Exciton Dynamics and Carrier Tunneling Processes in $Zn_{1-x}Cd_xSe$/ZnSe Asymmetric Double Quantum Wells

J. N. Zeng, I. Souma, Y. Amemiya and Y. Oka

Research Institute for Scientific Measurements, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-77, JAPAN

(Received: Feb. 3, 1997 Accepted: Feb. 20, 1997)

Abstract

We report dynamics of excitons and carriers in $Zn_{1-x}Cd_xSe$/ZnSe asymmetric double quantum wells by time-resolved photoluminescence studies. Tunneling times of electrons and holes through ZnSe barriers have been determined as a function of the barrier width by comparing the temporal characteristics of exciton luminescence with the solutions of the carrier population rate equations. The stimulated emission process in this system under high excitation density has also been studied. The stimulation is affected by the tunneling process.

1. Introduction

Blue-light emissions from wide band-gap quantum wells (QWs) of $Zn_{1-x}Cd_xSe$/ZnSe are the recent stimulating subjects on the application for short-wavelength semiconductor optical devices. On the other hand perpendicular transports of photoexcited carriers in superlattices and QWs have provided lots of physical phenomena such as Bloch oscillations, optical Stark effects and bistabilties and so on. An asymmetric double quantum well (ADQW) consisting of a couple of a narrow well (NW) and a wide well (WW) is the simplest system to study the perpendicular carrier dynamics. The tunneling process in ADQWs of the $Zn_{1-x}Cd_xSe$/ZnSe system will be, therefore, applicable to fast response devices in the blue light region.

In this paper, we study the carrier tunneling processes in the $Zn_{1-x}Cd_xSe$ /ZnSe ADQWs with various barrier widths by using the time resolved photoluminescence (TRPL).[1,2] We measure the time behavior of the luminescence of the NW and WW excitons in ADQW samples prepared by epitaxy methods and compare with a semiclassical model of tunneling for excitons and electron-holes. Stimulated emission processes in the WW under high optical excitation have also been studied and its relationship with the tunneling process is discussed.

2. Experimental

$Zn_{1-x}Cd_xSe$/ZnSe ADQW samples were prepared by the hot wall epitaxy or molecular beam epitaxy methods on GaAs (100) substrates. The ADQWs consist of two QWs of different well widths which is separated by the ZnSe barrier. The thicknesses of NW and WW (L_{NW} and L_{WW}) are 14 and 40 Å, respectively and the barrier thickness (L_b) is varied from 270Å to 34 Å. The Cd content x in $Zn_{1-x}Cd_xSe$ wells is 0.33. In the samples, 10 periods of the ADQW were synthesized in order to obtain higher PL intensities. Each ADQW is separated from the neighboring wells by the ZnSe isolation barriers of 400 Å. The widths of a ZnSe buffer and a cap layer are 0.5~1μm and 2000 Å, respectively in all samples.

PL and TRPL measurements were performed for the samples immersed in liquid helium. The excitation light source was the second harmonic of a Ti : sapphire mode-locked laser with a repetition rate of 76 MHz and 200 fs pulse duration. The excitation energy (3.2 eV) is much higher than the gap energy of the ZnSe barrier, which provides the injection of photo-generated carriers from the thick isolation barriers into NW and WW. TRPL was measured by using the time-correlated single-photon-counting method. The photoluminescence excitation (PLE) spectra were also measured by monitoring the WW exciton PL intensity.

3. Results and Discussion

PL spectra of three ADQW samples with L_b of 270 Å, 160 Å, 45 Å are shown in Fig. 1. The recombination luminescence of excitons located in the WW and NW can be seen at 2.52 and 2.66 eV. As L_b decreases, the PL intensity from the NW decreases dramatically, while the increase of PL intensity from the WW arises. The variation of intensities in the NW and WW indicates the occurrence of the carrier transfer from NW to WW with decreasing L_b, which is the process to exhaust the population of carriers and excitons in the NW. The tunneling of electron and hole or of excitons from NW to WW leads to a decrease of the NW exciton lifetime, while the population of a pair of electron-hole or an exciton contributes to the increase of the WW exciton luminescence. Unpaired electrons or holes in WW can not

