Effect of Coulomb enhancement on optical gain in (Zn,Cd)Se/ZnSe multiple quantum wells

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The optical-gain spectra of a 40-Å (Zn,Cd)Se/ZnSe multiple quantum well has been measured at various temperatures using a variable-stripe method. By comparison with a gain calculation including many-body effects, we have shown that gain in this structure arises from an electron-hole plasma (EHP) for temperatures higher than 140 K. Excellent agreement between experiment and theory allows us to demonstrate the significance of many-body effects such as carrier dephasing and Coulomb enhancement. The breakdown of the EHP model below 140 K indicates an increasing excitonic contribution. [S0163-1829(96)11748-3]

The physics of II-VI semiconductors has been investigated for many years but the recent announcement of a (Zn,Cd)Se/ZnSe laser diode with a room-temperature operating lifetime in excess of 100 h has led to renewed interest in the mechanism of gain and lasing in these materials. II-VI quantum wells (QW's) are quite different from standard III-V laser materials such as GaAs; the smaller dielectric constant and wider band gap leads to a greatly increased Coulomb interaction between electrons and holes. One effect of this enhanced Coulomb interaction is an increased exciton binding energy in II-VI QW's (of the order 40 meV) which can be larger than the LO phonon energy. Excitons are thus less susceptible to phonon ionization and have been predicted to play a greater role in the optical properties of II-VI QW's at room temperature.

The possibility of exciton-mediated gain and lasing in II-VI semiconductors has been studied theoretically[1] for bulk material and more recent experimental investigations in QW's have indicated that excitons do contribute to gain at low temperatures.[2–4] On the other hand, at high temperatures and carrier densities, the strong Coulomb interaction makes the distinction between excitons and an electron-hole plasma (EHP) less clear and several authors report that gain in II-VI diode lasers is due to an EHP.[5,6]

At low carrier concentrations, below the Mott density, electrons and holes form bound states, i.e., excitons, with distinct spectroscopic properties such as pronounced absorption peaks below the free carrier band. With increasing carrier density, screening and renormalization decrease the exciton binding energy until, above the Mott density, excitons are unbound forming an EHP. If the Coulomb interaction is strong, as it is in II-VI materials, unbound electrons and holes near the band edge are still highly correlated and can produce spectroscopic features similar to excitons. This is seen in other systems[7] where the high-density Coulomb enhancement peak in absorption is known as a Mahan exciton.

We have investigated gain and lasing in a 40-Å (Zn,Cd)Se/ZnSe QW as a function of temperature. By fitting the experimentally measured gain spectra with a many-body model, including the screened Coulomb interaction, we show that the mechanism for gain in these structures is completely described by a Coulomb-enhanced EHP model and that exciton effects are not significant, at least for temperatures above 140 K. Although gain originates from an EHP, the effect of Coulomb enhancement on gain and spontaneous emission in these structures is pronounced[8,9] and the implications for the design of II-VI devices such as laser diodes and light-emitting diodes are considered.

The sample was grown on a GaAs substrate by molecular-beam epitaxy and consists of five periods of 40-Å $Zn_{0.72}Cd_{0.28}Se$ QW's separated by 150-Å ZnSe barriers. A 1-μm ZnSe buffer layer surrounds the wells on the substrate side with a 0.3-μm ZnSe capping layer on the top. For gain measurements, the sample was cleaved into a 1.6-mm-wide bar and excited with a frequency-tripled Q-switched Nd:YAG laser at 355 nm; delivering 5-ns pulses at a repetition rate of 20 Hz. The sample exhibited room-temperature lasing at an excitation density of approximately 70 kW/cm^2, which compares well with measurements in other II-VI QW materials.[2,3]

