Stimulated Emission and Absorption of Highly Ordered Self-Organized PbSe Quantum Dots in Vertical Cavities

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Vertical cavities containing PbSe/PbEuTe superlattices with self-organized PbSe quantum dots, ordered in lateral as well as in vertical direction, are investigated by optical spectroscopy. Stimulated emission from the quantum dots, optically excited with 10 ns long pulses, is observed up to 140 K. The absorption spectrum of the superlattice is deduced from the width of the resonator modes, obtained by a transmission experiment. The quantum dot absorption can clearly be distinguished from the wetting layer signal.

1. Introduction

For III-V diode lasers, the introduction of quantum dots into the active zone of the laser allows to reduce the threshold current density [1], to increase the modulation band width, and to improve the temperature stability. This is the motivation for us to test quantum dot superlattices based on lead-salts as active material in vertical-cavity surface-emitting lasers for the mid-infrared. In self-assembled quantum dot superlattices, the elastic interactions between the growing dots on the surface and those buried within the previous layers often lead to the formation of long range correlations within the dot ensembles [2]. In lead-salt quantum dot superlattices, in particular, dot correlations inclined to the growth direction are observed, dependent on the superlattice period. This leads to a unique fcc-like ABCABC... vertical dot stacking sequence and a nearly perfect lateral ordering within the growth plane, corresponding to the formation of self-organized trigonal 3D lattices of dots [3].

While the ordering mechanism and the topography of the correlated PbSe quantum dot superlattices have been clarified [2], [3], there are almost no *optical* studies on PbSe quantum dots reported. Up to now, absorption measurements did not give conclusive results, due to the small optical density of the dots, and due to fact that absorption spectra of lead salt films are dominated by strong interference fringes. In this work, we study both the stimulated emission as well as the absorption of a lead salt quantum dot superlattice which is embedded between two dielectric bragg mirrors within a vertical cavity.

2. Sample Details

The high-finesse lead-salt microcavity samples filled with the PbSe quantum dot superlattice were grown by molecular beam epitaxy (MBE) onto (111) oriented BaF_2 substrates. The pyramidal shaped self-organized PbSe quantum dots were formed during Stranski-Krastanow heteroepitaxial growth of PbSe on PbEuTe due to the 5.4 % lattice mismatch [3]. The dot superlattices were grown at 360 °C deposition temperature by alternating 5 monolayers (MLs) of PbSe with 480 Å PbEuTe spacer layers on a PbEuTe buffer layer. Under these conditions, the dots in the superlattice are aligned in directions inclined by about 39° to the growth direction. An areal dot density of about 250 μm^{-2} (lateral separation 680 Å), an average dot height of 120 Å, and a base width of 300 Å with a relative full width at half maximum of the dot size distribution of about ±10 % is observed for superlattices with more than 60 bilayers [3]. The strong increase of the band gap energy of PbEuTe with increasing Eu content and the larger band gap of PbTe as compared to PbSe lead to a significant quantum confinement of the free carriers in the PbSe dots. In bulk material, the energy gaps at 4 K amount to 422 meV and 146 meV for $Pb_{0.95}Eu_{0.05}Te$ and PbSe, respectively.

The dot superlattices are sandwiched between two high-reflectivity Bragg mirrors. For sample 1 each mirror consists of 3 periods of quarter wavelength $EuTe/Pb_{0.94}Eu_{0.06}Te$ bilayers. For sample 2 the bottom mirror consists of 4 bilayers $EuTe/Pb_{0.94}Eu_{0.06}Te$ whereas the top mirror is grown with a higher Eu content of 20 %, to make it transparent for the pump laser wavelength of 1.9 µm. To achieve a reflectivity higher than 99% for this mirror 5 layer pairs are used. For sample 1 (2) we used a superlattice with 140 (236) periods grown on a 3 µm (1.8 µm) thick buffer layer and the designed center wavelength of the Bragg mirror is 4.3 µm (3.3 µm). The vertical cavity of sample 1 is 10 µm long whereas the length of the sample 2 cavity amounts 14 µm.

