Strong mode coupling in InP quantum dot-based GaInP microdisk cavity dimers

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Abstract. We report on strong mode coupling in closely spaced GaInP microdisk dimer structures including InP quantum dots as the active medium. Using electron beam lithography and a combination of dry- and wet-etch processes, dimers with inter-disk separations down to \(d < 100\) nm have been fabricated. Applying a photo-thermal heating scheme, we overcome the spectral mode detuning due to the size mismatch between the two disks forming the dimer. We observe signatures of mode coupling in the corresponding photoluminescence spectra with coupling energies of up to 0.66 MeV. With the aid of a numerical analysis, we specify the geometrical and physical factors of the microdisk dimer precisely, and reproduce its spectrum with good agreement.

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Coupled microcavity resonators, the so-called photonic molecules (PMs) [1], have gained much interest for their promising applications in both classical and quantum optics [2]. Among them are improved response time microcavity lasers [3, 4], producing directional light emission [5] and sensing of a molecule's optical dipole moment [6]. Also the influence of the geometry of the individual cavities on the inter-cavity coupling [7] and a quality factor enhancement with respect to uncoupled cavities has been addressed [8, 9].

Whispering gallery mode (WGM) supporting microcavities such as microspheres [10] and microdisks (MDs) [11] are well-suited candidates for the formation of PMs as their evanescent fields allow for coherent mode coupling between adjacent cavities [12]. PMs consisting of closely spaced WGM supporting cavities have been reported for a range of material systems, e.g. dye-doped polymer biospheres [13], silica disk microresonators [14], for spherical cavity dimers with a shell of CdTe nanocrystals [12] and doped with CdSe QDs [15].

For many applications precise control of the resonance wavelength and the resonator distance on the nanometer regime is required. Due to fluctuations in processing parameters the spectral resonances of the individual cavities cannot be predicted with the accuracy needed for a perfect coupling of the cavity modes. Therefore an accurate external control of the coupling is highly desired. Two recent publications report on such an external control based on individual heating of one of two closely spaced GaAs MD cavities containing InAs QDs as an active medium [16, 17]. In both experiments, the GaAs MDs were completely underetched and put on a sapphire substrate where they were subsequently placed next to each other by means of micro-manipulation to form dimer structures. The close spacing between the disks allows for the formation of bonding and anti-bonding PM supermodes that are no longer confined to a single cavity but expand over the entire dimer structure.

However, scalable photonic quantum technologies will be based on wafer integrated optical cavities to ensure stable, controllable and repeatable operation. In addition, cavity embedded quantum emitters are particularly intriguing as these systems would enable optical quantum simulators using well-controlled multipartite quantum systems [18, 19] or the generation of sub-Poissonian light from coupled cavity modes [20, 21].

In this paper, we demonstrate strong mode coupling in closely spaced InP quantum dot-based GaInP MD cavities grown on a GaAs wafer. While we use here the QD ensemble as an internal light source to probe the density of states of the WGMs, we envision the possibility of using similar systems, i.e. individual addressable QDs in coupled MD cavities, to mediate a coherent coupling between them via the super-modes of a PM [22]. By a photo-thermal
heating scheme, we overcome the spectral mismatch of WGMs originating from a slight size mismatch between the MD cavities of the dimer structure [16, 17]. We can tune the modes originating from different MDs into resonance, where we observe an avoided spectral crossing in the corresponding photoluminescence (PL) spectra, which is characteristic of the strong mode coupling in a PM. Our experimental findings are well reproduced by a numerical simulation. We identify the modes involved in strong couplings with their mode numbers and obtain the change of the refractive index in the process of experimental thermal heating.

2. Experimental: growth, structuring and the optical experiment

The structure under investigation in this work is based on an epitaxial heterostructure grown on a (100) GaAs substrate miscut toward the [111]A-direction by 6° using metal-organic vapor-phase epitaxy. Initially, a 100 nm GaAs buffer layer was deposited followed by a short period superlattice consisting of 30.5 pairs of 7 nm GaAs:Si and 7 nm of (Al0.5Ga0.5)As:Si terminated by 200 nm GaAs. The superlattice is employed to form a smooth surface for the subsequent layers. As a sacrificial layer for the aluminum-selective wet-etching process, 1 µm of (Al0.5Ga0.5)As:Si is included. For the QDs forming the active layer of the MDs 2.1 monolayers (ML) of InP were deposited with a growth rate of 1.05 ML s⁻¹ at a growth temperature of 710°C leading to a monomodal height distribution of the QDs with a structural surface density of ∼1.5 × 10¹⁰ cm⁻². Further information on the QD growth process can be obtained from [23].

