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Low-threshold ZnSe microdisk laser based on fluorine impurity bound-exciton transitions

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ABSTRACT

Impurity states in semiconductors, in which two long-lived ground states can be optically coupled to a single excited state, provide a powerful mechanism for applications including lasing without inversion, electromagnetically induced transparency, and optically addressable quantum memory for quantum information processing. We report low-threshold lasing from fluorine-doped ZnMgSe/ZnSe quantum wells in microdisk cavities. The lasing mechanism was studied by power-dependent photoluminescence spectroscopy. Lasing thresholds lower than $50\,W\,cm^{-2}$ were observed and the fraction of spontaneous emission contributed to the lasing modes was about $\beta = 0.03$–0.1.

1. Introduction

Quantum interference of the two optical pathways of an optical Λ-system, in which two long-lived ground states are optically coupled to a single excited state, provides a powerful mechanism for a number of useful applications. These applications include lasing without inversion [1] and electromagnetically induced transparency [2]. In semiconductors, a Λ-system may be naturally formed by a neutral donor via its electron-spin ground states and the lowest donor-bound-exciton state. The fluorine impurity in ZnSe is such a donor which features the critical characteristics of tight binding, bright emission, and low expected electron-spin decoherence due to a small number of substrate nuclear moments.

Early studies of lasing in ZnSe were limited because the material was not stable enough to be used in high-current or in high-excitation regimes [3,4]. However, when a high ratio of stimulated emission to spontaneous emission, known as the β-factor [5] is introduced by a high-*Q*, low-volume optical microcavity, low-threshold lasing is achievable.

Such lasing is seen in recent developments in microcavity quantum dot lasers [6]. Further, if the lasing medium was provided by discrete donor-bound-exciton transitions rather than by the continuum of free excitons, the coherence provided by the ground states of the donor spins could allow the possibility of lasing without inversion [1], further decreasing the input power requirements. Donor-bound-exciton transitions, coupled to a microdisk cavity with a high cooperative factor are also of substantial interest for basic research in the field of exciton–polariton coupling, which to date has only been investigated in resonators with quantum wells (QWs) [7] or quantum dots [8] as the active media.

Bound-exciton emission in ZnSe was observed many years ago [9], and the optical transitions of single, isolated nitrogen acceptors in ZnSe were recently measured [10]. We have studied the ^{19}F donor earlier in bulk samples [11]. Previous works on microcavity lasers in the wide band-gap II–VI semiconductors were focused on lasers based on CdSe quantum dots [12,13], or ZnCdSe [14] confined by quaternary alloys of ZnMgSSe and ZnCdSSe, which are lattice matched to a GaAs substrate.

To reduce alloy fluctuations, we confined a ZnSe QW only in ternary ZnMgSe barriers with low Mg concentration. Lasing due to confined free excitons seeded by excitonic molecules has been previously observed in similar systems [15,16]. Here we show lasing directly on discrete, bound-exciton transitions in samples with a fluorine δ-doped (^{19}F:ZnSe) layer at the centre of the QW.

2. Experimental

The ZnMgSe/ZnSe/^{19}F:ZnSe/ZnSe/ZnMgSe samples were grown by molecular beam epitaxy on GaAs-(0 0 1) substrates. For optimal

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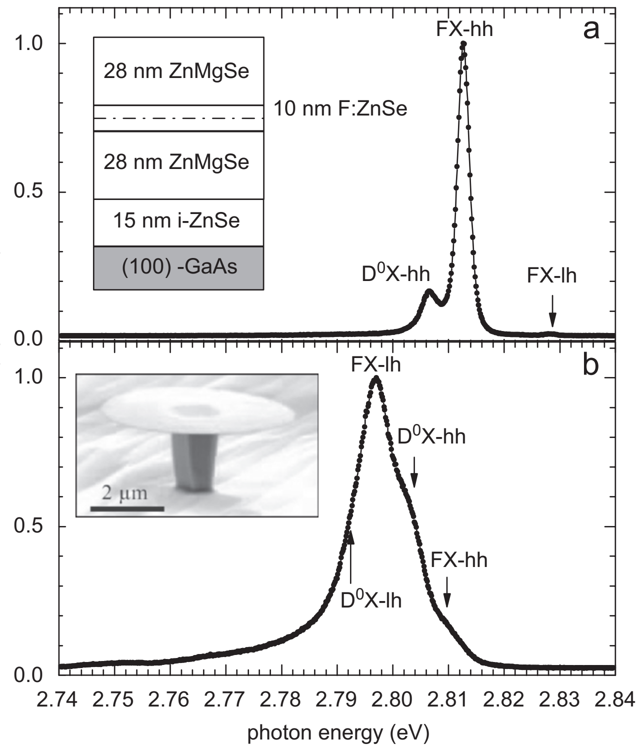

Fig. 1. PL spectra taken at 5 K. (a) PL for an unstructured F:ZnSe δ-doped QW sample as drawn in the inset. (b) PL of a 6 μm microdisk structure. Inset: Scanning electron micrograph picture of such a disk.

