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Investigations of strain in ZnMgSe/ZnSe microdisks by means of the micro-Raman imaging

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Strain in the semiconductor multilayer structures of lasing devices has an important influence on their optical properties. Micro-Raman spectroscopy is a suitable technique for non-destructive direct analysis of the structural properties of semiconductor structures. The strain distribution in ZnMgSe/ZnSe microdisks has been measured by micro-Raman spectroscopy. The frequency of the ZnSe longitudinal optical phonon shifts about 2.8 cm^{-1} towards lower frequencies from the centre to the edge of the disk. In the central region of the disk the ZnSe quantum well and the ZnMgSe barriers are pseudomorphic strained on GaAs while in the free stranding region the ZnSe quantum well is pseudomorphic with the ZnMgSe barriers. Despite the strain relaxation alongthe radial direction of the disk, the microdisk is free of defects as demonstrated by the absence of the transverse optical phonon lines. These findings confirm sufficiently high quality values in such strained microdisk cavities to facilitate their application in optoelectronic devices.

1 Introduction

Semiconductor microdisks (MDs) are promising for applications in photonics and quantum-information processing, such as low-threshold lasers [1, 2] and efficient solid-state based single photon emitters [3]. Compared to other optical micro cavities, MDs are easy to fabricate and yield strong confinement due to internal total reflection of the confined whispering gallery modes (WGMs) close to the disk boundary. Since its first introduction in 1992 [1], many efforts were made to improve the Purcell factor in the MD lasers in order to reduce the laser thresholds. In this context experimental implementations of MD lasers were realized with II/VI quantum well (QW) structures grown lattice matched on GaAs [4]. However, their quaternary composition results in alloy fluctuations, which have substantial impact on the optical properties (line width, lasing threshold) of the lasing devices. Alternatively, with strained ZnSe QW in ternary ZnMgSe layers with low Mg concentration the alloy fluctuations can be minimized. In this context low-threshold lasing was observed on donor bound exciton transitions in fluorine doped ZnMgSe/ZnSe structures [5,6]. These findings indicate sufficiently high Q-values in such strained MD cavities. Because of the strain release in microdisks the performance of microdisk lasers may be enhanced [7] due to band gap narrowing from the centre towards the disk edge where the WGMs are located. Raman spectroscopy (RS) is a suitable technique for non-destructive characterization of the structural properties of semiconductor MD heterostructures. In this paper we present a detailed study of the strain distribution in the MDs performed by high spatial and high resolution micro-RS, which unambiguously reveals a strain gradient in our microdisk resonators.

2 Experimental

Strained ZnMgSe/ZnSe/ZnMgSe multilayer structures were grown by molecular beam epitaxy on (001) GaAs substrates. For optimal interface properties, a 20 nm buffer of undoped ZnSe was first deposited on the substrate followed by 30 nm thick ZnMgSe barrier layers with a mole

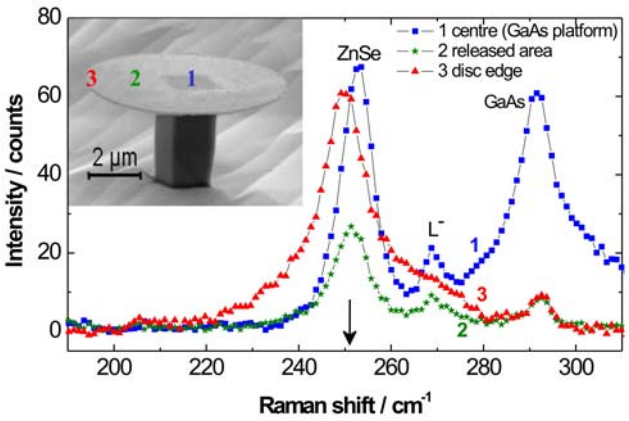

Figure 1 Raman spectra of the LO phonons recorded at thee different spots along the 8 µm diameter MD supported by GaAs post. Schematically Raman probing locations are shown in the insetted SEM image of such a disk. The arrow indicates the LO phonon frequency of bulk ZnSe.

fraction of magnesium of 0.09 and a 10 nm thick ZnSe QW. Photolithography was performed to define features of different dimensions (i.e. 1 to 10 µm in diameter). For the pattern transfer into the structure the samples were etched by reactive ion etching in an Oxford Plasmalab 80 Plus etch system. The undercut of the ZnMgSe/ZnSe on the substrate was achieved by selective wet chemical etching of the GaAs using a NaOH : H_2O_2 solution. The morphology of the free standing ZnMgSe/ZnSe MD structures was studied by scanning electron microscopy (SEM).

