Exciton-polaritons study in ZnO- based hybrid microcavities

Yu-Pin Lan[*a], Ying-Yu Lai[a], Si-Wei Huang[a], Jun-Rong Chen[a], Yung-Chi Wu[a], Shiang-Chi Lin[a],
Tien-Chang Lu[a], Shing-Chung Wang[a], and Wen-Feng Hsieh[a], Hui Deng[b]
[a]Department of Photonics, National Chiao Tung University, Hsinchu 300, Taiwan
[b]Department of Physics, University of Michigan, Ann Arbor, Michigan 48109, USA

ABSTRACT

The characteristics of exciton-polaritons in ZnO-based microcavities (MCs) are demonstrated with a large vacuum Rabi splitting due to large exciton binding energy and oscillator strength. The lower polariton branches (LPBs) can be clearly observed. For low temperature and large negative detuning conditions, a clear polariton relaxation bottleneck in bulk ZnO-based MCs has been observed in angle-resolved photoluminescence measurements from 100 to 353 K at different cavity-exciton detunings. The bottleneck is strongly suppressed with increasing the temperature and pumping power and reducing detuning. This observed results supposed to be due to more efficient phonon-assisted relaxation and a longer radiative lifetime of the polaritons. In addition, the linewidth broadening, blue-shift of the emission peak, and polarization of polariton lasing from below threshold to up threshold are also discussed.

Keywords: ZnO, microcavity, exciton-polariton

1. INTRODUCTION

Light-matter interaction is an interesting physical phenomenon. A simple way is to create a high-finesse optical cavity, which can be used to confine photons, and then put atoms in this cavity. When the cavity configuration is reduced to be close to, or below the dimension of the wavelength of light, it will be generally termed as "microcavity" [1]. Consider a semiconductor microcavity structure consisting of a active layer embedded between two high-reflectivity DBRs, the free electron-hole pairs will be created by exciting the active layer using laser sources or electrical injection. Furthermore, a quasi-particle, termed as exciton, is created if an excited electron-hole pair bound by the Coulomb interaction. This exciton has a similar function as the excited atom in a microcavity. When the exciton strongly interacts with the confined optical field of the semiconductor microcavity, it is possible for this system to be in the strong-coupling regime. If the rate of energy exchange between the cavity field and the excitons becomes much faster than the decay and decoherence rates of both the cavity photons and the excitons, an excitation in the system is stored in the combined system of photon and exciton. Thus the excitations of the system are no longer exciton or photon, but a new type of quasi-particles called the microcavity polaritons or exciton-polaritons. In the past decade, exciton-polaritons, the half-light/half-matter quasi particles generated from strong coupling between excitons and microcavity photons in semiconductor high-Q microcavities (MCs) have attracted much attention [2]. Due to their bosnic nature, exciton-polaritons with very small effective mass (10^{-8} that of hydrogen atom) increasing the critical temperature of condensation to cryogenic or even RT. On the other hand, the excitonic nature enables polaritons to interact with phonons, excitons, or polaritons and relax to their final state. The above mentioned properties have led to demonstration of various experimental results, including solid state Bose Einstein condensates (BEC) or ultra-low threshold polariton lasers [3-9], and polariton parametric oscillator [10].

*yplan@nctu.edu.tw

Physics and Simulation of Optoelectronic Devices XX, edited by Bernd Witzigmann,
Marek Osinski, Fritz Henneberger, Yasuhiko Arakawa, Proc. of SPIE Vol. 8255, 82551F
© 2012 SPIE · CCC code: 0277-786X/12/$18 · doi: 10.1117/12.908529

However, the exciton-polaritons were observed in CdTe [3] and GaAs [4, 5] MCs only at cryogenic temperatures due to the small exciton binding energies in these materials. In contrast, wide-bandgap semiconductor materials have larger exciton binding energies [11], and have emerged as candidates for high temperature polariton lasers. Room temperature (RT) polariton lasing has been demonstrated recently in bulk and multiple-quantum-well (MQW) GaN MCs [9, 10], yet the exciton binding energy in GaN is only comparable to the thermal energy at 300 K. Another wide-bandgap material, ZnO, possesses even larger oscillator strengths and exciton binding energies (~60 meV in the bulk layer) [12-14]. RT photon lasing in ZnO MCs has been reported very recently [15], but RT ZnO polariton lasers have only been discussed in theory [12, 16]. In this report, the strongly coupled characteristics in ZnO hybrid microcavity are present systematically.

