# Exciton-related lasing mechanism in ZnSe-(Zn,Cd)Se multiple quantum wells

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The processes involved in the stimulated emission by photopumping in (Zn,Cd)Se-ZnSe multiple quantum wells have been investigated at 77 K for a series of different well widths. It has been shown by means of photoluminescence-excitation spectroscopy that the confined excitons in the well play an important role in determining the lasing mechanism. The optical gain just above the lasing threshold is attributed to the recombination of an exciton accompanied by emission of one LO phonon. Far above threshold, inelastic exciton-exciton scattering processes contribute significantly to the gain.

#### I. INTRODUCTION

Progress in the growth technology of II-VI wide-gap semiconductors, to our knowledge, has opened the way to the first demonstration of blue-green laser diodes (LD's) based on strained (Zn,Cd)Se-Zn(S)Se quantum well structures.<sup>1</sup> More recently, the operation of blue LD has been reported in the lattice matched ZnSe-(Zn,Mg)(S,Se) system.<sup>2</sup> One of the most important objectives in this field is the stable operation of the laser diode under continuous wave (cw) mode at room temperature (RT), and since it has been shown that the gain of ZnSe quantum well is smaller than that of the GaAs under the same injection condition due to the larger effective masses and band gap<sup>3</sup>, it is essential to achieve the lowest threshold current density through the optimization of the laser structure itself. This can be facilitated using concepts which have been developed in III-V semiconductor LD's, such as choice of well-designed waveguide structures and the introduction of compressive or tensile stress to the wells.<sup>4</sup>

A detailed understanding of the lasing mechanism in II-VI semiconductors is very important for the design of laser structures. In III-V semiconductors (like GaAs or InP), it has been shown that recombination from the electron-hole plasma is the dominant process for the optical gain. Due to the large carrier densities present at the lasing threshold, the bound states of the excitons are screened out leaving only Coulomb-correlated continuum transitions. Consequently, in III-V LD's, apart from few exceptions, for example,<sup>5</sup> only band-to-band transitions have been included in gain spectra calculations. However, the situation is different for II-VI semiconductors because the Mott density, above which excitons are screened out, is estimated to be much larger than that in III-V semiconductors  $(7 \times 10^{16}/\text{cm}^3 \text{ in GaAs compared})$ with  $6 \times 10^{18} / \text{cm}^3$  in ZnSe). This differences arises primarily from the much smaller dielectric constant in ZnSe and the larger electron effective mass.

In this paper, we discuss the recombination processes involved in the stimulated emission at 77 K from a series of photopumped (Zn,Cd)Se-ZnSe multiple quantum well (MQW) structures with different well widths by means of photoluminescence excitation spectroscopy (PLE). Our intention is to optically probe the sample under lasing conditions. Specifically whether there remains a strong excitonic character to the absorption during lasing. We will do this by monitoring the PLE spectrum above threshold. We show that even above threshold there remains a strong excitonic character to the PLE spectrum so demonstrating that excitonic states play a central role in the lasing mechanism.

Historically, exciton-related lasing mechanisms have been studied for bulk II-VI crystals at low temperature.<sup>6</sup> Guillaume, Deveber, and Salvan<sup>7</sup> attributed the optical gain in CdS excited by an electron beam at 10 K to three exciton-related processes namely, the following.

(1) A low gain process owing to the annihilation of a free exciton with the emission of a photon and a LO phonon ( $E_x$ -LO phonon process),

(2) A medium gain process of exciton-exciton inelastic scattering.

(3) A high gain process involving exciton-electron scattering.

More recently, Newbury, Shazard, and Cammack<sup>8</sup> concluded that the exciton-exciton inelastic collision is the dominant stimulated emission process at 6 K in an optically pumped ZnSe epilayer. Exciton-related lasing processes have been investigated in MQW structures such as (Zn,Cd)Se-ZnSe,<sup>9–13</sup> ZnSe-Zn(S,Se),<sup>14</sup>(Zn,Cd)Se-Zn(S,Se),<sup>15</sup> and (Zn,Cd)S-ZnS.<sup>16</sup> Since the exciton binding energy is enhanced and also the exciton-LO phonon coupling is reduced in the quasi-two-dimensional confinement,<sup>17</sup> Ding *et al.*<sup>11</sup> have pointed out that excitons can play an important role even at RT.