Gain spectra were measured using the variable-stripe length method of Shaklee, Nahory, and Leheny.[10] A cylindrical lens was used to focus the exciting light to a rectangular stripe on the sample perpendicular to the cleaved facets. The stripe width was approximately 80 μm and the stripe length was varied using a calibrated slit. For accurate temperature control the sample was mounted on the cold finger of a variable-temperature liquid-nitrogen cryostat. Stimulated emission from the edge of the sample was dispersed in a 1-m spectrometer and detected using an optical multichannel analyzer. The edge emission intensity is given by

$$I(L) \propto \frac{(e^{g(\hbar\omega)L}-1)}{g(\hbar\omega)}, \tag{1}$$

where L is the length of the exciting stripe and $g(\hbar\omega)$ is the modal gain. A gain spectrum, $g(\hbar\omega)$ is thus calculated from the solution of

$$\frac{I(L_1)}{I(L_2)} = \frac{(e^{g(\hbar\omega)L_1}-1)}{(e^{g(\hbar\omega)L_2}-1)}, \tag{2}$$

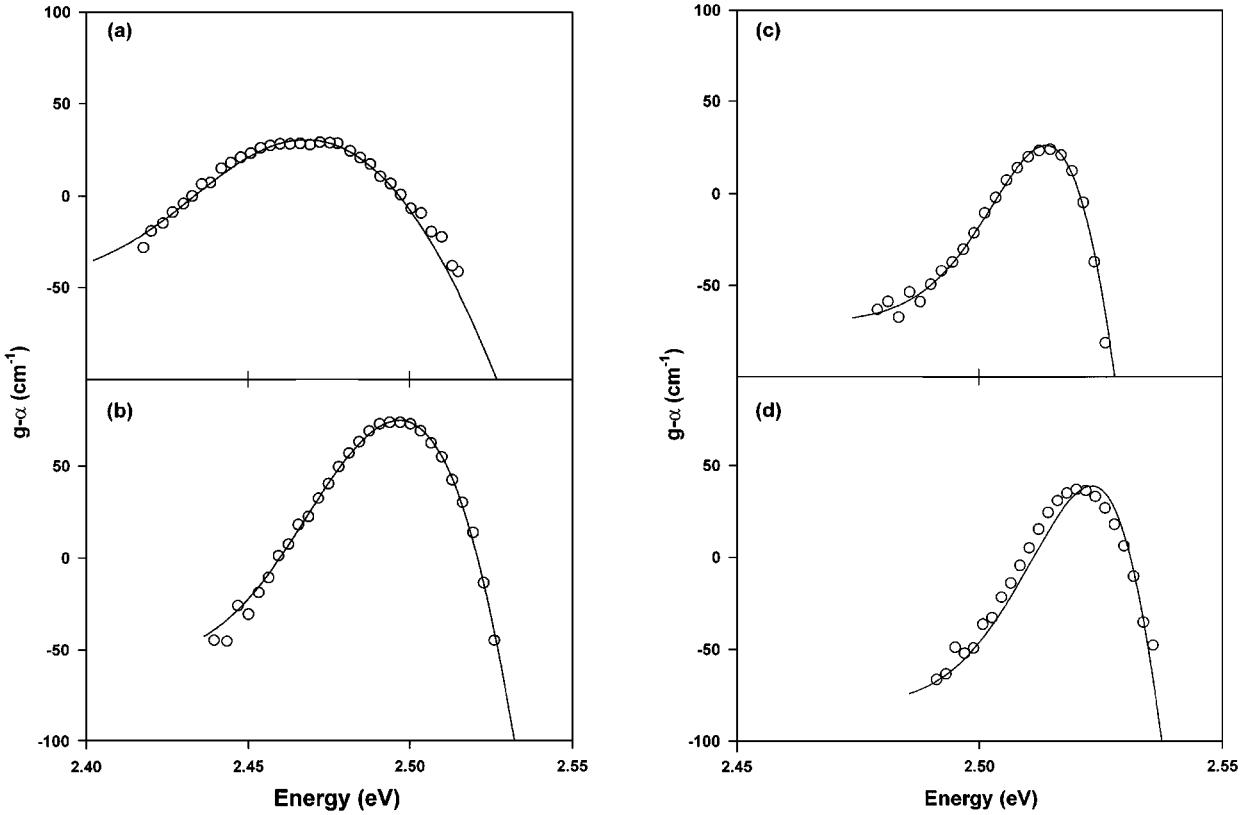

FIG. 1. Experimentally measured gain spectra for a $Zn_{0.72}Cd_{0.28}Se/ZnSe$ multiple QW (open circles) obtained at carrier temperatures of (a) 450 K, (b) 300 K, (c) 140 K, and (d) 110 K. The solid line shows the gain spectra calculated using a standard many-body model. A confinement factor of 0.04 was used for the fits in (a)–(c), whereas a value of 0.015 was necessary in (d).

