

Optical characterization of CdSe nanocrystals

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Abstract

We have investigated the optical properties of CdSe nanocrystals by using the transmittance and photoluminescence spectroscopy. The samples used in the optical experiment are nanocrystals of cylindrical rods and spheres dispersed in toluene. The transition features in the photoluminescence spectra agree with the transition edges observed in the transmittance spectra. Theoretical models based on the effective mass approximation with infinite barriers were established for both types of nanocrystals. The calculated transition energies and oscillator strengths are compared with the observed spectral features. Improved models with the finite barrier and shallow potential step to account for the capping contribute to minor corrections of the order of thousandths.

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1. Introduction

The size of nanocrystals is of the order of several to tens nanometers which is close to the de Broglie wavelength of the electron and hole. Physically, nanocrystal is one of the low-dimensional semiconductor systems called quantum dot. In the quantum wire or quantum well systems, there are one or two translational degrees of freedom and a large number of free electrons and holes due to the large number of density of states. In the quantum dot system, there is no translational symmetry, and all degrees of freedom are confined. Nanocrystals have atomic like electronic configurations, photon-induced phenomena and very different optical properties from the bulk crystals. From the atomic or molecular points of view, nanocrystals can be regarded as large molecules. They are usually dispersed uniformly in the transparent solid or liquid for characterization. Based on the special optical properties of quantum dots and nanocrystals, many applications have been developed [1–6]. The optical properties such as line shape analysis have not been well understood yet. In this report, we investigated the absorption spectra of CdSe rods and spheres. The

transition energies and oscillator strengths are calculated by the effective mass approximation models and can be used as the basis for line shape analysis in the future.

2. Experimental

The CdSe nanocrystal was synthesized using the CdO one-pot approach [7,8]. The reaction mixture of cadmium oxide (CdO), tetradecylphosphonic acid (TDPA), and trioctylphosphine oxide (TOPO) is heated up to 300 °C in argon, and turned into homogeneous solution. After introducing selenium and trioctylphosphine (TOP) solution, and reacting for several minutes, slowly cooled down to room temperature. The brown CdSe powder obtained after methanol rinsing could be dispersed in toluene and hexane. Transmission electron microscopy (TEM) pictures in Fig. 1 show the rod shape of the TDPA- and TOPO-capped CdSe nanocrystals.

CdSe nanocrystals were dispersed in toluene for transmission measurements. A 150 W xenon arc lamp was used as light source. The wavelength was selected by a quarter-meter monochromator. The probe light is detected by a Si detector after transmitted through the sample and was normalized by the bare, toluene only, light intensity. In the photoluminescence (PL) experiment, the excitation

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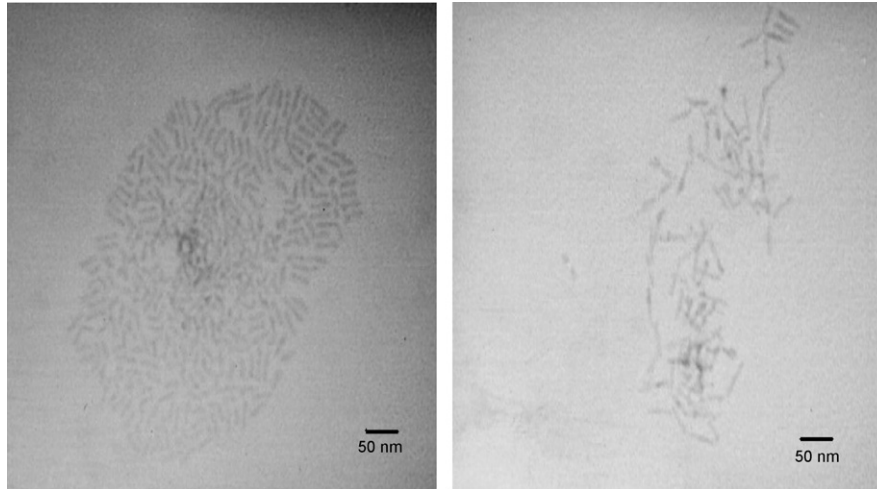


Fig. 1. TEM pictures of the 5×15 nm and 5×25 nm nanorods.

photons were provided by the 476 nm line of an argon-ion laser.

