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Self-assembled quantum dots in a liquid-crystal-tunable microdisk resonator

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ABSTRACT

GaAs-based semiconductor microdisks with high quality whispering gallery modes ($Q > 4000$) have been fabricated. A layer of self-organized InAs quantum dots (QDs) served as a light source to feed the optical modes at room temperature. In order to achieve frequency tuning of the optical modes, the microdisk devices have been immersed in 4 – cyano – 4′–pentylbiphenyl (5CB), a liquid crystal (LC) with a nematic phase below the clearing temperature of $T_C \approx 34\,^{\circ}C$. We have studied the device performance in the temperature range of $T = 20-50\,^{\circ}C$, in order to investigate the influence of the nematic–isotropic phase transition on the optical modes. Moreover, we have applied an AC electric field to the device, which leads in the nematic phase to a reorientation of the anisotropic dielectric tensor of the liquid crystal. This electrical anisotropy can be used to achieve electrical tunability of the optical modes. Using the finite-difference time domain (FDTD) technique with an anisotropic material model, we are able to describe the influence of the liquid crystal qualitatively.

1. Introduction

Semiconductor microcavity devices have been demonstrated to be of great value to control light-matter interaction in the past [1]. Their applicability reaches from micro- and nanostructured low-threshold laser structures over single-photon sources to optical qubits based on exciton–photon–polaritons [3,4]. For many of the aforementioned applications, however, it is mandatory to achieve spectral resonance between the emitter, typically a quantum dot or quantum well layer which is sandwiched into the sample structure, and the frequency of the optical mode. The emitter frequency is in many cases extremely difficult to control precisely in the fabrication process, especially in the case of self-organized quantum dot (QD) growth, which always results in a finite size distribution of the QDs. The frequency of the optical modes is determined by the dielectric constants of the used materials and the geometry of the devices. However, even in a material system like GaAs, where today advanced fabrication techniques are available, it is still not possible to control the fabrication of optical resonators to such a degree that spectral resonance to a single QD within the natural linewidth of the dot is possible without applying an additional step in order to tune both lines into resonance. In the past, different tuning techniques have been applied, ranging from temperature tuning [3,4] to adsorption of liquid nitrogen on the sample surface [2]. In this contribution, we will present an approach for tunable semiconductor microcavities based on liquid crystal devices, as originally suggest by Busch and John [5]. First experiments have studied the effect of temperature on such hybrid devices [6,7].

2. Experimental details

As a starting material, we use a GaAs-based heterostructure as shown in Fig. 1 grown by solid-source molecular beam epitaxy (MBE). On a semi-insulating GaAs substrate, a 300 nm silicon-doped GaAs back contact layer is grown. Subsequently, a 500 nm $Al_{0.33}Ga_{0.67}As$- layer is deposited. This layer serves as a sacrificial layer and is removed later in a selective etch step to suspend the above lying membrane layers. The membrane itself consists of 240 nm GaAs with an embedded layer of InAs self-assembled quantum dots in the center. The In-content of the quantum dots is optimized, so that the QDs show strong photoluminescence (PL) signal at room temperature, as shown in the top of Fig. 3. From this material, microdisks with radii of $r = 3\,\mu m$ are fabricated using the

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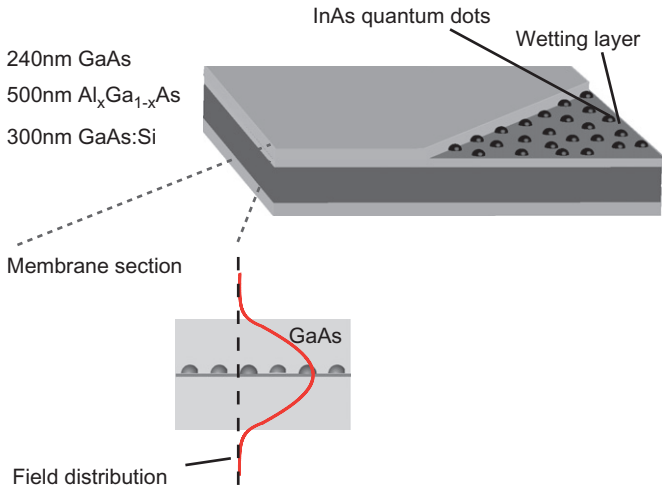

Fig. 1. Sample material for microdisk fabrication.

