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Manipulation of CdSe/ZnS quantum dots in room-temperature fluid with an inhomogeneous electric field and optical excitation

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Abstract

We successfully demonstrated the manipulation of CdSe/ZnS quantum dots with a diameter of nearly 6 nm in organic solvent at room temperature by applying an intense electric field (1.7 MV/m) under resonant optical excitation (the wavelength 532nm). From the time-variation of the quantum dot distribution monitored by fluorescence imaging, it was experimentally found that the quantum dots gather around the local maximum of the electric field intensity and the potential energy applied on the quantum dot under the optical excitation is estimated to be nearly 400 K, which is approximately 20 times larger than that expected with conventional dielectrophoresis. Such a large potential energy is considered to be due to the Stark effect of the exciton created in the optically excited quantum dot.

Keywords: quantum dot, dielectrophoresis, exciton, Stark effect, manipulation

I Introduction

Semiconductor quantum dots (QDs) with a diameter of several nm are drawing attention as one of interesting optical materials [1], since their electronic structures and optical properties significantly change with their size and environment [2]. For their future applications to optical devices, more precise control and/or selection of their characteristics is considered to be one of key factors. However, the manipulation of such a small neutral particle in room temperature fluid is still difficult.

The aim of our study is to develop an experimental method for selecting nm size quantum dots based on the difference in spectral characteristics. In liquid helium, the optical separation has been successfully demonstrated for CuCl quantum dots created by laser ablation [3]. For realizing similar optical separation for a variety of chemically synthesized quantum dots at normal temperature, we are developing a new experimental approach by using an inhomogeneous electric field and optical excitation.

The optical excitation of semiconductor quantum dots creates bound electron-hole pairs called “excitons” inside the particles. Among them, weakly bound excitons, which are more attractive for application due to their wide controllability of spectral characteristics, are known to show large AC Stark shift in their fluorescence spectrum. For example, CdSe/ZnS core-shell type quantum dots show the AC Stark shift as much as 60 meV (～700 K) for the excitation laser intensity of 2.5 kW/cm^2 [4]. We focused on this experimental result and tried to realize a novel scheme for manipulating quantum dots by using the Stark effect of the exciton.

To show the effectiveness of this manipulation scheme, in this work, we used the CdSe/ZnS quantum dot with a diameter of nearly 6 nm as a typical sample particle. For generating trapping potential much higher than room temperature, we applied an inhomogeneous external electric field, whose intensity was much higher than the optical electric field of the excitation laser beam.

The movement of the quantum dot was observed for more than 30 hours by monitoring the intensity distribution of the fluorescence from the quantum dots. As a result, it was successfully confirmed that the quantum dots gathered around the maximum point of the electric field strength. It was also confirmed that the quantum dot distribution returned to the homogeneous distribution after tuning off the applied electric field.

The depth of the trap potential is experimentally estimated from the equilibrium distribution of the quantum dots and is

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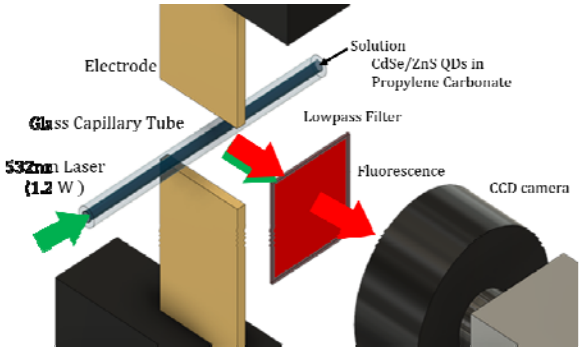

Figure 1. Experimental setup

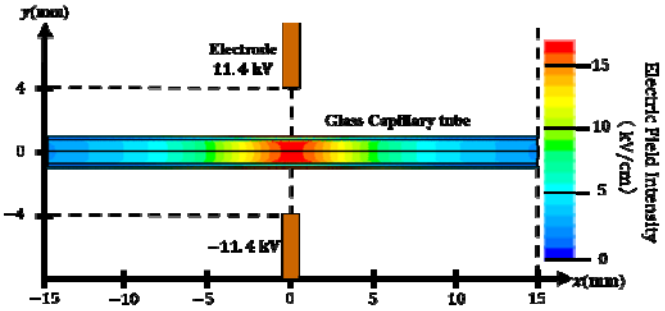

Figure 2. Electric field intensity $|E|$ in the capillary tube estimated from numerical calculation with the finite element method.

discussed in comparison with the one expected for conventional dielectrophoresis [5].