where L_1 and L_2 are different excitation stripe lengths. Gain spectra were calculated from edge emission spectra using stripe lengths of 600 and 400 μm.

Figures 1(a)–1(d) show the measured gain spectra for carrier temperatures of 450, 300, 140, and 110 K. In each case, the spectra were recorded at a nominal pump intensity of 1.5 I_{th}, where I_{th} was the threshold pump intensity. The spectra shown in Fig. 1 are the measured modal gain, i.e., the material gain of the sample multiplied by the confinement factor of the waveguide structure. There is little material waveguiding in the sample because the difference in the refractive index between the QW's and the ZnSe buffer is relatively small. Light is then mostly gain guided in these structures and so the confinement factor cannot be readily calculated. However, the fits in Figs. 1(a)–1(c) were all obtained using a confinement factor of 0.04, showing that the confinement factor does not change appreciably with temperature or energy. The scattering losses α in Fig. 1 are mainly due to poor waveguiding and scattering at interfaces in the sample. A value of 50–60 cm^{-1} was deduced from the low-energy tail of the gain spectra and confirmed by measuring the slope efficiency of lasing in samples of various cavity lengths.[11]

An important parameter in the calculation of the gain spectra is the carrier temperature. In our experiment carriers are excited high above the band gap by the pump wavelength of 355 nm, leading to significant carrier heating. The carrier temperature was found by fitting the high energy tails[12] of the front surface spontaneous emission spectra with a Boltzmann factor of the form

$$I(\hbar\omega) \propto e^{-\hbar\omega/k_B T_c}, \tag{3}$$

where k_B is Boltzmann's constant and T_c is the carrier temperature.

For example, Fig. 2 shows the front surface spontaneous emission spectrum obtained at a lattice temperature of 300 K and the same excitation density used for the measurement of the gain in Fig. 1(a). On a logarithmic scale, the linearity of the high-energy tail over several orders of magnitude shows that the carriers have reached good thermal equilibrium before they radiate. For lattice temperatures of 100, 230, and 300 K, the measured carrier temperatures were 140, 300, and 450 K, respectively, and these values were used in the gain calculations.

The solid lines in Fig. 1 show fits to the experimental gain spectra using many-body theory including a screened Coulomb potential. The gain spectrum was calculated from the imaginary part of the optical susceptibility[13]

$$\chi(\hbar\omega) = \sum_k d_k \chi_k, \tag{4}$$

where

$$[\hbar\omega - E_{e,k} - E_{h,k} + i\hbar\gamma]\chi_k$$

$$= -(1 - f_{e,k} - f_{h,k})\left[d_k + \sum_{k'} V_s(k-k')\chi_{k'} \right], \tag{5}$$

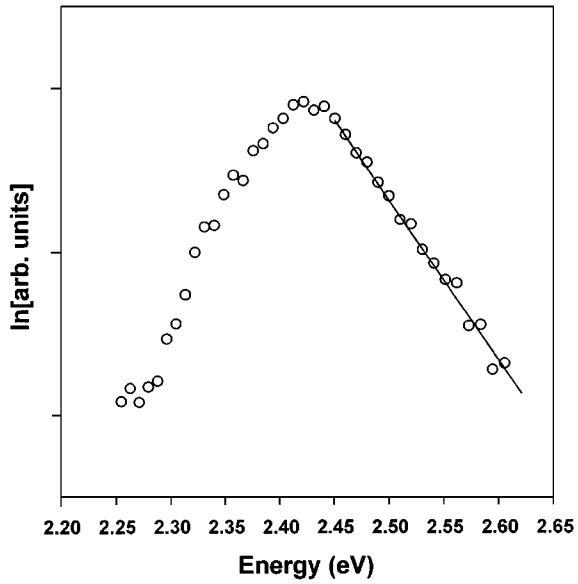

FIG. 2. Front surface spontaneous emission from the multiple QW (open circles) obtained at a lattice temperature of 300 K. The solid line shows the best fit to the high-energy side of the spectrum, giving a carrier temperature of 450 K.