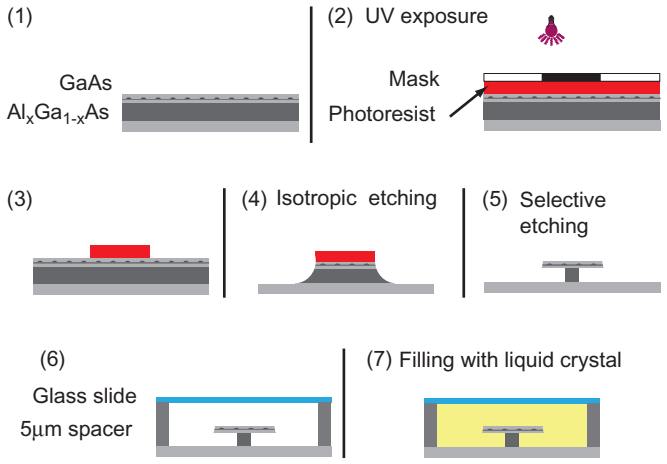

Fig. 2. Processing sequence for the formation of LC-filled microdisks.

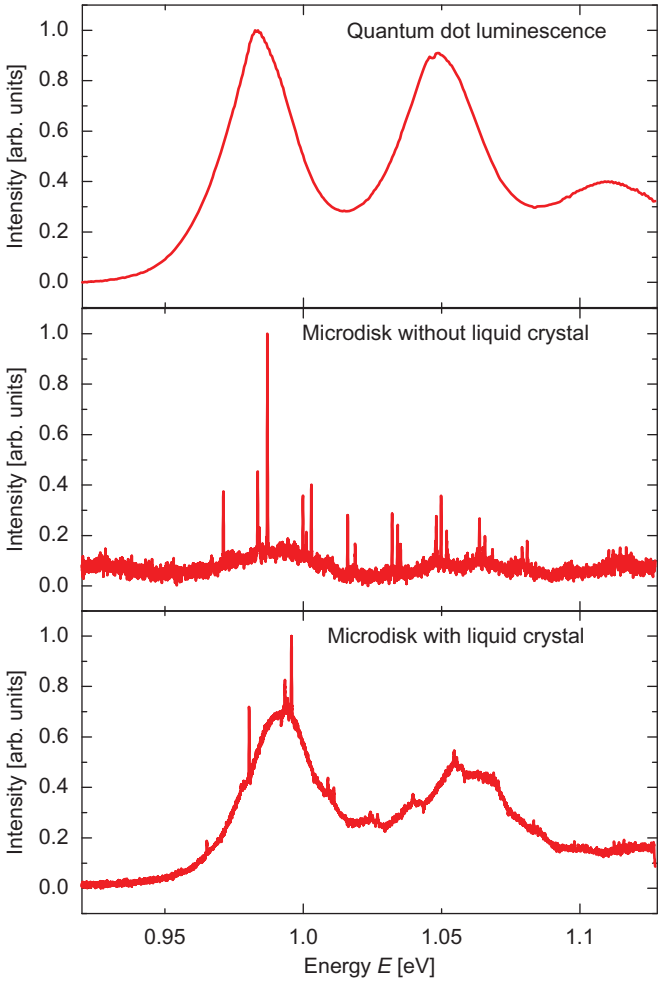

Fig. 3. PL spectra of (top) quantum dots at 300 K before sample fabrication, (middle) microdisk (with QDs) at 300 K and (bottom) microdisk immersed in liquid crystal.

process sequence shown schematically in Fig. 2. Firstly, using optical contact lithography circular mesas are defined using an isotropic etch step employing a HBr-based etchant. After resist removal, the structure is undercut by selectively etching part of the sacrificial $Al_{0.33}Ga_{0.67}As$ layer using a KI based etch [8]. To prepare the microdisk devices for the liquid crystal and promote the adhesion between the materials, the surface is pretreated in either MAP (*N*-mehtyl-3-aminopropyltrimethoxysilane) or PVA (polyvinyl alcohol). Then, a 5 µm thick spacer is added and the structure is immersed into the liquid crystal 4 – cyano – 4′–pentylbiphenyl (5CB). To contain the liquid crystal inside the device, the structure is covered by a thin glass slide. For the electric-field tunable devices, the structures were sandwiched between two slides coated with indium-tin-oxide (ITO). The samples are analyzed using micro-photoluminescence at room temperature using a Czerny-Turner monochromator with a focal length of 0.5 m and a liquid-nitrogen cooled InGaAs photodiode array. For optical excitation, a Ti:sapphire laser is used at a wavelength of $\lambda = 780$ nm.