3. Stimulated Emission

For optical pumping of the vertical cavity structure, the first Stokes shifted line of a Q-switched Nd:YAG laser, produced in a hydrogen Raman cell, was used, resulting in a wavelength of 1.907 µm. The pulse length was about 10 ns and the repetition rate 50 Hz. The laser beam was focused on the sample to a spot size of 320 µm and the emission was recorded by an InSb detector using a box-car technique. For sample 1 we observe stimulated emission at energies around 300 meV (4.1 µm). With increasing temperature the emission shifts to higher energies due to the temperature dependence of the PbSe band gap. Finally, the stimulated emission disappears at a temperature of 100 K, because the laser gain spectrum shifts away from the energy range where the Bragg interference mirror exhibits a sufficiently high reflectivity.

For sample 2 much higher operation temperatures should be accessible, since the center of the Bragg mirror stop band agrees with the expected emission wavelength of the PbSe quantum dots at room temperature. As shown in Fig. 1, the emission spectrum consists of up to five lasing modes, where the envelope of the laser lines corresponds to the laser gain spectrum. The gain spectrum again shifts to higher energies with increasing temperature. When the temperature increases from 10 K to 100 K the emission intensity increases by an order of magnitude. For higher temperatures the intensity drops again and at about 150 K no emission is observed any more. For sample 2, the quenching of the stimulated emission is not due to a detuning of the emission energy in respect to the Bragg mirror stop band center. Therefore, it must be concluded that some intrinsic processes are responsible for the disappearance of the laser emission at 150 K. To prove this we have investigated the stimulated emission of a reference sample with a bulk like PbTe layer as laser active medium, again designed for operation at room temperature.

For this sample we obtain much better results than with the quantum dots samples. By optically pumping laser emission is observed up to 45 °C and the minimum threshold is about 200 times smaller than for the quantum dot cavities [4]. This result shows that the PbSe quantum dots are not at all advantageous for laser emission compared to bulk PbTe, which might be an indication that the band alignment between PbSe and PbEuTe is not of type I, or that the luminescence efficiency of the PbSe dots is reduced due to Eu intermixing from the PbEuTe barriers. To decide this, further experimental results on the quantum dot superlattices are needed.