The MDs were characterized by confocal micro-Raman scattering in backscattering geometry $z(x,y,)\bar{z}$, where x = [100], y = [010] and z = [001] at room temperature. The investigations were performed on a commercial Raman spectrometer NTEGRA Spectra (NT-MDT Co., Moscow). The samples were excited by 473 nm CW laser radiation with an average power of 0.5 mW. The laser beam was collimated to 400 nm diameter. At this power level no significant warming of the sample was observed. This is inferred from RS experiments where the power level was varied between 0.08 and 8 mW. In this experiment no spectral shift (within the experimental resolution of our setup) of the GaAs LO phonon line was observed indicating that the sample warming due to the laser radiation is less than 20 K. The scattered light was focused onto a 100 µm pinhole at the entrance of a single-monochromator with transmitting efficiency up to 70%. It was equipped by reflective diffraction grating with 1800 lines per mm yielding a spectral resolution of 1 cm^{-1}. Signal detection was performed by a cooled low noise Andor CCD camera.

3 Results and discussion

We performed spatially resolved micro-RS measurements on ZnMgSe/ZnSe MD structures. Thereby the Raman shift of the ZnSe longitudinal optical (LO) phonons depending on the strain state of the lattice of the MD was investigated. Figure 1 shows a series of Stokes-Raman spectra recorded at different spots on a disk with 8 µm diameter. The Raman probing locations are shown in the insetted SEM image of such a disk. Several peaks are distinguished. The peak at about 292 cm^{-1} corresponds to the LO phonons of GaAs [8]. At 269 cm^{-1} a plasma-phonon coupled mode (L^-) from near-interfacial GaAs is observed [9]. GaAs LO and L^- modes were found in the centre (area 1) of the disk and with low intensity at the freestanding part (area 2 and 3) of the disk due to the scattering at the GaAs underneath the disk. The peak with a frequency of about 252 cm^{-1} is in a good agreement with the LO phonon mode of the ZnSe layer [10]. A small intensity contribution at about 250 cm^{-1} corresponds to the ZnSe-like phonon from ZnMgSe with 9% Mg mole fraction. In ternary compounds the spatial correlation length of phonon is reduced by potential fluctuations due to alloy disorder [11], which leads to significant broadening of the Raman lines and thus to a decrease of the intensity. For this reason the intensity of the ZnMgSe related Raman lines is considerably reduced and only ZnSe phonon modes are detected in the Raman spectra of our disk. The ZnSe LO phonon frequency in the centre (area 1) of the disk is at 252.8 cm^{-1}. At an intermediate area (area 2) and at the edge of the MD (area 3) corresponding ZnSe LO frequencies were observed at 251.5 cm^{-1} and at 250.0 cm^{-1}, respectively, yielding a frequency shift of ZnSe LO phonons of 2.8±0.5 cm^{-1} from the edge to the centre. For reference the arrow shows the frequency of ZnSe LO phonons at 252.0 cm^{-1} measured with a fully relaxed 2 µm thick "bulk like" ZnSe layer on (001) GaAs. The width of the LO ZnSe Raman line in the centre and at the intermediate area of the disk is comparable with the line width of strained ZnSe layers [12]. At the edge of the disk we observed a slight increase of the line width due to light scattering at the disk sidewalls.