Localization effects, due to the fluctuation of well widths, the inhomogeneity of the stress, or the alloy disorder (when the active layer is a solid solution) complicate the study of the lasing mechanisms. These localization effects depend on the structural quality of the sample, and can occasionally obscure a general understanding of the recombination process. Thus, one has to be particularly careful in the assignment of spectral features. Under high excitation conditions the energy levels change because of the many body effects such as phasespace filling, screening of the Coulomb interaction, and band-gap renormalization.

### **II. EXPERIMENTAL PROCEDURE**

The (Zn,Cd)Se-ZnSe MQW layers were grown in a VG Semicon molecular beam epitaxy system using conventional Knudsen cell sources of zinc, cadmium, and selenium on semi-insulating (Cr-doped) (100) GaAs substrates at a temperature of 280 °C. Details of the growth techniques have been published previously.<sup>18</sup>

The MQW structures consist of a ZnSe buffer layer (1.0  $\mu$ m), 15 cycles of Zn<sub>0.80</sub>Cd<sub>0.20</sub>Se wells and ZnSe barriers, and a ZnSe cap layer (0.20  $\mu$ m) and in these experiments, a series of samples with different well widths 15 Å, 30 Å, and 120 Å, and a constant barrier width of 80 Å were investigated (see Table I). Given an exciton Bohr radius (a<sub>B</sub>) of 35 Å in bulk Zn<sub>0.80</sub>Cd<sub>0.20</sub>Se, these well widths cover the range from 0.43a<sub>B</sub> to 3.4a<sub>B</sub>. Thus we span the transition from quasi-two-dimensional to bulk excitonic character.

The samples were cleaved into bars of 5 mm length and 500  $\mu$ m width. The Fabry-Perot cavity was formed by the natural facets of the sample bars. The laser samples were mounted on a copper cryostat whose temperature was held constant at 77 K. An Xe-Cl excimer laser (LAMBDA PHYSIK LPX-100) pumped tunable dye laser (LAMBDA PHYSIK FL2001) with dyes of Coumarin 120 and Coumarin 47 was used for the photopumping to cover the spectral range from 440 to 476 nm. The pulse duration of the pump laser is 5 ns which is much longer than the recombination time in the materials, so that the excitation was quasi-cw. The pump intensity was controlled using neutral density filters and was focused onto the sample surface using a cylindrical lens. The geometry of the measurement has been reported elsewhere.<sup>19</sup> The pulsed emission signal with a repetition rate of 15 Hz from the cleaved edge of the sample was focused into a Spex 0.6 m single grating monochromator using collection optics and was averaged using a boxcar integrator. The lasing in the  $TE_0$  mode from the structures under whole excitation condition was confirmed by means of polarization and beam pattern measurements.

### **III. RESULTS AND DISCUSSION**

Figure 1, curve (a), shows the cw PL spectrum from top face of the sample with a well width of 15 Å un-

TABLE I. Parameters of the MQW structures.  $L_W$  and  $L_B$  denote the the well width and the barrier width, respectively.

Sample	Well	Barrier	$L_W(\text{\AA})$	$L_B(\text{\AA})$
MQW 1	$Zn_{0.80}Cd_{0.20}Se$	ZnSe	15	80
MQW 2	$Zn_{0.80}Cd_{0.20}Se$	ZnSe	30	80
MQW 3	$\mathrm{Zn}_{0.80}\mathrm{Cd}_{0.20}\mathrm{Se}$	ZnSe	120	80

