

Recombination dynamics of localized excitons in a CdSe/ZnSe/ZnS_xSe_{1-x} single-quantum-well structure

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Optical properties of localized excitons have been studied in a highly strained CdSe quantum well with 1-ML thickness by employing time-resolved photoluminescence (PL) and nonlinear PL spectroscopy under various excitation conditions. At 20 K, the time-integrated PL from the well layer was peaked at 2.7276 eV with a linewidth of 21 meV under low excitation intensity ($0.11 \mu\text{J}/\text{cm}^2$). The lifetime ranged from 200 ps to 50 ps as the monitored photon energy was changed from the low-energy tail to the high-energy one. The behavior could be well understood as a result of exciton localization, which is induced by terraces and islands with units of ML thickness fluctuation lying at the interface between CdSe and ZnSe. An emission with the fast decay component was observed at the low-energy side of the peak (2.7168 eV) under higher excitation condition. The emission could be well resolved as a positive component by the nonlinear PL measurement, and originates from the many-body effect of localized excitons, which is probably attributed to localized biexcitons. [S0163-1829(96)07828-9]

I. INTRODUCTION

Ternary and quaternary II-VI alloy semiconductors are examples of materials with perturbed crystal potential. The perturbation results from microscopic, statistical variations of the alloy composition on a microscopic scale. This alloy disorder leads to a density of tail states of the form of $\exp[-E/E_0]^n$ extending down to the forbidden gap, where E is band energy and E_0 the characteristic energy for the density of states. The value of n is under debate and it has been reported to be from 0.5 to 2.0. The localization of excitons to these tail states locates the peak energy of photoluminescence (PL) on the lower-energy side than that of absorption (the so-called Stokes shift) and the PL linewidth is broadened.

Such phenomena are also induced in quantum-well (QW) structures with interfacial disorders. Microscopically, heterointerfaces between wells and barriers are pseudosmooth within certain lateral regions, which vary in size at small terraces or extended islands with their step heights being usually in monolayers (ML). If the region of islands is substantially larger than the in-plane diameter of excitons, PL spectra are composed of a few sharp emissions, which correspond to quantized energy levels determined by the well width.^{1,2} Such an interface has been achieved in III-V semiconductors by the advanced growth technique. However, the density of islands highly depends on growth conditions and, in most cases, transition energies are averaged within the volume of excitons due to the small size of islands. Consequently, unlike the localization caused by the effect of alloy disorder, the degree of localization strongly depends on the quality of the interface.

In recent years, intense studies have been carried out on excitonic properties of ZnCdSe/Zn(S)Se QW's in terms of

their usefulness for blue-green laser diodes.^{3,4} Since the well layer in the structures is under compressive strain, the lowest excitonic transition occurs between the $n=1$ conduction electron and $n=1$ heavy hole, generally designated as E_{x1hh} . The solid composition of Cd in Zn_xCd_{1-x}Se well layers is typically about 15–20% in the device structures. The PL linewidth of E_{x1hh} for such layers grown under the appropriate condition is as low as about 9 meV at low temperature, a value that agrees fairly well with the theoretical one derived from the alloy broadening effect.^{5,6} This suggests that the localization of the exciton in these structures is mainly caused by the effect of alloy broadening.

In CdSe/ZnSe QW structures, on the other hand, localization of excitons occurs only due to interface roughness. This enables us to investigate easily the properties of localized excitons focusing on the interface structures. The well width fluctuation of 1 ML in the CdSe/ZnSe QW with ultrathin well width (≤ 5 ML) results in the fluctuation of the electron confinement energy of the order of 100 meV. The strong spatial variation of the carrier confinement leads to significant localization of excitons to the tail states, and the optical and electrical properties of the structure are extremely sensitive to such kind of disorder. Accordingly, another feature of this structure is the large lattice mismatch between the well and the barrier. It has been reported that pseudomorphic growth of the CdSe layer on the ZnSe buffer without the formation of misfit dislocations is limited to a few ML's and the high PL efficiency is obtained in such pseudomorphic well layers.⁷ In this study, the CdSe single QW (SQW) with 1-ML thickness was embedded in a ZnS_xSe_{1-x}ZnSe/ZnS_xSe_{1-x} waveguide structure, which was coherently grown on the GaAs substrate. The use of the structure has two reasons: one is for the effective confinement of photo-generated carriers, and the other is that the flatness of ZnSe

