Toward polariton lasing in a zinc oxide microcavity: design and preliminary results.

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Abstract. We report the observation of the strong coupling regime in a half ZnO cavity. A large Rabi splitting of 130 meV is seen as well as a bottleneck in the exciton-polariton relaxation due to a shorter cavity-photon lifetime. The deposition of the top mirror is expected to yield a Q factor of 620 and the structure would then fulfill the main criteria required for room temperature polariton lasing to occur. This conclusion is supported by simulations based on the solution of Boltzmann equations leading to a polariton density threshold of $10^{16}$ cm$^{-3}$.

1. Introduction

The bosonic behaviour of particles has been widely studied since the first report of a Bose-Einstein condensate (BEC) in a cold rubidium gas [1]. However these properties appear at very low temperature and a heavy cooling is needed. To observe BEC at higher temperatures, a good candidate tends to be the cavity polariton. Firstly demonstrated by Weisbuch et al [2], the strong coupling regime (SCR) between excitons and photons in a planar microcavity (MC) yields to a mixed state with bosonic properties: the so-called exciton-polariton. For fifteen years, a wide range of semiconductors has been investigated to observe the BEC at low temperature [3] and the polariton lasing up to room temperature, mainly with GaN-based microcavities [4].

A promising candidate to obtain a fully functional polariton laser at room temperature (RT) should be the zinc oxide (ZnO), mainly due to the large binding energy of its excitons and their high oscillator strengths [5]. Recent works on ZnO microcavities have reported the SCR observation up to 410K [6-9]. The next step is now to obtain a polariton laser at RT but three main enhancements are currently needed:

(i) a high Rabi splitting which is predicted for thick cavities (three times larger than for GaN); this is necessary to obtain an efficient “polariton trap” and to decrease the lasing threshold [10].

(ii) a negative detuning between the excitons and the photonic mode; Faure et al [11] demonstrated the relaxation of the polariton in a lower energy state, with an important photonic part.

(iii) a high quality factor which is closely related to the photon lifetime into the cavity ($\tau_c$); recent calculations require Q greater than 500 ($\tau_c\approx0.1$ps) to achieve a reasonably low lasing threshold [10].
In this paper, we will first explain how we intend to fulfill the criteria detailed above to obtain a laser emission based on polaritons at RT in a bulk ZnO microcavity. We present experimental results obtained on a half-cavity (the ZnO-air interface acts as the top mirror) showing the SCR at room temperature with a large Rabi splitting. Finally we calculate the threshold density of the polariton lasing in this cavity, using the solutions of Boltzmann equations.

2. Cavity design and experimental setup

The cavity design is crucial to satisfy the previous criteria. A ZnO layer is then embedded between two distributed Bragg reflectors (DBR). The bottom one consists of 13 pairs of AlN and (Al,Ga)N with 24.5% of aluminium deposited on a Si(111) substrate by molecular beam epitaxy (MBE). To complete the structure, an 11 pairs dielectric mirror will be realized by plasma enhanced chemical vapor deposition (PECVD). These DBRs present a high reflectivity (close to 95% seen from the ZnO layer) which leads to a theoretical quality factor Q = 620. The ZnO layer is also deposited by MBE with a thickness of 220 nm, chosen to obtain a Rabi splitting as high as 150 meV (calculations).

Angle resolved reflectivity measurements were performed to observe the signature of the SCR [12] and photoluminescence (PL) experiments to study the behaviour under optical pumping. The pump consists of a cw-laser emitting at $\lambda_0=266$ nm (4.66 eV) with a power density of about $100$ W.cm$^{-2}$.

3. Observation of the strong coupling in a half-cavity

Figure 1 shows angle resolved reflectivity spectra measured on the half-cavity (the top mirror is replaced by air, with a poor reflectivity at the interface with ZnO). The DBR stop-band is centred on 3.22 eV with a width of 0.45 eV. The cavity mode is well defined close to 3.2 eV and the quality factor is equal to Q = 50. At both low and room temperature, the photonic modes (cavity mode, red line and Bragg mode, green line) have a similar behaviour: they shift toward higher energies with the increase of the incidence angle but near the excitons position, their move slows down due to the strong interaction between excitons and photons giving evidence of the polaritonic dispersion.

![Figure 1](image.png)

**Figure 1.** Angle resolved reflectivity spectra performed at low (left part) and room temperature (right) with an incidence angle varying from 5° to 75°. Dashed lines represent lower polariton branches (cavity, red and Bragg, green) and the energy of the excitons is shown with marks.

However, the observation of the upper polariton is inhibited by the high absorption of the ZnO continuum [8]. To determine the dispersion of all branches and thus evaluate the Rabi splitting, we use...
a transfer matrix calculation [13]. The refractive indices of AlN and (Al,Ga)N are well known [14] and
the only fitting parameters are the excitonic ones. As shown in ref [8], at low temperature the high
excitonic absorption prevents most of the photons to reach the cavity bottom for the excitonic
energies. As a consequence, the reflectivity in the [3.35-3.45 eV] region is similar to that of bulk ZnO.
The excitonic parameters which fit the experimental spectra are reported in Table 1.

