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Experimental verification of Förster energy transfer and quantum resonance between semiconductor quantum dots

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ABSTRACT

Quantum dot (QD) superlattices have the potential to provide new optical properties and functions based on interactions between adjacent QDs. Two types of interactions occur between the QDs: energy transfer (ET) from small-sized QDs to large QDs and resonant coupling between QDs with equal eigenenergies. Since ET and resonant coupling strongly depend on the distance between QDs, it is critical to precisely control the distance to understand the interaction mechanism. In this review, we describe that the distance between QDs can be controlled with an accuracy of 1 nm by a layer-by-layer method and further explain the mechanisms of ET and resonant coupling between adjacent QDs.

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1. Introduction

In semiconductor quantum dots (QDs) with sizes smaller than 10 nm, quantum effects arise because of the finite size of the QDs [1,2]. As a result, the QDs exhibit physical/chemical properties and functions different from those of the bulk crystal [3–6]. Thus, semiconductor QDs are considered as a promising new functional material. Notable properties of semiconductor QDs include the ability to control the photoluminescence (PL) energy by changing the QD size and high PL efficiency [7–12]. Since the optical properties of semiconductor QDs depend on their size, it is extremely important to prepare QDs with uniform size. Furthermore, since the surface-to-volume ratios of QDs are much larger than those of bulk crystals, the surface structure greatly affects the emission characteristics of the QDs [7,8]. Thus, controlling both the size and surface structure of QDs is critical.

Most techniques for preparing QDs can be categorized as physical methods involving molecular beam epitaxy [13,14] or chemical syntheses of colloidal QDs using chemical reactions in solution [7–12,15,16]. The PL efficiency of QDs can be enhanced by preparing the QDs with core/shell structures [7,8,12]. To date, high-quality, size-tunable, and monodispersed colloidal QDs have been demonstrated.

While randomly dispersed QDs have been the focus of most past studies, QD super lattices, in which the QDs are regularly arranged, have the potential to provide new optical properties and functions based on the interactions between adjacent QDs [17–19]. Two types of interactions occur between adjacent QDs: excitation energy transfer (ET) from small-sized QDs to large QDs and resonant interactions between QDs with equal eigenenergies. In Ref. [20], Klimov and colleagues reported on resonance ET between CdSe QDs. This inspired studies on QD-based ET processes [21,22]. ET between QDs has generally been attributed to the Förster mechanism [20–22], which is responsible for ET between organic molecules [23]. However, the appropriateness of the Förster model for explaining ET between QDs. The question remains. However, the appropriateness of the Förster model for explanation of ET between QDs has not been verified. Furthermore, although many studies on ET between QDs have been reported, reports on resonance interactions between QDs are limited.

Since ET and resonant interactions strongly depend on the distance between QDs, it is critical to precisely control the distance between QDs to clarify their interaction mechanism. The layer-by-layer (LBL) method can be used to control the distance between QDs [24–26] because (1) it is possible to laminate QDs dispersed in an aqueous solution on a substrate layer by layer, and (2) the interlayer distance can be controlled by the thickness of the spacer

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(electrolyte polymer) layer. Multilayered structures of semiconductor QDs such as CdTe [21,27], CdSe [28,29], and CdS [30,31] have been fabricated via LBL assembly.

In this review, we describe the precise control of interlayer distance in QD layered structures using the LBL method and discuss the mechanisms of ET and resonant interaction between QDs with a focus on the authors' research results. CdS and CdTe QDs were the focus of studies on ET and resonant interaction, respectively.

2. Synthesis and optical properties of CdS QDs

CdS QDs can be synthesized by injecting H_2S gas (0.2 mmol) into 100 ml of an aqueous solution containing 0.2 mmol $Cd(ClO_4)_2$ and 0.2 mmol sodium hexametaphosphate, a dispersion agent for colloids [32]. Fig. 1(a) shows the absorption and PL spectra of as-grown CdS QDs. In the absorption spectrum, the absorption structure is observed on the high-energy side of the band gap energy of ~2.5 eV in the bulk crystal, but the spectrum is very broad because of the wide size distribution of the QDs. In the PL spectrum, the defect-related PL band is the main PL band, and band-edge PL is very weak. These spectra are typical of conventional CdS QDs. In Ref. [33], Peng and colleagues have reported the synthesis of CdS QDs whose size is uniform and band-edge PL is observed as the main PL band by the hot-injection method.

