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Gain mechanism below Mott transition and VCSEL operation in a ZnO microcavity

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Abstract

The VCSEL operation of a ZnO planar microcavity under optical pumping is demonstrated from $T=80$ K to 300 K. It is observed as the cavity switches from the strong coupling to the weak coupling regime at high carrier density. A gain-related transition is observed below 240 K, which appears before breaking the strong coupling. This shows that exciton scattering processes are involved in the gain process.

Keywords: Zinc oxide; microcavity; strong coupling, VCSEL

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Blue and ultraviolet operating micro-lasers are attracting a strong interest due to the wide range of foreseen applications (display, optical data storage) and to the need for a deeper understanding of their operation mechanism. They are based on GaN and ZnO semiconducting materials and various kinds of optical resonators are presently investigated : Fabry-Perot modes in planar microcavities as well as in nanowires [1, 2]; whispering gallery modes in microwires [3] and microdisks [4]; cavities in photonic crystal membranes [5]; random lasing in ZnO powders [6] and waveguides [7]. Three main schemes are used in order to obtain gain : (i) an electron-hole plasma is involved in recently demonstrated GaN VCSELs [8] and ZnO nanowire lasers [9]; (ii) exciton scattering processes are claimed to mediate gain in ZnO nanowires; (iii) when excitons and photons are in the strong coupling regime up to high densities, polariton lasing is observed, as demonstrated in GaN planar microcavities [10]. The frontier between those three regimes is not yet clearly established in the case of ZnO, and the exciton-mediated processes, which are specific to II-VI materials and have been thoroughly studied 40 years ago in bulk samples (see [11] for a critical review), are still the object of controversies in microlasers.

In the present work, we demonstrate VCSEL operation from 80 K to 300 K in a ZnO planar microcavity which exhibits a strong exciton-photon coupling at low exciton density, and switches into the weak coupling regime at lasing threshold. The gain mechanism is proved to be exciton-mediated at threshold, as shown by the coexistence of strong exciton-photon coupling and a gain-related transition when the cavity mode is brought to large negative detuning.

The microcavity consists in a $5\lambda/4$ ZnO active layer embedded between a (Ga,Al)N/AlN bottom DBR and a (Si₂O₃)/Si₃N₄ top DBR. The nitride DBR and the ZnO layer are grown by MBE on a Si(111) substrate, whereas the dielectric DBR is realized by PECVD [12, 13]. The number of pairs of each mirror is respectively 13 and 12, leading to a nominal quality factor of about 500 for the cavity mode.

The microcavity exhibits a strong exciton-photon coupling at low and room temperature. The study of the angle-resolved reflectivity of the half cavity, without the top dielectric DBR, revealed a large Rabi splitting of 130 meV at T=300K [13]. A similar value (120 meV) is deduced from the analysis of the angle-resolved photoluminescence of the full cavity. The quality factor of the cavity depends on the size of the excitation spot : it is about 300 for $\sim 100\mu\text{m}$ spots, and reaches 450 for $\sim 2\mu\text{m}$ spots. It is therefore locally close to the nominal value, but some in-plane disorder induces inhomogeneous broadening on length scales smaller than 100 μm [14].

The emission of the microcavity has been studied under strong excitation density. The cavity is excited by a OPO (Opotek Vibrant UV-HE-LD, pulse duration: 3 ns; repetition rate 10 Hz, spot diameter 50 μm) at 3.78eV, and an incidence angle of 30°, corresponding to the first Bragg mode on the high energy side of the stop-band. Figure 1 presents a series of PL spectra with increasing excitation power density at T=300K. At low excitation density, the spectrum consists in a sharp peak at 3.20 eV related to the lower polariton branch, and a broad peak centered around 3.29 eV, attributed to ZnO exciton transitions. As the excitation density is increased above a threshold value of $0.8 \text{ J}\cdot\text{cm}^{-2}$, a second sharp peak appears, which grows non-linearly. Its energy corresponds to the estimated value for the bare photon mode, in the absence of strong coupling with excitons. This peak is attributed to lasing, i.e. to the VCSEL operation of the microcavity. No clear narrowing of the peak around threshold is observed, probably

due to the inhomogeneous broadening of the photon modes within the excitation spot and to the non steady-state operation within each 3ns long excitation pulse.