coherently grown on a $\text{ZnS}_x\text{Se}_{1-x}$ buffer is superior to that grown on a strain-relaxed ZnSe layer.

More recently there has been increasing interest in lasing processes in disordered semiconductors in terms of the fact that localized excitons can easily contribute to the optical gain due to their low density of states.^{3,8} In many studies, however, many-body effects of localized excitons have been ignored as a first-order approximation. This is probably because of the difficulty in the assignment of new emissions, which may appear as a function of photoexcitation intensity, caused by the inhomogeneous broadening effect such as alloying and/or interfacial disorder.

In this paper, effects of high excitation on the dynamical behavior of localized excitons in the CdSe SQW have been assessed by employing linear and nonlinear luminescence spectroscopy.

II. EXPERIMENTAL PROCEDURE

The CdSe/ZnSe/ZnS_xSe_{1-x} separate confinement single quantum well (SC-SQW) structure described above was grown by molecular beam epitaxy on a Zn-doped *p*-type GaAs substrate oriented to (100). The growth of the SC-SQW structure proceeded as follows.

First, a 0.85- μm ZnS_{0.07}Se_{0.93} cladding layer was deposited, followed by a 30-nm ZnSe buffer layer. A CdSe SQW (the active layer) is sandwiched between ZnSe waveguide layers (50 nm in each). Finally, a 0.15- μm ZnS_{0.07}Se_{0.93} capped layer was grown. The mean thickness of the well was set to 1 ML. The value was estimated according to the growth rate and PL energy of the CdSe layer. This estimation has been assured by the previous results for the growth of CdSe on ZnSe,⁹ where the CdSe thickness was estimated by the reflection high-energy electron diffraction oscillation and the cross section transmission electron microscopy.

Time-resolved PL measurements were carried out using a frequency-doubled beam of an Al₂O₃:Ti laser pumped by a cw Ar⁺ laser, and a photon counting method with a synchroscan streak camera in conjunction with a 25-cm single-grating monochromator. The wavelength (WL), the pulse width (PW), and the repetition rate (RR) of the excitation beam were 390 nm, 1.7 ps, and 82 MHz, respectively. The incident excitation energy density was tuned by the variable neutral density filter. The time resolution of the detection system is about 20 ps. Accordingly, the exponential decay time can be estimated within the accuracy of 5 ps by the deconvolution technique.

In order to investigate high excitation effects, nonlinear PL spectroscopy was performed by the population mixing technique, where two excitation beams were modulated at different frequencies ($\Omega=831$ Hz, $\Omega=1000$ Hz) by a chopper and focused at the same spot on a sample. The nonlinear PL component was obtained by detecting the sum-frequency element ($\Omega_1+\Omega_2=1831$ Hz) of the total PL signal using a lock-in technique. In this study, the second harmonic light of the amplified laser output (WL, 400 nm; PW, 200 fs; RR, 250 kHz) was used as an excitation source, and the delay between the excitation pulse pairs was set to zero. The principle of this spectroscopy is based on the fact that the PL contains a substantial nonlinear component in the radiative process. For example, the negative nonlinearity is observed

