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Longitudinal-optical-phonon-assisted energy relaxation in self-assembled CdS quantum dots embedded in ZnSe

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The energy relaxation processes of CdS self-assembled quantum dots (QDs) embedded in ZnSe were investigated. Longitudinal-optical (LO)-phonon resonant structures were discerned in the photoluminescence spectra under the CdS selective excitation, especially under nearly resonant excitation conditions. Strong energy selectivity among the Gaussian distributed energy levels of the QDs are observed in energy relaxation processes, in which excitons are dominantly injected into the QDs whose ground state energies with respect to the excitation energy are equal to the multi-LO-phonon energy of ZnSe. Detection-energy dependent photoluminescence excitation (PLE) measurements also suggest that the coupling strength between the excitons and LO phonons is strongly size-dependent, i.e., coupling is strongly enhanced in smaller QDs. In addition, type-II band alignment of the CdS/ZnSe heterostructure is experimentally confirmed directly by PLE measurements. © 2002 American Institute of Physics. [DOI: 10.1063/1.1504161]

I. INTRODUCTION

Self-assembled quantum dots (QDs) have attracted considerable interest due to their zero-dimensional features based on the atomlike density of states and their study has remarkably progressed in various material systems such as group IV,^{1,2} III-V,³⁻⁶ III-V nitride,⁷⁻⁹ and II-VI^{10,11} semiconductors. Among them, the group II-VI semiconductors are preferable for realizing the practical optoelectronic devices that operate in the visible spectral range due to their wide band gap energies. From a viewpoint of basic physics, stronger exciton-longitudinal-optical(LO)-phonon coupling, i.e., the Fröhlich interaction¹²⁻¹⁴ and oscillator strength¹⁵ of the group II-VI semiconductors yield a strong coupling with phonons and photons, which will provide unique physical phenomena. As we reported very recently, an observation of Rabi splitting from the group II-VI QDs¹⁶ in a microcavity structure¹⁷ is among them. Self-assembled QDs, however, have a large inhomogeneous broadening due to their inherent size distribution, which will crucially affect the device operation. In order to realize practical devices with high performance and to enhance our knowledge of the basic physics, it is very important to understand the energy relaxation mechanism in the inhomogeneously distributed energy levels. To date, limited research on interdot energy relaxation processes in group II-VI self-assembled QDs have focused on the CdSe/ZnSe system.^{18,19} On the other hand, CdS is an attractive material for the optoelectronic devices that operate in the green wavelength region where the InGaN system is inaccessible due to the phase segregation issue.²⁰ Furthermore, due to the absence of a wetting layer as in the Stranski-Krastanow growth mode, CdS QDs grown on ZnSe more clearly reveals the problem in comparison to the CdSe/ZnSe system where the two-dimensional layer plays a significant role in the energy relaxation processes in QDs ensembles.¹⁸

In this article, we describe the interdot energy relaxation processes of CdS self-assembled QDs embedded in ZnSe. In the energy relaxation processes among the inhomogeneously distributed energy levels in CdS QDs, a strong energy selectivity was observed. In this respect, dominant exciton injection occurred into the QDs whose ground state energies in relation to the excitation energy match the multi-LO-phonon energy of ZnSe. The size-dependent energy relaxation studied by photoluminescence excitation (PLE) measurements revealed that the LO-phonon-related energy relaxation was shown to be strongly enhanced in case of the smaller QDs. Type-II band lineup of the present CdS/ZnSe system was also directly confirmed by means of PLE measurements as well as nonresonant photoluminescence (PL) measurements.

II. EXPERIMENTS

CdS QDs on ZnSe barrier layers were fabricated by metalorganic chemical vapor deposition on semi-insulating GaAs(001) substrates. In order to minimize the root mean square roughness of the 60-nm-thick ZnSe surface below 2 nm, the growth temperature was optimized at 300 °C. Dimethyl cadmium and di-tertiarybutyl sulfide were used as precursors to grow the CdS QDs. Their flow rates were 0.16 and 0.49 $\mu\text{mol}/\text{min}$, respectively. For the growth of the ZnSe layers, di-ethyl zinc and di-tertiarybutyl selenide were used. Before and after the growth of the CdS QDs, H₂ purging of the growth chamber was done to prevent the formation of undesirable alloying. In this experiment, the CdS QDs were five-stacked for obtaining sufficient PLE signal intensities. Each CdS layer was separated by 60 nm ZnSe intermediate layers and the topmost QDs were left open to the air for atomic-force microscope (AFM) measurements. A series of samples with the average QD heights of 1.7, 2.4, 3.3, and 6.6 monolayers (ML) were prepared. The average dot height was obtained by *in situ* optical monitoring with a He-Ne laser.

