Influence of the miniband on emission mechanism in $Zn_{1-x}Cd_xSe/ZnSe$ quantum wells

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The stimulated emission has been investigated in $Zn_{1-x}Cd_xSe/ZnSe$ quantum wells with a constant sample width of 60 Å and barrier widths from 47 Å to 500 Å. A redshift of the stimulated emission peak with increasing excitation intensity was observed in samples with barrier widths narrower than 350 Å, and the stimulated emission mechanism in these samples is found to be dominated by exciton-exciton scattering. In contrast, no redshifts were observed for the sample with a barrier width of 500 Å, and the stimulated emission in this sample appears to be dominated by biexcitons. Time-resolved spectroscopy measurements were also performed on samples with barrier width of 47 and 500 Å below the threshold of stimulated emission, and it was found that the exciton lifetime decreases with increasing exciton energy for both of the samples. With a ten times smaller barrier width in the 47 Å sample, the lifetime of high energy exciton is significantly shorter than that of the other sample, while the low energy lifetime becomes longer. This implies that with a narrower barrier width of the quantum wells the excitons move faster to lower energy regions. The narrower the barrier, the easier it is for excitons to move through the miniband. This freedom of movement increases the exciton concentration at low energies in localized areas in the wells. High exciton concentration made exciton-exciton scattering more probable and reduce the stimulated emission threshold value. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2769791]

I. INTRODUCTION

The semiconductor laser emitting blue-green light was first produced by Haase *et al.* in 1991 with $Zn_{1-x}Cd_xSe/ZnSe$ quantum wells (QWs).[1] Since then, this system has been studied extensively,[2–4] because of the possibility of application for high density optical recording, solar cells, optical communications as well as full-color displays. However, the optical gain in the $Zn_{1-x}Cd_xSe/ZnSe$ system is usually lower than that in the $GaAs/Ga_{1-x}Al_xAs$ system due to the shorter radiative lifetime resulting from the larger energy gap.[5] Consequently, it requires a larger current for lasing, which induces defect creation. Therefore, for the $Zn_{1-x}Cd_xSe/ZnSe$ system, it is difficult to obtain cw operation for a long time at room temperature.

It is well known that at room temperature the lasing mechanism in $GaAs/Ga_{1-x}Al_xAs$ heterostructures is dominated by the electron-hole plasma (EHP) recombination. In *II–VI* bulk semiconductors, several mechanisms have been proposed for the stimulated emission effect of the band edge luminescence, such as recombination due to exciton-electron scattering,[6,7] excitonic molecule recombination,[8] exciton-exciton scattering,[9,10] EHP recombination,[11,12] and exciton-longitudinal optical (LO) phonon scattering.[13] Ding *et al.* have interpreted the stimulated emission process of the excitons in $Zn_{1-x}Cd_xSe/ZnSe$ multiple quantum wells (MQWs) with a 500 Å barrier and a 60 Å well by the phase space filling (PSF) effect.[14,15] Kawakami *et al.* have reported an exciton-LO phonon interaction mechanism[16] in superlattices with a constant barrier width (L_b) of 80 Å and well widths (L_w) varying from 15 Å to 120 Å; Kreller *et al.*[17] and Kozlov *et al.*[18] have reported that the stimulated emission in a single quantum well is dominated by the biexcitons.

We have reported the exciton-exciton scattering mechanism[19] and electron-hole plasma recombination[20] in *II–VI* superlattices at low temperatures. We also proposed that the stimulated emission mechanism in *II–VI* semiconductor is dominated by the electron-hole plasma recombination at room temperature.[21,22] We notice that Ding *et al.*[14] and Kreller *et al.*[17] used samples with barrier width of 500 Å and 800 Å in the experiment, respectively, which are wider than our barrier widths (47 Å–120 Å). To obtain a unified picture of the stimulated emission processes in the $Zn_{1-x}Cd_xSe/ZnSe$ MQWs, the stimulated emission process was observed from samples with the same well widths while barrier widths is changed from 47 Å to 500 Å. Moreover, Ding *et al.* and Kreller *et al.* used excitation intensities (I_{ex}) up to only a few kW/cm^2, while our I_{ex} ranged had up to several MW/cm^2. Further, we investigated time-resolved spectroscopy measurements in order to clarify the lasing mechanism of blue emission. As a result, we found that the intrawell migration rate for all samples is equivalent because the well width for all samples is the same. However, the

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interwell migration probability is decreased when the barrier width increases, and there is almost no interwell migration in the sample with barrier width of 500 Å.