interface properties, a 15 nm buffer of undoped ZnSe was first deposited on the substrate, followed by 28 nm of ZnMgSe with Mg content of about 10%. The ZnSe QW was 10 nm thick (see Fig. 1a). For doped samples, the QW was δ-doped in the centre using a ZnF_2 evaporation cell with a net molecular flux of approximately $2\times10^9\,cm^{-2}\,s^{-1}$, equivalent to a sheet donor concentration of $8\times10^9\,cm^{-2}\,s^{-1}$. Low temperature photoluminescence (PL) spectra of the ^{19}F-doped samples were measured at 5 K, using pulsed excitation by a frequency doubled mode-locked Ti:sapphire laser operated at 409 nm wavelength and focused down to a spot size of about 15 μm. Microdisks with diameters of 3 and 6 μm were defined by photolithography, reactive ion etching and by wet chemical etching of the underlying GaAs substrate.

3. Results and discussion

Fig. 1a shows the PL spectrum of a ZnMgSe/ZnSe multilayer structure with a F:ZnSe δ-doped region in the centre of the ZnSe QW. The emission at 2.812 eV corresponds to recombination of heavy-hole free excitons (FX-hh) from the compressively strained ZnSe QW (lattice mismatch of $f_{ZnSe} = -0.25\%$); this energy is consistent with the finite barrier model in Refs. [17,18]. Weak light-hole free-exciton (FX-lh) emission at 2.829 eV is indicated by the arrow in Fig. 1a. The lower energy peak seen at 2.806 eV in Fig. 1a is due to the heavy-hole donor-bound-exciton (D^0X-hh) recombination as confirmed by the expected 6 meV separation from the corresponding FX state. The widths of these peaks indicate only small inhomogeneous broadening, likely due to strain effects. However, the broadening is still less than typically seen in quantum dot ensembles.

The PL spectrum of a microdisk made from the same multilayer structure is presented in Fig. 1b. The inset shows a scanning electron micrograph picture of the microdisk. The absence of cracks and lateral deformation indicates a homogeneous release of strain along the radial direction of the disks; however, due to the volume ratio of ZnMgSe and ZnSe, the ZnSe QW is likely to be tensile strained on ZnMgSe in the periphery of the disks. This results in a substantial band-gap narrowing effect along the radial direction of the disk from the centre to the edge that enhances the charge carrier transfer to the periphery, where the optical whispering gallery modes (WGMs) are localized. As a result of the tensile strain, the relative positions of the FX-lh and FX-hh energies are reversed as indicated by the arrows in Fig. 1b. Both states and their corresponding D^0X-transitions contribute to the PL. The transition energies are still consistent with the finite barrier model reported in Refs. [17,18] if a remaining induced tensile lattice mismatch of the QW of about $f_{ZnSe} = +0.2\%$ were considered in the free standing region of the disk. The FX-hh and D^0X-hh transitions at 2.810 and 2.804 eV are only slightly shifted and broadened; these peaks stem from PL close to the centre of the disk. The D^0X-hh luminescence is now stronger than the FX-hh luminescence due to recombination of FX at the surface. In the relaxed periphery, the FX-lh emission moves to lower energies of about 2.798 eV; the D^0X-transitions again appear about 6 meV lower than their corresponding FX transitions, but are substantially broadened due to the strain gradient across the disk.

The peaks, corresponding to WGMs, are not visible in low-power PL. We believe that absorption due to lower energy transitions in the inhomogeneously broadened distribution