f_c and f_v are the electron and hole Fermi functions, V_s is the screened two-dimensional Coulomb potential,[14] d_k is the momentum-dependent optical transition-matrix element,[15] and $E_{e,k}$ ($E_{h,k}$) is the renormalized[16] electron (hole) energy at the momentum k. The temperature and carrier density-dependent dephasing rate γ (which corresponds to the homogeneous broadening) was calculated as described in Ref. 9 including carrier-carrier and carrier-phonon scattering. The free carrier result, where the Coulomb interaction has been neglected, is calculated by setting $V_s=0$ in Eq. (5). Details of the band-structure calculation are given in Ref. 9. All material parameters for these calculations were taken from Ref. 17.

We found excellent agreement between experimental and theoretical gain curves for carrier temperatures between 450 and 140 K. Typical fits in this range are shown by the solid lines in Figs. 1(a)–1(c). The theoretical gain spectra were fitted to the data by calculating the gain for a particular separation in the quasi-Fermi level. It should be noted that the separation of the quasi-Fermi level and the confinement factor (found to be constant for temperatures above 140 K) are the only fitting parameters in the calculation, since both the carrier temperature and the material composition have been measured experimentally. Although our model includes band-gap renormalization, it was impossible to predict the wavelength dependence of the gain, and so the calculated spectra in Fig. 1 have been shifted rigidly in energy to obtain a fit. This, however, does not affect the shape of the gain spectrum since band-gap renormalization is not significantly energy dependent. Any conclusions we draw from fits to experimental gain spectra in this paper do not depend on a precise knowledge of the band-gap energy.

The width and the shape of the gain spectra on the low-energy side are dependent on the homogeneous and inhomogeneous broadening. The inhomogeneous linewidth (10 meV) due to well width and compositional fluctuations was

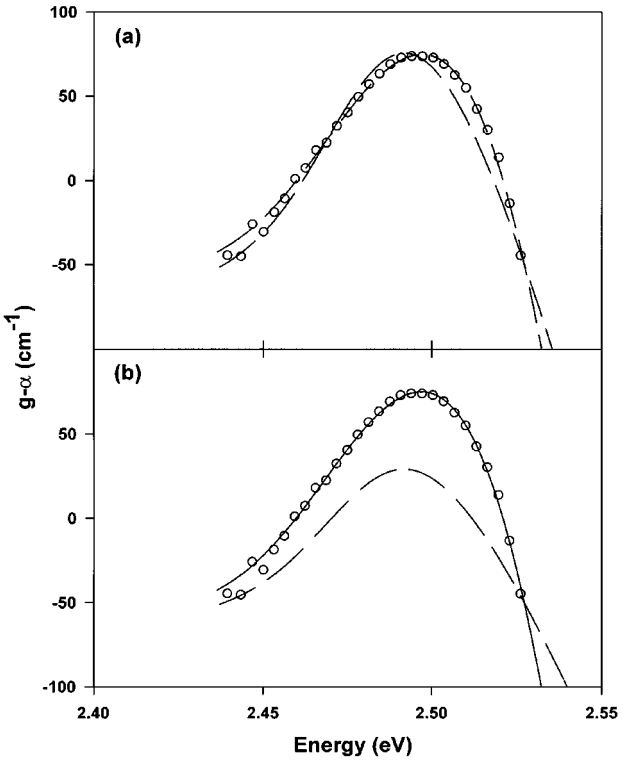

FIG. 3. (a) Best fits to the experimentally measured gain spectrum (open circles) at a carrier temperature of 300 K using the many-body theory and a confinement factor of 0.04 (solid line) and the free carrier theory using a confinement factor of 0.07 (dashed line). (b) As (a) but both calculated spectra scaled by the same confinement factor (0.04).

