Acoustic driving effect on radiative decays of excitons in ZnSe/ZnS single quantum wells

O. A. Korotchenkov,[a] A. Yamamoto, and T. Goto
Department of Physics, Graduate School of Science, Tohoku University, Sendai 980, Japan

M.-W. Cho and T. Yao
Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980, Japan

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We report that acoustic driving of ZnSe/ZnS quantum wells can yield $\geqslant 30\%$ shortening of the fast component (on the order of 10^2 ps) of exciton radiative decays and relatively enhanced tail emissions at greater instants. The shortening is attributed to the driving-induced relocalization of excitons to lower-energy states while the enhancement is indicative of the drift diffusion of mobile populations in the driving electric field. These results suggest that the radiative recombination predominantly occurs from localized excitons. © *1999 American Institute of Physics.* [S0003-6951(99)02521-8]

The kinetics of excitons in II–VI semiconductor microstructures have been the subject of intense research in recent years due to the prominent features of excitonic effects of both the fundamental and applied relevance.[1–3] Since many aspects of device performance are governed by the dynamics of excess carriers, influence of the exciton kinetics by external parameters is desirable. Application of an electric field perpendicular to the layer of a quantum well (QW) dramatically affects exciton recombination lifetimes[4] and provides a very convenient means of altering the dynamics of photoexcited carriers. The passage of a surface acoustic wave (SAW) through a QW structure can yield μs prolonged recombination lifetimes due to the confinement of spatially separated electron–hole pairs to the moving piezoelectric potential of the SAW,[5] so that the SAW may be used to drag excess carriers that can subsequently recombine. It is the objective of this study to explore the SAW potential in order to affect the fast radiative decays of excitons in QWs taken with picosecond time-resolved luminescence techniques.

The ZnSe single QWs of 3, 5, and 7 monolayer (ML) thickness were sandwiched between ZnS cap and barrier layers grown on a GaAs (001) substrate. The linear dimensions of the samples ranged from 4 to 7 mm. Details of the growth procedure and the sample structure have been previously described.[6,7] An acoustic driving was employed by mounting the sample on a 41° Y-cut, *X*-propagating $LiNbO_3$ delay line,[8] where the SAW was generated by applying ≈ 10 MHz frequency voltage U to the interdigital transducer. The sample-piezoelectric substrate system was acoustically mismatched so that charge carriers in the QW were driven by a piezoelectric field of the SAW which penetrated into the QW sample. The acoustic power was estimated from the radiation conductance of the transducer yielding ≈ 1.1 W/cm at $U = 45$ V.

The optical excitation was produced by picosecond pulses of a wavelength tunable second harmonic light of a Rhodamine 6G dye laser pumped by a mode-locked YAG laser. A repetition rate of the pulses was 76 MHz. The excitation light was focused onto the sample with a spot diameter of no greater than 150 μm and the peak excitation power was less than 120 mW. The data presented here were taken mainly with a 4.0 eV laser light. This energy is above the band gap of ZnSe (2.8 eV) and ZnS (3.8 eV), so a part of the carriers generated in ZnS will then be captured by the ZnSe QW. The samples were immersed in liquid helium. After dispersing the QW photoluminescence (PL) by a subtractive double monochromator, a Hamamatsu streak camera was used, and the instrumental temporal resolution amounted to ≈ 60 ps.

As a point of reference, Fig. 1 shows the time-integrated PL spectra of our samples. Each spectrum consists of a peak arising from the recombination of excitons confined in the QW positioned at about 3.052, 3.097, and 3.138 eV for the 7, 5, and 3 ML wells, respectively. The linewidth increase in the 3 ML QW is most likely a consequence of a large energy fluctuation of localized excitons that results from enhanced heterointerfacial disorder.

Dotted curves in Fig. 2 show the decay curves of the PL all taken at the peak energies of the spectra exhibited in Fig. 1. The observed decays for the 7 and 5 ML QWs [Figs. 2(a) and 2(b), respectively] are described well by fast exponential behaviors for an intensity range of about an order of magni-

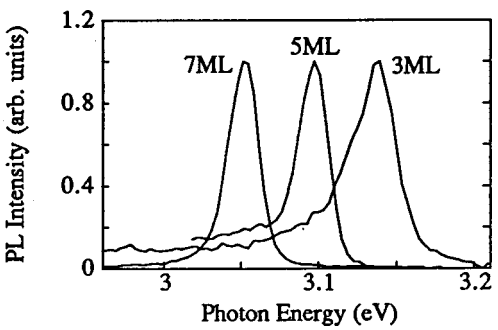

FIG. 1. Comparison of the time-integrated PL spectra of the QWs at 4.2 K, excited at 4.0 eV. The spectra are scaled arbitrarily.