The QDs are embedded in a separate-confinement heterostructure (SCH) to optimize both electronic and optical confinement. The SCH is formed by two sequences of 10 nm of (Ga0.5In0.49)0.5P0.5, 59 nm of ((Al0.5Ga0.5)0.51In0.49)0.5P0.5, 35 nm of ((Al0.2Ga0.8)0.51In0.49)0.5P0.5 and finally 40 nm of (Ga0.51In0.49)0.5P0.5 all lattice matched to GaAs and symmetrically arranged around the QD layer. This structure is a result of a numerical optimization of the cavity design with regard to a smoother vertical mode confinement on the basis of a transfer matrix formalism. The active layer together with the SCH constitutes the actual disk layer, whereas remnants of the sacrificial layer form the posts that support the disks.

In order to fabricate high-quality MDs with a well-defined sub-100 nm gap in between, electron beam (Ebeam) lithography was used to define the patterned mask for the subsequent etch. A hydrogen silsesquioxane-based Ebeam resist XR-1541 by Dow Corning Co. was exposed to a high-energy (100 kV) electron flux in an Elionix Ebeam lithography system. The exposed area then forms amorphous silicon oxide and serves as an etch mask in the subsequent inductively coupled plasma reactive ion etch. In the etch system a hydrogen bromine (HBr)-based plasma was generated and directed toward the patterned wafer piece using a bias to directionally etch on the non-masked region, forming circular pillars with very smooth and vertical side walls. The etch depth was chosen to be approximately 1 µm to ensure sufficient exposure of the sacrificial material to the following wet chemical etch. After the dry etch and removing of the exposed resist, an undercut etch was performed by soaking the sample into Transene aluminum etchant type A (a solution of phosphoric acid, nitric acid and acetic acid) which dissolves only the high-aluminum-content sacrificial material, providing the optical isolation between the MD and the substrate. Scanning electron microscopy (SEM) images of the fabricated structure are presented in figure 1, where two 2 µm-diameter MDs are placed close to each other with a gap of approximately 90 nm.

For the optical experiments the sample was mounted in a helium flow cold finger cryostat as depicted in figure 2(a). The sample temperature with this elongated cold finger can be set
Figure 1. (a) SEM pictures of a processed dimer structure taken from the side. (b) SEM picture taken from the top revealing MD diameters of $\sim 2.12 \, \mu\text{m}$ and inter-disk separation of $\sim 90 \, \text{nm}$.

Figure 2. (a) Cryostat scheme with optical access to the sample via two axes from the top and from the side, respectively. (b) Schematic diagram of the optical experiment displaying the excitation/detection beam path being directed toward one of the disks of a dimer. This alignment can be checked via a top view of the sample in the cryostat acquired via the detection of the light collected by the monitoring objective displayed in subfigure (a). The top part of subfigure (b) illustrates the scanning of the MD into the focus of the Gaussian beam of the excitation/detection beam path.

within a range of $T = 15$–$300 \, \text{K}$. The cold finger is mounted on three referenced stepping motor-driven translation stages inside the recipient for both the lateral sample positioning ($x$- and $y$-directions) and an adaption of the focal plane ($z$-direction) with respect to the
‘excitation/detection’-objective with a minimum step size of approximately 100 nm. Our cryostat geometry allows us to optically access the sample in perpendicular directions (z and y) via two 50× microscope objectives with a working distance of 17 mm. The horizontally mounted objective is part of a reflective micro-PL setup, i.e. the laser excitation and the collection of the PL signal are performed through the same horizontal beam path. As illustrated in the lower part of figure 2(b) the sample is mounted in a way that the horizontal beam path points at the sides of the MD dimers, while the position of the excitation laser spot can be monitored with the vertically mounted objective by collecting stray light from the top of the sample, which is subsequently detected via an imaging charge-coupled device (CCD). The PL signal collected through the horizontal objective is directed into the detection beam path by a non-polarizing beam splitter and sent to an imaging spectrometer with a focal length of 550 mm equipped with an LN$_2$-cooled CCD camera recording the dispersed PL signal.