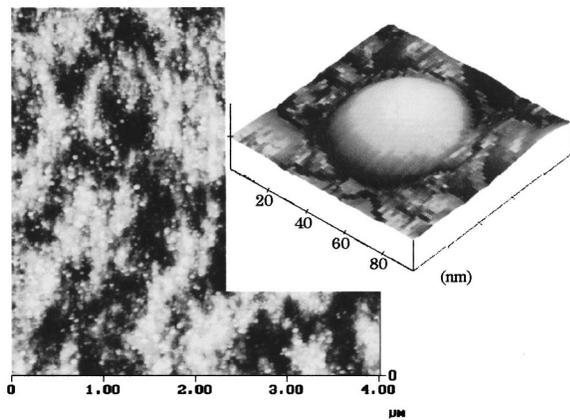


FIG. 1. Typical AFM images of the topmost layer of 3.3 ML height CdS dots grown on the ZnSe surface. Spatial distribution and density of QDs can be recognized as small white points in the $4\ \mu\text{m}\times 4\ \mu\text{m}$ image. The dome-shaped morphology of the CdS QDs is clearly resolved in the magnified picture.

For the study of the optical properties of these samples, the 325 nm line of He–Cd was used for the nonresonant excitation source, where the excitation energy was far above the ZnSe band gap. A multiline Ar⁺ laser was also used for the CdS QDs selective excitation. PLE spectra were taken with a monochromatic light from a tungsten halogen lamp. Throughout this work, the optical measurements were carried out at 16–36 K.

III. RESULTS AND DISCUSSION

An AFM image of the sample surfaces after the 3.3 ML CdS deposition is shown in Fig. 1. In the $4\ \mu\text{m}\times 4\ \mu\text{m}$ image, the spatial distribution of the QDs on the ZnSe surface can be overviewed, and the dome-shaped morphology of CdS QDs is clearly resolved in the magnified picture. Figure 2 shows the size distribution of the CdS dots for the 2.4, 3.3,

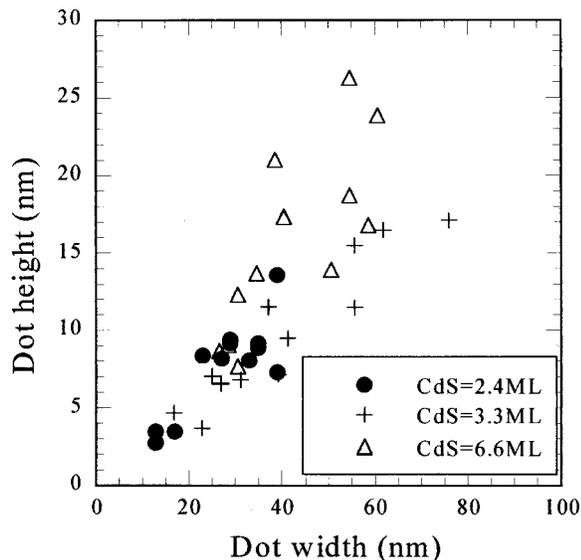


FIG. 2. Size distribution of CdS dots with the average thickness of 2.4–6.6 ML (from *in situ* monitor). The aspect ratio of the observed dot structures is approximately 4, which is independent of the dot size. The CdS QDs with an average thickness of 1.7 ML is not clearly resolved.

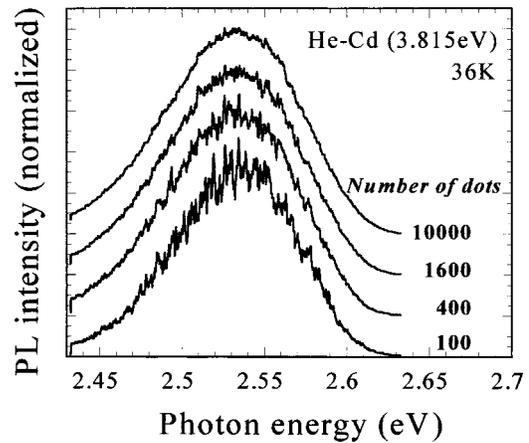


FIG. 3. Detection area dependence of micro-PL spectra of the CdS dots for a 1.7 ML sample measured at 36 K. With the decreasing number of dots introduced into the detector, the sharp lines become well deconvoluted from the Gaussian PL line shape, which unambiguously indicates the quantum dot formation.