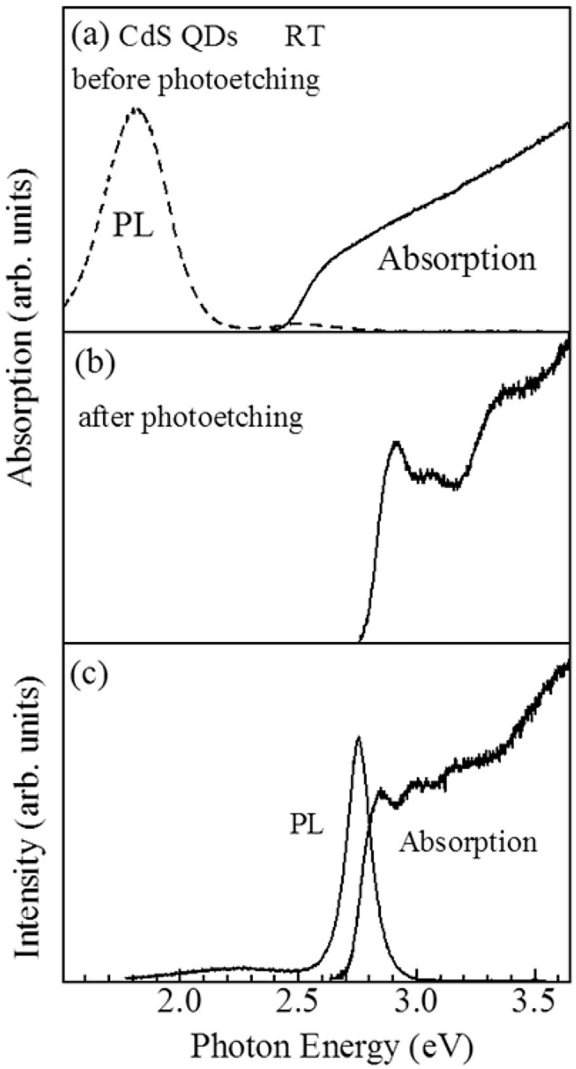

Fig. 1. (a) Absorption and PL spectra of as-grown CdS QDs. (b) Absorption spectrum of CdS QDs after size-selective photoetching. (c) Absorption and PL spectra of surface-modified CdS QDs after photoetching.

The wide size distribution can be remedied using size-selective photoetching [33,34]. When CdS is irradiated with light under dissolved oxygen, electrons and holes are developed, resulting in the dissolution of CdS via the following chemical reactions in the vicinity of the surface [34].

$$CdS \rightarrow CdS\left(nh^+, ne^-\right)$$

$$CdS\left(nh^+, ne^-\right) + 2O_2 \rightarrow Cd^{2+} + SO_4^{2-}$$

Specifically, first, a sample is irradiated with monochromatic light corresponding to the absorption edge. Among the QDs of different sizes, the QDs whose exciton energies are resonant with the irradiation-light energy are photoetched. As the exciton energy of the QD increases with a decrease in the QD size, the QDs to be photoetched become smaller by increasing the irradiation-light energy. This process results in a narrow size distribution, and the QDs with uniform size can be finally obtained [33,34]. The photoetching method is very simple, it only irradiates the CdS QD solution with monochromatic light combining Xe lamp and interference filter.

Fig. 1(b) shows the absorption spectrum of CdS QDs after size-selective photoetching. Photoetching narrowed the QD size distribution, causing a clear absorption peak to appear in the absorption spectrum. The average diameter of the QDs and the width of the size distribution were estimated from the absorption spectrum via line-shape analysis to be 4.5 nm and 5%, respectively [35].

Fig. 1(c) shows the absorption and PL spectra of surface-modified CdS QDs after photoetching. The pH of the sample solution after photoetching was adjusted to the alkaline range, and an aqueous solution of cadmium ions was added to modify the surface. As a result, the surfaces of the CdS QDs were covered with $Cd(OH)_2$ layers [34,36], and band-edge PL was observed as the main PL band. Furthermore, the QD size could be controlled by the wavelength of the light used for irradiation during photoetching. CdS QDs with mean diameters of 4.5 and 5.0 nm were used as energy donors (D) and acceptors (A), respectively.

3. Synthesis and optical properties of CdTe QDs

CdTe QDs can be synthesized by a hydrothermal method as described in the previous paper [11,37]. Briefly, freshly prepared NaHTe solution was injected into the solutions of $Cd(ClO_4)_2 \cdot 6H_2O$ and *N*-acetyl-*L*-cysteine (NAC) at pH 7.0. A molar ratio and concentration of monomers of Cd: Te: NAC = 1.0: 0.2: 1.2 and $[Cd^{2+}] = 20$ mM were used. Then, 10 mL of the solution was loaded into an autoclave and incubated at 200 °C for a specified period. The QD size can be controlled by the reaction time as discussed in Refs. [11] and [37].

Fig. 2 shows the absorption and PL spectra of the CdTe QDs prepared at 200 °C for various reaction times. As reaction time increased, the absorption peak energy shifted toward the lower-energy side. Thus, the mean size of the CdTe QDs could be controlled by changing the reaction time. The lowest exciton energy originated from the 1s−1s electronic transition in CdTe QDs, where the diameter d is given by Ref. [38]

$$E(d) = 1.596 + \frac{1}{0.137d^2 + 0.206} \quad (\text{eV}). \tag{1}$$

Using Eq. (1), the mean diameters were estimated from the

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