II. DISCUSSION

MQWs samples of the $Zn_{1-x}Cd_xSe/ZnSe$ system were grown on [100] GaAs substrates by the hot wall epitaxy method. Samples were composed of 50 periods of $Zn_{1-x}Cd_xSe/ZnSe$ ($x=0.35$) layers with well width are fixed at 60 Å and barrier widths are changed from 47 Å to 500 Å. The concentration of Cd (x) was confirmed by x-ray diffraction of the $Zn_{1-x}Cd_xSe$ crystal. The value of x from the x-ray diffraction was consistent with the calculated result from the peak energy of photoluminescence. We grew a single crystal of $Zn_{1-x}Cd_xSe$ after every 3–4 samples of superlattice to make sure that the concentration of Cd was unchanged.

The stimulated emission process has been studied in MQWs samples using the Shaklee method:[23] In order to get a longer lasing cavity, corresponding to the length of our laser line (~3 mm), we used the edge emission method for our analysis. The samples were excited by a pulsed nitrogen laser beam, which had a rectangular shape (0.1×3 mm^2) and was normal to the surface. Stimulated emission in a direction parallel to the surface was detected at a cleaved edge of the sample. The light was directed through a monochromator and then detected by a photomultiplier. The signal from the photomultiplier was analyzed by a boxcar integrator and recorded by the computer.

The time-resolved spectra were measured by a multichannel photomultiplier. The pumping source was the second harmonic (395 nm) of a Ti-Sapphire laser. The emission signal from the surface of the sample was focussed into a monochromator then detected by the photomultiplier. The signal from the photomultiplier was preamplified before entering a discriminator. A delayed reference signal from the laser was compared with the signal from the discriminator. The signal difference was introduced into a time-to-amplitude converter, then to a multichannel analyzer, and recorded in a computer. The system had a time resolution of about 30 ps.

Figure 1(b) shows the stimulated emission spectra at different I_{ex} from a sample with $L_w=57$ Å and $L_b=47$ Å. The X band represents the recombination of heavy hole excitons. A band we call P appears on the low energy shoulder of the spontaneous emission band (X band). Since the emission intensity (I_{em}) of the P band shows a superlinear dependence ($I_{em}\propto I_{ex}^{2.3}$) (Ref. 22) on the excitation I_{ex}, we attribute this band to the stimulated emission. As the I_{ex} increases further, the exciton density becomes higher than the Mott transition density (5.1×10^{12} cm^{-2});[15] a new emission band appears at about the same energy as the P band and shifts to the lower energy side with increasing I_{ex}. This band, which we call the N band, can be distinguished from the P band by the dependence of its I_{em} on I_{ex}, linearlike spontaneous emission at low excitation.[22] Due to the redshift, the constant FWHM, the superlinear relation for I_{em} with I_{ex}, and the fact that P bands can be well fitted by the exciton-exciton scattering model,[20] we ascribe P bands to exciton-exciton scattering. We attribute the N band, which is observed at high I_{ex}, to spontaneous emission. Because of the broadening of the FWHM of this band with I_{ex}, the redshift, the linear dependence of I_{em} on I_{ex}, and the fact that N bands can be well fitted by the electron-hole plasma recombination[21] model, we conclude that N bands are dominated by electron-hole plasma recombination. Figure 1(a) shows the stimulated emission spectra in a sample with $L_w=60$ Å and $L_b=500$ Å. Band B appears on the low energy shoulder of the spontaneous emission band (X band). Since I_{em} shows a superlinear relation to the $I_{ex}(I_{em}\propto I_{ex}^{1.65})$, we also ascribe this band to stimulated emission. However, the peak energy of emission bands at different I_{ex} remains unchanged. This indicates that the stimulated emission mechanism in this sample is different from the mechanism in the sample shown in Fig. 1(b).