[a]Present address: Faculty of Physics, Kiev University, Kiev 252022, Ukraine; electronic mail: olegk@tower.ups.kiev.ua

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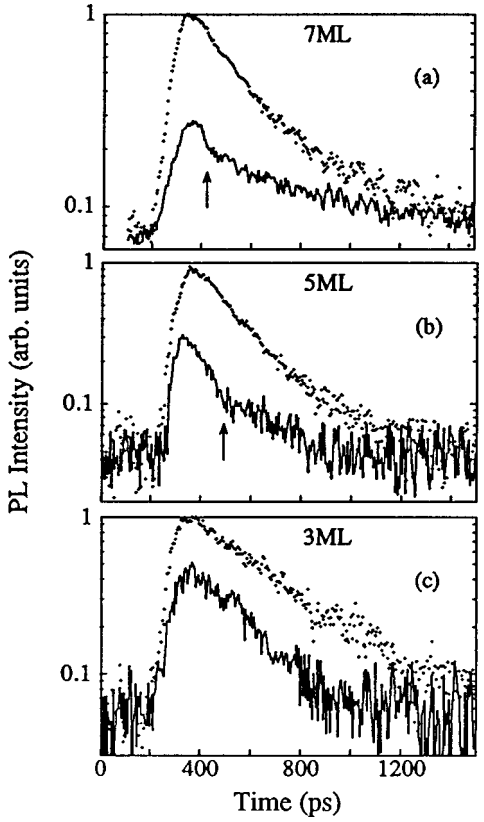

FIG. 2. Transient PL intensities from the QWs at photon energies 3.052, 3.097, and 3.138 eV for the 7 ML (a), 5 ML (b), and 3 ML (c) wells, respectively, following picosecond excitation at 4.000 eV at time $t \approx 200$ ps. The dotted and solid curves correspond to $U=0$ and 45 V, respectively.

tude and exhibit long-lived nonexponential tail emissions which exist on a time scale of nanoseconds, and with an intensity ~ 1 decade lower than the fast PL processes. For the 3 ML QW, the decay curve closely resembles a single exponential over the range from about 400 to 1400 ps in Fig. 2(c).

The time-integrated intensity gradually decreases above some threshold value of the driving amplitude as illustrated by closed circles in Fig. 3 for the 5 ML QW. The changes of the transient PL intensities with the driving are exhibited by solid lines in Fig. 2. Obviously, for increasing driving amplitude the fast-decaying PL becomes much faster accompanied by a relative increase in the long-lived decays at time instants greater than that marked by arrows in Figs. 2(a) and 2(b). From the slopes of the fast-decaying parts of the curves, the fast PL processes in the 7 and 5 ML wells may be quantified by their decay times τ_f, yielding data presented in Fig. 4. Also shown in Fig. 4 are the decay times τ_f in the 3 ML QW fitted to a one-exponential function.

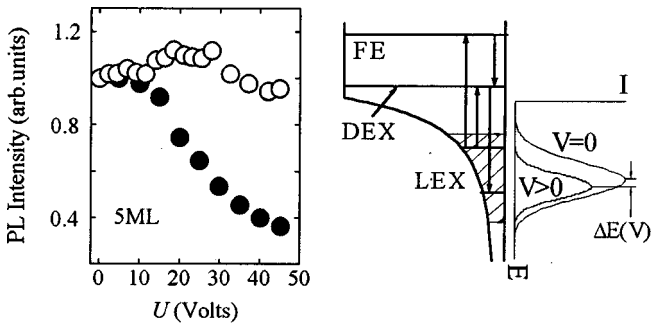

FIG. 3. Time-integrated PL intensities of the 5 ML QW vs driving amplitude excited at 4.0 eV (closed circles) and 3.5 eV (open circles). The levels of localized (LEX), delocalized (DEX) excitons, and free electrons (FE) are sketched in the potential well shape approximated by a QW background and a trapping potential well. The resulting PL spectra $I(E)$ are centred around the energy of a nominally deep localized state. The hatched area illustrates the distribution of localized states in the well.

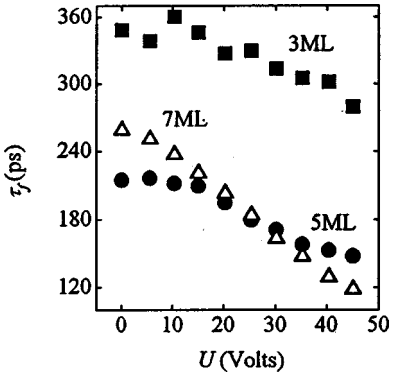

FIG. 4. Exciton decay times τ_f vs driving amplitude for the 3, 5, and 7 ML QWs.

The quenching of the PL may be due to ionization of excitons by the driving electric field. It is known that a field of about $F_i = E_x/ea_B$, where E_x, a_B, and e are, respectively, the exciton binding energy, effective Bohr radius, and electron charge, can ionize an exciton. In a three-dimensional medium, $E_x^{3D} \approx 15$ meV and $a_B^{3D} \approx 60$ Å, thus giving $F_i^{3D} \approx 2.5 \times 10^4$ V/cm. To obtain an upper bound of ionization fields in a QW system, we supposed $E_x^{2D} \approx 2E_x^{3D}$ and $a_B^{2D} \approx 0.6a_B^{3D}$. Then $F_i^{2D} \approx 8 \times 10^4$ V/cm.