The temperature dependence of the observed spectra around threshold provides valuable information on the operation mechanism of the laser. Figure 2.a presents the power dependence of the PL spectrum at $T=80$ K. At low excitation density, the spectrum now consists in one sharp peak at 3.22 eV related to the lower polariton branch, a weaker and broader transition at 3.37 eV at the energy of free excitons, and an additional sharp peak at 3.35 eV. This last feature is also present under continuous wave (CW) excitation and presents a slight angular dispersion. It is a higher order mode of the cavity ($7\lambda/4$ instead of $5\lambda/4$), strongly coupled to excitons, as predicted in previous theoretical work on ZnO microcavities [15]. It is the signature of the strong variation of the refractive index nearby the excitonic transition in ZnO [16, 17], i.e. the large excitonic oscillator strength. A similar effect has been observed in ZnO nanowire lasers, where the free spectral range strongly decreases nearby the exciton energy [1, 2]. As the temperature is increased, the exciton homogeneous broadening attenuates the variation of the refractive index : the $7\lambda/4$ mode is broader but still visible at 240 K (Figure 2.b), and disappears at 300 K (Figure 1).

As the excitation power is increased, a new transition (G) appears in the PL spectrum, shifting from 3.305 to 3.28 eV. Its temperature dependence is plotted on Figure 3 and compares well with previously reported stimulated emission features in ZnO, which are attributed to exciton-exciton scattering processes at low temperature and low density, and gain in the electron-hole plasma (EHP) at higher density (see [11] for an exhaustive review). This peak does not disperse with angle nor with detuning and is therefore not related to the cavity. It could be either due to amplified spontaneous emission (ASE), or

to random lasing in the plane of the cavity, as recently reported for ZnO waveguides [7], which would scatter in the vertical direction due to interface roughness and cracks.

The energy of LPB and $7\lambda/4$ modes at low density, and of the lasing mode at threshold, are also reported on Figure 3. The lasing mode only weakly shifts with temperature, as expected for a photon mode only sensitive to the refractive index. The LPB and $7\lambda/4$ modes partly follow the Varshni law for ZnO exciton, which supports the fact that they are in the strong coupling regime.

Let us now discuss into details the role of excitons and the electron-hole plasma in the gain mechanism. Figure 3.c presents the power dependence of the PL spectrum for a larger negative detuning compared to figures 3.a and 3.b. No lasing is observed in the same excitation range, but the stimulated emission feature is as strong as for smaller detunings, albeit to far from the cavity mode to observe lasing. More interestingly, the LPB and $7\lambda/4$ modes are kept sharp over the whole density range, which tends to prove that the strong coupling regime of excitons with photons is maintained at the highest carrier density when lasing is not observed. Within the investigated density range, the electron-hole system is thus kept below the Mott transition to an electron-hole plasma, and the gain at 3.3 eV results from P-band exciton scattering processes [18, 19]. It has to be noticed that it is impossible to directly estimate the carrier density, since it depends on the carrier lifetime in the ASE or random laser regimes, which is unknown. On the contrary, when lasing is observed (Fig. 3.a and 3.b), the LPB and $7\lambda/4$ modes rapidly broaden above threshold and the system transits into the weak coupling regime. We can therefore conclude that (i) gain through a degenerate EHP, which is the standard gain mechanism in III-V VCSELs, can be discarded in our ZnO VCSEL at $T=80\text{K}$; (ii) the onset of lasing induces a fast decoherence for excitons, leading to a

transition into the weak coupling regime. The microcavity behave similarly up to $T=240$ K, however we cannot conclude about the gain mechanism at $T=300$ K.

As a conclusion, we have demonstrated a ZnO VCSEL operating from 80 K to room temperature. Below $T=240$ K, the gain mechanism implies exciton-scattering processes, as proved by the coexistence of the strong coupling regime and a gain-related transition. This is a novel mechanism for VCSEL operation, which usually rely on gain in a degenerate EHP. This supports the recent interpretation of gain mechanism in ZnO nanowires [2] in terms of exciton scattering processes.