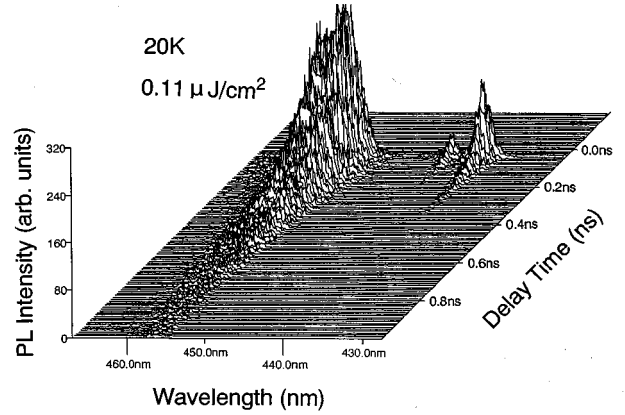


FIG. 1. Three dimensional representation of the time-resolved PL spectrum taken from the CdSe/ZnSe/ZnS_xSe_{1-x} SC-SQW structure under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$. Emissions from ZnSe and ZnS_xSe_{1-x} layers decay rapidly, and the emission from the CdSe QW dominates in intensity. This indicates that photogenerated carriers are effectively captured to the well.

in the vicinity of the linear-PL peak under high excitation conditions due to the band-filling effect. Accordingly, positive nonlinearity is occasionally detected at either the higher- and/or lower-energy side of the negative peak as a result of the formation of hot excitons and/or bimolecular excitons (biexcitons).

III. RESULTS AND DISCUSSION

A. Time-resolved PL from a CdSe QW, ZnSe waveguides, and ZnS_xSe_{1-x} cladding layers

Figure 1 shows a time-resolved PL spectrum from the CdSe/ZnSe/ZnS_xSe_{1-x} SC-SQW structure under the excitation energy density (I_{ex}) of $0.11 \mu\text{J}/\text{cm}^2$. The spectrum is composed of three parts, located at about 437, 440, and 458 nm, which correspond to emissions from ZnS_xSe_{1-x} cladding layers, ZnSe waveguiding layers, and a CdSe SQW, respectively. Lifetimes of PL from ZnS_xSe_{1-x} and ZnSe layers are as small as 47 and 25 ps, respectively. Time-resolved signals from ZnSe layers and a CdSe SQW reach the maximum after time delays of several ps and 55 ps, respectively with respect to the maximum of the signal from ZnS_xSe_{1-x} cladding layer. These results indicate that recombination processes in both ZnSe and ZnS_xSe_{1-x} layers are not dominated by radiative transitions but by transfer processes of photogenerated carriers to the CdSe SQW. Moreover, an emission from the CdSe well is dominant in spite of a very thin thickness (1 ML) of the active layer.

It had been recognized that the well layer thickness of 1 ML was too small to effectively collect carriers. Kolbas, Lo, and Lee¹⁰ have pointed out, however, that the efficiency of carrier collection in Al_xGa_{1-x}As/GaAs SQW's is not determined by the well width but by the spatial extent of the wave function (Ψ). The full width at half maximum (FWHM) of probability densities ($|\Psi|^2$), at first, decreases with well width and then increases for very thin wells due to the spreading of the wave function out of the well. In our structure, the FWHM of $|\Psi|^2$ of the $n=1$ electron and heavy

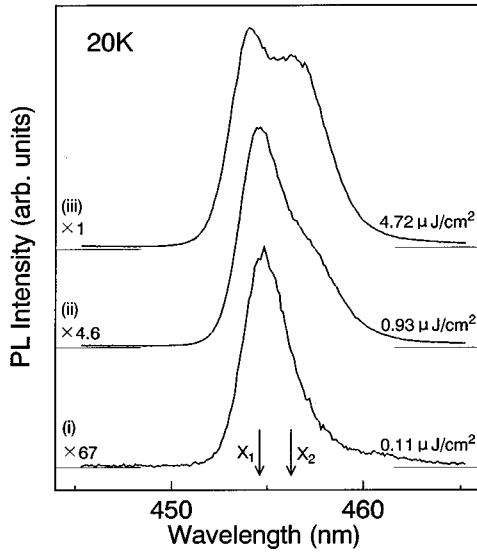


FIG. 2. Time-integrated PL spectra obtained under three different I_{ex} values of 0.11, 0.93, and $4.72 \mu\text{J}/\text{cm}^2$. Under the lowest excitation ($I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$), a broad excitonic emission (denoted by X_1) is observed. With increasing excitation density a shoulder (denoted by X_2) grows on the low-energy side of the X_1 . The energy difference between the X_1 and the X_2 peaks is estimated to be about 11 meV.