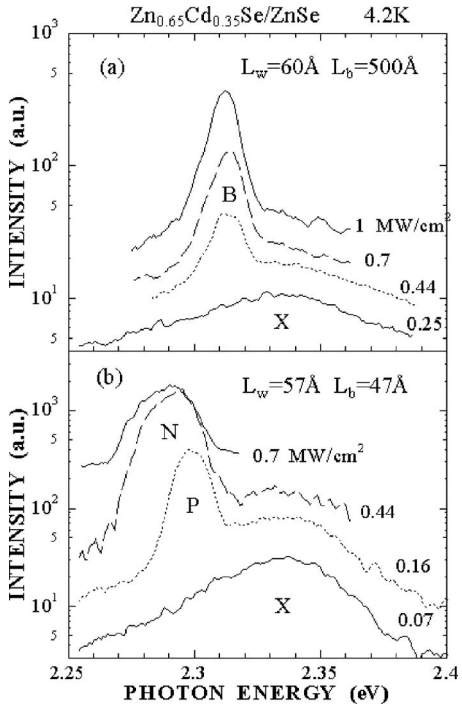

FIG. 1. Stimulated emission spectra for the sample of $Zn_{0.65}Cd_{0.35}Se/ZnSe$ MQWs with (a): $L_w=60$ Å and $L_b=500$ Å and (b): $L_w=57$ Å and $L_b=47$ Å at different excitation intensities.

Stimulated emission spectra was observed in samples with $L_w=60$ Å and $L_b=47$ Å, 89 Å, 170 Å, 350 Å, and 500 Å. The redshift of stimulated emission bands for these samples is shown in Fig. 2, where the redshift represents the energy difference between the P band which just occurred and disappeared at different I_{ex}. The redshift increases as L_b decreases. The energy shifts for samples with $L_b=47$ Å and 89 Å were significantly larger than those compared to the other samples. We suggest that the redshift is related to the miniband width since the miniband becomes very narrow when the barrier width is larger than 150 Å. In fact, the energy shift of the sample with $L_b=500$ Å, having negligible miniband width, is close to zero.

The time-resolution spectra for two samples with $L_b=47$ and 500 Å have been taken at 4.2 K. Figure 3(a) shows the result for the sample with $L_w=60$ Å and $L_b=500$ Å. The circle indicates the lifetime of excitons at different energies

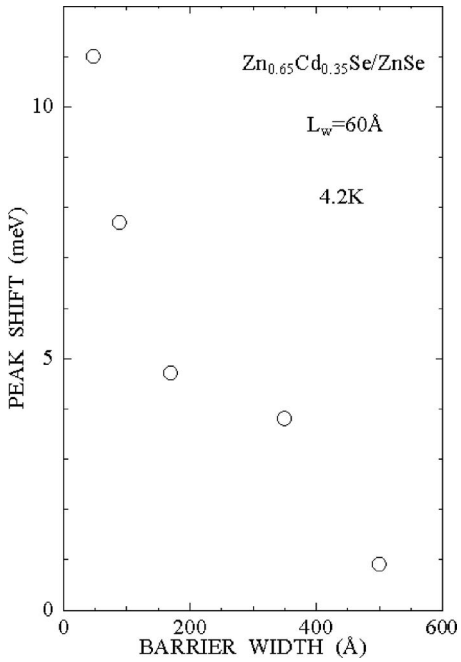

FIG. 2. Redshifts are shown as a function of barrier widths for samples of L_w=60 Å and L_b=47, 89, 170, 350, and 500 Å.

calculated by a deconvolution method from the time-resolution spectra. The lifetime of excitons on the high energy side of the emission band is about 200 ps; then it gradually increases to 400 ps on the low energy side of the emission band. This phenomenon suggests the localization of excitons caused by the inhomogeneity of the alloy concentration and fluctuations of the order of a monolayer at the interfaces. Figure 3(b) shows the lifetimes of excitons for the sample with L_w=60 Å and L_b=47 Å. The lifetime of excitons on the high energy side of the emission band is about 100 ps (which is shorter than the lifetime of excitons in the sample with L_b=500 Å). On the other hand, the lifetime of excitons increases to 500 ps at low energies of the emission band; this is longer than the lifetime of excitons in the sample with L_b=500 Å.