and 6.6 ML samples. The CdS QDs with an average thickness of 1.7 ML is not identified by the AFM measurement due to insufficient spatial resolution and the residual roughness of the ZnSe surfaces. The density of the CdS dots is in the range of 2×10^9 – $8\times 10^9/\text{cm}^2$ and the aspect ratio is approximately 4 which is independent of the dot size. In order to reveal the dot formation for the 1.7 ML sample, detection area dependent micro-PL measurements were carried out and the results are summarized in Fig. 3. In this micro-PL experiment, only the number of dots introduced into a detection system was systematically changed.²¹ As the dot number decreased, sharp lines start to clearly appear on the Gaussian-shaped PL spectrum and the peak-valley ratio also increased. This deconvolution property from a broad PL line shape was highly reproducible and the energy positions of the sharp lines changed when shifting the excitation position on the sample surface. These observations prove that the size and/or position dependencies of the PL features were clear manifestations of the CdS dot formation.

Prior to the study of an energy relaxation process for the present CdS/ZnSe QDs, the nonresonant optical properties for the CdS QDs will be discussed. Normalized nonresonant PL and PLE spectra of the series of the samples measured at 16 K are summarized in Fig. 4. In the PLE measurements, the detection energies were set at the PL peaks for each sample, but the spectrum was not recorded for the 6.6 ML sample due to the extremely weak PL intensity. In the PL spectra, the luminescence from the CdS QDs region with full width at half-maximum of around 100 meV was observed. It is found that the PL peak energy is blueshifted when the dot size decreases. The ZnSe luminescence was hardly observed in all samples, which indicates a high exciton injection efficiency into the QDs region with fast energy relaxation. In the PLE spectra, beside the steep increase of PLE intensity at around 2.8 eV due to the onset of the ZnSe fundamental absorption, broad peak structures were also detected. These broad peaks are partly attributed to the low spectral resolution of 12 meV due to the wide slit widths of the monochromators obliged by the small signal intensity. However, the

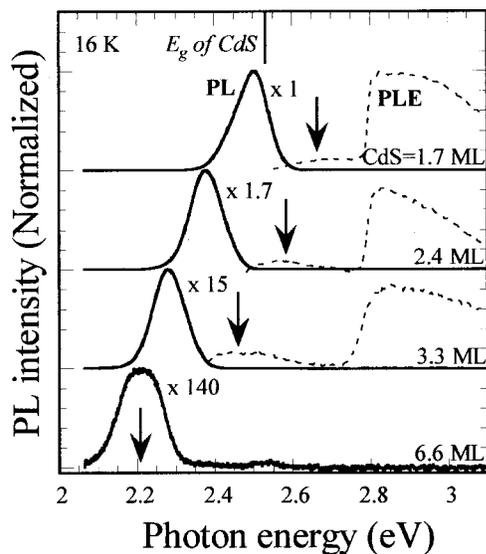


FIG. 4. Normalized PL (solid lines) and PLE (dashed lines) spectra of a series of samples under nonresonant excitation conditions. Downward arrows are calculated transition energies based on the type-II band alignment. Clear blueshift of peak energies with a reduction in the dot size is observed. Strong suppression of PL intensity with the increasing dot size is also noticeable.

linewidth of these peaks are much larger than the spectral resolution, then the systematic shift of the broad peaks to the higher energy with the decreasing average dot size strongly implies the contribution of the QDs in the PLE spectra. Figure 5 shows the dependence of the PL peak energies as a function of the CdS average thickness. This PL blueshift feature, which is analogous to the previously reported article,²² is attributed to the quantum size effect of the CdS QDs. In Fig. 4, it should be stressed that the PLE peak energy decreases across the energy gap of CdS of 2.53 eV with the increasing dot size, which is unusual for type-I heterostructures. This behavior is explained by the type-II band align-