References

- [1] V. V. Ursaki, V. V. Zalamai, I. M. Tiginyanu, A. Burlacu, E. V. Rusu, and C. Klingshirn, *Appl. Phys. Lett.* **95**, 171101 (2009), <http://link.aip.org/link/?APL/95/-171101/1>.
- [2] M. A. Zimmler, F. Capasso, S. Müller, and C. Ronningf, *Semiconductor Science and Technology* **25**, 024001 (2010), <http://stacks.iop.org/0268-1242/25/i=2/a=024001>.
- [3] L. Sun, Z. Chen, Q. Ren, K. Yu, L. Bai, W. Zhou, H. Xiong, Z. Q. Zhu, and X. Shen, *Phys. Rev. Lett.* **100**, 156403 (2008), <http://link.aps.org/abstract/PRL/v100/-e156403>.
- [4] H. W. Choi, K. N. Hui, P. T. Lai, P. Chen, X. H. Zhang, S. Tripathy, J. H. Teng, and S. J. Chua, *Appl. Phys. Lett.* **89**, 211101 (2006), <http://link.aip.org/link/?APL/89/-211101/1>.
- [5] M. Arita, S. Ishida, S. Kako, S. Iwamoto, and Y. Arakawa, *Appl. Phys. Lett.* **91**, 051106 (2007), <http://link.aip.org/link/?APL/91/051106/1>.
- [6] J. Fallert, R. J. B. Dietz, J. Sartor, D. Schner, C. Klingshirn, and H. Kalt, *Nat Photon* **3**, 279 (2009), <http://dx.doi.org/10.1038/nphoton.2009.67>.
- [7] P.-H. Dupont, C. Couteau, D. J. Rogers, F. H. Teherani, and G. Lerondel, *Appl. Phys. Lett.* **97**, 261109 (2010), <http://link.aip.org/link/?APL/97/261109/1>.
- [8] Y. Higuchi, K. Omae, H. Matsumura, and T. Mukai, *Applied Physics Express* **1**, 121102 (2008), <http://apex.jsap.jp/link?APEX/1/121102/>.
- [9] J. Fallert, F. Stelzl, H. Zhou, A. Reiser, K. Thonke, R. Sauer, C. Klingshirn, and H. Kalt, *Opt. Express* **16**, 1125 (2008), <http://www.opticsexpress.org/abstract.cfm?URI=oe-16-2-1125>.
- [10] S. Christopoulos, G. B. H. von Högersthal, A. J. D. Grundy, P. G. Lagoudakis, A. V. Kavokin, J. J. Baumberg, G. Christmann, R. Butte, E. Feltn, J.-F. Carlin, et al., *Phys. Rev. Lett.* **98**, 126405 (2007), <http://link.aps.org/abstract/PRL/v98/e126405>.
- [11] C. Klingshirn, R. Hauschild, J. Fallert, and H. Kalt, *Phys. Rev. B* **75**, 115203 (2007), <http://link.aps.org/doi/10.1103/PhysRevB.75.115203>.

- [12] S. Faure, C. Brimont, T. Guillet, T. Bretagnon, B. Gil, F. Medard, D. Lagarde, P. Disseix, J. Leymarie, J. Zúñiga-Pérez, et al., Appl. Phys. Lett. **95**, 121102 (2009), <http://link.aip.org/link/?APL/95/121102/1>.
- [13] F. Médard, D. Lagarde, J. Zúñiga-Pérez, P. Disseix, J. Leymarie, M. Mihailovic, D. D. Solnyshkov, G. Malpuech, E. Frayssinet, S. Sergent, et al., of Physics: Conference Series **210**, 012026 (2010), <http://stacks.iop.org/1742-6596/210/i=1/a=012026>.
- [14] G. Christmann, D. Simeonov, R. Butte, E. Feltin, J. F. Carlin, and N. Grandjean, Appl. Phys. Lett. **89** (2006).
- [15] S. Faure, T. Guillet, P. Lefebvre, T. Bretagnon, and B. Gil, Phys. Rev. B **78**, 235323 (2008), <http://link.aps.org/abstract/PRB/v78/e235323>.
- [16] Y. S. Park and J. R. Schner, of Applied Physics **39**, 3049 (1968), <http://link.aip.org/link/?JAP/39/3049/1>.
- [17] M. Cobet, C. Cobet, M. R. Wagner, N. Esser, C. Thomsen, and A. Hoffmann, Appl. Phys. Lett. **96**, 031904 (2010), <http://link.aip.org/link/?APL/96/031904/1>.
- [18] C. B. A la Guillaume, J.-M. Debever, and F. Salvan, Phys. Rev. **177**, 567 (1969), <http://link.aps.org/doi/10.1103/PhysRev.177.567>.
- [19] B. Hönerlage, C. Klingshirn, and J. B. Grun, phys. stat. sol. (b) **78**, 599 (1976), <http://dx.doi.org/10.1002/pssb.2220780219>.

FIGURES CAPTIONS

Figure 1 : a) Photoluminescence at $T=300$ K and a detection angle of 0° , as a function of the excitation power density; the lower polariton branch, lasing mode and exciton energies are indicated; b,c) Amplitude and energy of the two observed transitions

Figure 2 : Photoluminescence as a function of the excitation power density for various temperature and detuning conditions; the excitation power density increases from 0.4 to 4 J.cm^{-2} , similarly to Figure 1. The lower polariton branch (LPB), lasing mode (L), gain feature (G), $7\lambda/4$ polariton mode ($7\lambda/4$) and exciton energy (X) are indicated.

Figure 3 : Energies (symbols) of the observed transitions as a function of temperature : lower polariton branch (LPB), lasing mode (L), gain feature (G), $7\lambda/4$ polariton mode ($7\lambda/4$), and exciton (X); the Varshni law for zinc oxide excitons and the reported values for the gain feature [11] are shown as green and blue lines respectively.