obtained from the 10-K heavy-hole exciton absorption resonance. Inhomogeneous broadening was incorporated into the calculation as a Gaussian convolution of the individual optical transitions. The high-energy side of the gain spectrum is particularly sensitive to the carrier temperature and the strength of the Coulomb interaction. The use of an experimentally determined carrier temperature in the calculation means that we are successfully predicting the magnitude of the Coulomb enhancement for all three gain spectra.

To date most calculations[18] of optical gain in II-VI materials have neglected the Coulombic attraction between electrons and holes. To illustrate the pronounced effect of Coulomb enhancement we also calculated gain neglecting the Coulomb interaction. Figure 3(a) shows the best fit to the data with this straightforward model for a carrier temperature of 300 K. For the different models to make physical sense, the transparency points at the same carrier density must coincide with the experimentally measured value. For this reason, the peak energy of the gain spectrum calculated using free carrier theory does not coincide with the measured value. In fact, it was impossible to account for both the peak energy and the transparency point using this theory. The effect of the Coulomb enhancement can be seen on the high-energy side of the gain spectrum, where the slope of the gain around the transparency point is dramatically changed. We could not account for the experimentally measured slope using a free carrier theory. In addition, to find the best fit in Fig. 3(a) using the free carrier theory, an increased confine-

ment factor of 0.07 was used. If the same confinement factor is used in both calculations, as is shown in Fig. 3(b), the significance of Coulomb enhancement is seen more clearly with the peak of the gain increased by more than 100% by including Coulomb enhancement, and therefore any predictions made using the free carrier theory will underestimate the gain.

It has been shown that for a given value of threshold gain the spontaneous emission is increased by the Coulomb enhancement[9] leading to an increase in the threshold current density. On the other hand, Coulomb enhancement produces a larger value of gain for a smaller separation in the quasi-Fermi levels and so reduces carrier leakage. It has been shown[19] that the reduction of carrier leakage significantly decreases the threshold current of II-VI diode lasers at room temperature. Such competitive effects show that a thorough understanding of Coulomb enhancement is essential for the development and optimization of blue-green optoelectronic devices.

It is well known that with increasing temperature, the higher carrier densities required for gain reduce the effect of Coulomb enhancement due to an increase in carrier screening.[16] Less is known about the low-temperature and low carrier density behavior of the Coulomb enhancement to the EHP optical transitions. By modeling the gain as a function of temperature, we have observed the breakdown of the EHP model for (Zn,Cd)Se QW structures. Our model successfully predicts the experimentally measured gain for carrier temperatures as low as 140 K. Below this temperature, it is impossible to account for the measured gain. Figure 1(d) shows the best fit obtained for a carrier temperature of 110 K. Again, we were unable to simultaneously account for the peak position and the transparency point using this model; furthermore, the confinement factor was 0.015, a value 60% lower than that used for the successful fits indicating that the EHP model fails to predict the magnitude of the gain below 140 K. As the lattice temperature decreases, the LO-phonon population also decreases and it becomes increasingly likely for excitons to exist long enough to contribute to the optical gain of the material. In light of recent evidence[2,3] for excitonic lasing below 200 K, the sudden breakdown of the EHP model for carrier temperatures below 140 K suggests that in our samples excitons contribute strongly to gain below this temperature.

In conclusion, we have experimentally measured gain spectra in a 40-Å (Zn,Cd)Se/ZnSe multiple QW at various temperatures. By comparison with a gain calculation including many-body effects, we have shown that gain in this structure arises from an EHP for temperatures higher than 140 K. Below this temperature, the EHP model breaks down, indicating an increasing excitonic contribution at low carrier temperatures. Our results show that the many-body effects such as Coulomb enhancement and carrier scattering contribute significantly to the optical properties of these materials and that these effects are an important consideration in the design of II-VI laser diodes.

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