2. ZnO HYBRID MICROCAVITY

2.1 Sample fabrication

The hybrid microcavity structure consists of a bulk ZnO $3\lambda/2$ thick cavity sandwiched between a bottom 30-pair AlN/$Al_{0.23}Ga_{0.77}N$ DBR and a top 9-pair dielectric SiO_2/HfO_2 DBR. The AlN/AlGaN DBR was grown by MOCVD on a 3μm thick GaN buffer layer on *c*-plane sapphire substrate. During the growth, TMGa and TMAl were used as group III source materials and NH_3 as the group V source material. After thermal cleaning of the substrate in hydrogen ambient for 5 min at 1100 °C, a 30-nm-thick GaN nucleation layer was grown at 520 °C. The growth temperature was raised up to 1040 °C for the growth of 3μm GaN buffer layer. Then, the AlN/AlGaN DBRs were grown under the fixed chamber pressure of 100 Torr similar to the previous reported growth conditions. The bulk ZnO $3\lambda/2$ thick cavity was grown on AlN/AlGaN DBR by pulsed-laser deposition (PLD) system, which is commonly adopted for the growth of ZnO epi-films. The beam of a KrF excimer laser (λ = 248 nm) was focused to produce an energy density ~5–7 $J . cm^{-2}$ at a repetition rate 10 Hz on a commercial hot-pressed stoichiometric ZnO (99.999 % purity) target. The ZnO films were deposited with a growth rate ~0.5475 $Å . s^{-1}$ at a substrate temperature of 600 °C and a working pressure ~ 9.8×10^{-8} Torr without oxygen gas flow. Finally, the 9-period SiO_2/HfO_2 dielectric DBR was deposited by dual electron-beam gun evaporation system to complete the microcavity structure.The schematic sketch of the ZnO-based microcavity structure is shown in Figure 1(a). The measured RT reflectivity of upper DBR, RT PL intensity of the ZnO cavity layer in a half cavity regime, and measured RT reflectivity of the bottom DBR are shown in Figure 1(b)-(d) respectively. The larger stop bandwidth of the upper dielectric DBR compare to the bottom epitaxial DBR is due to the larger index difference between the SiO_2 and the HfO_2 layer.

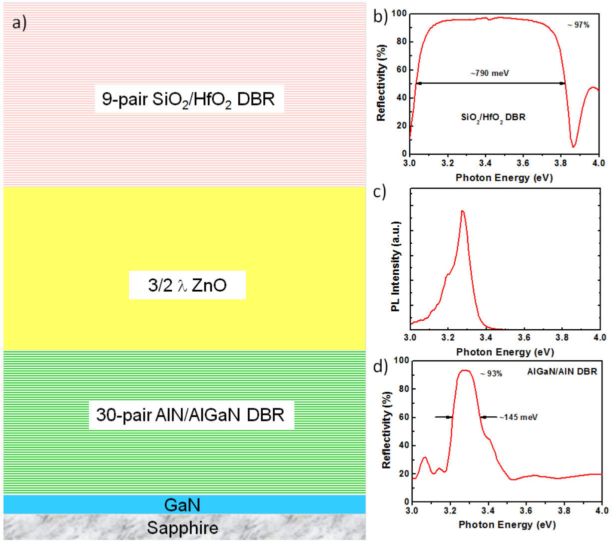

Figure 1. (a) The ZnO-based hybrid microcavity schematic diagram. (b) The measured RT reflectivity of upper DBR. (c) RT PL intensity of the ZnO cavity layer. (d) The measured RT reflectivity of the bottom DBR.

The RT reflectivity and μ-PL spectral from the full hybrid microcavity is shown in Figure 2. The narrowed PL linewidth of full cavity structure about 1.73nm at the 383nm is results from PL signal of the half cavity which is filtered by a narrow cavity mode of the full MC structure. The calculated cavity quality factor Q is about 221 when the pump spot size is about 3μm. It's noteworthy that the cavity dip was strongly dependent on the sample position due to the thickness fluctuation of the ZnO cavity layer and the bottom DBR. Such behavior also provides various choices of the different photon-exciton detuning in our sample.