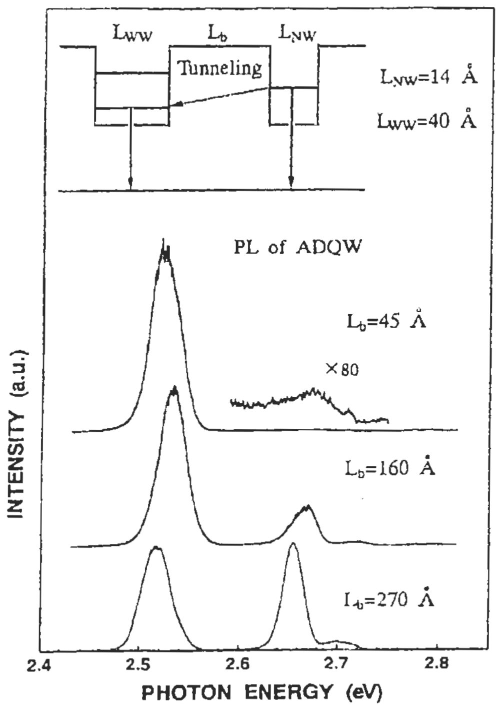

Fig. 1 Photoluminescence spectra of the ADQW samples.

contribute to the PL in WW. Therefore the enhancement of PL intensity from the WW with decreasing barriers indicates the tunneling of electron-hole pairs or that of excitons.

Fig. 2 displays the PL and PLE spectra for the samples with L_b = 34 and 270 Å, respectively. The PLE spectrum for the WW exciton luminescence shows different behavior in the samples with L_b = 34 and 270 Å. In the case of the sample of 270 Å -barrier (Fig. 2b), no PLE peak is observed corresponding to the NW exciton PL peak. This result shows that the NW excitons do not contribute to the emission from the WW, which indicates the lack of tunneling of the NW excitons through the barrier. On the other hand, in the case of the sample with 34 Å -barrier, the PLE peak appearing at the

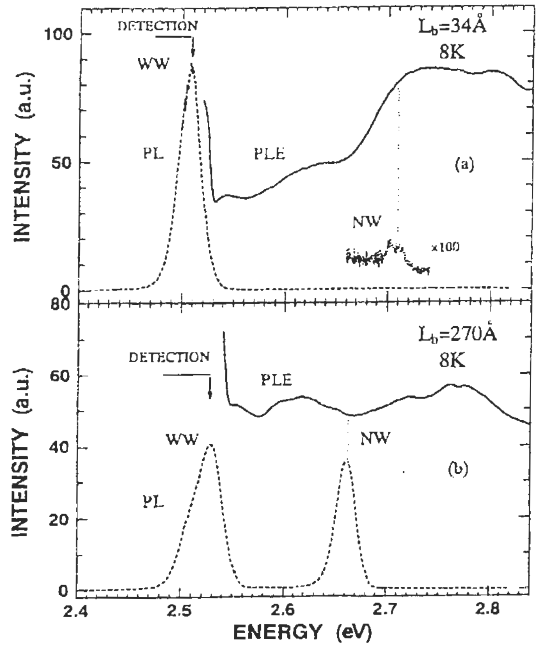

Fig. 2 PL and PLE spectra of the ADQW samples.

NW exciton energy implies that the NW excitons directly contribute to the WW exciton PL, which is due to the tunneling of excitons or pairs of electron and hole[3].

Fig. 3 shows the result of the temporal behavior of the PL peak intensities of NW and WW for 34 and 160 Å -barrier samples. Circles and squares represent temporal intensity changes for the WW and NW luminescence, respectively. The solid curves are the results of calculation by using solutions of the population rate equations for the excitons. Simple rate equations are introduced to evaluate the temporal variation of the PL intensities of the NW and WW, as follows:

$$\frac{dn_0(t)}{dt} = -\frac{n_0(t)}{\tau_0} - \frac{n_0(t)}{\tau_1} - \frac{n_0(t)}{\tau_2} \tag{1}$$

$$\frac{dn_1(t)}{dt} = \frac{n_0(t)}{\tau_1} - \frac{n_1(t)}{\tau_{NW}} - \frac{n_1(t)}{\tau_t} \tag{2}$$

$$\frac{dn_2(t)}{dt} = \frac{n_0(t)}{\tau_2} + \frac{n_1(t)}{\tau_t} - \frac{n_2(t)}{\tau_{WW}} \tag{3}$$

