**Single quantum dot imaging and spectroscopy in high magnetic fields**

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## I INTRODUCTION

Semiconductor nanocrystal quantum dots (QDs) have many interesting size-dependent properties (1). With diameters on the molecular scale, they can be considered "artificial atoms," having atomic-like (i.e. discrete) emission spectra, which are tunable across the visible spectrum by changing QD size. They are widely studied for a range of applications, including biological tagging, the development of highly efficient light sources, and potentially for quantum computing. Photoluminescence spectroscopy of quantum dots provides a means of examining properties of single quantum confined excitons (electron-hole pairs). Observations of the PL of a single dot within a strong magnetic field would allow observation of spin (Zeeman) splitting and field effects on spectral lines.

Single dot spectroscopy within a high magnetic field requires a means of isolating and viewing a single dot, and the construction of an instrument to house the experiment within the confines of a high-field superconducting magnet. The center of the magnetic field is a distance of 1 to 1.5 meters from the top of the magnet, thus presenting a special challenge for stimulating and viewing single dots with a free-space laser beam. The challenge arises because viewing single dots in a high-field magnet requires a microscope several meters in length and operable at cryogenic temperatures, in which the sample is positioned remotely from outside the magnet. This paper details the preliminary stage of such an experiment: the building of a long distance microscope capable of viewing single quantum dots.

## II SETUP

Fig. 1 shows the experimental set up used for viewing QDs on the optical table. A 10 mW frequency doubled YAG laser with wavelength of 532 nm was used, followed by a 532 nm band pass filter to remove any remaining 1064 nm light. A diverging lens expanded the beam, and a 2" convex lens mounted on a translating stage controlled the beam diameter at the focus. At one extreme of travel, the 2" lens created a collimated beam, producing a spot of minimum size on the sample, and at the other extreme it created a beam that converged 0.5 m in front of the objective, producing a larger spot on the sample. In this manner it was possible to adjust the intensity of the excitation beam incident on the sample. A 40x Olympus achromatic objective with a numerical aperture of 0.65 was used. The sample was mounted on an XYZ translation stage. An ocular lens of a long focal length focused the projected image onto a 384 by 576 pixel CCD chip. A 532nm notch interference filter blocked reflected laser light for viewing PL, and could be replaced by neutral density filters for focusing the laser beam. The distance between the beam splitter and the objective is 1 m in anticipation of this section eventually being enclosed in a probe inside a magnet. The effective magnification of this setup is 230.

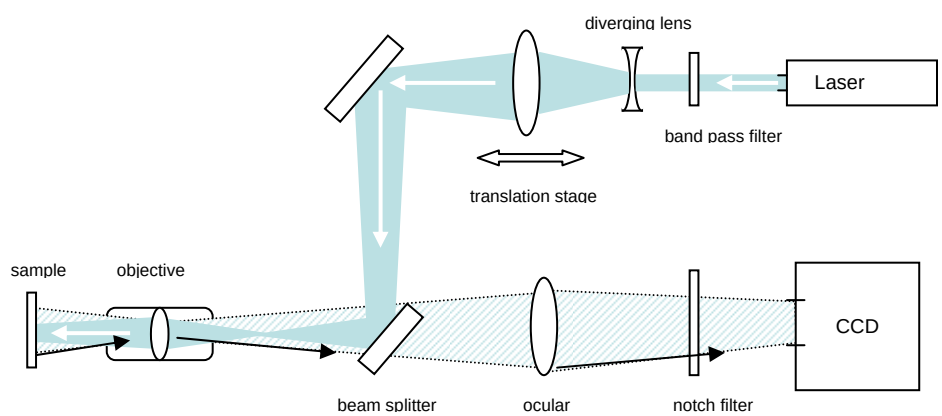


Figure 1: Experimental setup for microscope.

Samples were prepared using 55 Å diameter CdSe quantum dots which luminesce at 620 nm at room temperature. The dots were diluted in hexane and thinly spread on a fused silica slide. The distance between the sample and the objective was fixed at the focal length of the objective by collimating the incoming beam and adjusting the sample position to minimize the size of the reflected laser beam spot as seen on the CCD. The minimum spot size is diffraction limited, the diameter given by $1.22\lambda/(2NA)$ where NA is the objective numerical aperture. This is calculated as 0.52 µm for this setup, but the CCD has a resolution of 0.11 µm, giving a minimum spot size of 0.55 µm. A differential micrometer capable of fine adjustments was used to control sample translation along the optical axis, as the depth of focus for this objective and laser combination is 0.80 µm.

