Exciton-Polariton Lasing in Selenide Micropillars

Polariton lasing in high-quality Selenide-based micropillars in the strong coupling regime

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We have designed and fabricated all-epitaxial ZnSe-based micropillars exhibiting the strong coupling regime between the excitonic transition and the confined cavity modes. At cryogenic temperatures, we demonstrate pulsed, single transverse mode polariton lasing operation in the micropillars. Owing to their high crystalline quality, low sidewall roughness, and a quality factor in excess of 5000, we find a particularly low optical excitation threshold, ranging from $P_{\text{thr}} = 2.11 \mu J/cm^2$ for a diameter of 10 $\mu m$ to $P_{\text{thr}} = 2.41 \mu J/cm^2$ for a diameter of 1.5 $\mu m$. These values are comparable or lower than what is reported so far in planar microcavities made of other large bandgap materials, or in GaAs micropillars. By a careful analysis of $P_{\text{thr}}$ versus diameter we could trace back the conservation of a low polariton lasing threshold in small diameter micropillars to a sidewall roughness length scale as low as $(2 \pm 1) \text{ nm}$.

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In the last two decades exciton-polaritons in microcavities (MCs)\textsuperscript{1\textendash}13 have attracted considerable interest, mostly because with their half-light half-excitonic nature, they behave accordingly to a weakly interacting driven-dissipative bosonic quantum fluid\textsuperscript{3, 18}. From an applied quantum optics point-of-view, polaritons also offer very interesting perspectives. It has been shown theoretically that by squeezing the polaritonic wavefunction into a small volume, like in micropillars, the polaritonic nonlinearity can become large enough to play the role of a quantum filter for light, without resorting to single emitters like atoms or quantum dots\textsuperscript{3}.\textsuperscript{17}

Refined etching and microstructuring techniques have been developed for GaAs-based microcavities, allowing the fabrication of high quality micropillars\textsuperscript{14\textendash}16, mesas\textsuperscript{2} as well as advanced polaritonic circuits elements like waveguides, interferometers, optical gates\textsuperscript{17\textendash}19 and lattices with direct applications for quantum simulation\textsuperscript{12, 13}. This approach is likely to be successful in the upcoming years, however, for practical use, its drawback is to be stuck to cryogenic temperatures. A way around this problem is the use of large bandgap materials, where the exciton binding energy is larger, and hence stable at room temperature. But the price to pay is twofold: less mature etching techniques, and a weaker nonlinearity. Interestingly, V. Savona et al. have shown that the quantum regime of the nonlinearity can be reached even with small polaritonic nonlinearity by exploiting quantum interferences in a coupled micropillar pair geometry\textsuperscript{12, 13}.

With this idea in mind, ZnSe or CdTe based microstructures in the strong coupling regime have a great potential: On the one hand, they have an excitonic binding energy in principle large enough to be able to maintain the strong coupling regime at room temperature.\textsuperscript{13} On the other hand, keeping in mind that the excitonic nonlinearity scales like $a_B$, the Bohr radius\textsuperscript{12}, the latter is still large enough in these materials to provide a non-negligible nonlinearity.\textsuperscript{13} Finally, ZnSe has the advantage over CdTe to exhibit a polaritonic optical transition in the blue region of the spectrum ($440$ nm versus $740$ nm respectively), thus allowing to squeeze the polaritonic wavefunction into smaller diameter as we will see further on.

In this letter, as a first step along this route, we report on the fabrication of high quality ZnSe-based micropillars in the strong coupling regime. We find discretized transverse polariton modes, that are well resolved spectrally and in momentum space, and display a quality factor in excess of 5000. Upon weak pulsed excitation, polariton lasing is achieved at cryogenic temperature, with a particularly low excitation threshold and small blueshift for a non-GaAs material.

The investigated microcavity is similar to that used in a another work\textsuperscript{20} it consists of a 16.5-fold lower distributed Bragg reflector (DBR), a $3\lambda$-cavity with $5 \times 3$ ZnSe quantum wells (QWs) placed in the antinodes of the electric field, and a 14-fold upper DBR. The high-index material of the DBRs consists of ZnMgSSe and the low-index material is a short period superlattice of MgS/ZnCdSe (further growth details available elsewhere\textsuperscript{21, 22}). Previous X-ray diffraction and transmission electron microscopy investigations show that the structure is fully lattice matched to the GaAs substrate and reveals a low defect density compared to state of the art II-VI material\textsuperscript{23}. A set of micropillars with diameters of $\phi = 10$ $\mu m$, $5$ $\mu m$, $3.2$ $\mu m$, $2.2$ $\mu m$, and $1.5$ $\mu m$ have been fabricated in this microcavity using focused ion beam etching. A sketch of the structure is shown.