Figure 1

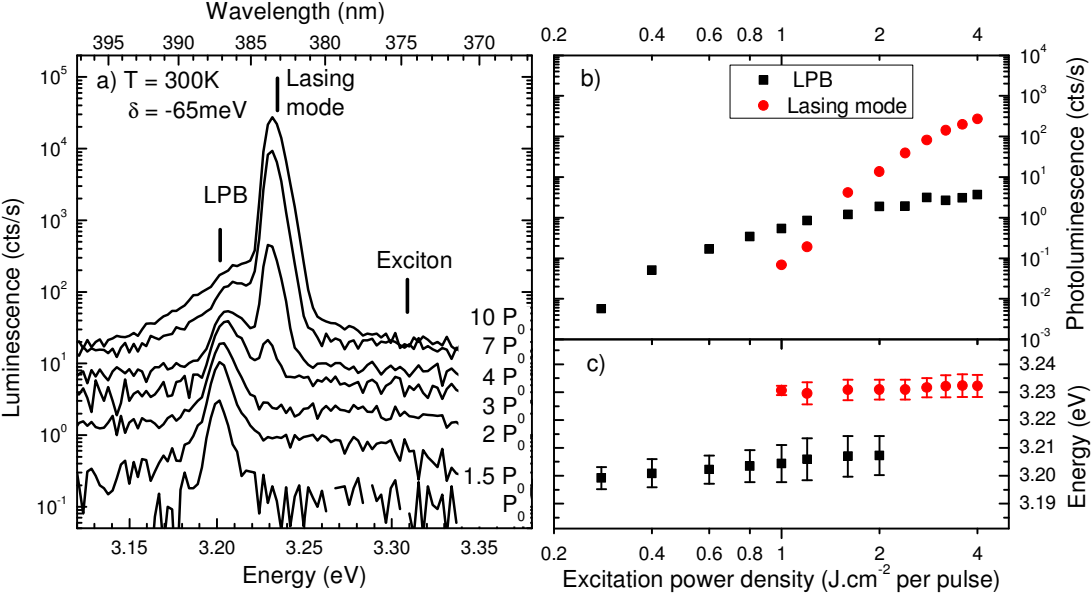


Figure 2

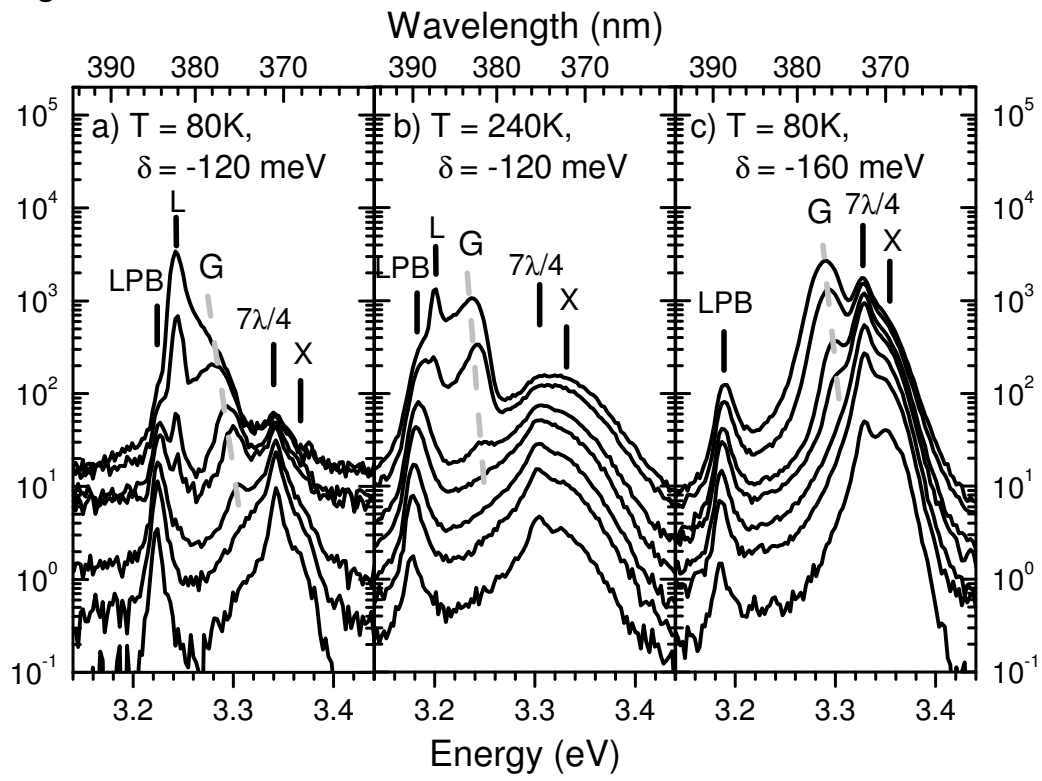


Figure 3

