A comparison between experiment and theory on few-quantum-dot nanolasing in a photonic-crystal cavity

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Abstract: We present an experimental and theoretical study on the gain mechanism in a photonic-crystal-cavity nanolaser with embedded quantum dots. From time-resolved measurements at low excitation power we find that four excitons are coupled to the cavity. At high excitation power we observe a smooth low-threshold transition from spontaneous emission to lasing. Before lasing emission sets in, however, the excitons are observed to saturate, and the gain required for lasing originates rather from multi-excitonic transitions, which give rise to a broad emission background. We compare the experiment to a model of quantum-dot microcavity lasers and find that the number of excitons that must be included to fit the data largely exceeds the measured number, which shows that transitions involving the wetting layer can provide a surprisingly large contribution to the gain.

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1. Introduction

Lasers can deliver coherent, narrow-band, and single-mode light and have become ubiquitous in contemporary technology. Nanometer-scale lasers may offer significant advantages because of large spectral separation of modes, compact sizes, and because the Purcell effect enables lowering the lasing threshold [1]. Nanolasers may find applications in many areas including chip-to-chip optical communication [2, 3], on-chip optical signal processing [4, 5], and bio-chemical sensing [6, 7].

By virtue of the Purcell effect, the fraction of spontaneously emitted photons being emitted into the cavity mode, i.e., the β-factor, can be enhanced by either increasing the Q-factor or decreasing the mode volume of the cavity, which also increases the stimulated-emission rate [8]. Ultimately, the β-factor may approach unity [9], which implies that no threshold appears in the emitted intensity as a function of excitation power. Self-assembled quantum dots (QDs) embedded in photonic-crystal cavities provide a seemingly ideal setting to study nanolasing because of the simultaneous confinement of excitons and photons at the nanoscale [10]. Few-QD lasing in this system was first demonstrated by Strauf et al. [11] and single-QD lasing was recently reported [12, 13]. However, a stark discrepancy between atomic laser models and experiment was also found [11]: lasing was observed even when the excitons were strongly detuned from the cavity. This non-resonant coupling has been the subject of intense experimental [14–18] and theoretical [16, 19, 20] investigations and it is now understood that the dominant mechanisms are phonons at small detunings [18, 21] and, at large excitation powers and/or large detunings, multi-excitonic configurations due to hybridization with the wetting-layer states [16, 17], which may also be explained as Auger processes involving wetting-layer states [22]. It should be noted that some of the observed effects are particular to specific types of QDs and/or excitation schemes, e.g., QDs grown by droplet epitaxy [23] or positioned growth [21] do not have a wetting layer and have correspondingly different carrier-trapping dynamics.

Traditionally, laser oscillation is modelled by rate equations [24], which in their standard form do not include neither the modified spontaneous and stimulated emission due to the cavity nor the many-body effects in the QDs. It is only in recent years that a semiconductor model for QD-based microcavity lasers has been developed [25–28]. However, a quantitative comparison between this model and photonic-crystal nanolasers at the few-QD level is still missing. In this work, we demonstrate few-QD nanolasing in photonic-crystal nanocavities and compare it to a semiconductor-laser model. The comparison to theory shows that our experimental results cannot be attributed to usual intra-QD exciton transitions. Rather, the gain orginates from wetting-layer-mediated processes, which lead to a surprisingly high gain.

2. Measurements of few-quantum-dot lasing

We have studied nanocavities obtained by leaving out three holes in a triangular-lattice photonic-crystal, i.e., L3-cavities [29] as shown in the inset of Fig. 1(a). The photonic-crystal lattice constant was 260 nm and the hole radius 66 nm. The membranes were 154 nm-thick and contained a single layer of embedded InAs QDs grown by the Stranki-Krastranov method with a density of 80 μm^{-2} and inhomogeneous broadening of 50 nm resulting in approximately 1 QD per 1 nm-bandwith in the area of the cavity. We characterized the samples by confocal diffraction-limited microphotoluminescence spectroscopy at a temperature of 10 K. Figure 1(a) shows the emission spectrum of one cavity obtained with high-power above-band excitation at a wavelength of 800 nm; the six modes (denoted M1-M6) characteristic of the L3 cavity are efficiently excited by using the QD-ensemble as an internal broadband light source. Among all the cavity modes, the fundamental mode, M1, is the mode of our interest for laser oscillation since it has the highest Q-factor and the smallest mode volume.