hole is calculated to be 4.5 and 4.0 nm, respectively, which are that values also achieved with the well width of about 4 nm.¹¹ It should be also noted that the ZnSe waveguiding layers (100 nm) act as the reservoir of photogenerated carriers, which promotes the collection of them to the lowest-energy level of the well.

The FWHM of PL from the CdSe SQW is about 21 meV. The broadening originates from the fluctuation of well layer thickness within the volume of the exciton. This probably indicates that there exist some regions without the CdSe layer grown, and that the QW is divided into some parts in the lateral direction. If this is the case, it is necessary to take into account the effect of lateral confinement for the calculation of energy level.

B. Time-integrated PL

Figure 2 shows the time-integrated PL spectra at 20 K taken from the CdSe well layer under the I_{ex} values of (i) 0.11, (ii) 0.93, and (iii) $4.72 \mu\text{J}/\text{cm}^2$. The PL at the lowest excitation density shown in curve (i) is dominated by the recombination of localized excitons (denoted by X_1), whose peak is situated at 454.61 nm (2.7276 eV). The X_1 peak gradually shifts to higher photon energy with increasing excitation densities. This is because of the filling of the density state, which is easily induced for localized excitons lying at the tail state. This phenomenon is demonstrated by the nonlinear luminescence measurement described below.

With raising the excitation density up to $0.93 \mu\text{J}/\text{cm}^2$, a shoulder appears on the low-energy side. When further increasing excitation density, the shoulder grows more rapidly than the X_1 line, and a new peak (denoted by X_2) appears at 456.41 nm (2.7168 eV). The energy difference between X_1 and the X_2 is estimated to be about 11 meV.

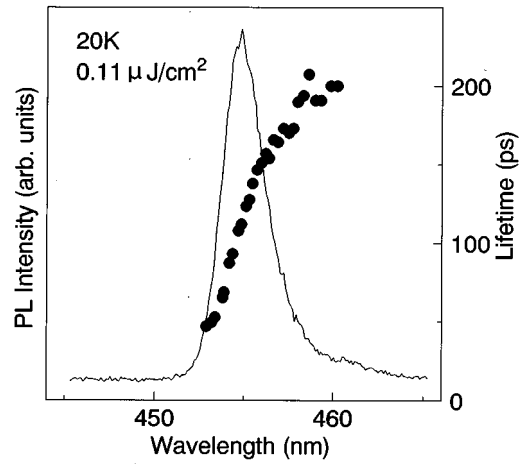


FIG. 3. Lifetimes (closed circles) and time-integrated PL spectrum for the sample studied here obtained under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$. The values of lifetime increase monotonously with decreasing photon energy.

C. Decay and lifetime

Figure 3 shows the lifetimes of the emission as a function of photon energy, together with the time-integrated PL spectrum measured under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$, the lowest excitation condition. Figure 4 depicts PL decay curves monitored at four different emission energies. The decay spectra are well fitted by single exponential curves. The PL intensity reaches the maximum after a certain time after pulsed excitation. The rise time as well as the lifetime increase with decreasing monitored photon energy. These behavior can be understood as the effect of exciton localization,¹²⁻³⁰ where decay of excitons is not only due to radiative recombination but also due to transfer processes to the low-energy tail by the assistance of acoustic phonons.