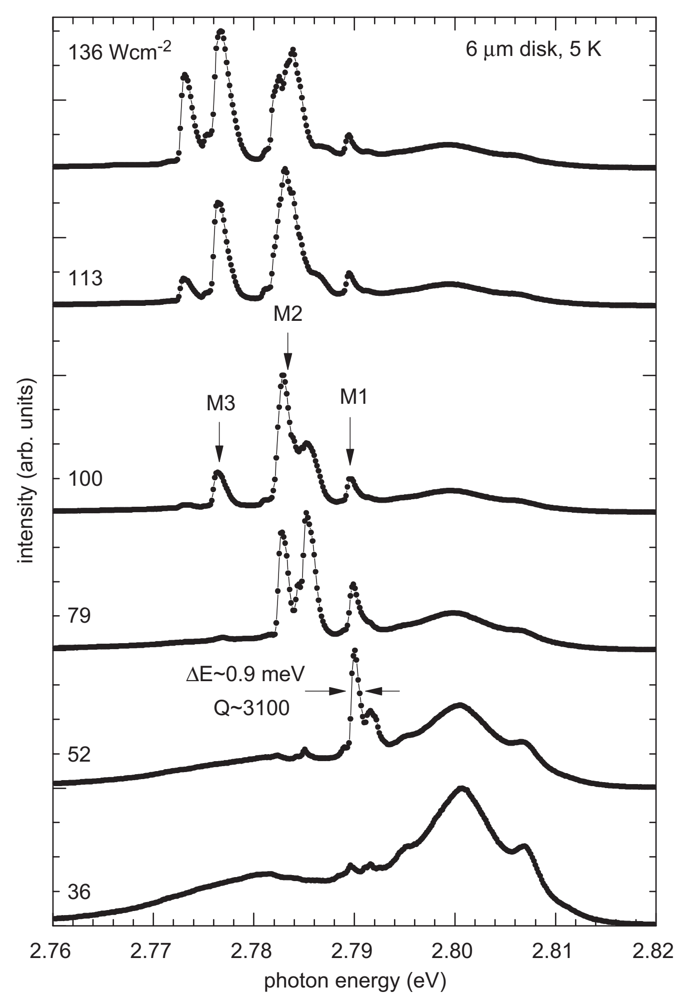

Fig. 2. Power-dependent PL spectra and lasing of a microdisk with 6 μm diameter. The excitation density was varied between 36 and 136 W cm^{-2}. For low excitation, the first WGM (mode M1) emerges at 2.79 eV and has a cavity-Q of about 3100. Further modes (M2 and M3) are observed with increasing excitation density.

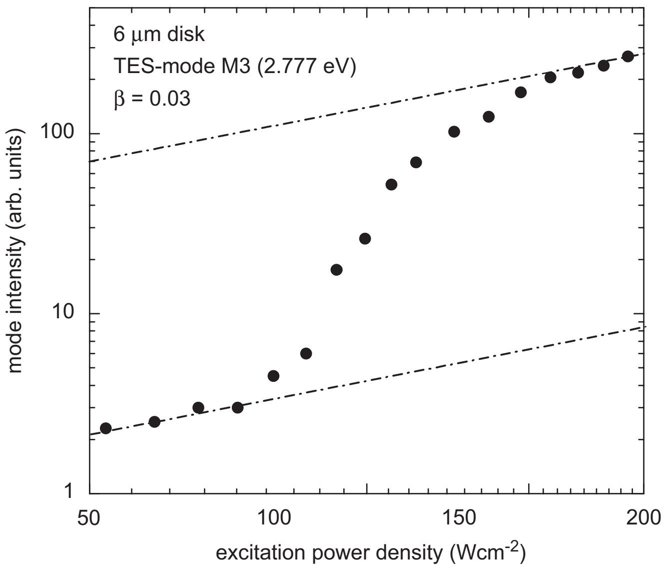

Fig. 3. Integrated intensity of the TES lasing mode M3 at 2.777 eV vs. the excitation power density. The onset of lasing is observed at a threshold of about 100 W cm^{-2}.

damps the cavity-*Q*. However, at higher excitation powers, these transitions become saturated and the cavity modes appear.

Fig. 2 shows a series of PL spectra taken from a microdisk with 6 μm diameter and measured with different average excitation intensities varying between 36 and 136 W cm^{-2}. For small power densities exceeding 36 W cm^{-2}, the superlinear increase of a group of two sharp peaks close to 2.790 eV is observed. These peaks correspond to WGMs of the microdisk excited by D^0X-lh emission. The line width of the dominating mode (M1 in Fig. 2) indicates a cavity-*Q* of 3100. Additional groups of modes emerge at the energies 2.783 eV (mode M2) and 2.785 eV for pump power densities larger than 52 W cm^{-2}. Further modes appear at 2.777 eV (mode M3) and 2.773 eV for pump power densities larger than 100 W cm^{-2}, respectively. These modes are too closely spaced to correspond to the free spectral range of the lowest order WGMs; multiple modes are seen more likely due to higher order radial modes of the disk. The strain gradient and slight warping of these disks prevent a simple quantitative evaluation of their spacing.

At moderate excitation density, the main intensity of the primary mode M1 is transferred to mode M2 and for high-excitation density, to mode M3. The energies of both modes M2 and M3 are resonant with the calculated D^0X-hh and the D^0X-lh two electron satellite (TES) transition energies [19]. The WGMs are only observed in the lower energy part of the spectra corresponding to emission from donor-bound excitons and their related transitions, resulting from the band gap narrowing at the periphery of the disk.

Fig. 3 shows the power dependence of mode M3 at 2.777 eV as a function of the pump power. A clear lasing threshold is seen at about 100 W cm^{-2}. The ratio of the slope of output power vs. input power indicates the fraction of spontaneous emission coupled into the lasing mode to be $\beta = 0.03$. Lasing was also observed in 3 μm microdisks revealing threshold power densities less than 20 W cm^{-2} for the D^0X-lh TES transition. The corresponding β-factor measured with 3 μm microdisks is 0.1 as shown in Ref. [20].

4. Conclusions

Microdisk cavities with 3 and 6 μm diameter were fabricated with pseudomorph strained and fluorine δ-doped ZnMgSe/F:ZnSe/ZnMgSe QWs. Although strain relaxation occurred in the free standing region of the microdisks, a high cavity-*Q* larger than 3000 is obtained. The appearance of low-threshold lasing in δ-doped ^{19}F:ZnSe QW microdisks indicates that ensembles of fluorine donors have been successfully coupled to a cavity with cooperativity factors high enough to achieve low-power density thresholds less than 100 W cm^{-2}. Such a reduced threshold also facilitates some schemes for quantum communication and quantum computation based on donor-bound-exciton transitions. The reduction of inhomogeneous broadening by preserving the uniform strain acquired during MBE growth may also allow such applications as lasing without inversion.

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