3. Results

In the fabrication of the dimers the diameters of the two disks unavoidably differ on a nm scale. This results in variation of the emission wavelength/energy for WGMs of identical azimuthal and radial mode number from disk to disk. We therefore need a technique to tune MD dimer modes into resonance to achieve strong mode coupling. In our experiments, we adapt the technique presented in [16, 17], in which a laser is used to both optically excite and heat the MDs. For this purpose, we deploy a diode pumped solid-state laser system emitting at $\lambda_{\text{exc}} = 532$ nm ($E_{\text{exc}} = 2.33$ eV) well above the GaInP barrier material band gap at $E_{\text{GaInP}}(T = 15$ K) $\sim 1.99$ eV. To achieve a resonant coupling between the disks of a dimer, we point the excitation beam path to the smaller of both the disks as depicted in figure 2(b) and increase the optical power absorbed by the disk by vertically (y-direction) scanning the respective disk, stepwise into the focus of the laser. With an increasing scanning step number, we increase the temperature in the respective disk, which results in an increase of the refractive index of the disk layer and thus in a red-shift of the mode emission. A drawback of this technique stems from the Gaussian intensity profile of the focused laser beam, resulting in a nonlinear correlation between the scanning step and the absorbed optical power and therefore the refractive index change. Nevertheless, the scheme allows for the heating of one MD of a dimer and, therefore, provides a means of demonstrating the fundamental feasibility of strong mode coupling in our structures.

Following the above outlined procedure, we scanned a dimer consisting of 2 $\mu$m diameter MDs through the focus of the excitation beam path while the beam path points at one of the disks. For every scanning step a spectrum was recorded. The top part of figure 3(a) displays normalized spectral data recorded at the beginning of the scanning series, i.e. at $T \sim 15$ K. The spectrum contains six dominant lines labeled M1–M6 with increasing wavelengths (decreasing emission energies). At the high applied excitation powers ($P > 100$ W cm$^{-2}$), we can readily assign them to mode emission. The bottom part of figure 3(a) displays a logarithmic color map composed of a scanning step series of normalized spectra. With increasing the scanning step number in the mode pair M1–M2 initially exhibiting $\lambda_{\text{M1}} = 657.50$ nm ($E_{\text{M1}} = 1.8857$ eV) and $\lambda_{\text{M2}} = 658.09$ nm ($E_{\text{M2}} = 1.8840$ eV), respectively, we observe a redshift for M1 whereas such a redshift is not perceivable for M2. Between the scanning steps 12 and 17 M1 and M2 are close to resonance and the scanning step dependence of the two lines gradually changes.
Figure 3. (a) The top graph displays normalized spectral data for the first scanning step, i.e. for $T \sim 15$ K, including multiple spectral lines labeled M1–M6 with increasing wavelength (decreasing emission energy). The bottom part contains a logarithmic color map composed of a scanning step series of normalized spectra. (b) Spectra for the scanning steps 1, 12, 15 and 23 corresponding to the initial situation, the anti-crossing between M3 and M4, the anti-crossing between M1 and M2 as well as between M5 and M6 and after the anti-crossings, respectively.

While the line initially labeled as M1 now shifts slower and finally completely loses its redshifting behavior, the opposite is observed for M2. This results in an avoided crossing between M1 and M2 which is characteristic of strong mode coupling in a PM [16, 17]. At higher scanning step numbers the redshift saturates and is even reversed when the maximum overlap between the disk and the excitation beam is transgressed and the temperature in the heated disk subsequently decreases.

In the multiplet including the lines M3–M6 one can perceive two different situations. The pairs M3–M4 ($\lambda_{M3} = 660.93$ nm ($E_{M3} = 1.8759$ eV), $\lambda_{M4} = 661.35$ nm ($E_{M4} = 1.8747$ eV)) and M5–M6 ($\lambda_{M5} = 661.78$ nm ($E_{M5} = 1.8735$ eV), $\lambda_{M6} = 662.17$ nm ($E_{M6} = 1.8724$ eV)) exhibit the above-described avoided crossing while M3 and M6 cross each other. Figure 3(b) displays the spectra for the initial condition (step 1), for the avoided crossing of M3 and M4 (step 12), for the avoided crossings of both M1 and M2 as well as M5 and M6 and finally after the anti-crossings. Especially for the mode pairs M1–M2 and M5–M6 we observe a switching in the line widths before and after the avoided crossing, which is again characteristic of a strong mode coupling. In the case of the pair M3–M4, due to the initially very similar line shape, this effect is not directly perceivable due to a strong fluctuation in the line widths.