The CCD shutter was controlled and data taken using WinSpec/32 software. Dilute dot samples required integration times of 1.5 to 4.0 seconds, resulting in PL intensities of 30 to 50 counts above baseline. The thermoelectrically cooled CCD picked up approximately 1 count per second of dark noise. Ambient light added another 2.5 counts per pixel per second.

For use in an experiment in a magnet, this setup will be on an optical table located above the magnet, and the objective and sample will be inside a probe in the magnet bore. A mirror will be used to direct the beam perpendicular to the table top, directing it down the probe.

## III RESULTS

Fig. 2 shows a series of three sequential images, taken with 2 s exposure time. Distinct bright spots are seen, with a diffraction-limited diameter of 0.55 µm. Some spots are constant, while others, such as those highlighted, are seen to appear and disappear. This blinking behavior is a known characteristic of single nanocrystal quantum dots, and is taken as evidence that the images are indeed PL from single dots.

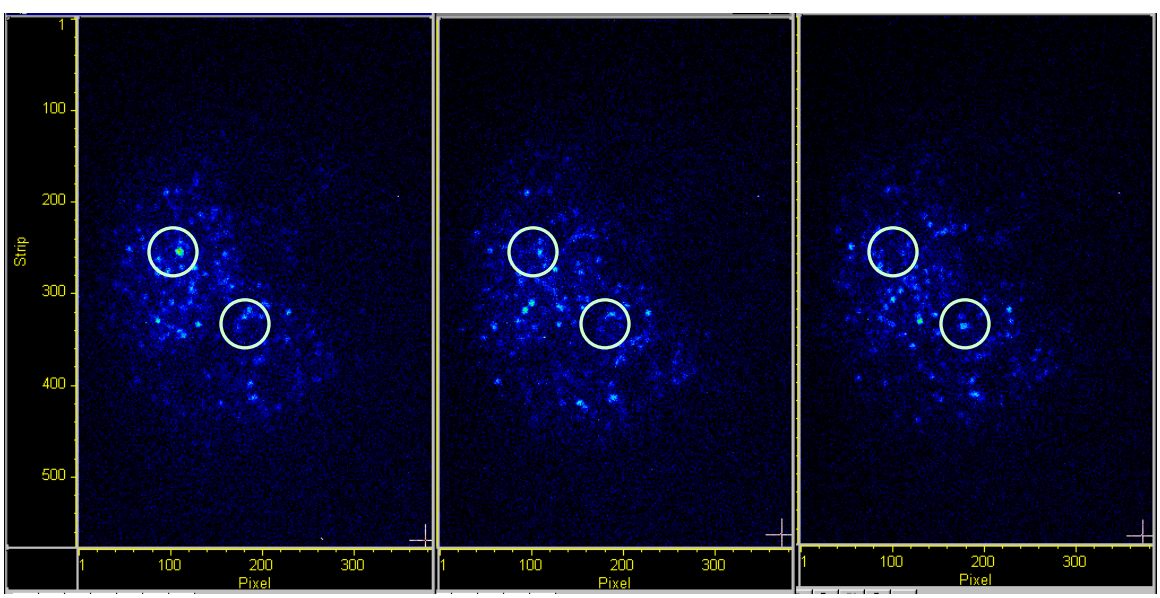


Figure 2: Sequential images taken in WinSpec of dilute quantum dot sample, with exposure time of 2 seconds.

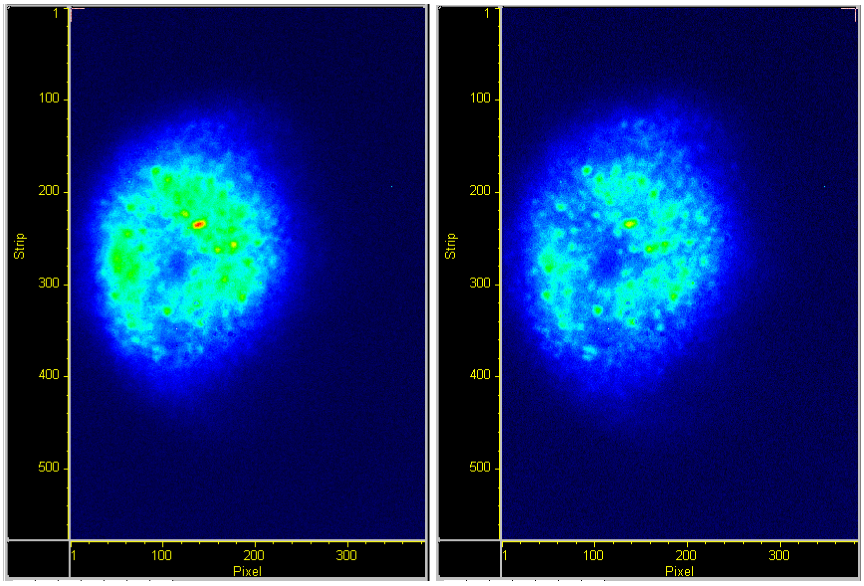


Figure 3: Images taken in WinSpec imaging mode, with exposure time of 1.5s. The two images were taken 1minute apart.