der low power excitation ( $10 \text{ mW/cm}^2$ ). Since the lighthole valence band in the well is lowered in energy with respect to the heavy-hole band due to the compressive strain and confinement, the emission peak at 2.7411 eV originates from the radiative recombination of the confined n = 1 heavy-hole excitons (designated as  $E_{x1hh}$ ). Reflectivity and cw-absorption measurements show that the Stokes shift of this peak is 3.0 meV. Figure 1, curves (b)-(d), are the emission spectra from the cleaved facet as a function of increasing excitation intensity. The excitation photon energy is 2.8171 eV in this case. At an excitation intensity of  $0.079 \text{ kW/cm}^2$ , the spectrum has a single peak whose position is same as that in the cw PL. An emission band designated as L appears in the low energy side of the main peak at about 1.0  $kW/cm^2$  and grows superlinearly with increasing excitation intensity. The superlinear increase and spectral narrowing of the Lband indicates the initiation of stimulated emission, and occurs at an excitation intensity of  $10 \text{ kW/cm}^2$ . We note that the emission peak of the lower band  $(E_{x1hh})$  does not shift with the increase of the excitation intensity and is still be observable above the lasing threshold so that the lasing line is at a substantially lower energy than the cw PL peak. The energy separation between the cw PL and the lasing peak at the threshold is 24 meV.

The lasing peak intensity of this sample as a function of excitation wavelength is shown in Fig. 2 for various pump intensities. The photoluminescence excitation



FIG. 1. Emission spectra taken from the  $Zn_{0.80}Cd_{0.20}Se$ -ZnSe MQW whose well width is 15Å. (a) PL from the top surface under the cw excitation of  $10mW/cm^2$ . (b)-(d) Emission from the cleaved edge under pulsed photo (2.8171 eV) excitation of (b) 0.079 kW/cm<sup>2</sup>, (c) 6.3 kW/cm<sup>2</sup>, and (d) 12.5 kW/cm<sup>2</sup>.

spectrum, curve (a), is very similar to the low power cw absorption, and consists of energy levels of the ZnSe barriers (and/or ZnSe cladding layer) and confined levels in the  $Zn_{0.80}Cd_{0.20}Se$  wells. The peak at 2.7450 eV is assigned  $E_{x1hh}$  and the peak at 2.7618 eV originates from the n = 1 light-hole exciton  $(E_{x1lh})$ . With increasing excitation intensity, the excitonic peaks become less distinct, and there is a small shift to higher energy and a decrease in the exciton transition strengths. The net shift of the peak is the result of several effects, namely, the exciton-band filling (a blueshift), a reduction of the exciton binding energy due to the exciton self-screening (a blueshift), and the renormalization of the band gap (a redshift). Similar, small blueshifts have been seen in GaAs/(Al,Ga)As MQW's under short pulse excitation which creates a population of  $excitons^{20,21}$  and are attributed to exciton-exciton interactions. The blueshift is observed only for the  $E_{x1hh}$ ; the shift for  $E_{x1hh}$  is essentially zero. A complete theoretical analysis of the many-body interactions is needed for a quantitative understanding of the peak shifts and bleaching.

It is important to clarify the energy shifts observed. To be consistent with the notation in III-V semiconductors we define the Stokes shift as the difference between the absorption (or PLE) and the emission energies. We also define  $\Delta E_{\text{laser}}$  to be the energy difference between the  $E_{x1\text{hh}}$  peak in the PLE and the lasing line. Thus  $\Delta E_{\text{laser}}$ changes with excitation intensity.

The  $\Delta E_{\text{laser}}$  value at the lasing threshold (denoted



FIG. 2. Lasing peak intensity taken from the MQW  $(L_W=15 \text{ Å})$  as a function of excitation wavelength under excitation intensity of (a)  $1.0I_{\rm th}$ , (b)  $2.0I_{\rm th}$ , (c)  $4.0I_{\rm th}$ , and (d)  $16.0I_{\rm th}$ , respectively. In this case,  $I_{\rm th}$  is the threshold intensity under the resonant excitation condition to the  $E_{x1hh}$  line.