3. Results and discussion

Fig. 2 presents the absorption spectra from the transmittance spectroscopy of CdSe nanorods. The absorption edges are blue shifted as the size of the nanocrystal is reduced. The PL spectra of CdSe nanocrystals are also shown in Fig. 2. The transition features in the PL spectra agree with those observed in the absorption. Optical transition features blue shifted as the size of the nanocrystal becomes smaller due to quantum-confinement effect.

For the nanorod, we start with a rigid cylinder effective mass model with infinite potential barriers. The wave functions have cylindrical symmetry; the radial component is a Bessel function while the z component is a sinusoidal function as follows:

$$\varphi_{qst} = \sqrt{\frac{2}{\pi b [J'_m(K_{mn}a)]}} J_m(K_{mn}\rho) \sin(k_q z) e^{im\phi}, \quad (1)$$

where q , m , and n are quantum numbers, $J_m(K_m\rho)$ is the Bessel function, and $J'_m(K_{mn}a)$ is the first derivative of the Bessel function at radius a . Solving the rigid boundary condition equations would generate the allowed wave vectors $k_q = n_q\pi/b$ and $K_{mn} = y_{mn}/a$, where y_{mn} are roots of Bessel function $J_m(K_{mn}a) = 0$. Eigenenergies may be obtained from the energy–momentum relation:

$$E_{qmn} = \frac{\hbar^2}{2M} (K_{mn}^2 + k_q^2) = \frac{\hbar^2}{2M} \left[\left(\frac{y_{mn}}{a} \right)^2 + \left(\frac{n_q\pi}{b} \right)^2 \right]. \quad (2)$$

The transition energy will be the sum of the bulk gap energy, electron subband energy, and hole subband energy. The bulk band gap energy and the effective masses are taken from the literature [9]. Fig. 3 shows the variation of transition energies versus rod length for different radii. The transition energy is more sensitive to the radius than to the

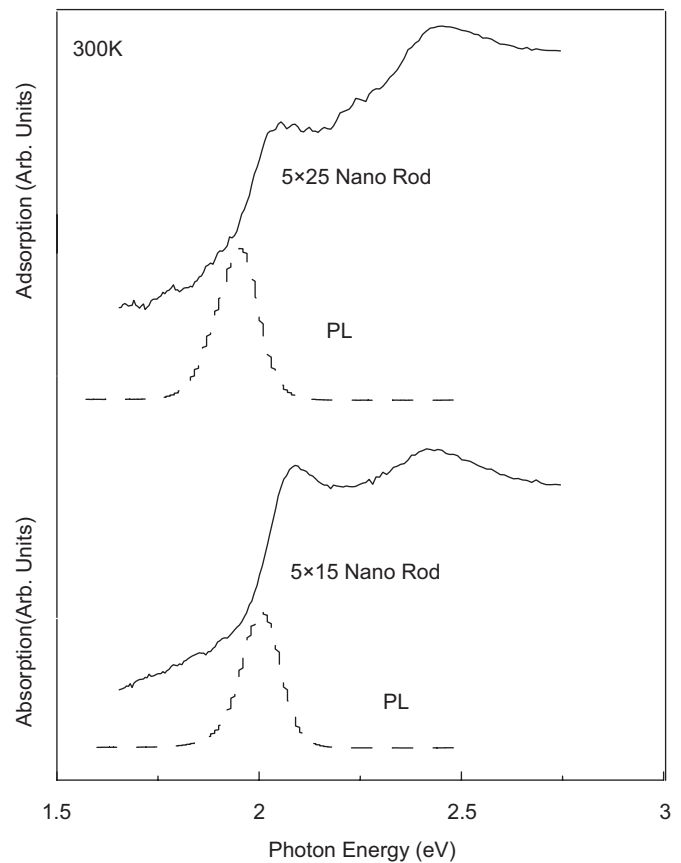


Fig. 2. Absorption and PL spectra of CdSe nanorods.

length. The calculated major transition energies are compared with the observed spectra in Fig. 4. Vertical bars indicate the position of theoretically calculated transition energies, and their lengths represent the oscillator strengths $f \equiv (2m_0\omega/3\hbar) |\langle \phi_m | r | \phi_\ell \rangle|^2$ obtained by integrating the dipole matrix element.

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