Lifetimes obtained have been well fitted by the theoretical decay time $\tau(E)$ as a function of emission energy as follows:

$$\tau(E)^{-1} = \tau_r^{-1} \{1 + \exp[(E - E_{me})/E_0]\}, \quad (1)$$

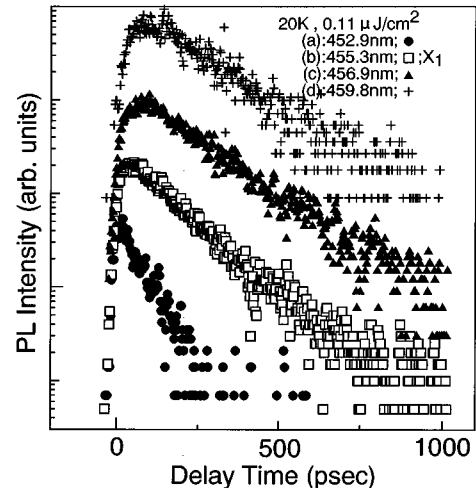


FIG. 4. PL decay curves from the CdSe SQW under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$ obtained under various monitored photon energies.

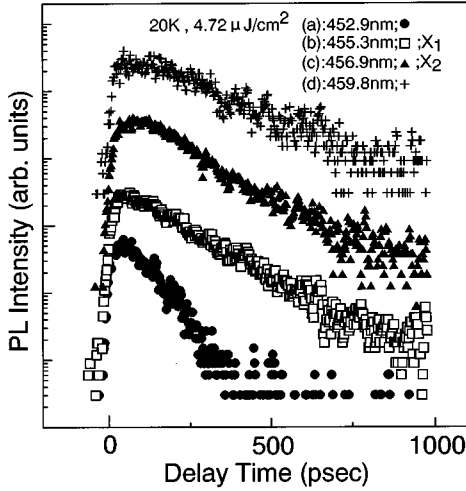


FIG. 5. PL decay curves from the CdSe SQW under $I_{\text{ex}}=4.72 \mu\text{J}/\text{cm}^2$ obtained at various monitored photon energies. Decay curves obtained in the vicinity of the X_2 are well fitted by the double exponential function.

where E_0 is the characteristic energy for the density of states, τ_r is the radiative lifetime, and E_{me} is defined by a definite energy for which the decay time equals the transfer time.¹² The best fit has been obtained using the following parameters: $E_0=8.5$ meV, $\tau_r=209$ ps, and $E_{me}=2.7267$ eV. It should be noted that such a fitting under low excitation conditions has also been made for localized excitons in CdSe/ZnSe SQW's grown by a self-limiting monolayer epitaxy⁷ and that a similar E_0 value has been obtained. E_0 represents the degree of localization. The value of 8.54 meV is larger than the values reported for CdS_xSe_{1-x} (Refs. 12–21) and Zn_xCd_{1-x}S (Refs. 27 and 31) alloys and for Zn_xCd_{1-x}Te/ZnTe (Refs. 25 and 26), and CdTe/ZnTe (Refs. 22 and 24) QW's. This implies that the large localization of excitons is rather a universal characteristic in the ultrathin CdSe/ZnSe SQW system.

Figure 5 shows decay curves monitored at various emission energies under $I_{\text{ex}}=4.72 \mu\text{J}/\text{cm}^2$, the highest excitation condition. It was found that the PL transient $[I(t)]$ in the vicinity of the X_2 line decays with a double exponential profile shown as

$$I(t) = A_f \exp(-t/\tau_f) + A_s \exp(-t/\tau_s), \quad (2)$$

where τ_f and τ_s are the decay times of the fast and the slow components, respectively. Fitted values of τ_f , τ_s , and A_f/A_s are potted in Fig. 6 as a function of monitored emission energy. Both the slow and the fast decay components increase with decreasing photon energy, indicating that both the X_1 and the X_2 lines are related to localized excitons. The tendency of the slow components is almost the same as that of lifetimes obtained under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$. The fast components are estimated to be roughly half as small as the slow components, probably suggesting that the X_2 emission originates from the biexcitonic transition, where a biexciton recombines radiatively, leaving a photon and an exciton. This is because the formation rate of biexciton concentration is proportional to the square of exciton concentration after a certain time of pulsed excitation.^{32,33} The situation, however,