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So far, from the overall scanning step dependence and the anti-crossing/crossing behavior of the observed modes, we can conclude that the modes M1, M3 and M5 originate from the heated disk, whereas M2, M4 and M6 are located in the disk outside the focus of the excitation beam path. Furthermore, we conclude that the pairs M1–M2, M3–M4 and M5–M6 each represent modes with the same azimuthal and radial mode number but located in different disks. Previous publications [16, 17] have shown that such mode pairs are most likely to form the bonding and anti-bonding supermodes of an MD PM, while the modes with differing mode numbers are less likely to be strongly coupled, which manifests in the observed crossing.

To analyze the observed line anti-crossings, we apply a phenomenological 2×2 matrix model. According to the model which is described in detail in [24], the eigenenergies of a coupled mode system as a function of a real parameter \( P \) are given by

\[
E_{\pm} = \frac{E_1(P) + E_2(P)}{2} - i\frac{\Gamma_1 + \Gamma_2}{4} \pm \frac{1}{16} \sqrt{\left(\frac{\Gamma_1 - \Gamma_2}{2}\right)^2 + 2i\left(E_1(P) - E_2(P)\right)^2}.
\]

Note that, to rely on experimentally accessible quantities, in equation (1) the loss rates \( \gamma_j \) have been substituted by the modes’ full-width at half-maximum (FWHM) \( \Gamma_j = 2\gamma_j \). The real parts of \( E_{\pm} \) correspond to the experimentally observed mode energies. From these values we extract \( E_j \) which correspond to the mode energies of the uncoupled system. \( g \) is the coupling energy and the two MDs of the dimer and their corresponding modes are indicated by the index \( j = 1, 2 \). Approaching resonance, i.e. for \( E_1(P) \approx E_2(P) \), we can distinguish two cases. For \( 4g > |\Gamma_1 - \Gamma_2| \), we observe an avoided crossing in the real parts of \( E_+ \) and \( E_- \). These real parts physically correspond to the energies of the anti-bonding and bonding supermode of a PM. For the opposite case, i.e. for \( 4g < |\Gamma_1 - \Gamma_2| \), there is no strong mode coupling, resulting in a crossing of the real values of \( E_{\pm} \) and therefore the experimentally observed mode energies. To fit equation (1) to our experimental data, firstly we identify the real parameter \( P \) as the scanning step number from the scanning step PL series. A natural approach would be to find an identity yielding the temperature corresponding to a particular scanning step. This procedure founders on the intricacy to precisely determine the optical power absorbed in the MD for a particular scanning step. Therefore, secondly, we have to find the dependence of the \( E_j \) identified as the modes’ off-resonant emission energies of the scanning step number. This dependence could well be approximated by third/fourth-order polynomials. Thirdly, we identify the \( \Gamma_j \) as the FWHM of the modes at low excitation/heating powers. Thus, \( g \) remains the last free fitting parameter.

In figure 4, we present fits to the anti-crossings identified in figure 3. Following the above-described procedure of determining \( E_j(P) \) and \( \Gamma_j \) where \( j = 1 \) (j = 2) corresponds to the high (low) energy line at the initial experimental conditions and using the coupling energy \( g \) as the fitting parameter, we could well reproduce the experimental data. The solid black (red) lines represent the real parts of the eigenenergies \( E_{\pm} \) of the coupled system, whereas the black-(red-) dashed lines represent the eigenenergies \( E_j \) of the uncoupled system. The modes M3 and M4 are clearly uncoupled for the initial experimental conditions manifested by \( E_j = \text{Re}[E_{\pm}] \). In this case, the fitting yields a coupling energy \( g_{\text{M3–M4}} = 164 \pm 10 \mu\text{eV} \). The mode pairs M1–M2 and M5–M6 are subject to a certain degree of coupling already at the initial conditions, which manifests in \( E_j \neq \text{Re}[E_{\pm}] \) for the beginning of the scanning step series. In these cases, the coupling energies were determined to much larger values of \( g_{\text{M1–M2}} = 660 \pm 50 \mu\text{eV} \) and \( g_{\text{M5–M6}} = 510 \pm 50 \mu\text{eV} \). The larger errors are due to a greater uncertainty in the determination of the parameter dependences \( E_j(P) \).
4. Numerical analysis

The experimental spectra of the optical dimer are theoretically and numerically addressed. In the process of establishing a theoretical model, the geometrical and physical factors of the dimer are precisely specified, such as the difference of two MDs’ diameter and the refractive index of the dimer as a function of the scanning step, and each mode involved in strong couplings is identified with its radial and azimuthal mode numbers. The result of the numerical analysis reproduces the experimental spectrum reasonably. Theoretical and numerical analyses are undertaken with the following steps.