The SAW electric field amplitude is $F_{\mathrm{SAW}} = k\varphi_0$, with k the wave number of the SAW and φ_0 the amplitude of the electric potential produced by the SAW which can be obtained from earlier studies.[9] We find $F_{\mathrm{SAW}} \approx 6 \times 10^3$ V/cm at $U = 45$ V. This is clearly too small to be consistent with the picture of direct ionization of excitons in electric fields of the SAW.

The impact ionization threshold, determined by $m^* \mu^2 F_{\mathrm{impact}}^2 = 2E_x^{2D}$ (μ and m^* being, respectively, the electron mobility and interband effective mass), is found to be $\approx 3.8 \times 10^3$ V/cm. The threshold field $F_{\mathrm{SAW}} \approx 1.2 \times 10^3$ V/cm at $U = 10$ V determined in the 5 ML QW (closed circles in Fig. 3) is in fair agreement with F_{impact}. Therefore, the assumption here is that impact ionization dominates the PL quenching. Also, the supporting evidence is that the quenching is substantially suppressed by exciting PL with a 3.5 eV laser light (open circles in Fig. 3). As this energy is well below the band gap of ZnS, the excess free carrier density in a thin QW is remarkably decreased, thus suppressing the sweeping out of carriers by the driving field.

The data of Fig. 3 clearly exclude possible thermal effects of the driving. Indeed, the thermal quenching is expected to be independent of the energy of excited light which is not the case in Fig. 3. Moreover, for a radiative recombination dominant system, an increase in the radiative lifetime with temperature is expected for the QW excitons, as has been verified previously in a separate measurement of the 5

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ML QW.[7] In marked contrast, as shown in Fig. 4, acoustic driving produces primarily a τ_f decrease.

Based on the premises above, the following scenario applies:

(1) In the absence of the driving, the PL line is composed of the components arising from excitons with various lateral sizes. The radiative decay time τ_R is of the form[10]

$$\tau_R \propto \frac{a_B^2}{L^2} \propto \frac{1}{E_x^{2D} L^2}, \tag{1}$$

where the coherence length of the exciton L is determined by lateral dimensions of the randomly distributed interface localized states. The 3 ML QW has a much higher density of interface defects compared to that in the 5 and 7 ML wells. Then the τ_R value in Eq. (1) is replaced by a longer lifetime determined by a smaller L, and this is manifested by enhanced τ_f in the 3 ML QW (Figs. 2 and 4).

(2) As U is increased the localized excitons may be detrapped into the mobile states or spatially separated individual electrons and holes (upward arrows in the sketch of Fig. 3) by the mechanism discussed above.

(3) These mobile populations are then free to diffuse and to drift in the moving electric field of the SAW, so they may become trapped at nonradiative defects or relocalized in the lower-energy states (downward arrows in Fig. 3) with a subsequent increase in the binding energy E_x^{2D} in Eq. (1). Physically, the τ_f decrease may be partitioned into the trapping and relocalization components. However, it is unlikely that nonradiative traps dominate the effect on τ_f. Indeed, the 3 ML QW has the highest density of interface defects as opposed to the smallest change of τ_f in the 3 ML QW; see Fig. 4. Meanwhile, because of a poorer interface quality, the relocalization of excitons to the lower-energy sites by the drift-diffusion motion might have not been important in a thin QW, and this is supported by the absence of a nonexponential tail PL in Fig. 2(c). In contrast, the driven-induced drift diffusion in the 7 and 5 ML QWs is illustrated by enhanced nonexponential decays in Figs. 2(a) and 2(b).

(4) Significantly, the line shapes of the spectra in Fig. 1 remained almost independent of U, while their peak positions were redshifted $\Delta E \leqslant 10$ meV with applying the driving, i.e., the spectral changes sketched in Fig. 3 were observed. We therefore conclude that the radiative recombination predominantly occurs from localized excitons, and the PL spectra are indicative of the distribution of localized states. In the 3 and 5 ML QWs, the magnitude of the shift ΔE is quantitatively consistent with the τ_f decrease (Fig. 4) anticipated from Eq. (1), whereas the reduction in τ_f is somewhat underestimated in the 7 ML well. Whether this is due to nonradiative processes remains to be seen.

To conclude, the radiative decays in ZnSe QWs may be effectively varied by acoustic driving. It is argued that $\sim$30% shortening of the fast-decaying component is due to relocalization of excitons to lower-energy states in the driving electric field, while relatively enhanced tail decays correspond to the drift-diffusion motion of delocalized populations imposed by the driving. The data suggest that the radiative recombination predominantly occurs from localized excitons.

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