### Table 1. Experimental excitonic parameters for ZnO at low temperature.

| Exciton   | Energy (eV) | Inhomogeneous broadening (meV) | Oscillator strength (meV\(^2\)) |
|-----------|-------------|--------------------------------|---------------------------------|
| A Exciton | 3.3705      | 5.0                            | 250 000                         |
| B Exciton | 3.378       | 5.0                            | 180 000                         |

Using these parameters and the transfer matrix formalism, reflectivity spectra are calculated for
each incidence angle at room temperature. The results are reported on Figure 2 as a dispersion curve
(energy versus angle). Experimental results (reflectivity, full dots) and calculations are in good
agreement. Without ZnO excitons, we determine the dispersion of the uncoupled cavity mode (dashed
line, estimated detuning -60 meV). By removing the band to band contribution, it is then possible to
deduce the upper polariton branch energy leading to a Rabi splitting of 130 meV at room temperature
for an incidence angle around 40° (160 meV at low temperature).

Figure 2. Energy as a function of the incidence angle showing the polariton dispersion;
experimental points are reported as circle (reflectivity) whereas the lines correspond to
transfer-matrix calculations.

Figure 3. PL spectra as a function of the detection angle; false colours correspond to the
emission intensity. A bottleneck appears around 40° on the lower polariton branch with an
excitation power of about 100W.cm\(^{-2}\).

4. Optical injection

The half-cavity has also been studied under non-resonant optical injection at room temperature. Figure
3 shows angle resolved PL spectra measured for various detection angles. The luminescence intensity
is reported using a colour scale varying from blue to red (low to high emission). The lower polariton
dispersion is well defined with a maximum of emission at 3.25 eV and a detection angle around 40°.
This is the expression of a bottleneck effect due to an inefficient (no thermalization) relaxation of polaritons along the lower branch.

The simulation of polariton relaxation in the complete microcavity has been carried out by solving the semi-classical Boltzmann equations [10]. We have considered the pulsed pumping case, using the cavity parameters. The dependence of the maximal occupancy of the ground state on the injected density is shown in Figure 4. One can clearly observe a non-linear threshold around a density of 2x10^{16} cm^{-3} (two orders of magnitude smaller than the Mott density) which suggests that polariton condensation at room temperature is likely to take place in this structure. The inset shows the polariton distribution function for two different particle densities. The polaritons are well thermalized both below and above threshold, with a distribution function having clearly a Bose character above threshold, evidencing exciton-polariton condensation in the ground state.

Figure 4. Calculated maximal occupancy of the ground state as a function of the particle density injected by the pump pulse. Inset: the polariton distribution function corresponding to the maximal occupancy of the ground state below (dashed line) and above threshold (solid line).

5. Conclusion

In this paper, we observe the strong coupling regime in a bulk ZnO half-cavity with a large Rabi splitting (130meV) at room temperature. However polariton life-time is too short to achieve a complete relaxation and thermalization. This condition should be obtained after the deposition of a dielectric mirror above the described half-cavity. Further calculations assert a low threshold density (10^{16} cm^{-3}) for polariton lasing in the whole cavity.

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References

[1] Anderson M H, Ensher J R, Matthews M R, Wieman C E et al. 1995 Science 269 198
[2] Weisbuch C, Nishioka M, Ishikawa A and Arakawa Y 1992 Phys. Rev. Lett. 69 3314
[3] Kasprzak J, Richard M, Kundermann S, Baas A et al. 2006 Nature 443 409
[4] Christopoulos S, von Hogersthal G B H, Grundy A J D, Lagoudakis P G et al. 2007 Phys. Rev. Lett. 98 126405
[5] Chichibu S F, Uedono A, Tsukazaki A, Onuma T et al. 2005 Semicond. Sci. Technol. 20 S67
[6] Shimada R, Xie J, Avrutin V, Ozgur U and Morkoc H 2008 Appl. Phys. Lett. 92 011127
[7] Schmidt-Grund R, Rheinlander B, Czekalla C, Benndorf G et al. 2008 Appl. Phys. B 93 331
[8] Medard F, Zuniga-Perez J, Disseix P, Mhailovic M et al. 2009 Phys. Rev. B 79 125302
[9] Sturm C, Hilmer H, Schmidt-Grund R and Grundmann M 2009 New J. Phys. 11 073044
[10] Johne R, Solnyshkov D and Malpuech G 2008 Appl. Phys. Lett. 93 211105
[11] Faure S, Brimont C, Guillet T, Bretagnon T, Gil et al. 2009 Appl. Phys. Lett. 95 121102
[12] Houdre R, Weisbuch C, Stanley R P, Oesterle U et al. 1994 Phys. Rev. Lett. 73 2043
[13] Medard F, Zuniga-Perez J, Frayssinet E, Moreno J C et al. 2009 Photonics Nanostruct. Fundam. Appl. 7 26
[14] Antoine-Vincent N, Natali F, Mhailovic M, Vasson A et al. 2003 J. Appl. Phys. 93 5222