We suggest that the shift of the LO phonon peaks of ZnSe to higher frequencies arises from in-plane compressive strain in the ZnSe layers due to the lattice mismatch between the GaAs substrate and the layer. It can be modelled as follows. The lattice parameter of ZnSe (0.5669 nm) is larger than that of GaAs (0.5654 nm) [13]. Therefore the pseudomorphic epitaxial growth of ZnSe on (001) GaAs will produce a biaxial compression of the unit cell of ZnSe along the *x* and *y* directions which is parallel to the grown surface. The nonzero components of the strain tensor in such a layer are $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon = (a_{||} - a_{ZnSe})/ a_{ZnSe}$ and $\varepsilon_{zz} = \{2S_{12}/(S_{11} + S_{12})\}\varepsilon$, where a_{ZnSe} represents the lattice parameter of bulk ZnSe, $a_{||}$ is the in-plane lattice parameter of the epitaxial layer and S_{ij} are the elastic compliance constants of ZnSe. The strain ε is negative for compressive strain. Under these conditions we obtain for the shift of LO phonon frequency ω of the strained layer given by the expressions in Ref. [14] and [15] using $\omega_0 = 252.0$ cm^{-1} the relationship $\Delta\omega = \omega - \omega_0 = -428.3\varepsilon$, with ω in units of cm^{-1}. This relation reveals a blue shift of ω for negative ε, which is in good agreement with the experimentally measured shift of the ZnSe LO phonon as shown in Fig. 1. Consider-

ing the volume ratio of ZnMgSe to ZnSe in our disks, the ZnSe QW is likely to be tensile strained on ZnMgSe in the periphery of the freestanding material. If we suppose that ZnSe is compressively strained on GaAs at the post position (area 1) and tensile strained on ZnMgSe at the edge (area 3) of the disk, we obtain $\varepsilon = -6.44\times10^{-3}$ and hence $\Delta\omega = 2.76$ cm^{-1}. The lattice parameter of ZnMgSe is calculated using Vegard's law to 0.56906 nm and the Mg concentration was obtained from high resolution x-ray diffraction measurements. The calculated value for the LO phonon shift is in agreement with the measured value supporting the model of a relaxed free standing layer system in the edge region of the disk.

We observed no transverse optical (TO) phonon Raman line across the whole area of the disk. Olego and Cardona [16] performed a detailed study of TO phonon RS. They found that the existence of the TO phonon line in the RS spectra of (001) oriented samples is related to defects in the samples. Thus the absence of TO phonon lines in our Raman spectra is a strong indication that the density of extended defects in the disk area is negligible.

The distribution of strain in the micro-cavity (MC) is depicted in Fig. 2, where the Raman shift of the ZnSe LO peak is plotted vs. position on the disk (the intensity of the ZnSe LO Raman line outside the disk area is zero). Three different regions of strain are distinguished: a) at the intersection of the GaAs post we find clear evidence that the ZnMgSe/ZnSe QW is pseudomorphic strained with the lattice parameter of the GaAs post, b) in a region which covers about the inner-half of the freestanding part of the disk we observe a phonon frequency which is almost identical to that of relaxed (bulk like) ZnSe, c) the edge region with a width of about 2 µm shows a radial symmetric distribution of tensile strain which is due to the equilibrium distribution of strain in the ZnMgSe/ZnSe QW with a common lattice parameter.

4 Conclusions

We have demonstrated that dry and wet etching techniques can be used to fabricate defect free ZnMgSe/ZnSe microdisks. The strain in these ZnSe based microstructures was investigated by micro-Raman scattering experiments. The measurements reveal a redshift of about 2.8 cm^{-1} shift of the ZnSe LO phonon frequency. Modelling of the experimental results shows that the shift can be explained by the homogeneous release of strain along the radial direction of the disk from the centre (ZnSe compressively strained on GaAs) to the edge (ZnSe tensile strained on ZnMgSe). The absence of the ZnSe TO Raman line in the spectra indicates that strain relaxation in the structure is not accompanied by the generation of extended defects. Our results demonstrate an effective preparation process of defect free microdisks by under-etching II-VI compound strained multilayers.

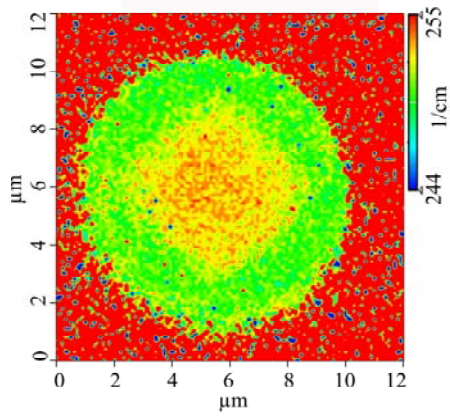

Figure 2 Distribution of strain in the micro-cavity (MC).

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