II Experiment

Figure 1 shows our experimental setup. The dilute propylene carbonate solution of CdSe/ZnS quantum dots (Aldrich, Lumidot 640) was sealed in a glass capillary tube with an inner diameter of 1.5 mm. The capillary tube with the solution was placed between two electrodes, and the inhomogeneous electric field E, whose maximum intensity was approximately 17 kV/cm, was applied to the solution along the center axis of the capillary tube. The values of $|E|$ and $|E|^2$ along the center axis, which were numerically calculated with the finite element method, are shown in Fig.2 and Fig.3, respectively. Simultaneously, a resonant laser beam (the wavelength 532 nm, the power 1.2 W) was introduced along the center axis of the capillary tube. The laser beam was loosely focused around the maximum point of the electric field in the capillary tube (the focal waist size ~ 0.3 mm). The spatial distribution of the quantum dot was monitored by observing fluorescence from the quantum dot with a CCD camera. To remove scattered light from the observation, long-pass glass color filters (> 580 nm) were placed in front of the CCD camera.

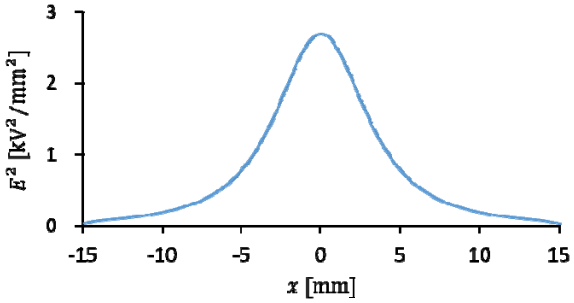

Figure 3. Spatial distribution of the value of $|E|^2$ along the center axis of the capillary tube (theoretical result calculated with the finite element method).

III Results and Discussion

Figure 4 shows the observed time variation of the quantum dot distribution on the center axis of the capillary tube. After applying the electric field and the resonant laser beam, the quantum dot moved toward the maximum point of the electric field. The quantum dot distribution became almost constant in approximately 30 hours. It was also confirmed that the

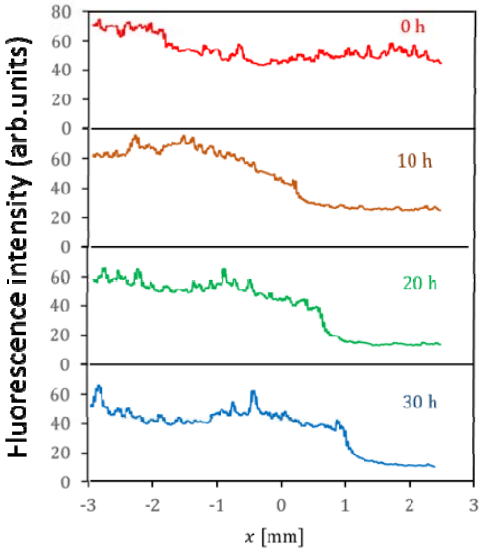

Figure 4. Change of the quantum dot distribution observed after applying the electric field and the resonant laser beam,

observed equilibrium distribution returned to a nearly uniform distribution as seen before applying the electric field.

From the observed difference in quantum dot density between two locations x = 3 mm and x = 0 mm, the potential depth was estimated to be roughly 400 K. This value is approximately 20 times larger than the one expected with conventional dielectrophoresis [5,6] for the maximum electric field intensity of 17 kV/cm.

IV Summary

We succeeded in the manipulation of CdSe/ZnS quantum dots with a diameter of nearly 6 nm in organic solvent at room temperature. For such a small nanocrystal, neither conventional dielectrophoresis nor optical trapping was valid in practice at room temperature. In this work, we gave one of effective experimental schemes to overcome this problem for the first time. Although this scheme is expected to be effective for other semiconductor quantum dots having weakly bound excitons, detailed physical mechanism generating such a large interaction energy remains unclear, further study of which is needed to establish and apply this novel manipulation technique for a wide range of nanoparticles.

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