Spectroscopy plays a key role in multiphoton microscopy, augmenting the spatial information obtained via imaging with information about local chemical environments. Quantum dots, which are of increasing interest as laser sources, biological markers to replace dyes, and components of high efficiency solar cells, exhibit changes in their fluorescence spectrum when a second carrier, known as a biexciton, is promoted to the excited state. These shifts and additional peaks are on the order of several nanometers and are very weak, requiring high spectral resolution, high sensitivity, and narrow emission bandwidth to observe. These characteristics must be measured on single quantum dots, so as to not average out the spectral and temporal properties of the fluorescence. More easily observed are the spectral shifts due to different degrees of quantum confinement in quantum dots. As confinement increases with a decrease in dot size, the bandgap of the dot will increase from that of the bulk.

We have implemented a simple prism-based spectrometer fabricated with “off the shelf” optics specifically for multiphoton microscopes that has no slit, requires no filtering of the output, and yields a spectral resolution that varies between 0.3 nm and 2.2 nm across the detector. Unlike traditional grating-based spectrometers, high spectral resolution is not dictated by a slit, but rather by the dispersion of the prism and the beam size. Thus, our spectrometer employs the full acceptance of a high-numerical aperture objective without the loss associated with focusing through a slit.

We characterize the performance of this spectrometer by examining single CdSe quantum dots. Detecting emissions from single emitters tests the sensitivity limit—a function of detector gain and system throughput. The emission exhibits amplitude modulation known as “blinking”, which is thought to be associated with dot charging associated with the excitation and relaxation cycle. The spectrum in the slide was acquired with below band gap light. Excitation involves a non linear two-photon process. The spectrum shows a strong dependence on the polarization direction of the incident exciting light. This implies the existence of anisotropy in the shape, surface defect structure, or capping groups, which leads to differences in the character of the excited state under the two conditions. Now that the system is functional and has been tested on prototypical CdSe quantum dots, work is focused on silicon nanostructures. These have been passivated with different capping ligands and the goal of the study is to gain insights into the effect of ligand on defect state and carrier dynamics.