3. Results and discussion

Firstly, the influence of the liquid crystal on the resonant cavity modes was investigated. The results are shown in Fig. 3. Before microdisk formation, the PL spectra of the quantum dots show a characteristic orbital shell structure with the s-, p- and d-shell clearly visible. The microdisk devices fabricated from this sample material show a series of sharp optical modes, originating from the Whispering Gallery Modes (WGM) of different radial order. The azimuthal quantum numbers of the optical modes are typically in the range between $M = 40$ and 50 due to the comparably large diameter of the disk. The observed quality factors are in the range of $Q = 4000-4500$, very close to the spectral limit of the used spectrograph/camera setup. After immersion of the device in the liquid crystal, the optical modes are significantly shifted spectrally, and the number of observed radial modes has significantly decreased. This is due to the fact that because of scattering and absorption of the laser light the excitation conditions at the sample are significantly changed from the reference measurements. However, the observed linewidth of the optical modes is not significantly changed within the instrumental limits.

In the next step, the influence of temperature tuning on the optical spectra was investigated. Therefore, the sample was first heated above the clearing temperature, then cooled down to $T = 295$ K and the LC molecules were aligned vertically using an external electric field. Then, the sample temperature was slowly raised up to $T = 325$ K while at each temperature, micro-PL spectra were recorded. The results for a selected spectral region are shown in Fig. 4. The dashed line indicates the initial position of a selected optical mode at $T = 300$ K for clarity. One can see that up to a temperature of $T = 307$ K, a drastic shift of the optical

mode frequency occurs, with a maximum shift of $\Delta E = 6.5$ meV. For temperatures above 307 K a much smaller shift in the opposite direction is observed. This behaviour can be explained with the nematic–isotropic phase transition of the liquid crystal, which occurs around 304 K and leads to a change in the dielectric function from unaxial optical anisotropy (birefringence) to an isotropic medium. The difference between the extraordinary/ordinary refractive index and the refractive index in the isotropic phase for 5CB is as large as $\Delta n = 1.55$ [11], which is the reason for the observed strong optical shift to higher energies. The subsequent shift to lower energies at a much lower temperature tuning rate is due to the change in the GaAs refractive index as a consequence of the thermal lattice expansion. These experiments allow the direct monitoring of the LC phase transition using photonic devices and demonstrate that LCs greatly enhance the tunability of semiconductor-based nanophotonic devices.

In the next step, the sample was sandwiched between ITO-covered glass slides. Between these electrodes, AC voltages up to 50 V could be applied. The use of AC voltages avoids electromigration of the liquid crystal molecules at elevated electric fields. Before the experiments, the sample was heated above the clearing temperature, in order to remove any remaining alignment along the vertical axis. However, it should be noted that after cooling down, there is still some remaining alignment in the parallel direction to be expected, as the molecules favor this alignment direction on the pretreated GaAs surfaces. After the sample has been cooled down below the clearing temperature, the voltage was slowly increased and the micro-PL spectra recorded simultaneously. The results are shown in Fig. 5. The recorded spectra clearly demonstrate the tunability of the optical modes by application of a suitable voltage, up to about 45 V, when the effect becomes saturated. Voltages above that range do not lead to a significant change of the mode frequencies any more, so that one can assume that the dipole-like molecules are nearly completely aligned along the electric-field defined vertical direction.

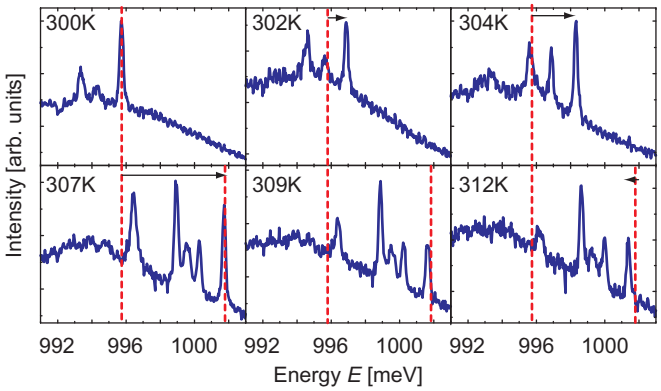

Fig. 4. Temperature dependent mode shift induced by reorientation of the liquid crystal.