When the sample was moved so that a clean portion of the slide that had not been smeared with dot solution was viewed an otherwise identical set of exposure conditions resulted in a blank readout showing only dark and ambient noise.

Note that Fig. 3 is brighter and shows a higher concentration of dots than Fig. 2. This is a function of the dilution of the QD during sample preparation and is not yet controllable due to unknown concentrations of the original QD solution. Preparing a sample dilute enough to see individual dots is a process of trial and error. The fact that the dots in Fig. 3 are brighter may be because they are not single dots but are aggregates of dots instead.

A probe for conducting this experiment inside a magnet will consist of two concentric tubes connected by a threaded section such that one can turn, and therefore translate, inside the other. The central tube will house the microscope objective, and the outer one will hold the dot sample. The focus will be adjusted by turning the inner tube, changing the distance between sample and objective. At the top of the tubes shall be a vacuum-tight optical window so that light can be sent into and collected from the probe.

The microscope described here requires that the sample be moved in order to locate and position a desired dot at the center of the field of view so that its image falls on the entrance slit of a spectrometer. Such remote translation within the probe is a central challenge in a probe design. The solution offered here is a piezoelectric tube scanner, as described by Binnig and Smith (2). The scanner consists of a single tube of piezoelectric ceramic with the outer diameter axially sectioned into four equal area electrodes and a single electrode on the interior surface (Fig. 4). The tube is radially polarized so that a voltage applied to one electrode causes that segment to expand, causing the tube to bend away from that electrode. An equal and opposite voltage is applied to the opposing electrode; alternately it can be grounded.

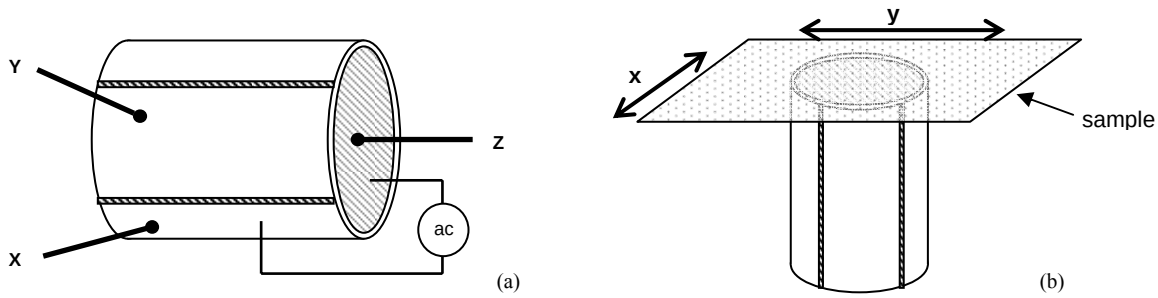


Figure 4: (a) Illustration of a piezoelectric tube scanner with axially segmented outer electrodes and a single interior electrode. (b) Scanner tube with sample.

In this manner, small XY movements on the order of a few nanometers per Volt are effected. For large translations, a slip-stick method is used. As shown in Fig. 4b the sample is placed on top of the tube. When the tube is bent slowly the sample with move with the tube; when the tube is bent rapidly the sample will remain stationary due to inertia (akin to the trick of yanking a tablecloth out from under plates and dishes.) Successive fast and slow translations, driven by a saw-tooth voltage, can move the sample any arbitrary distance. Rough positioning can be achieved this way, with a full 2D range of motion possible by vectoral addition of X and Y movements.

Fixed inside the outer tube of the probe such a scanner could be easily controlled, and the sample translated with a fine degree of accuracy.

In conclusion, the microscope built can successfully image single quantum dots at room temperature at a distance of 2.5 meters. Before this set up can be used to perform spectroscopy the PL signal-to-noise ratio needs to be increased. This could be aided by using a liquid nitrogen cooled CCD, thus decreasing the dark noise, and working to eliminate all ambient light. Different samples of quantum dots should also be explored as they may luminesce more efficiently.

## REFERENCES

1. A. P. Alivisatos, Science **271**, 933 (1996)
2. G. Binnig and D. P. E. Smith, Rev. Sci. Instrum. **57**, 1688 (1986).