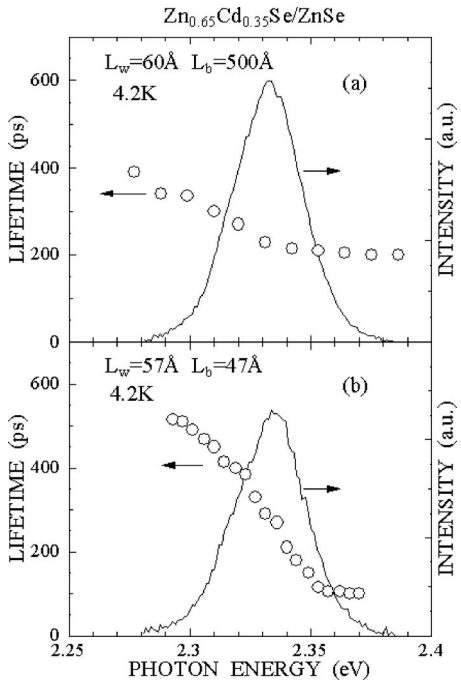

FIG. 3. Lifetime of excitons (circle) are shown as a function of the photon energy for the sample of (a): L_w=60 Å and L_b=500 Å and (b): L_w=57 Å and L_b=47 Å. The solid curve represents the spontaneous mission spectra.

We use the exciton-exciton scattering model to explain the redshift of the *P* band. In the superlattice, an inhomogeneous energy level will be formed by the alloy disorder in the well and variations in the well width due to monolayer fluctuations at the interfaces. Since the exciton energy is higher for narrower wells and lower for wider wells, the exciton energy fluctuates in the nonuniform well layer. Alloy disorder makes the alloy concentration vary in a well causing additional fluctuations of the exciton energy. The entire fluctuation of the exciton energy in the superlattice is called the localization energy. For narrower wells, the localization energy is dominated by the monolayer fluctuations while alloy disorder dominates the fluctuation of the exciton energy in the wider wells.[24] Excitons can be localized in this inhomogeneous energy level. For an inhomogeneous volume greater than that of the exciton, the exciton will move inside the localization region, and the exciton-exciton scattering can occur if the exciton density is high. Under these conditions phase space filling is relevant; that is after two excitons are scattered, the first exciton will jump to a higher, unoccupied localization energy level, and the other exciton will lose the energy gained by the first exciton and emit a photon. At high exciton densities, the first exciton can jump only to a higher localization energy level in accordance with the limitation of the PSF effect. Thus the photoluminescence excitation spectrum, as a result of exciton-exciton scattering, shows a blueshift.[16] Therefore, as the I_{ex} becomes stronger, the exciton will emit from the lower energy.

In Fig. 1(b), the *P* band and the *N* band from the sample with L_b=47 Å also are dominated by exciton-exciton scattering and electron-hole plasma recombination, respectively. The emission mechanism for the sample in Fig. 1(a) is different from that in Fig. 1(b). Although I_{em} of the *B* band in Fig. 1(a) shows a superlinear relation to I_{ex}, the relation for I_{em} with $I_{ex}(I_{em} \propto I_{ex}^{1.65})$ is different from both that of the *P* band $(I_{em} \propto I_{ex}^{2.3})$ and the *N* band $(I_{em} \propto I_{ex}^{1.0})$, and the peak energy of the stimulated emission band shows no redshift. We postulate that the emission mechanism of *B* bands in the sample with L_b=500 Å is dominated by the biexciton. Since the 1.65 power dependence of I_{ex} is consistent with the result of the biexciton emission.[25,26] The peak energy of the *B* band (stimulated emission) in Fig. 1(a) is shifted by 17 meV with respect to the peak energy of the *X* band (spontaneous emission). We assign this energy difference to the biexciton binding energy, which is between the biexciton binding energy of 23 meV measured by Yamada[27] and 11 meV from Yamaguchi.[28]

Kreller *et al.*[29] reports that the absorption of the biexciton also shows a blueshift, which indicates that biexcitons in the quantum well also obey the PSF effect. According to the PSF effect, the lower energy levels in the localization area are gradually occupied by biexcitons, and additional biexcitons can only lie at higher energy levels. However, the dis-

sociation of biexcitons takes place at the lowest energy of the localization area. When biexcitons are dissociated, the energy difference from stimulated emission band to the emission band of the exciton equals the biexciton binding energy. Thus, the stimulated emission of biexcitons shows a constant peak energy as I_{ex} increases.