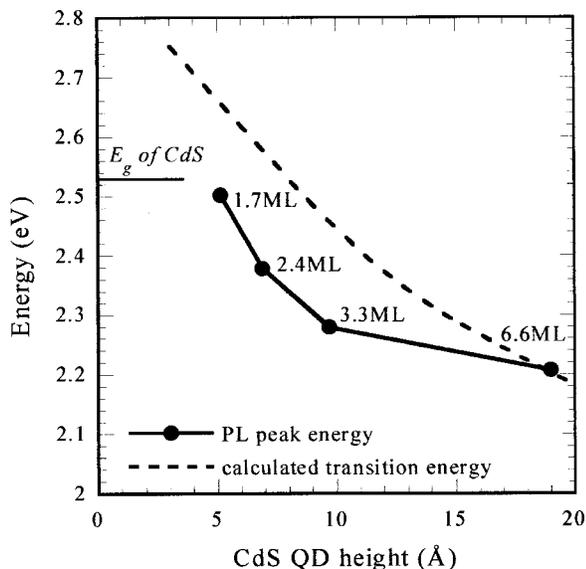


FIG. 5. PL peak energies as a function of CdS average thickness. The solid line is a visual guide. Dashed line indicates the calculated transition energy.

ment in which the transition between the ground level of the CdS quantum dot (conduction band) and the top of the valence band of the bulk ZnSe is taken into account.²³ A transition energy calculation assuming a one-dimensional confinement along the growth direction and the valence band offset of 670 meV between ZnSe and CdS²⁴ was then carried out. The resultant size dependence of the transition energy is shown by the dashed line in Fig. 5. The estimated transition energies for each sample are also indicated as downward arrows in Fig. 4, which agrees very well with the detected PLE peaks. This result is a direct confirmation of the type-II band alignment of the present CdS/ZnSe heterostructure^{24,25} based on the PLE measurements. It should be noted that the contribution of the direct transition in the CdS QDs to the observed spectra is excluded because its transition energy, including the one-dimensional confinement, is higher than the above calculation by 670 meV. A large Stokes shift between the PL and PLE peaks is also observed due to the dot size inhomogeneity, which is typical for self-assembled QDs systems. In Fig. 4, a significant reduction in the PL peak intensity of more than two orders of magnitude was observed when the dot size increases from 1.7 to 6.6 ML. The type-II band alignment also explains this quenching with the increasing dot size. The increased spatial separation of electrons and holes for larger dots leads to the lower oscillator strength due to the smaller overlap integral of the wave functions as theoretically predicted in type-II heterostructures.²⁶

In order to investigate the energy relaxation process in the energy level distribution of the CdS QDs, PL measurements under CdS selective excitation were carried out using an Ar⁺ laser with three different excitation energies (E_{ex}) of 2.627, 2.666, and 2.707 eV. In this experiment, the sample with the average CdS thickness of 1.7 ML was used. The PL spectra with respect to the excitation energies are presented in Fig. 6. The PL spectrum with nonresonant excitation using a He-Cd laser (3.814 eV) is also shown at the top. The dashed lines represent the fitted Gaussian line shapes to reproduce the spectra. When the excitation energy is lowered, deviation from the Gaussian line shapes and subpeak structures, which are superimposed on the higher energy side of the smooth Gaussian-line shaped luminescence, become distinguishable. The extracted peak energies of the observed subpeak structures are plotted as a function of the excitation energy in Fig. 7. The solid line is an energy reference indicating the excitation energy, which is an origin to measure the energy relaxation after the photoexcitation. The energy spacing between the dashed lines corresponds to the period of the subpeak structures. From this figure, the period of the observed subpeaks is about 30.5 meV, which is well coincident with the ZnSe LO-phonon energy (E_{LO}) of 31 meV (Ref. 27) rather than that of 37 meV (Ref. 28) in CdS. Therefore, the origin of these subpeak structures can be assigned to the LO-phonon resonance in the exciton energy relaxation processes that dominants in ZnSe layers. As previously mentioned, electrons and holes are spatially separated in the present type-II heterostructures; electrons (holes) are predominantly confined in the CdS (ZnSe) layers. Thus, the observed energy relaxation involved in the LO-phonon emission in the ZnSe layers implies that a significant portion of