 $\Delta E_{\rm laser}^{\rm th})$  of this sample is much larger ( $\Delta E_{\rm laser}^{\rm th}=27.1~{\rm meV})$  than the Stokes shift (3 meV). This value is close to both the LO-phonon energy of  $\approx 30~{\rm meV}$  and the exciton binding energy of  $\approx 35~{\rm meV}$ . Two possible processes can be responsible for the lasing. One is the LO-phonon assisted recombination of excitons and the other is is the exciton-exciton scattering process. The phonon assisted process is

## $exciton \longrightarrow LO phonon + photon$

and the energy balance for the process is given by

$$\hbar\omega_L = E_g - E_{\rm ex} - E_{\rm LO} + E_{\rm kin}^{\rm ex},\tag{1}$$

where  $E_g$  is the band gap,  $E_{\text{ex}}$  is the exciton binding energy,  $E_{\text{LO}}$  is the LO-phonon energy, and  $E_{\text{kin}}^{\text{ex}}$  is the kinetic energy of the exciton. In this case

$$\Delta E_{\text{laser}}^{\text{th}} = E_{\text{LO}} \tag{2}$$

assuming that  $E_{kin}^{ex}$  is negligibly small.

Recent investigations of Raman spectra of  $\operatorname{Zn}_x \operatorname{Cd}_{1-x}$ Se mixed crystal have shown that LO-phonon spectrum versus solid composition (x) is a single mode type, and the  $E_{\text{LO}}$  of the  $\operatorname{Zn}_{0.80}\operatorname{Cd}_{0.20}$ Se alloy is estimated to be about 30 meV.<sup>22</sup>

If this process is the mechanism of the stimulated emission then first the  $\Delta E_{\text{laser}}^{\text{th}}$  value should be equal to  $E_{\text{LO}}$ and should not depend on the well width  $(L_W)$ . Secondly no redshift of the lasing line should be observed with increasing the photopump intensity.

The exciton-exciton scattering process can be described as follows:

 $exciton(A) + exciton(B) \longrightarrow electron + hole + photon.$ 

The energy conservation condition can be written as

$$\hbar\omega_P = E_g - 2E_x - \delta E_{\rm kin} + E_{\rm kin}^{\rm ex(A)} + E_{\rm kin}^{\rm ex(B)}, \qquad (3)$$

where  $\delta E_{\rm kin}$  is the kinetic energy of the free electron and hole resulting from this process. If the lowest conduction and the highest valence bands are unfilled and the kinetic energy of the excitons are small enough at the lasing threshold, the  $\delta E_{\rm kin}$  value is expected to be zero. Therefore,

$$\Delta E_{\text{laser}}^{\text{th}} = E_{\text{ex}}.$$
(4)

With increasing excitation intensity, the conduction and valence bands begin to be filled with free carriers. In the three-dimensional case, the highest filled states of the carriers  $(E_{3D}^{e}$  for the electron and  $E_{3D}^{h}$  for the hole) at cryogenic temperature can be expressed as a function of the free electron (hole) density  $(N_{car})$  as follows:<sup>8</sup>

$$E_{3D}^{e(h)} = \frac{1}{m_{e(h)}^*} \frac{h^2}{2} \left(\frac{3}{8\pi}\right)^{2/3} N_{car}^{2/3}, \tag{5}$$

where  $m_e^*$  and  $m_h^*$  are the electron and hole effective masses, respectively. Newbury, Shazard, and Cammack deduced  $\delta E_{\rm kin}$  assuming that all carriers relax completely to the bottom of their respective bands.

$$\delta E_{\rm kin} = E_{\rm 3D}^e + E_{\rm 3D}^h \propto N_{\rm car}^{2/3}.$$
 (6)

Since this is the bimolecular process,  $N_{\rm car}^{2/3}$  is expected to be proportional to the square of the excitation intensity  $(I_e)$ . Therefore

$$\Delta E = \delta E_{\rm kin} + E_{\rm kin}^{\rm ex(A)} + E_{\rm kin}^{\rm ex(B)} \tag{7}$$

$$\simeq \delta E_{\rm kin} \propto I_e^{1/3},$$
 (8)

where  $\Delta E$  represents the redshift of the lasing line.