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FIG. 1. (Color online) (a) Reflectivity as a function of energy and angle of the planar MC adjacent to the micropillars. The middle and lower polariton branches are well identified. The solid line is a three-oscillator fit of the dispersion. Calculation parameters: $\hbar\Omega_R = (32 \pm 2)$ meV, $\hbar\Omega_h = (13.0 \pm 1.5)$ meV (light hole exciton Rabi splitting), $n_c = 2.15 \pm 0.15$ (cavity background index), $E_{hh} = 2812 \pm 1$ meV, $E_{lh} = 2831$ meV, $E_c = 2822.5 \pm 2.0$ meV (bare cavity energy). (b) Measured PL emission intensity below threshold as a function of energy and angle of the planar sample (linear color scale from black to white). (c)-(g) SEM images of the prepared micropillars. (h)-(l) Measured PL emission intensity below threshold for micropillars with diameters of 10.0 µm, 5.0 µm, 3.2 µm, 2.2 µm, and 1.5 µm respectively. (m) Sketch of a ZnSe MC micropillar.

in Fig. 1 m, while the actually obtained micropillars are shown in Fig. 1 c-g. A multistep etching protocol has been developed to minimize the etching damages on the micropillar sidewalls.

Within an angle-resolved micro-reflectivity measurement, we first characterized the planar microcavity properties. A typical spectrum is shown in Fig. 1 a. A three coupled oscillator model fit (involving the 1s heavy-hole excitons and the 1s light hole excitons) yields a heavy hole exciton Rabi splitting $\hbar\Omega_R = (32 \pm 2)$ meV. In the region of the micropillars, a detuning $\delta = E_C - E_X = +(10.5 \pm 3.0)$ meV is found.

FIG. 2. (Color online) Measured polariton PL intensity (log. color scale) of the 2.2 µm micropillar as a function of energy and emission angle below (a) and above (b) the polariton lasing threshold ($P_{th} = 2.16 \mu J/cm^2$).

We then realized microphotoluminescence (PL) measurements of each micropillars, using a frequency-doubled picosecond-pulsed Ti-Sapphire laser, focused by a microscope objective into a spot approximately 12 µm in diameter. Excitation is carried out non-resonantly through the first high-energy high transmission mode of the Bragg mirrors at $E_{Laser} \approx 2950$ meV. To avoid heating of the sample, the laser is chopped by an acousto-optic modulator with a duty cycle of 5% at 500 kHz. The PL signal is detected by Fourier-transform spectroscopy, and spectrally resolved in a spectrometer. The sample was kept in a Helium-flow cryostat at a constant temperature of $T = 6$ K.

The PL of the micropillars in the weak excitation regime is shown in Fig. 2 a-l, alongside the PL of the nearby planar microcavity (Fig. 2 b). Owing to the narrow linewidth of polaritons, transverse state discretization is well resolved up to a diameter of 10 µm. We also observe the expected blueshift of the polariton ground state (that centered around $k_\parallel = 0$) for increasing lateral confinement of the polaritonic mode within the micropillars. This blueshift reaches 4.2 meV for the (smallest) micropillar diameter of 1.5 µm. The strong coupling regime is thus unambiguously conserved in these micropillars since the bare confined cavity mode is more than 25 meV above the polariton ground state in the planar microcavity. Qualitatively the confined polariton states patterns in momentum space are in excellent agreement with the theoretical ones calculated by V. Savona et al. This points towards the fact that the micropillars are virtually free from optical defects, that would otherwise deform that pattern.

Fig. 2 shows the PL spectra of a 2.2 µm diameter micro-}

polariton lasing takes place in the lowest energy polariton mode $m_1$. The emission pattern in momentum space is shown below (Fig. 2a) and above (Fig. 2b) threshold in logarithmic colorscale. Despite the very high population contrast, we can still see the first polariton excited state above threshold, with an unchanged momentum contrast, we can still see the first polariton excited state above threshold, with an unchanged momentum contrast. A two-body interaction induced blueshift of 1 meV is observed at threshold, i.e. an order of magnitude lower than that expected in the case where the strong coupling regime would break up.