The emission mechanism is different between samples in Figs. 1(a) and 1(b). To verify how the mechanism changed from the exciton-exciton scattering to the biexction dissociation with variation of L_b, we present the redshift of P bands vs L_b in Fig. 2. The energy shift of the stimulated emission band is large when L_b is small; it means that excitons can move through the miniband into the neighboring wells. This allows the exciton to migrate into regions with more inhomogeneous energy levels. Then, at the lower energy level in the localization area, excitons are collected from higher energy areas in neighboring wells. Consequently, the density of excitons increases at the lower energy levels in the localization area. The probability of excitons move through the miniband also makes the localization energy broader in the inhomogeneous area. A high density of the excitons makes the scattering between excitons more probable and the biexciton is difficult to occur because of the screening effect between electrons and holes. At a much higher I_{ex}, one exciton thus can shift to a much higher energy level, and the other exciton will emit from a much lower energy level.[20] Therefore, the redshift is large.

In the sample with $L_b > 150$ Å, the tunneling of excitons is weak because the miniband width is very small. Only a few excitons can migrate to the inhomogeneous energy levels in neighboring wells, and, as a consequence, the energy width of occupied states is much narrower than that in the sample with $L_b=47$ Å. Therefore, the redshift is much smaller than that in the sample with $L_b=47$ Å. In the sample with $L_b=500$ Å, the width of the miniband is close to zero, thus no tunneling effect occurs, and excitons are confined in individual wells. Since there is no tunneling effect, excitons are unable to collect in the lowest energy regions produced by inhomogeneity. Consequently, the density of excitons is not as high as in the narrow well samples. Thus the screening effect between electrons and holes is not strong enough to separate biexcitons at this intense I_{ex}.

The above considerations can be verified by the results of time-resolution spectra as shown in Figs. 3(a) and 3(b). In the superlattice, the photoluminescence occurs at the low energy of the absorption band. After excitation, excitons will form and migrate to the localization area in a time of several hundred femtoseconds. At that, excitons will recombine and continue to relax to lower energy levels. In Fig. 3(a), the lifetime gradually becomes longer on the low energy side of the emission band. Comparing the lifetimes in Figs. 3(b) and 3(a), we find that on the low energy side the lifetime in Fig. 3(b) is much longer than Fig. 3(a). However, on the high energy side the lifetime in Fig. 3(b) is shorter than in Fig. 3(a). In Fig. 3(b), the migration effect increases the density of excitons at low energies, which raises the lifetime of excitons at the low energy side of the emission band and decreases the exciton density at the high energy side. Therefore, the lifetime of excitons on the low energy side of the emission band in Fig. 3(b) is larger than that in Fig. 3(a). Migration through the miniband increases the exciton density and extends the lifetime of the excitons. The intrawell migration rate for all samples is equivalent because the well width for all samples is the same. However, the interwell migration probability is decreased when the barrier width increases, and there is almost no interwell migration in the sample with barrier widths of 500 Å. For exciton accumulation, although intrawell migration has more efficiency than interwell migration, the sample with barrier width of 500 Å has the lowest efficiency compared with other samples due to the fact there is no interwell migration inside. On the contrary, there is higher exciton density in the sample with barrier width of 47 Å due to its higher interwell migration efficiency. In the case of this high density of excitons, the dissociation of biexcitons occurs due to the screening effect between biexcitons in the sample in Fig. 3(b), and exciton-exciton scattering occurs as shown in Fig. 1(b).