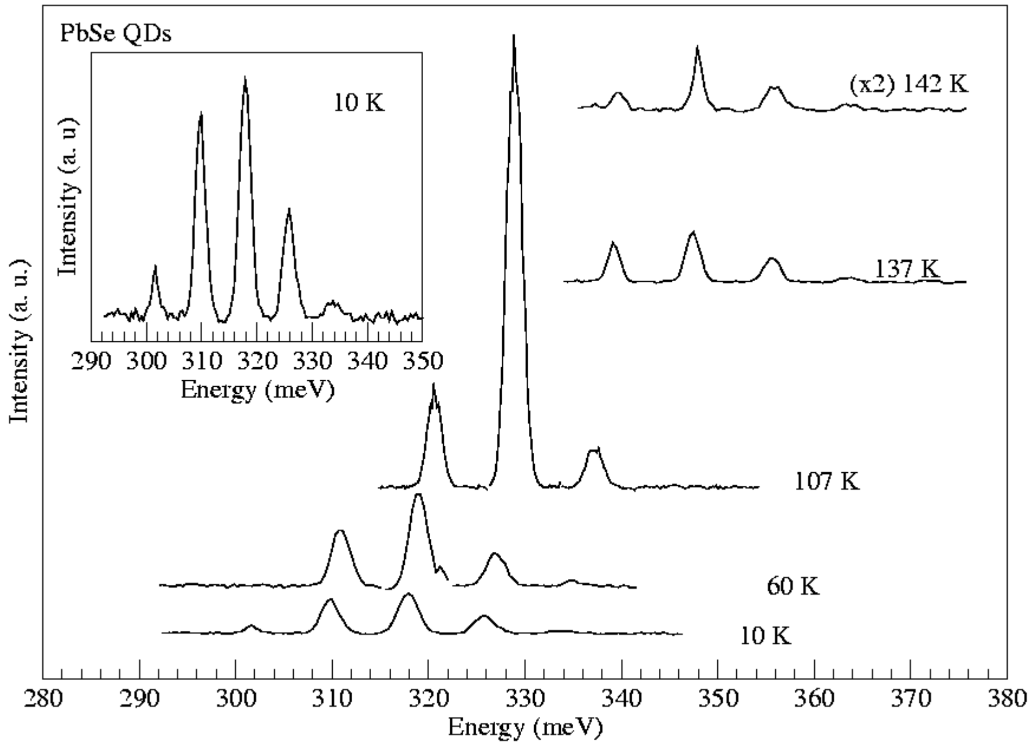

Fig. 1: Emission spectrum of sample 2 for various temperatures. The inset shows the 10 K spectrum with enhanced sensitivity to demonstrate the 5 laser modes observed at low temperatures.

4. Q-dot Absorption

To deduce the absorption of the quantum dots from a conventional PbSe/PbEuTe quantum dot superlattice by a transmission experiment is not possible, due to the small optical density of the quantum dots. Furthermore, the transmission spectrum of a single lead-salt layer predominantly shows strong interference fringes due to multiple reflections on the sample surface and the layer to substrate interface. When the superlattice is placed in a vertical cavity, however, the absorption is strongly enhanced and thus can be detected. In particular, the absorption causes a damping and a broadening of the cavity resonances. The extinction coefficient is directly proportional to the difference between the width of the damped resonance w and that of the resonance without damping w_0, normalized to the energy of the resonance ν [5]. So the extinction coefficient κ can be determined from a transmission experiment at the resonance energies. The proportionality factor between $(w_0 - w)/\nu$ can be obtained from a simulation of the transmission spectrum by the use of the transfer matrix method [5].

The results shown in Fig. 2 are obtained for sample 1. The transmission spectrum of this sample exhibits 17 cavity resonances within the stop band of the Bragg mirror. From the transmission experiment at low temperatures the width of the damped resonances is determined, while that of the undamped resonances is obtained from a measurement at room temperature. This can be done because at room temperature the PbSe quantum dot absorption is shifted to higher energies, out of the Bragg mirror stop band. The extinction coefficient in Fig. 2 shows a small peak around 240 meV due to quantum dot absorption of the longitudinal valley, whereas at 310 meV a stronger absorption peak is observed due to a transition within the oblique valleys. At 330 meV a step-like onset of κ is observed caused by absorption of the wetting layers. Most interestingly, the peak absorption of the quantum dots is slightly weaker than that of the two-dimensional wetting layer, in contrast as it would be expected from the peak-like density of states of the quantum dots. This might be an additional reason why the performance of the quantum dot laser is worse than that of the reference laser sample with a bulk-like active zone.

The stimulated emission of sample 1 at 10 K is shown in the inset of Fig. 2. The spectrum shows two laser modes with the orders m = 28 and m = 29. Fore these modes indeed the peak of the quantum dot absorption is deduced, confirming that lasing in the vertical-cavity sample 1 is caused by the quantum dots and not by the wetting layer.

