

Formation of Localized States by Well-Barrier Fluctuation in a II-VI Superlattice and Excitonic Stimulated Emission

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A well-defined exciton absorption peak from a localized state was observed for the first time. The localized state was formed by fluctuation of well-barrier interfaces in a ZnSe/ZnSSe superlattice structure. Stimulated emission was observed from the localized state up to 100K, and the physical origin is discussed from the temperature dependence of the stimulated emission peak. This opens the possibility to form a natural quantum box structure by controlling the well-barrier fluctuation.

1. INTRODUCTION

In II-VI semiconductors, excitonic properties are expected to play a prominent role in optical transitions due to the larger exciton binding energy ($\sim 20\text{meV}$ in bulk ZnSe) in comparison to III-V semiconductors ($\sim 4\text{meV}$ in bulk GaAs). Excitonic stimulated emission was discussed in a ZnCdSe/ZnSe quantum well (QW) structure from the observation of the parallel shift of a stimulated emission peak relative to an exciton absorption peak.¹⁾ Similar parallel shift was observed in a ZnSe/ZnSSe QW structure, but the physical origin of the stimulated emission was found to be an electron-hole plasma.²⁾

For realizing stimulated emission, high excitation is necessary to achieve population inversion to have a positive gain. Resultant high density of carriers screen the coulomb interaction necessary to form excitons. One solution to reduce the screening effect is to form localized states to confine excitons three dimensionally. Spatial separation of excitons by the localized states will reduce the screening effect and this will stabilize excitons to higher populations. Three-dimensional confinement also will increase the exciton binding energy, which will further stabilize excitons up to higher temperature.

In this paper, formation of a well-defined localized state employing heterointerface fluctuations is reported. An exciton absorption peak from the localized state was observed in photocurrent spectra measured on a ZnSe/ZnSSe superlattice (SL) prepared by metalorganic chemical vapor deposition (MOCVD). Stimulated emission was observed from the localized state with the low threshold of 6kW/cm^2 at 36K and was observed up to the measured 160K. This opens the possibility of realizing blue semiconductor lasers based on the excitonic optical gain from well-defined localized states.

2. EXPERIMENTAL

SL samples were prepared at 515°C by MOCVD using diethylzinc (DEZn), diethylsulfide and diethylselenide as source gases. The SL consisted of 157 periods of a 2-nm-thick ZnSe well and a 2-nm-thick $\text{ZnS}_{0.18}\text{Se}_{0.82}$ barrier grown on a (100) GaAs substrate. Growth interruption of 30s was inserted at the heterointerfaces, during which DEZn was kept flowing. The flow condition during the interruption was determined to reduce the mid-gap luminescence. In the SL samples thus prepared, band-edge luminescence dominated.

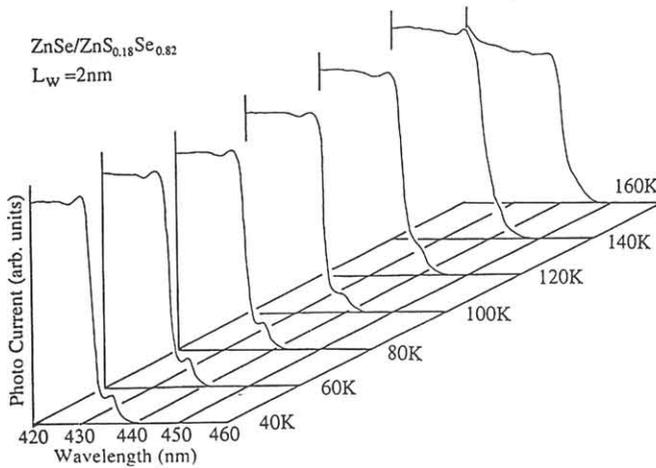


Fig. 1 Temperature dependence of PCS.

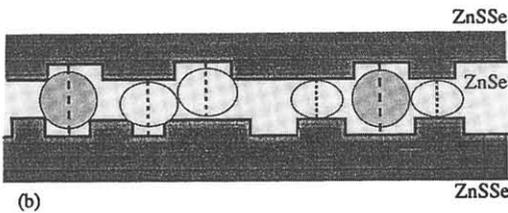


Fig. 2 Schematic to show the localized excitons (shaded circles) formed by one-monolayer fluctuation of the heterointerfaces.

The absorption spectra of the SL were examined by measuring photocurrent spectra (PCS). The details of the PCS measurements on a variety of ZnSe/ZnSSe SL's will be reported elsewhere,³⁾ but sharp heavy-hole (hh) and light-hole (lh) free-exciton (FE) absorption peaks were observed for the well width larger than 5nm. The blue shift of the FE peaks, when the well width was reduced, was in reasonable agreement with the calculation considering the quantum confinement and the strain effect.³⁾

In the PCS measured on the present SL shown in Fig. 1, two distinct absorption peaks were observed at the low temperature. The higher energy peak was assigned as the hh-FE absorption peak based on the above studies.