where n_0, n_1, n_2 are the exciton population densities in the ZnSe barriers, NW and WW, respectively and τ_t is the tunneling time of excitons from NW to WW. τ_0, τ_{NW}, τ_{WW} are the lifetimes of excitons in ZnSe barriers, NW and WW, respectively. τ_1^{-1} and τ_2^{-1} represent the transfer rates of excitons from ZnSe barriers to NW and WW. For the sample with L_b=270 Å, almost same PL intensities from NW and WW were observed as shown in Fig. 1. Since the barrier between the NW and WW is thick enough to localize excitons in the two wells independently. The lifetimes of the NW and WW are measured to be 240 ps and 270 ps. These lifetimes are used for fitting of the decays in thinner L_b samples. From the changes of the decay times of NW and WW in the ADQW samples, the tunneling time is determined by the best fitting. The result of the thick barrier width sample of 160 Å (at the lower portion of Fig. 3.) indicates that the lifetime and intensity of the NW excitons are similar to those of the WW excitons and the tunneling time is the order of 10^6 ps. As L_b decreases up to 34 Å (at upper portion of Fig. 3), the slope of the decay curve of NW declines markedly and its intensity also reduces by 2 orders. The tunneling time is determined as 25 ps, which is the carrier tunneling through the barrier.

In order to compare the obtained results with calculation, we use a semiclassical model for computing the tunneling times for excitons, electrons and holes. A model of electron tunneling gives following tunneling time:

$$\tau_t = L_{NW} \frac{[V_0 + (r-1)E]^2}{16rE(V_0 - E)} \sqrt{\frac{2m_W}{E}} \exp\left\{2L_b \sqrt{\frac{2m_b(V_0 - E)}{h^2}}\right\} \quad (4)$$

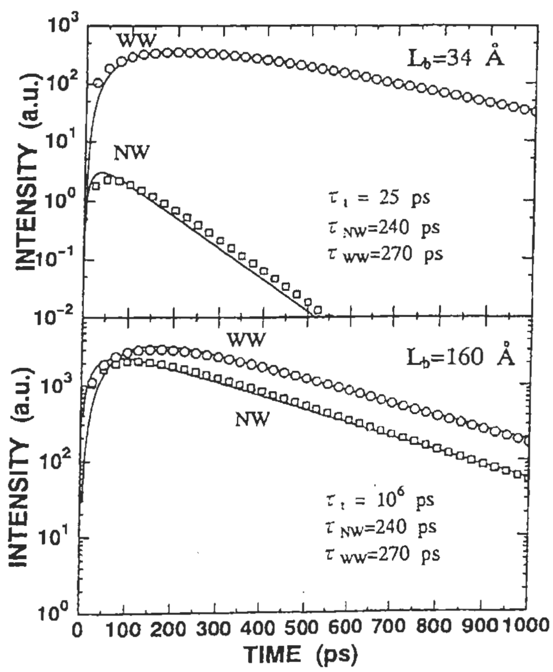

Fig. 3 Time variations of the WW and NW exciton luminescence

where E and V_0 are the electron energy and potential height. r is the ratio of m_w and m_b, which are the electron effective masses of well and barrier. τ_t changes exponentially with L_b. For the excitonic tunneling, the eq. (4) should be modified by using the reduced mass m^* of the exciton in place of m_w and m_b, and the energies of excitons in NW and ZnSe barriers in place of E and V_0. We assume r = 1 here. Therefore following expression is obtained for exciton tunneling:

$$\tau_t = L_{NW} \frac{V_0^2}{16E(V_0 - E)} \sqrt{\frac{2m^*}{E}} \exp\left\{2L_b \sqrt{\frac{2m^*(V_0 - E)}{h^2}}\right\} \quad (5)$$

Fig. 4 shows the dependence of tunneling times on barrier thickness, where double circles indicate experimental tunneling times. Solid, Dotted, and dashed lines represent the calculated results of the semiclassical model, corresponding to the dependence of exciton, electron and hole, respectively. The difference of the tunneling time for electrons and holes is about 1 order for the thin barrier, while, for the thicker barrier, the difference becomes larger. The discrepancy arises from the different effective masses of electron and hole. The tunneling time of exciton given by eq. (5) locates in the middle of the electron and hole tunneling time, which indicates the faster tunneling of the exciton than the hole. Here the exciton reduced mass, which is smaller than the effective masses of electron and hole, can