In order to investigate how many exciton states are coupled to the M1-cavity mode, we

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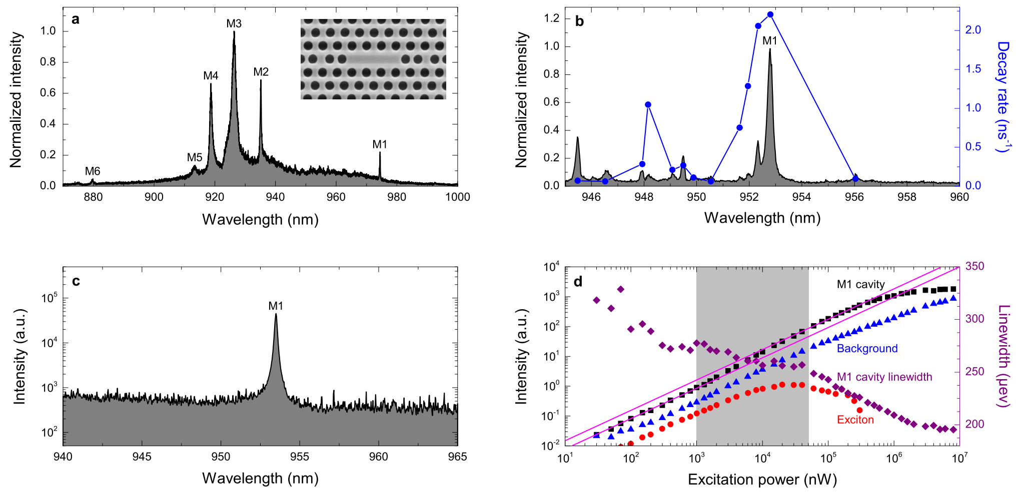

Fig. 1. Few-QD nanolasing. (a) Spectrum of an L3 cavity with embedded QDs at high-power (1 mW) above-band excitation showing the characteristic cavity modes denoted M1-M6. The inset shows a scanning-electron micrograph of the sample. (b) Spectrum (black curve; left axis) around the M1-cavity obtained with M6-resonant excitation at 100 nW showing the M1 cavity mode and a number of excitonic peaks whose decay rates (blue circles; right axis) have been measured using time-resolved spectroscopy. (c) Spectrum around M1 obtained with M6-resonant excitation at 100 µW showing the M1-cavity peak on a pronounced background. (d) Integrated intensity as a function of excitation power for the M1-cavity mode (black squares), an exciton peak (red circles), and the background (blue triangles). The smooth s-shaped transition between linear regions (magenta lines) of the cavity power dependence is a characteristic signature of high-β lasing. Evidently, the exciton saturates around threshold (gray region) and cannot provide the gain for the laser, which is provided by the background. The linewidth (purple diamonds; right axis) shows a small narrowing consistent with high-β lasing.

applied a low excitation power of 100 nW (measured before the microphotoluminescence objective) and recorded the spectrum around M1. Here and in the following we employ M6-cavity-resonant excitation in order to reduce undesired excitation of QDs outside the cavity. The decay rates of the exciton peaks were obtained by time-resolved spectroscopy. The result is shown in Fig. 1(b), where many single-exciton lines together with the M1-cavity mode are clearly observed. Four exciton lines are found to have significantly enhanced decay rates (faster than 0.5 ns^{-1}) due to the cavity as compared to the remaining excitonic states that are strongly inhibited by the band gap of the photonic crystal [30]. The power dependence of the four lines showed that they were due to single excitons.

The results shown in Fig. 1(b) seem to indicate that only four exciton states feed the cavity but this conclusion is incorrect because lasing sets in only at much higher excitation powers. For an excitation power of 100 µW, the excitons are saturated and the spectrum is dominated by the cavity mode on a broad background as shown in Fig. 1(c). The background is attributed to multi-excitonic transitions involving the wetting layer [1, 16, 22]. We show below that at this excitation power the laser is above threshold.

The excitation-power dependence of the different spectral features is shown in Fig. 1(d). The M1-mode exhibits an s-shaped variation with excitation power bounded by linear regions and, for high excitation power, saturation, which is characteristic of high-β lasing with a smooth

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threshold around the region marked in gray in Fig. 1(d). At high power levels the emission saturates. All four excitons begin to saturate for an excitation power of approximately 9 μW; experimental data for one of them is shown in Fig. 1(d). This means that the excitons saturate below the threshold, indicating that they are not (solely) providing the gain for lasing. The background integrated over a spectral range corresponding to the linewidth of the cavity (measured off resonance), on the other hand, does not saturate, which could indicate that the gain is actually provided by the background, i.e., the multi-excitonic states. This assertion is corroborated by our theoretical analysis described later. The fact that the cavity saturates before the background could originate from the limited rate of carrier scattering into the spectral region relevant for the cavity mode; a detailed microscopic theory including wetting-layer states would be needed to verify this but unfortunately such a model does not exist at present. The cavity linewidth decreases with increasing excitation power but only a modest linewidth narrowing is observed because for high β, a significant fraction of the photons in the cavity are spontaneously emitted resulting in a reduced coherence as compared to macroscopic low-β lasers [1, 26]. An often encountered challenge in experimentally probing QD-nanolasers is that their properties may resemble that of light-emitting diodes because their coherence time can be very short and their input-output curve exhibiting only a slight s-shape as opposed to a clear threshold. In particular, one could speculate that biexcitons, which can have a strong impact on the input-output curve [31], could give rise to the observed effects. However, we note that biexcitons cannot explain the linewidth behavior in our experiment.