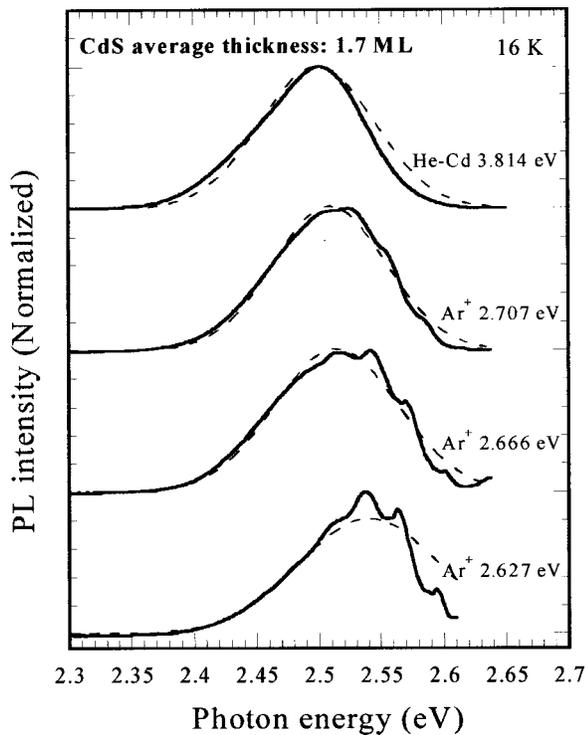


FIG. 6. PL spectra of QDs with average CdS thickness of 1.7 ML under CdS selective excitation. Excitation energies are 2.627, 2.666 and 2.707 eV, and the result of the nonresonant excitation is also shown. With a lower excitation energy, subpeak structures superimposed on the higher energy side of the broad Gaussian peak become apparent. Dashed lines show fitted Gaussian line shapes. The blueshift of the Gaussian PL peak and the distortion of the spectra are also observed by lowering the excitation energy.

the exciton wave function spreads out in the ZnSe layers in real space. Therein, energy relaxation takes place. It is expected that the QDs with the energy level E_{QD} , which satisfy the relationship of $E_{\text{ex}} - E_{\text{QD}} = n \cdot E_{\text{LO}}$, where n is the num-

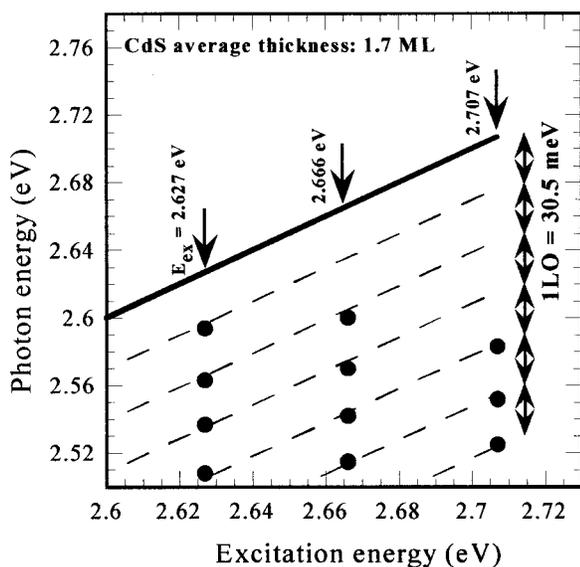


FIG. 7. Extracted peak energies of the observed subpeak structures under three excitation energies. The period of the observed subpeaks is about 30.5 meV, which is well coincident with the LO-phonon energy of ZnSe. The origin of these subpeaks is attributed to the LO-phonon resonance in the exciton energy relaxation that significantly occurs in ZnSe layers.

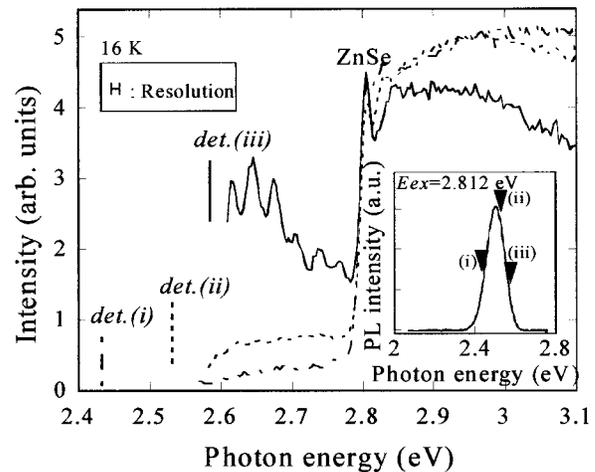


FIG. 8. PLE spectra under three detection energies of (i) 2.431 eV, (ii) 2.530 eV, and (iii) 2.583 eV. Corresponding energy position on PL spectrum is shown in the inset. Distinguished resonance up to six orders of LO-phonon resonant structures are observed for the 2.583 eV detection energy. Lowering the detection energy (equivalent to detecting the larger dots) shows strong damping of the phonon resonance and the relative intensity is also suppressed. It is suggested that the smaller QDs have a stronger exciton-LO-phonon coupling.