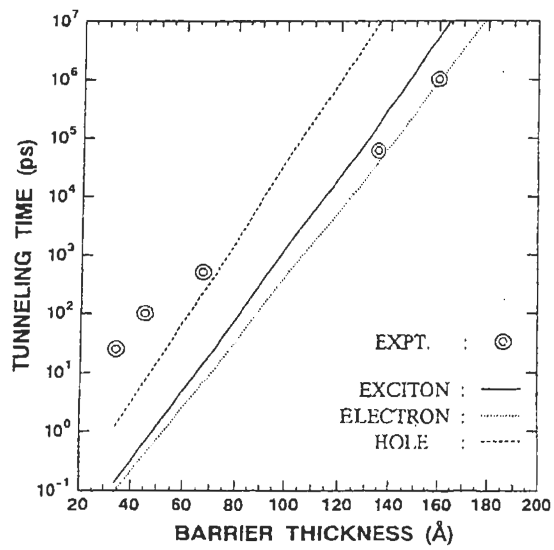

Fig. 4 Tunneling times of carriers and excitons as a function of the barrier thickness

induce slower tunneling of excitons than that of the electrons but faster tunneling than the holes. This fact means that an electron penefrating into the barrier assists the hole tunneling by the Coulomb interaction. The L_b dependence of tunneling time of the present results shows a slower gradient than the calculation.

In the semiclassical model, the energy difference of two levels is assumed to be zero. However, a mismatch of the exciton ground state energy for NW and WW actually exists as 140 meV. Therefore, in the nonresonant condition, the energy-relaxation by phonons between two ADQW levels partly affects the tunneling rate of the carriers.[2,6] The scattering of carriers by LO and acoustic phonons in the ZnSe system at 4.2 K are 2 ps and about the order of 100 ps, respectively. The phonon assisted tunneling process has to be taken into account to compare the theoretical tunneling times with experimental values. The fluctuation of the ADQW interface also affects the dependence of tunneling time.

Stimulated emission effect[4,5] under high optical excitation has also been investigated in this system. Fig. 5 shows the emission spectra

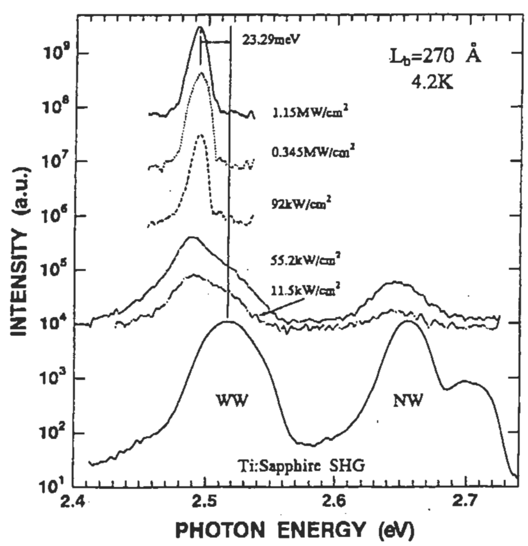

Fig. 5 Stimulated emission spectra in the ADQW

of 270 Å -barrier ADQW sample for various optical excitation densities. When the excitation intensity reaches to 11.5 kW/cm^2, a stimulated emission peak is observed in lower energy side of the WW exciton band with a shift of 23 meV. The spectrally integrated intensity changes quadratically with the excitation intensity, which is ascribed to the exciton-exciton collision process. With decreasing the barrier width, the low threshold excitation intensity and high values of superlinear dependence are realized, which are due to the tunneling process of carriers in the ADQWs.

4. Summary

We have investigated the tunneling processes in $Zn_{1-x}Cd_xSe/ZnSe$ ADQWs with different barrier widths by TRPL measurements. Present experimental results shows the predominant tunneling of electron-hole pairs in the thinner barrier sample and the tunneling time varies from 10^6 ps to 25 ps for the change of barriers from 160 Å to 34 Å. Stimulated emission effect under high optical excitation has also been investigated in this system. With decreasing the barrier width, the tunneling process induces the lower threshold level for the stimulations.

References

1. K. Hieke, W. Heimbrodt, I. Lawrence, W. W. Ruhle, Th. Pier, H, -E. Gumlich, D. E. Ashenford and B. Lunn Solid State Commun. **93**, 257 (1995)
2. B. Deveaud, A. Chomette, R. Ferreira and G. Bastard, Phys. Rev. B **42**, 7021 (1990)
3. F. C. Michl, R. Winkler and U. Rossler, Solid State Commun. **99**, 13 (1996)
   J. Y. Jen, T. Tsutsumi, I. Souma and Y. Oka, Jpn. J. Appl. Phys. **32**, suppl.32-3, 728 (1993)
4. Y. Jen, T. Tsutsumi, I. Souma and Y. Oka, Jpn. J. Appl. Phys. **32**, L1542 (1993)
5. R. Ferreira and G. Bastard, Phys. Rev. B **40**, 1074 (1989)
   S. Ten, F. Henneberger, M. Rabe, N. Peyghambarian, Phys. Rev. B **53**, 12637 (1996)