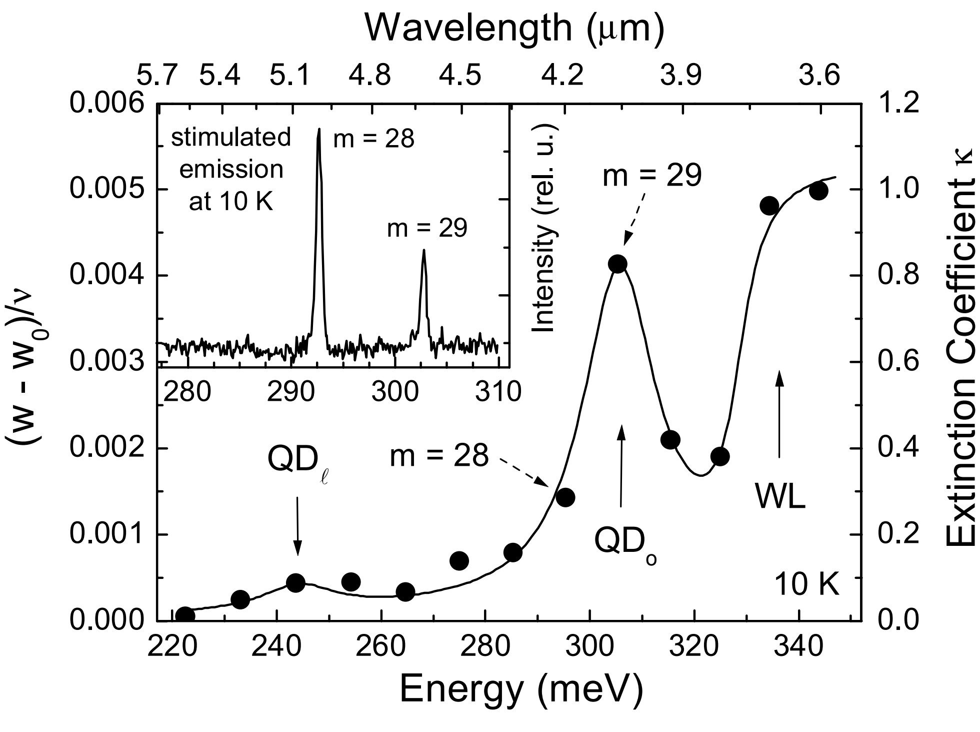

Fig. 2: Absorption spectrum from a PbSe quantum dot superlattice deduced from the width of the cavity resonances of a vertical cavity (sample 1). The dot absorption can be clearly distinguished from the wetting layer. The stimulated emission observed for this sample is shown in the inset. The emitting modes m = 28 and m = 29 agree with the absorption peak caused by the quantum dots.

3. Summary

We investigated the stimulated emission and absorption of vertical cavities containing PbSe/PbEuTe superlattices with self-organized PbSe quantum dots which are ordered in lateral as well as in vertical direction. Under optical excitation with 10 ns long pulses, stimulated emission in the mid-infrared spectral range is obtained from the quantum dot superlattice up to a temperature of 140 K. The absorption spectrum of the superlattice is deduced from the width of the resonator modes, as measured by transmission experiments. The quantum dot absorption can clearly be distinguished from the wetting layer signal, despite their small optical density. Therefore, the measurement procedure demonstrated here for PbSe quantum dots might be applied also for other systems with small optical densities in the near future.

References

[1] D.L. Huffaker, G. Park, Z. Zhou, O.B. Shchekin, D.G. Deppe, 1.3 mu m room-temperature GaAs-based quantum-dot laser, *Appl. Phys. Lett.* **73**, 2564 (1998)

[2] V. Holy, G. Springholz, M. Pinczolits, G. Bauer, Strain induced vertical and lateral correlations in quantum dot superlattices, *Phys. Rev. Lett.* **83**, 356 (1999)

[3] G. Springholz, V. Holy, M. Pinczolits, G. Bauer, Self-organized growth of three-dimensional quantum-dot crystals with fcc-like stacking and a tunable lattice constant, *Science* **282**, 784 (1998)

[4] J. Fürst, H. Pascher, T. Schwarzl, M. Böberl, W. Heiss, G. Springholz, G. Bauer, Mid-infrared IV-VI vertical-cavity surface-emitting lasers with 0-. 2- and 3-dimensional systems in the active regions, submitted to *Appl. Phys. Lett.*

[5] T. Schwarzl, W. Heiss, G. Springholz, H. Krenn, T. Fromherz, A. Raab, I. Vavra, Mid-infrared absorption of PbSe/PbEuTe quantum dot superlattices in high-finesse IV-VI microcavities, submitted to *Phys. Rev. B*