ber of LO-phonon emissions, will have a large exciton population due to efficient energy relaxation by the LO-phonon emission. In this sense, the LO-phonon works as a size filter of the QDs, in which the excitons are significantly injected into the QDs whose ground state energies with respect to the excitation energy are equal to the multi-LO-phonon energies. Furthermore, QDs which satisfy the above expression with a smaller n are likely to have a prominent resonance structure. The LO-phonon resonant features with a higher order will be blurred by the contribution of other energy relaxation channels such as acoustic-phonon²⁹ and/or Auger-like processes.³⁰ The blueshift of the Gaussian line shaped PL peak with a lower excitation energy was also observed in Fig. 6. The blueshift and distortion of the PL lineshapes from the Gaussian mentioned above are interpreted as follows; the faster and well-defined LO-phonon assisted population is realized at smaller QDs since a lower number of LO-phonon involvements, “ n ,” is required to relax down to the ground states of the QDs.

For the further investigation of the LO-phonon relaxation processes in the CdS QDs ensemble, detection energy dependent PLE measurements were performed. The PLE spectra are not only an indication of absorption but also crucially influenced by the exciton energy relaxation down to the detection window. Therefore, the size-dependent energy relaxation efficiency can be examined by selecting the energy of the detection window.^{31,32} Figure 8 shows the PLE spectra of the sample with an average CdS thickness of 1.7 ML at three detection energies of (i) 2.431 eV, (ii) 2.530 eV, and (iii) 2.583 eV with the corresponding energy positions on the PL spectrum shown in the inset. The intensity of each spectra was normalized at the ZnSe absorption edge for comparison. When the detection energy is set at 2.583 eV, the distinguished resonance structures are unambiguously found to dominate the shape of the PLE spectrum. The period of

the peak intervals is almost identical to that observed in the PL spectra under CdS selective excitation. Therefore, these resonance structures stem from the LO-phonon mediated exciton energy relaxation processes. Up to six orders of LO-phonon resonant structures are clearly resolved below the ZnSe absorption edge, which will hinder the appearance of the higher order resonance. Lowering the detection energy, which is equivalent to probe the larger QDs among the size distribution, results in a strong damping of the phonon resonance. The suppression of the relative intensity also occurs. Other samples with a larger average dot size did not show the LO-phonon resonance features in the PLE spectra regardless of the detection energy.

In the ZnCdS/ZnS superlattice system, the observation of clear oscillatory structure due to the LO phonon emission in the PLE spectra was reported, in which the detection energy dependence analogous to our result was shown.³³ It is suggested that the exciton localization played an important role in the detection energy dependence of the oscillatory structure in the PLE spectra. On the other hand, in the present case of QDs, an energy transfer process into the lower energy states is expected to be less dominant due to the strong exciton confinement and spatial separation between the QDs. Therefore, the obtained results can be interpreted as a strongly size-dependent energy relaxation efficiency mediated by the exciton-LO-phonon interaction, i.e., smaller QDs will induce a larger exciton-LO-phonon coupling. The size dependence of the exciton-LO-phonon interaction (Fröhlich interaction) in spherical QDs in glass is reported for GaAs and CdSe QDs.³⁴ A variational calculation showed monotonic reduction of the Fröhlich interaction by reducing the dot size. In addition, the calculation predicts that the coupling between electrons and holes will vanish when the dot radius tends to be zero since the charge distribution will become identical. In the present case of CdS/ZnSe, however, the type-II band lineup is likely to make the physics different. For a full account of the underlying physics of the observed size dependence, further studies from both theoretical and experimental aspects are required. The type-II band alignment of the CdS/ZnSe system, nonetheless, must play a very important role in the observed size dependence of the LO-phonon mediated energy relaxation efficiency. Overlap of the electron and hole wave functions will strongly depend on the dot size in the type-II systems since it is determined by the spatial overlap of exponential tails in the vicinity of the heterointerfaces. As the dot size gets larger, due to the reduced penetration of the wave function into the adjacent layers, the contribution of the exponential tails rapidly decreases. It is suggested that this size-sensitive excitonic character will cause a drastic change in the exciton-LO-phonon interaction in the present CdS/ZnSe system.

IV. CONCLUSION

The size dependence of the transition energy and energy relaxation processes of excitons in CdS self-assembled quantum dots embedded in ZnSe layers were discussed. The size dependence of the PLE spectra clarified that the CdS/ZnSe is categorized as a type-II band lineup. Photoluminescence un-

der CdS selective excitation and detection-energy dependence of the PLE measurements revealed that the exciton energy relaxation via ZnSe LO-phonon emission was more efficient for the smaller QDs.

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