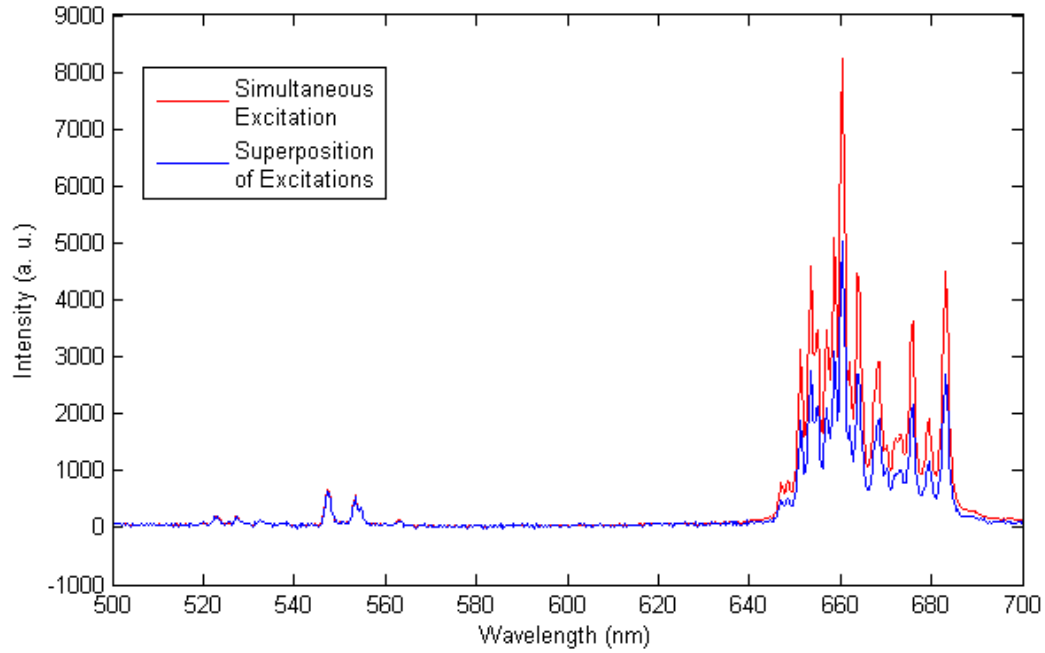

Figure 9: Up-conversion luminescence of Yttrium Oxysulfide doped with Erbium and Ytterbium when excited by our laser diodes separately compared to the superposition of its response to individual laser diode

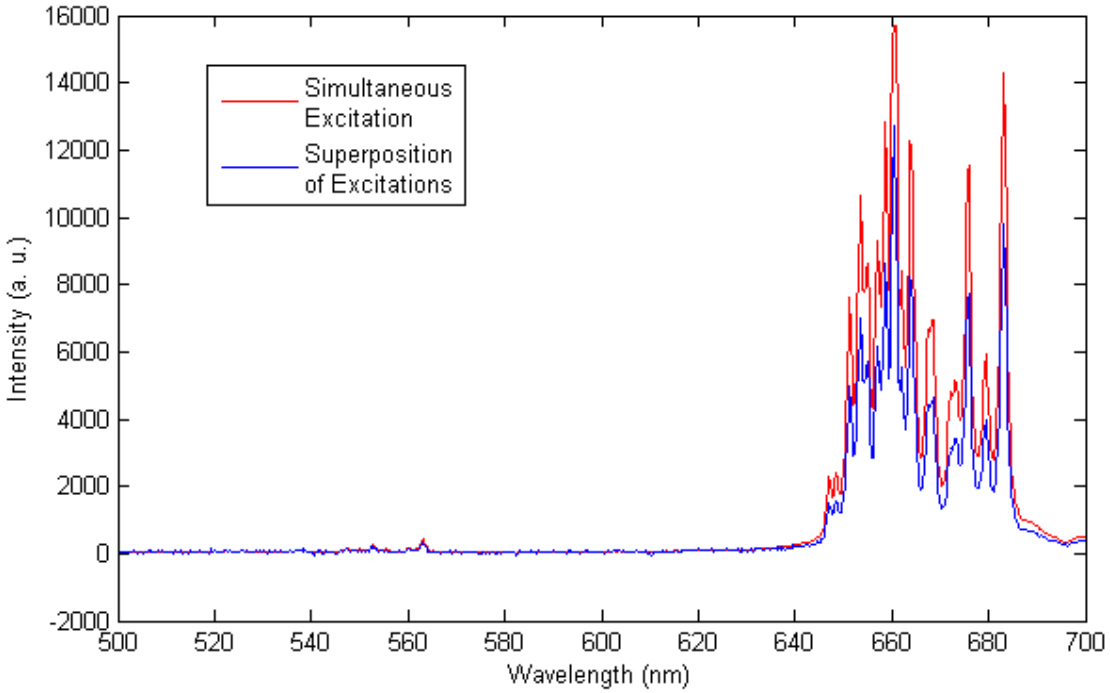

Figure 10: Up-conversion luminescence of Yttrium Oxide doped with Erbium and Ytterbium when excited by our laser diodes separately compared to the superposition of its response to individual laser diode

4. Conclusions and Moving Forward:

This summer yielded a number of very promising advances. The fact that these different up-conversion mechanisms interact in a non-linear fashion suggests that this system matches the requirement for a system to be controllable. Therefore, we should be able to use this system to prove that incoherent control can be successfully performed. Also, our collaboration with Intelligent Material Solutions has granted us access to a variety of samples which will enable us to probe the underlying mechanisms of up-conversion. The summer has paved the foundation for a series of experiments which could have significant implications. We will be able to examine quantum control and up-conversion in a novel fashion which could provide major advances in both fields. We are now poised to effectively run experiments which will help to generalize quantum control and increase our understanding of the phenomenon of up-conversion.