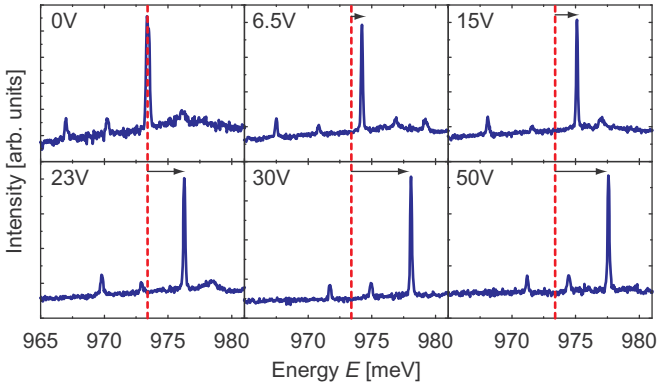

Fig. 5. Electric field dependent PL spectra.

The resonant cavity modes embedded in an anisotropic dielectric environment were simulated using an in-house developed finite-difference time-domain FDTD [9] code with standard CPML "open" boundaries. Subcell accuracy was achieved using a Finite Integration Technique (FIT). An ultrashort broadband pulse from a magnetic dipole source was used in order to excite the TE-like modes of the semiconductor cavity. The MIT "Harminv" software package based on the filter diagonalization method [10] was used to efficiently extract the frequency and decay rate of the cavity modes from the simulated electric fields.

In order to accommodate the effect of continuous reorientation of the optical axis, the simulations were performed for three extreme cases, i.e., for the case of the extraordinary optical axis in the *xy*- plane (parallel alignment of the LC molecules on the GaAs surface), for the case of full vertical alignment of all LC molecules, i.e., the extraordinary direction perpendicular to the sample direction, and for an intermediate partially disordered state, which was obtained by a linear interpolation of the dielectric tensors between the two abovementioned cases. The results are shown in Fig. 6. One can see that the results resemble the experimental findings for the case of electric field tuning qualitatively, if one indeed assumes that the molecules were fully aligned along the microdisk surface, as expected after the chemical pretreatment. However, the numerically predicted shift is about half the experimental observed shift, when directly comparing modes of similar azimuthal quantum number and frequency. This observed discrepancy between the experimental and theoretical results can be due to multiple reasons. Firstly, the intricacies of the fabrication process, especially the contact lithography, lead to fabrication imperfections that cannot completely be avoided, thus resulting in deviations from a perfect circular shape of the microdisk to start with, leading to scattering effects and thus increasing the fraction of the evanescent electric field that can interact with the liquid crystal. Moreover, the sidewall profile of the fabricated microdisk exhibits a slight sidewall angle due to the used HBr process. This slightly breaks the mirror symmetry in the *z*-direction and leads to an increased localization of the field in the lower part of the disk, closer to the edge. These effects can also enhance the sensitivity of the optical modes to the liquid crystal in the direct environment.

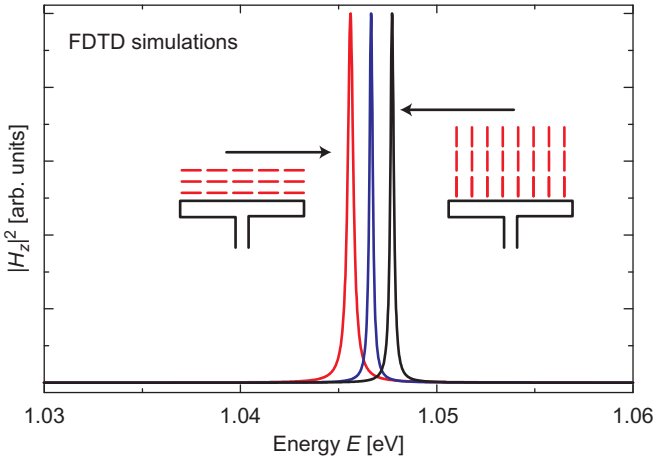

Fig. 6. Finite differences time-domain simulations of the microdisk devices embedded in an anisotropic dielectric material showing a mode shift induced by the orientation of the liquid crystal.

4. Conclusion

The experiments discussed above show that liquid crystal tunable semiconductor microcavity devices have great potential for applications at room temperature. Especially for tunable nanolaser devices this technique is highly attractive. A combination with photonic crystal defect cavities should allow for even more sophisticated devices due to the additional degree of freedom photonic crystals offer.

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