Firstly, the effective refractive index is estimated. Since the optical dimer in experiment is fabricated to be very thin, we assumed that all the modes in the spectrum have no excitation in the $z$-direction; hence the dimer can be treated as a two-dimensional object in numerical and theoretical analyses. Then, by computing the ground state of its wave guide modes the effective refractive index can be derived. Strictly speaking, the approach treating the MD as a wave guide has an intrinsic limit, because WGMs are formed around the boundary of an MD. However, this approach has shown reasonable agreement with experiments in many works and accordingly has been used broadly \[25, 26\]. In the derivation, we averaged the refractive indices of the vertical layers of MD, and used this value as the refractive index of the plate.

Secondly, the mode numbers of the mode in the spectrum and the refractive index of the dimer are specified by the comparison of the experimental spectrum with the analytically calculated spectrum of a single MD. For the spectrum of the single MD, the radius of the MD is set as measured by SEM ($R = 1060$ nm). Then, taking the estimated refractive index in the previous step as an initial condition, the refractive index of the single MD is finely adjusted, and at each value of the refractive index the spectrum of a single MD is computed in the vicinity of the modes in the experimental spectrum and compared with them. For this comparison, the splitting of each mode pair is used as another criterion besides the wavelengths of the modes. Since the magnitude of splitting increases with the radial mode number of modes involved in the coupling, the radial mode numbers should be assigned in the order of M4, M6 and M2. With respect to the two criteria, the splittings and the wavelengths of the modes, the best agreement is
obtained at the refractive index 3.35 for the azimuthal and radial mode numbers (20, 3) of M6, (28, 1) of M4 and (17, 4) of M2. The WGM resonances of our MD structures have previously been shown to exclusively exhibit transversal electric polarization [27]. We therefore arrive at the mode assignment as stated in figures 5(a)–(c).

Thirdly, using the identified mode numbers and the refractive index, the optical dimer is numerically modeled and analyzed. As in the experimental setup described in the previous section, different radii, \( R \) and \( 0.993R \), are assigned to the two microcavities of the optical dimer. The difference of two radii is deduced from the spectrum at scanning step \( P = 0 \), where the refractive indices of two microcavities are supposed to be the same. As was done in the experiment, the refractive index of the smaller microcavity is increased step by step, and at each step the spectrum of the dimer is computed. The refractive index is obtained close to the value estimated by averaging layers of the sample. At \( n = 3.3524 \), the spectrum demonstrates three anti-crossings. By comparing the splittings of them with those in the experimental data, the gap between two microcavities is adjusted to \( d/R = 0.06 \). This corresponds to an effective inter-disk separation \( d = 64 \) nm and is within the range of values determined by SEM measurements on the sample.

For the last step, the spectrum evolution with a changing refractive index is numerically calculated by applying the ‘boundary element method’ [28] to solve Maxwell’s equation for the

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\]
identified geometry. In this process, material absorption is not considered, because we verified that resonant wavelengths are almost unchanged by the absorption, although their widths are broadened. As the spectral range investigated in this work is narrow enough, chromatic dispersion is not taken into account either. Then, by curve fitting of the smoothly connected lines around the anti-crossings in figure 3, the change of the refractive index is obtained as a function of the scanning step (figure 5(e)). For the fitting, we use the function \( n(P) = (P/a_1) a_2 + 3.35 \), where \( P \) is the scanning step in the experiment, \( n \) is the effective refractive index and \( a_1 \) and \( a_2 \) are fitting parameters. We thereby obtain the numerically calculated spectrum evolution with respect to the experiment scanning step displayed in figure 5(d). Despite the overall agreement of the spectrum, a discrepancy of around 1 nm is seen in M1 and M2. We speculate that this is caused by the effect of the boundary and the pillars of the MDs on the WGMs.

5. Summary

In summary, we report on the fabrication of closely spaced InP quantum dot-based GaInP MD dimer structures. Using Ebeam lithography and a combination of dry and wet etching, we succeed in processing dimers with an adjustable inter-disk separation. To overcome the unavoidable fluctuations in MD diameters in the dimers, we use a laser excitation scheme that allows us to individually heat one MD of a dimer. Thus, by the observation of characteristic mode emission energy anti-crossings, we can show strong mode coupling and therefore the formation of PM supermodes in our MD dimers. The experimental findings could be well reproduced applying both a phenomenological \( 2 \times 2 \) matrix model and a numerical \textit{ab initio} analysis based on the boundary element method.

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