Carrier Dynamics Study of Cd$_x$Zn$_{1-x}$Se/ZnSe (x = 0.2) Multiple Quantum Wells

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In this paper, nonlinear optical properties of Cd$_x$Zn$_{1-x}$Se/ZnSe (x = 0.2) multiple quantum wells were studied by low temperature steady-state and transient photoluminescence at high excitation densities. The biexciton transition was observed on the low energy side of the exciton transition. Based on the characteristics of stimulated emission observed in similar structures, we suggest the biexciton transition as the mechanism for stimulated emission. Optical degradation was also studied by room temperature photoluminescence using femtosecond laser pulses as the excitation source. The results confirm the formation of nonradiative recombination centers with a saturating degradation effect after about 10 min of exposure.

Key words: Biexciton, carrier dynamics, CdZnSe/ZnSe, exciton, multiple quantum wells, optical degradation

INTRODUCTION

CdZnSe/ZnSe quantum well structures have become promising candidates for the fabrication of light emitting devices in the blue-green spectral range. Recent demonstrations of continuous wave CdZnSe/ZnSe separate confinement heterostructure (SCH) diode lasers at room temperature show great promise. However, the device lifetime is limited to only an hour, due to pre-existing defects and subsequent degradation of the material. Given the sophisticated dislocation dynamics of the II-VI material, the control of defects and associated device degradation poses a major challenge to the commercial realization of such devices. Besides device performance issue, a fundamental question arises concerning the mechanism of stimulated emission in these structures. Due to the large exciton binding energy of these structures (40 meV), which is larger than the LO phonon energy (30 meV), strong and distinct exciton absorption peaks can be observed even above room temperature. The robustness of exciton transitions challenges the conventional degenerate electron-hole plasma (EHP) theory for stimulated emission. Experimental evidence indicates that the conventional EHP model is definitely not the gain mechanism in these structures at cryogenic temperatures. Therefore, investigating the gain mechanism is of great interest not only from the standpoint of many-body physics but also for practical device development.

This paper consists of two parts: the first part presents results from nonlinear optical study of CdZnSe/ZnSe multiple quantum wells (MQWs) by low temperature photoluminescence (PL) and addresses the issue of gain mechanism; the second part presents the results of an optical degradation study on the same structure by room temperature PL.

EXPERIMENTAL

The CdZnSe/ZnSe MQW samples used in this study were fabricated by molecular beam epitaxy (MBE). The sample structure consists of a ZnSe single crystal
substrate, a 2 µm ZnSe buffer layer, and ten periods of undoped Cd$_{0.2}$Zn$_{0.8}$Se/ZnSe MQWs with well (CdZnSe) thickness of 50 Å and barrier (ZnSe) thickness of 200 Å. Both steady-state and transient PL measurements were conducted at 10K. A time-resolved technique called population mixing was used to measure the carrier lifetime. The experimental setup is described elsewhere. The second harmonic from a modelocked Ti:Sapphire regenerative amplifier (400 nm, 280 fs, 250 kHz) was used as the excitation source. The sample was maintained at 10K in a closed-cycle helium cooled cryostat. The temporal resolution is determined by the pulse width of the laser (280 fs).

Figure 1 shows the PL spectra of the sample taken at different excitation intensities. An emission line appears on the lower energy side of the exciton and becomes more pronounced at higher excitation densities. We attribute this line to the biexciton transition. The biexciton transition has been observed in similar MQW structures grown on GaAs substrates by the same group of authors previously. A biexciton consists of two excitons with opposite internal spins coupled by Coulomb interaction. The dissociation of a biexciton involves the emission of a photon, leaving behind a free exciton. Typical characteristics of biexcitons are: superlinear PL intensity dependence on excitation power, faster decay lifetime than excitons, cross-circular polarization transition dependence, and reverse Boltzmann spectral lineshape. Assuming Gaussian lineshape for both transitions, the integrated areas of excitons and biexcitons were calculated for each curve. The dependence of the biexciton intensity on the exciton intensity gives approximately $I_{\text{biexciton}} \propto I_{\text{exciton}}^2$, which follows the expected superlinear intensity dependence on excitation density. The energy separation between the exciton and biexciton lines is about 10 meV, much larger than that observed in GaAs QWs (1–2 meV).

The transient behavior of biexcitons was verified by population mixing experiments. Figure 2 shows the decay curves for the exciton and biexciton. The reciprocal of the slope of each curve yields the corresponding carrier population lifetime. The biexciton exhibits a much shorter decay lifetime than the exciton (98 ps vs 263 ps). This behavior is due to the fact that the formation of biexcitons from excitons is a recursive process and the density of biexcitons is roughly proportional to the square of the exciton density. The coupled dynamics of excitons and biexcitons was well explained theoretically by Kim et al. by solving thermodynamic equations of exciton and biexciton populations.

The polarization dependence was verified by repeating the steady-state experiment at the highest excitation density with two beams cross-circularly polarized first and then co-circularly polarized. In the cross-circularly polarized configuration, the spectra in Fig. 1 are unchanged. However, in the co-circularly polarized configuration, the biexciton lines are not observed, indicating that no biexciton transitions are taking place. The biexciton transition thus indeed follows the polarization selection rule. This polarization dependence of the biexciton transition results from the two-photon absorption nature of the biexciton: excitation of both $\sigma^+$ and $\sigma^-$ excitons is necessary for the subsequent formation of a biexciton from the interaction of these two excitons.

**DISCUSSION**

**Electronic Structure Analysis**

Based on the above evidence, the formation of biexcitons in the MQW samples is confirmed with a