We note that the relation mentioned above is not proper if the kinetic energies of excitons involved in the process  $[E_{\rm kin}^{\rm ex(A)}, E_{\rm kin}^{\rm ex(B)}]$  cannot be omitted or the carriers are scattered to the higher wave number (k) states to satisfy the k-conservation rule. However, in the case of photopumped lasing in a ZnSe epilayer, the redshift follows this relation  $(\Delta E \propto I_e^{1/3})$  even up to pump intensities ten times threshold.<sup>8</sup> In the two-dimensional case we obtain

$$E_{2\mathrm{D}}^{e(h)} = \frac{1}{m_{e^{\parallel}(h^{\parallel})}^{*}} \frac{h^{2}}{2} \frac{1}{2\pi} N_{\mathrm{car}},$$
(9)

$$\Delta E \propto I_e^{1/2},\tag{10}$$

where  $m_{e\parallel}^*$  and  $m_{h\parallel}^*$  are the in-plane effective masses of electron and hole in the well layers, respectively. If the exciton-exciton scattering is the process for the lasing in the MQW structures at the threshold, the  $\Delta E_{\text{laser}}^{\text{th}}$ value corresponds to the binding energy of quasi-twodimensional excitons which varies with the  $L_W$ . Figure 3 shows the exciton binding energy  $(E_B)$  of n = 1heavy-hole exciton  $(E_{x1\text{hh}})$  in the ZnSe-Zn<sub>0.80</sub>Cd<sub>0.20</sub>Se-ZnSe quantum well as a function of  $L_W$  calculated using



FIG. 3. Calculated binding energy of excitons confined in the  $Zn_{0.80}Cd_{0.20}Se$  QW as a function of well width, assuming that the in-plane effective mass of the heavy hole in  $Zn_{0.80}Cd_{0.20}Se$  is (a) 1.46 $m_0$  and (b) 0.50 $m_0$ , respectively.

a two parameter variational approach. The band offsets necessary for this approach have been estimated using the method of Van de Walle,<sup>23</sup> assuming that only the  $Zn_{0.80}Cd_{0.20}Se$  layers are compressively strained by a lattice mismatch of 1.3%. The conduction and heavy-hole band offsets ( $\Delta E_c$ ,  $\Delta E_{v-hh}$ ) were calculated to be 168 meV and 66 meV, respectively. It is noted that no or very small light-hole band offset  $(\Delta E_{v-lh})$  was expected in this calculation. The exciton binding energy of  $E_{x1hh}$ has been done for the different in-plane masses of heavyhole  $[(a)m_{hh\parallel}=1.46m_0, (b)m_{hh\parallel}=0.50m_0]$ . As far as we are aware there exists to date no comprehensive measurements of the valence subband dispersion in these materials. However, the actual in-plane mass would be between the two. The  $E_B$  value at  $L_W=15$  Å is estimated to be in the range between 37 meV and 45 meV which is substantially larger than the observed  $\Delta E_{\text{laser}}^{\text{th}}$  value (27.1 meV). If exciton-exciton scattering is responsible of the lasing action  $\Delta E_{\text{laser}}^{\text{th}} = E_{\text{ex}}$ , and should follow the well width dependence of the binding energy. Furthermore with increasing photopump intensity there should be a square-root dependency on the photopump intensity.

Consequently, as far as the energy value is considered, the exciton-LO phonon scattering process is more probable for the stimulated emission at the threshold pumping intensity. In order to test this assignment further, the well width dependence of the lasing transition has been studied.

Figures 4 and 5 are the pulsed emission from the cleaved edge as a function of excitation intensity obtained from ZnSe-Zn<sub>0.80</sub>Cd<sub>0.20</sub>Se MQW's whose well widths are 30 Å and 120 Å, respectively. The stimulated emission is observed at 2.6818 eV ( $L_W$ =30 Å) and 2.6021 eV ( $L_W$ =120 Å); the energy position varies due to the reduced quantum confinement.