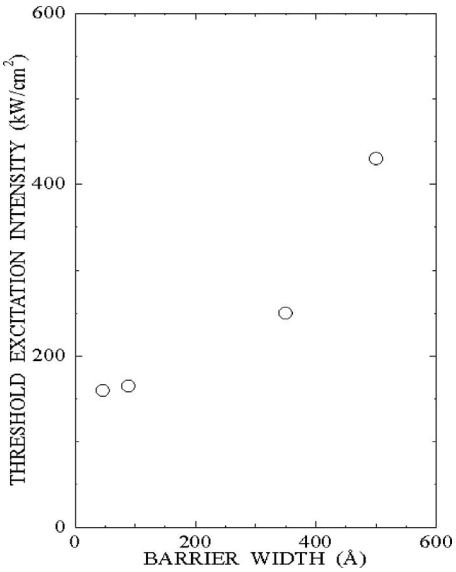

FIG. 4. Threshold excitation intensity are shown as a function of the barrier width for samples of $L_w=60$ Å and $L_b=47$, 89, 350, and 500 Å.

The threshold excitation intensities of stimulated emission for these samples were plotted as a function of barrier width as shown in Fig. 4. The threshold excitation intensity decreased as the L_b decreased. As L_b decreases, the increasing probability of migration for excitons through the miniband makes the density of exciton increase. The higher density of excitons causes the threshold excitation intensity of the stimulated emission to decrease.

In the case of weak I_{ex}, the emission band is dominated by the exciton recombination. For exciton recombination, the X band is related to a spontaneous emission, and shows a constant peak energy and constant band width as I_{ex} increases, and I_{em} have a linear relation with I_{ex}. As I_{ex} increases a little (small intense I_{ex}), on the low energy shoulder of the X band, a new emission band B appears, which is a stimulated emission due to I_{em} depends on the 1.65 power of I_{ex}. The B band shows constant peak energy and constant bandwidth as I_{ex} increases.[30,31] As I_{ex} becomes more intense

(medium intense I_{ex}), the exciton-exciton scattering occurs.[32,33] In the case of exciton-exciton scattering, I_{em} of the P band depends on a quadratic power with I_{ex}, therefore, the P band is also a stimulated emission band. P band shows a redshift, and the bandwidth is a constant. In the case of small and medium intense excitation, both biexciton and exciton obey the PSF rule. The electron-hole plasma occurs in a more intense I_{ex} (strong I_{ex}). The I_{em} of N band shows a linear relation with I_{ex} and N band shifts to the low energy and the bandwidth becomes broaden as I_{ex} increases. In the narrow barrier, we observed the X, P, and N band, and in the proceed experiment, we also observed the B band by gradually changed I_{ex} with smaller quantities.[34] For the wide barrier, we believed, with more intense I_{ex}, the P band and N band will be observed.

III. RESULTS

We have shown that the stimulated emission is dominated by the exciton-exciton scattering in the narrow barrier MQWs under low temperature. As I_{ex} increases, according to the PSF effect, one exciton will scatter to a higher energy level, and the other exciton will emit from a lower energy level in accordance with energy conservation. This explains the redshift of the stimulated emission as I_{ex} increases. The stimulated emission intensity in the exciton-exciton scattering model shows a superlinear dependence on I_{ex}. For wide barrier QWs, biexcitons are easier to occur under low temperature. The constant peak energy of stimulated emission B bands is shifted relative to the emission bands of excitons by a biexciton binding energy, and the stimulated emission intensity is proportional to the 1.65 power of I_{ex}.

When I_{ex} becomes strong, the density of excitons is increased. As the density of excitons increases, biexcitons are formed. In narrower barrier widths, excitons can migrate through the miniband and thus increase the exciton density. The resulting screening tends to inhibit the formation of biexcitons. Thus exciton-exciton scattering occurs in samples with narrow barrier widths. As I_{ex} increases further, the density of excitons becomes greater than the Mott transition density. The exciton thus separates to an electron and a hole. Since the density of electrons and holes is very high, electron-hole plasma is formed. The I_{em} of the electron-hole plasma depends linearly on I_{ex}. The emission spectra will gradually broaden and shift to lower energy due to the interaction between electrons and holes.

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