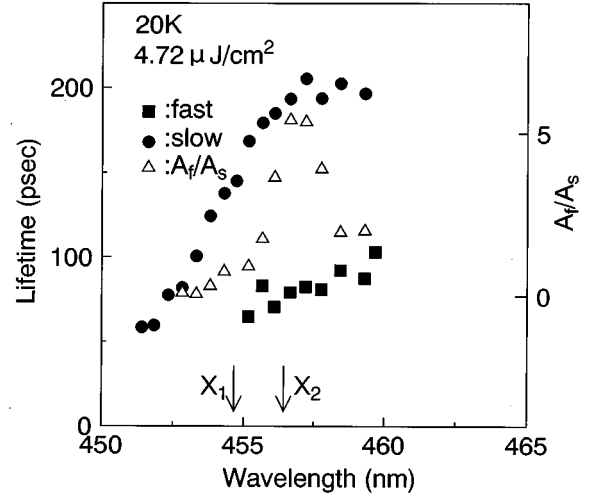


FIG. 6. Slow and fast components of lifetimes (τ_s and τ_f), and A_f/A_s under $I_{\text{ex}}=4.72 \mu\text{J}/\text{cm}^2$. The tendency of the slow components is almost the same as that of lifetimes obtained under $I_{\text{ex}}=0.11 \mu\text{J}/\text{cm}^2$ (Fig. 3). The fast components are roughly half as small as the slow components.

is complicated in this case because the effect of transfer processes has to be considered in the localized excitonic states. Consequently, for the quantitative analyses, it is necessary to construct the model of population dynamics of localized excitons and localized biexcitons.

PL intensity, I_{PL} , changes with I_{ex} as in the following equation. Neglecting the terms higher than third order, I_{PL} is written as

$$I_{\text{PL}} = kI_{\text{ex}}^n \approx aI_{\text{ex}} + bI_{\text{ex}}^2. \quad (3)$$

The signal detected by the nonlinear PL spectroscopy is the second term (b) proportional to I_{ex} .

Figure 7 shows the nonlinear PL spectra under three different excitation intensities. The positive signal means the superlinear component, while the negative signal corresponds to the sublinear one. Under low values of I_{ex} , the positive signal is observed in the whole spectral range, and the spectra shape is almost the same as the time-integrated linear PL. The origin is probably due to the saturation of residual nonradiative centers by the photogenerated carriers. Increasing I_{ex} more than about $0.31 \mu\text{J}/\text{cm}^2$, the positive signal at the X_2 line grows more than that at the X_1 line. Under the excitation intensity of $I_{\text{ex}}=4.72 \mu\text{J}/\text{cm}^2$, the nonlinear signal at the X_1 line changes its sign to show negative nonlinearity because of the band-filling effect of localized excitons. However, the positive signal at the X_2 line remains to grow even at the excitation density, indicating that the X_2 line originates from the many-body effect of localized excitons such as localized exciton-exciton collisions or localized biexcitons.³⁴

D. Time evolution of PL spectra

Figures 8 and 9 show the time evolution of the luminescence spectra measured as a function of time after pulsed excitation (t_d) under two different excitation intensities of 0.11 and $4.72 \mu\text{J}/\text{cm}^2$, respectively. Under the excitation