PLE spectra of the lasing line obtained at the MQW's of  $L_W=30$  Å and  $L_W=120$  Å are shown in Fig. 6 and 7, respectively. For the sample of  $L_W=30$  Å, well-defined excitonic peaks are observed at 2.7132 eV for the  $E_{x1hh}$  and 2.7365 eV for the  $E_{x1hh}$ , respectively, for the excitation intensity just above the lasing threshold. The  $\Delta E_{\text{laser}}^{\text{th}}$  value of this sample is 31.4 meV. For the sample of  $L_W=120$  Å, the  $E_{x1hh}$  peak located at 2.6336 eV is much broader and weaker, and disappears if the excitation intensity is raised above  $1.5 \times I_{\text{th}}$ . The  $E_{x1hh}$  peak cannot be observed. This may be as a result of the weaker, nearly three-dimensional, exciton and/or the partial relaxation of the misfit strain. The  $\Delta E_{\text{laser}}^{\text{th}}$  value of this sample is 31.5 meV.

Figure 8 shows the  $\Delta E_{\text{laser}}^{\text{th}}$  and the Stokes shifts of the cw PL as a function of well width. The  $\Delta E_{\text{laser}}^{\text{th}}$  data lie between 27.1 meV and 31.5 meV, and do not show any distinct dependence on the well width. This is is consistent with the lasing mechanism at threshold being LO-phonon mediated rather than due to exciton-exciton scattering. The Stokes shifts are in the range between 3.0 and 7.7 meV, which is substationally smaller than the  $\Delta E_{\text{laser}}^{\text{th}}$ . Therefore, it is expected that the localization of excitons to the alloy disorder and/or the fluctuation of the well width is not directly related to the lasing mechanism in the series of samples investigated here.



FIG. 4. Emission spectra taken from the cleaved edge of the MQW sample ( $L_W=30$  Å) under pulsed photo (2.7365 eV) excitation of (b) 0.063 kW/cm<sup>2</sup>, (c) 6.3 kW/cm<sup>2</sup>, and (d) 20.0 kW/cm<sup>2</sup>, respectively.

This is in contrast to some recent work<sup>10–12</sup> in which it was convincingly argued that the lasing mechanism arose from an inhomogeneously broadened band edge. This was demonstrated by showing the emergence of the stimulated emission from the peak of the luminescence. In our samples the stimulated emission does *not* emerge from the peak of the cw luminescence but at some 30 meV lower energy. The difference is due to the presence of a large apparent Stokes shift in their samples which they attribute to reabsorption. As the cw PL from the top surface and the low intensity pulsed PL from the cleaved facet show peaks at the same energy (see Fig. 1) we are confident that reabsorption is not shifting the peaks in our sample.

Thus we believe the LO-phonon mechanism is responsible for the lasing at threshold in our samples but not necessarily in theirs.

The dependence of the  $E_{x1hh}$  excitation peak, the lasing energy, and the  $\Delta E_{\text{laser}}$  on the excitation intensity for the MQW sample ( $L_W=30$  Å) are shown in Fig. 9. In this sample, the excitonic peak can be observed up to about  $I = 50 \times I_{\text{th}}$  which is the largest among the three MQW's. The lasing energy drops rapidly by about 3 meV if the excitation power is increased from  $1.0 \times I_{\text{th}}$  to  $1.5 \times I_{\text{th}}$ , remains constant in the range be-





FIG. 5. Emission spectra taken from the cleaved edge of the MQW sample ( $L_W$ =120 Å) under pulsed photo (2.7668 eV) excitation of (b) 3.2 kW/cm<sup>2</sup>, (c) 20.0 kW/cm<sup>2</sup>, and (d) 80.0 kW/cm<sup>2</sup>, respectively.

FIG. 6. Lasing peak intensity taken from the MQW  $(L_W=30 \text{ Å})$  as a function of excitation wavelength under excitation intensity of (a)  $1.0I_{\rm th}$ , (b)  $2.5I_{\rm th}$ , (c)  $5.0I_{\rm th}$ , (d)  $7.9I_{\rm th}$ , and (e)  $11.2I_{\rm th}$ , respectively. In this case,  $I_{th}$  is the threshold intensity under the resonant excitation condition to the  $E_{x1hh}$  line.