3. Comparison between experiment and theory

We model our data by a microscopic QD-laser model introduced by Gies et al. [25]. Within the cluster-expansion technique, equations of motion for the photon number in the cavity and for carrier occupation numbers are derived, which in a consistent way include higher-order correlations, such as carrier-photon and photon-photon correlations. This method has been used to study input-output characteristics as well as photon statistics in QD-based microcavity systems [32] allowing to extract, e.g., the β-factor. It has also been used to estimate the effective number of QD-excitons emitting into the lasing mode. Here we use this model to estimate the effective number of excitons that would be needed to provide the gain in the absence of multi-excitonic transitions. While this is not a rigorous approach, we believe that this is the most useful procedure, since a fully microscopic model for QD-nanolasers including the inhomogeneously broadened QD ensemble and the wetting-layer carrier dynamics is not available at present.

The input-output curve resulting from the procedure outlined above is shown in Fig. 2, where the theoretical curves have been scaled with arbitrary but constant factors along both axes to take into account the excitation and detection efficiency of our experimental setup. Evidently, the gain provided by four excitons is insufficient to reach the lasing threshold. To quantitatively provide an insight into the relative strength of the excitonic and multi-excitonic gain, we model the multi-excitonic gain as coming from effective excitonic emitters. We therefore increased the number of excitons in the model, assuming a light-matter coupling strength equal to the average of the values obtained for the four excitons observed (9.8 μeV). We found a good agreement between the experiment and the model by using 120 excitons resulting in a β-factor of 0.4. This points to the fact that lasing in this device is indeed rather driven by the strong background emission observed directly in Fig. 1(c) than by actual intra-QD transitions. This is consistent with Fig. 1(d), where the exciton emission quenches whereas the integrated background emission shows a region of superlinear increase. The background is too spectrally wide to stem from phonon coupling and indicates rather transitions involving the continuum in the wetting layer and thus not excitons but rather multi-excitonic states [16, 17, 22]. We conclude

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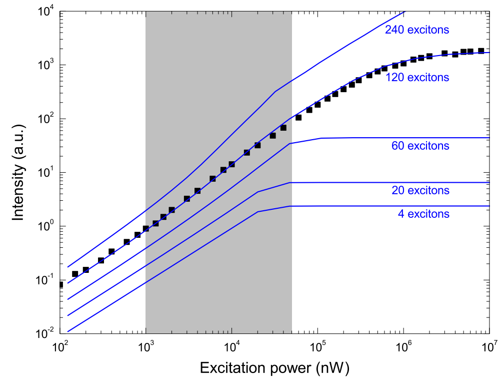

Fig. 2. Comparison of the experimental (black squares) and theoretical (blue lines) input-output curves. Clearly, the model cannot fit the data for small numbers of emitters although only four excitons were found to be coupled to the cavity at low excitation power. By varying the number of emitters in the model as indicated in the plot we find that 120 emitters must be included to fit the experiment. The theoretical curves have been vertically offset for clarity.

that a gain equivalent to that of 120 excitons is required to model our data, which, surprisingly, is 30 times larger than the number of excitons coupling to the cavity at low excitation power. The actual number of electron-hole states involved in the lasing oscillation may be orders of magnitude larger but with correspondingly lower average light-matter coupling strengths. The development of more sophisticated microscopic models is required to obtain a deeper quantitative understanding of the detailed nature of the role of multi-excitonic transitions in nanolasers.

4. Conclusion

In conclusion, we have studied lasing from photonic-crystal nanocavities with few embedded QDs and compared the experimental data to a semiconductor model for QD-based microcavity lasers. The power dependence of the excitons, cavity, and background shows that only the background can provide the gain required to observe lasing. This is confirmed by the quantitative analysis showing that a few excitons cannot provide enough gain for the lasing in such nanolasers and in fact the number of emitters needed is much higher than the number of excitons coupled to the cavity below threshold. Thus, our study shows that the gain in nanolasers containing few QDs grown by self-assembly is mainly provided by multi-excitonic transitions involving wetting-layer states.

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