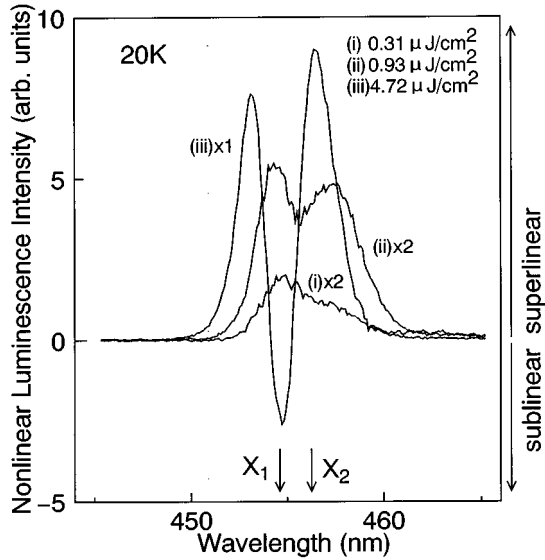


FIG. 7. Nonlinear PL spectra under three different I_{ex} values of (i) $0.31 \mu\text{J}/\text{cm}^2$, (ii) $0.93 \mu\text{J}/\text{cm}^2$ and (iii) $4.72 \mu\text{J}/\text{cm}^2$. The upward direction on the vertical axis means the superlinear dependence, while the downward direction corresponds to the sublinear dependence.

intensity of $0.11 \mu\text{J}/\text{cm}^2$, the PL peak at $t_d=0$ ps is located at 454.4 nm (2.728 eV), and then shifts towards a lower photon energy with delay time at a rate of -3.0×10^7 eV/s.³⁵ This behavior can be understood as a transfer process of excitons to the tail state as discussed in the previous section. The PL spectrum slightly narrows with time from $t_d=0$ to 40 ps. A shoulder appears and grows at about 458 nm (2.71 eV) after $t_d=$ about 40 ps. The component is overlapped with the main peak (X_1) after $t_d=400$ ps, and no peak shift was observed after then. This indicates that the density of tail states [$g(E)$] is not exactly the form of

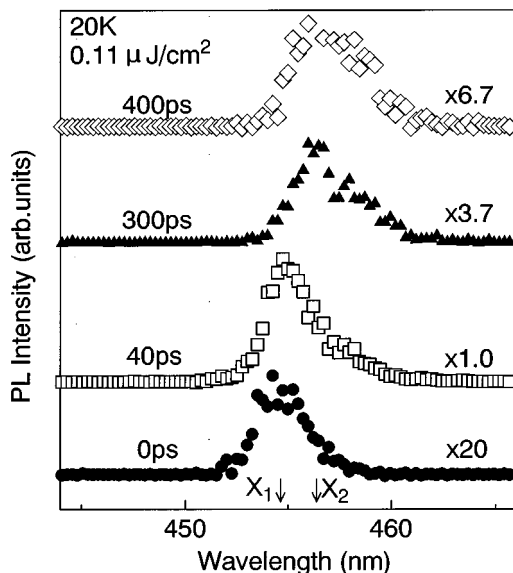


FIG. 8. PL spectra measured as a function of delay time under $I_{ex}=0.11 \mu\text{J}/\text{cm}^2$. Because of the transfer process of localized exciton, PL peaks shift towards lower photon energy with time delay.

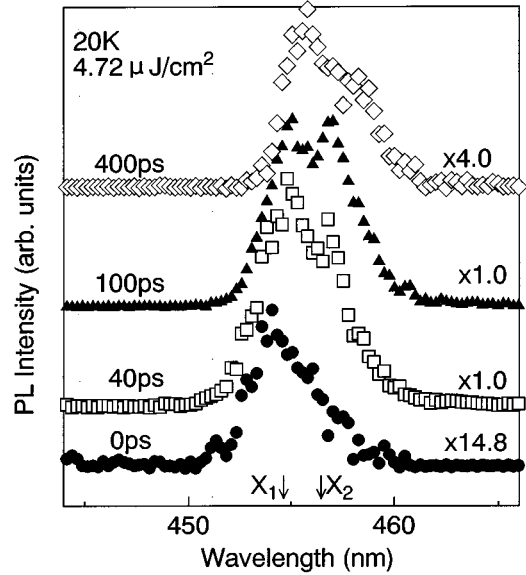


FIG. 9. PL spectra measured as a function of delay time under $I_{ex}=4.72 \mu\text{J}/\text{cm}^2$. The X_2 line does not appear just after excitation. It begins to grow at about t_d equal to several tens ps, and then reaches to the maximum at $t_d=100$ ps.