FIG. 7. Lasing peak intensity taken from the MQW  $(L_W=120 \text{ Å})$  as a function of excitation wavelength under excitation intensity of (a)  $1.0I_{\rm th}$ , (b)  $2.0I_{\rm th}$ , and (c)  $4.0I_{\rm th}$ , respectively. In this case,  $I_{\rm th}$  is the threshold intensity under the resonant excitation condition to the  $E_{x1hh}$  line.

tween  $1.5 \times I_{\rm th}$  and  $8.0 \times I_{\rm th}$ , and tends to decrease again above  $I = 8.0 \times I_{\rm th}$ .

In order to analyze the redshift of the lasing peak (designated as  $\Delta E$ ), the detailed data of  $\Delta E$  are plotted as a function of  $(I - I_{\rm th})$  in a log-log scale [(Fig. 10(b)].



FIG. 8. Stokes shift of the cw PL (open square) and the  $\Delta E_{\text{laser}}^{\text{th}}$  value (closed circle) as a function of well width. Data obtained at Zn<sub>0.85</sub>Cd<sub>0.15</sub>Se-ZnS<sub>0.08</sub>Se<sub>0.92</sub> MQW ( $L_W$ =50 Å) are also shown with triangular mark (Ref. 15).



FIG. 9. The dependence of the  $E_{x1hh}$  excitation peak (open circle), the lasing peak (open square), and the  $\Delta E_{\text{laser}}$  value (closed circle) on the excitation intensity observed in the MQW ( $L_W=30$  Å).

The data obtained in the ZnSe epitaxial layer at 10 K are also plotted [Fig. 10(a)] for comparison.<sup>8</sup>  $\Delta E$  of the ZnSe epitaxial layer is proportional to the  $(I - I_{\rm th})^{1/3}$  in the range from  $I - I_{\rm th} = 2.0 \times I_{\rm th}$  to  $10 \times I_{\rm th}$ . This has been interpreted as the lasing occurs as a result of the annihilation of an exciton via exciton-exciton inelastic scattering. In the case of the MQW sample, we expect from Eq. (10)  $\Delta E \propto (I - I_{\rm th})^{1/2}$ . As can be seen from Fig. 10(b)  $\Delta E \propto (I - I_{\rm th})^{1/2.4}$  if  $I - I_{\rm th}$  is above about  $7 \times I_{\rm th}$ . Thus we believe that the dominant lasing mechanism above about  $7 \times I_{\rm th}$  is inelastic exciton-exciton scattering. This is further supported by the fact that above about  $7 \times I_{\rm th}$  the PLE spectrum begins to blueshift in accord with the exciton-exciton scattering mechanism.<sup>20,21</sup>



FIG. 10. Redshifts of the lasing line  $(\Delta E)$  observed in the MQW  $(L_W=30 \text{ Å})$  as a function of  $I - I_{\text{th}}$ . Data obtained in the ZnSe epilayer (Ref. 5) are also shown for the comparison.

### **IV. CONCLUSIONS**

PLE spectra of the stimulated emission in the ZnSe-Zn<sub>0.80</sub>Cd<sub>0.20</sub>Se MQW's have been investigated at 77 K with the series of different well widths. It has been shown that the  $\Delta E_{\text{laser}}^{\text{th}}$  value is substantially larger than the Stokes shift of the cw PL and equal to the LO-phonon energy. Furthermore it is independent of the well width. Even under lasing conditions, excitonic peaks are clearly seen in the PLE spectra confirming the importance of excitonic transitions in the lasing process. We conclude that exciton-LO phonon scattering is the dominant process for the optical gain in the range from  $I = I_{\text{th}}$  to