These two peaks, together with the lh-FE peak, were also observed in an excitation photoluminescence (PL) spectrum measured at low temperature. The energy separation of the hh and lh FE peaks was calculated to be 38.6meV assuming the common exciton binding energy between the hh and lh excitons and was in reasonable agreement with the measured peak separation of 35.6meV.

The lower-energy absorption peak shown in Fig. 1 was clearly observed up to 100K and could be identified up to 160K. Extension of the Urbach tail from the higher-energy states disturbed the observation of this peak at higher temperature. Figure 2 is the simplified schematic to explain the observed exciton absorption peak in the lower energy. When the terraces with the width of nearly exciton Bohr radius are formed by the fluctuation of the heterointerfaces, localized states are formed if the terraces in the neighboring interfaces are located at the same position as shown by the shaded exciton region in Fig. 2.

The other exciton states in the same figure will appear in the higher energies than that of the localized states. The maximum energy separation in this example will correspond to the difference of two monolayers.

Comparison of the temperature dependence of the peak positions between PL and PCS is shown in Fig. 3. The lower-energy peaks corresponding the localized excitons coincided with each other between PL and PCS. The observation of the discrete localized exciton peak in the PCS in Fig. 1 together with this peak coincidence indicates the formation of the well-defined localized excitons as schematically shown by the shaded circles in Fig. 2.

The hh-FE peak in the higher energy shown in Fig. 3 gives some energy difference between PL and PCS. The exciton states in the higher energy will have the spatial distribution of the energy level by the spatial exciton distribution schematically shown in Fig. 2, and this will give the observed energy difference between luminescence and absorption. The closed squares in Fig. 3 are the temperature dependence of a stimulated emission discussed in the following.

The SL sample with cleaved facets was excited with a pulsed dye laser excited with a N₂ laser to examine the stimulated emission

properties. Figure 4 shows the light output vs excitation characteristic measured from the cleaved facet at 36K. The cavity length of this sample is 400 μ m, and the excitation wavelength was 421nm. The measured lasing threshold was 6kW/cm². The stimulated emission was observed at \sim 438nm as shown in the inset of Fig. 4.

For studying the physical origin of the stimulated emission, the temperature dependence of the stimulated emission peak was examined

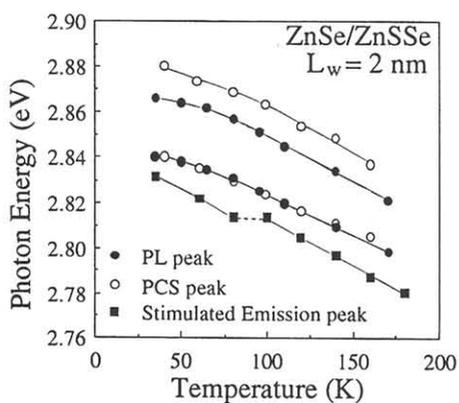


Fig. 3 Comparison of exciton peaks in PL and PCS and the stimulated emission.

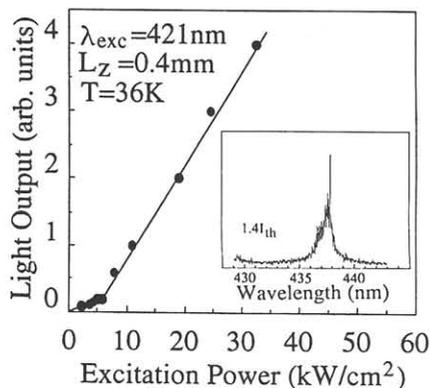


Fig. 4 Lasing light output measured at 36K.

and is shown in Fig. 3. The stimulated emission peak was close to the localized exciton peak. The energy separation between the stimulated emission peak and the localized exciton PL peak below 100K was linearly proportional to the temperature. The luminescence mechanism that is consistent with this characteristic is the exciton-electron scattering. This gives the energy separation of $1/2(M/m)kT$ from the free exciton,⁴⁾ where M is the total exciton mass and m is the electron mass. The estimated slope was 2.6kT, which is close to the theoretical value of 2.3kT calculated with the hh mass of 0.6 and the electron mass of 0.17.⁵⁾

3. SUMMARY

In summary, formation of well-defined localized states due to well-barrier fluctuation in the ZnSe/ZnSSe SL structure was observed by the exciton absorption and luminescence measurements. Stimulated emission from the localized states was observed up to 100K. The stimulated emission mechanism was found to be the exciton-electron scattering from the temperature dependence of the stimulated emission peak. Well-defined localized states reported in this paper show the possibility to realize a quantum box structure by controlling the well-barrier fluctuations in SL structures.

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