$\exp[-E/E_0]$ but rather a form of separately distributed features depending on the distribution of islands and terraces in the QW.

Under $I_{ex}=4.72 \mu\text{J}/\text{cm}^2$, the PL spectrum is nearly the same as that under $I_{ex}=0.11 \mu\text{J}/\text{cm}^2$, and no X_2 line is observed just after the excitation ($t_d=0$ ps). The X_2 line begins to grow at t_d equal to several tens of ps, and reaches the maximum at $t_d=100$ ps, and then decays more rapidly than the X_1 line. The dynamics of localized excitons under high excitation conditions can be described as follows. Photogenerated excitons relax to the states of lower energy and reach the local minima. In such a situation, excitons are localized spatially within the well layer due to the break of the momentum preservation, and many-body effects such as a formation of biexcitons or exciton-exciton scattering are easily produced compared to the case of flat QW's without the effect of localization. It should be noted that a similar scenario has also been proposed by Kreller *et al.*³⁶ in the $\text{Zn}_x\text{Cd}_{1-x}\text{Se}/\text{ZnSe}$ system.

The energy difference between X_1 and X_2 is about 11 meV at $t_d=100$ ps. The origin of the X_2 being due to inelastic collision of localized exciton, this energy difference should correspond to the binding energy of localized excitons. The exciton binding energy for bulk CdSe and ZnSe is 13 and 18 meV,^{37,38} respectively. We calculated the exciton binding energy (E_{ex}) of the CdSe/ZnSe QW's by the variational method using the trial wave function of the 1S exciton as

$$\psi_{1S} = \exp\left(-\frac{1}{\lambda}\sqrt{x^2+y^2+\eta(z_e-z_h)^2}\right).$$

The result shows that the maximum E_{ex} (about 34 meV) is achieved at the well width (L_w) of 3 ML, and that the E_{ex} is estimated to be 30.5 meV at $L_w=1$ ML. Although analyzing the E_{ex} value in the strongly localized system is dif-

difficult, it is expected that the E_{ex} value will be increased compared to the case of nonlocalization effect, due to the enhanced confinement of wave functions. Thus the process of inelastic collision can be excluded. It is very difficult to estimate theoretically the biexciton binding energy in the system where the penetration of wave functions to the barrier has to be taken into account. Nevertheless, judging from the relationship of decay times between the X_1 and X_2 , we could conclude that the radiative recombination of localized biexcitons is the most probable process for the X_2 .

IV. CONCLUSIONS

Optical properties of localized excitons in a highly strained CdSe QW with about 1-ML thickness have been studied at 20 K by employing time-resolved PL and nonlinear PL spectroscopy under various excitation conditions. The time-integrated PL from the well layer was peaked at 2.7276 eV with the linewidth of 21 meV under low excitation inten-

sity ($0.11 \mu\text{J}/\text{cm}^2$). The lifetime ranged from 200 to 50 ps as the monitored photon energy was changed from the low-energy tail to the high-energy one. These features could be interpreted by the model of exciton localization, which is induced by terraces and islands lying at the interface between CdSe and ZnSe layers.

An emission with the fast decay component was observed at the low energy side of the peak (2.7168 meV) under higher excitation condition. The component was also detected as a superlinear signal in nonlinear PL spectra, and was attributed to a dense excitonic effect such as localized biexcitons.

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