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- <sup>1</sup> M.A. Haase, J. Qiu, J.M. DePuydt, and H. Cheng, Appl. Phys. Lett. **59**, 1272 (1991).
- <sup>2</sup> H. Okuyama, T. Miyajima, Y. Morinaga, F. Hiei, O. Ozawa, and K. Akimoto, Electron. Lett. **28**, 1798 (1992).
- <sup>3</sup> D. Ahn, T. K. Yoo, and H. Y. Lee, Appl. Phys. Lett. **59**, 2669 (1991).
- <sup>4</sup> J.M. Gaines, R.R. Drenten, K.W. Haberern, T. Marshall, D. Mensz, and J. Petruzzello, Appl. Phys. Lett. **62**, 2462 (1993).
- <sup>5</sup> H. Haug and S. W. Koch, Phys. Rev. A **39**, 1887 (1989).
- <sup>6</sup> H. Haug and S. Koch, Phys. Status Solidi B 82, 531 (1977).
   <sup>7</sup> C.B. Guillaume, J.M. Deveber, and F. Salvan, Phys. Rev.
- 177, 567 (1969). <sup>8</sup> P.R. Newbury, K. Shazard, and D.A. Cammack, Appl.
- Phys. Lett. 58, 1065 (1991).
  <sup>9</sup> H. Jeon, J. Ding, A.V. Nurmikko, H. Luo, N. Smarth, and J. K. Furdyna, Appl. Phys. Lett. 59, 1293 (1991).
- <sup>10</sup> J. Ding, H. Jeon, T. Ishihara, A.V. Nurmikko, H. Luo, N. Samarth, and J. Furdyna, Surf. Sci. 267, 616 (1992).
- <sup>11</sup> J. Ding, H. Jeon, T. Ishihara, M. Haherott, A.V. Numikko, H. Luo, N. Samarth, and J. Furdyna, Phys. Rev. Lett. 69, 1707 (1992).
- <sup>12</sup> J. Ding, M. Haherott, T. Ishihara, H. Jeon, and A.V. Numikko, Phys. Rev. B 47, 10528 (1993).

 $7 \times I_{\rm th}$ .

At higher excitation levels, exciton-exciton inelastic scattering comes to dominate the gain.

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- <sup>13</sup> K. Ando, A. Ohki, and S. Zembutsu, Jpn. J. Appl. Phys. 31, L1362 (1992).
- <sup>14</sup> Y. Kuroda, I. Suemune, Y. Fujii, and M. Fujimoto, Appl. Phys. Lett. **61**, 1182 (1992).
- <sup>15</sup> Y. Kawakami, B.C. Cavenett, K. Ichino, Sz. Fujita and Sg. Fujita, Jpn. J. Appl. Phys. **32**, L730 (1993).
- <sup>16</sup> Y. Yamada, Y. Masumoto, J.T. Mullins, and T. Taguchi, Appl. Phys. Lett. **61**, 2190 (1992).
- <sup>17</sup> N.T. Pelekanos, J. Ding, M. Hagerrot, A.V. Nurmikko, H. Luo, N. Smarth, and J.K. Furdyna, Phys. Rev. B 45, 6037 (1992).
- <sup>18</sup> K.A. Prior, J.M. Wallace, J.T. Hunter, S.J.A. Adams, M.S. Haines, M. Saoudi, and B.C. Cavenett, J. Cryst. Growth **101**, 176 (1990).
- <sup>19</sup> Y. Kawakami, S. Yamaguchi, Y-h. Wu, K. Ichino, Sz. Fujita, and Sg. Fujita, Jpn. J. Appl. Phys. **30**, L605 (1991).
- <sup>20</sup> N. Peyghambarian, H.M. Gibbs, J.L. Jewell, A. Antonetti, A. Migus, D. Hulin, and A. Mysyrowicz, Phys. Rev. Lett. 53, 2433 (1984).
- <sup>21</sup> D. Hulin, A. Mysyrowicz, A. Antonetti, A. Migus, W.T. Masselink, H. Morkoç, H.M. Gibbs, and N. Peyghambarian, Phys. Rev. B **33**, 4389 (1986).
- <sup>22</sup> H.J. Lozykowski and V.K. Shastri, J. Appl. Phys. **69**, 3235 (1991).
- <sup>23</sup> C.G. Van de Walle, K. Shahzad, and P.J. Olego, J. Vac. Sci. Technol. B 6, 1350 (1988).