![Image](https://via.placeholder.com/150)

**FIG. 3.** (Color online) (a) Integrated PL intensity, (b) emission energy and linewidth measured on the lowest energy emission mode $m_1$ of the 2.2 $\mu$m-micropillar as a function of the excitation power. The pink region on the plots right side materializes the power range $P$ above the lasing threshold $P_{\text{thr}}$=2.16 $\mu$J/cm$^2$. Inset: PL spectrum measured on a 2.2 $\mu$m-micropillar with an excitation power of $P$=1.60 $\mu$J/cm$^2$.

A detailed analysis of the polariton emission in this small diameter micropillar across the lasing threshold is shown in Fig. 3. Fig. 3a shows that at the polariton lasing onset, a two orders of magnitude increase of the emission from the ground state is observed within 7% of excitation power increase around $P_{\text{thr}}$. Moreover, as shown in Fig. 3b, the linewidth decreases from $\Delta E_{m_1} = 0.56$ meV (full width at half-maximum) at low excitation power to $\Delta E_{m_1}^{th} = 0.40$ meV at threshold, and then increases again at higher power due to two-body interactions induced decoherence. This is the typical signature of polariton stimulated relaxation. The limited linewidth narrowing is the result of pulsed excitation and time-integrated detection. The measured blueshift versus excitation power is shown in Fig. 3b. It increases linearly below (starting from $P \sim 0.74 P_{\text{thr}}$), at, and above the threshold without notable slope change over the whole range. This is the expected behaviour when the latter is due to two-body scattering, involving excitons below threshold, and excitons and polaritons above threshold. This is unlike the weak coupling regime, where the electron-hole reservoir density gets clamped by the stimulation at and above threshold, resulting in a sharp slope change at threshold, i.e. a saturation of the blueshift upon increasing power further above threshold. These features match what has been reported already in GaAs micropillars by Bajoni et al and clearly show that the micropillars lase in the strong coupling regime.

The striking aspect of these Selenide micropillars is that the polariton lasing threshold is quite low as compared to state-of-the-art reports in planar microcavities. We have summarized some representative literature data in Tab. 1 obtained in planar GaAs and CdTe microcavities, where similar excitation conditions were used (non-resonant pico- or femto-second pulse laser). We find that in spite of the fact that we are dealing with ZnSe compounds, in spite of the FIB etching step, and of the small diameter of the micropillars, our excitation threshold is comparable or even lower. We attribute this result to the combination of a high optical quality factor of the planar microcavity, plus to a low damage of the micropillar sidewalls.

To verify this latter point, we have measured the excitation threshold versus the micropillar diameter. We indeed expect that the larger the optical loss rate, the larger the threshold. The results are show in Fig. 4. We observe that the diameter needs to be really small to observe a clear increase of the threshold with respect to that in the planar cavity.

In order to understand this behaviour quantitatively, we used a simple three level laser rate equation model (an excitonic reservoir, the lower polariton branch ground state, and the empty cavity). Assuming steady-state for simplicity, the excitation power at threshold reads

$$P_{\text{thr}}(R) = \gamma(R) + \gamma_{nr} \sqrt{\frac{\gamma(R)}{2G}}, \quad (1)$$

where $\gamma(R) = \gamma_0 + \gamma(R)$ is the radius-dependent radiative polariton escape rate, $\gamma_0$ is the polariton radiative decay rate, $\gamma_{nr}$ is the reservoir non-radiative loss rate and $G$ is the (non-stimulated) reservoir to polariton relaxation rate, driven by two-body scattering. The contribution of the sidewall roughness to the optical losses, has been studied in detail by Painter et al. It is shown there that $\gamma(R) \propto (R \lambda_0^2/V_r)^{-1}$, where $\lambda_0$ is the polariton emission wavelength, and $V_r$ is the average "grain
FIG. 4. (Color online) Polariton lasing threshold normalized by the planar threshold as a function of the pillar diameter. A modest increase of the lasing threshold is well described by the planar threshold as a function of the pillar diameter.

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