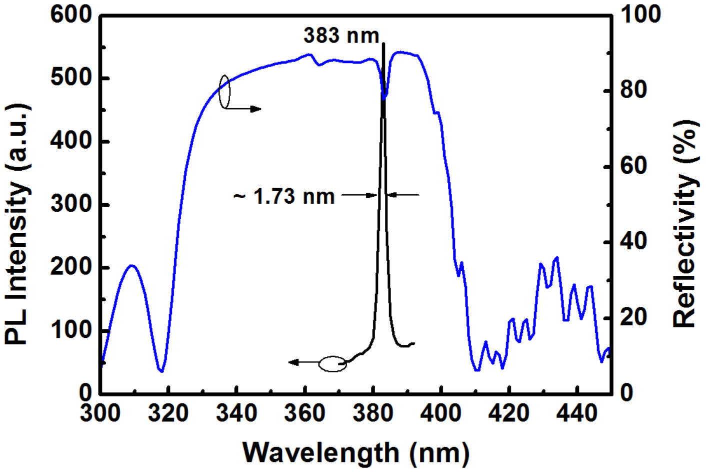

Figure 2. RT reflectivity and μ-PL spectra from the full hybrid microcavity

2.2 Measurement results

Polariton decays in the form of emitting a photon with the same $k_{//}$ and totoal energy $\hbar\omega = E_{LPB,\ UPB}$. The one-to-one correspondence between the internal polariton mode and the external out-coupled photon mode provides great convenience to experimental access to the strong coupling system. The external emitted photons carry direct information of the internal polaritons, such as the energy dispersion, population per mode, and statistics of the polaritons. Consequently, we could probe the internal properties of polaritons by collecting the emitted photons. Each in-plane photon mode couples only with an exciton state with the same $k_{//}$, which is related to the external angle of incidence θ via $k_{//} = k \sin\theta$. The polariton dispersion and associated phenomena can thus be studied in angular-dependent experiments. Therefore, the angle-resolved PL is a common optical system to measure the polariton dispersion curves. Figure 3 shows the schematic sketch of an angle-resolved PL system. The excitation source of the PL measurements is a 355 nm Nd:YVO_4 pulse laser at a 1kHz repetition rate and a pulse duration of 0.5 ns. The pumping laser spot size on the sample surface was about 60 μm in diameter at 60° incident angle. The PL emission light from the sample surface was collected using a UV optical fiber with 600 μm core mounted on a rotating stage with an angular resolution of ~1°, and detected by a liquid nitrogen cooled charge-coupled device attached to a monochromator (iHR 320, HORIBA scientific Inc.) with a spectral resolution of about 0.2 nm.

To probe the characteristics of strong exciton-photon coupling in the ZnO microcavity structure, RT angle-resolved reflectivity measurements were performed by using a two arm goniometer and a xenon lamp as a white light source. Figure 4a and 4b show the experimental exciton-polariton angle-resolved reflectivity spectral at different position of the sample, the reflectivity dips in the reflectivity spectral represent the position of the LPB. The black and blue dash in the Figures 4a and 4b depict the bared exciton mode and the fitted LPB of the MC respectively. The different exciton-polariton dispersion curves due to different detuning values can be easily observed by comparing Figure 4a and 4b. The LPB dips were then extracted to Figure 5a and 5b for further analyze. An identical vacuum Rabi splitting value of about 68meV is estimated at the resonant angle of 34° and 27°, respectively, due to the two different detuning values. The large value of vacuum Rabi splitting reveals a high potential of ZnO MC to be the mostly promising candidate for practical polaritonic devices.

In the experiment, the strong polariton relaxation bottleneck has been observed in bulk ZnO-based microcavities at low temperature by performing angle-resolved PL measurements. The polariton relaxation from bottleneck to low k states can be enhanced with increasing temperature, pumping power and reducing detuning. RT polariton lasing in a ZnO-based hybrid MC measurements show the linewidth broadening with pumping power and average polarization is not specific. We also obtain polariton lasing at 353k.

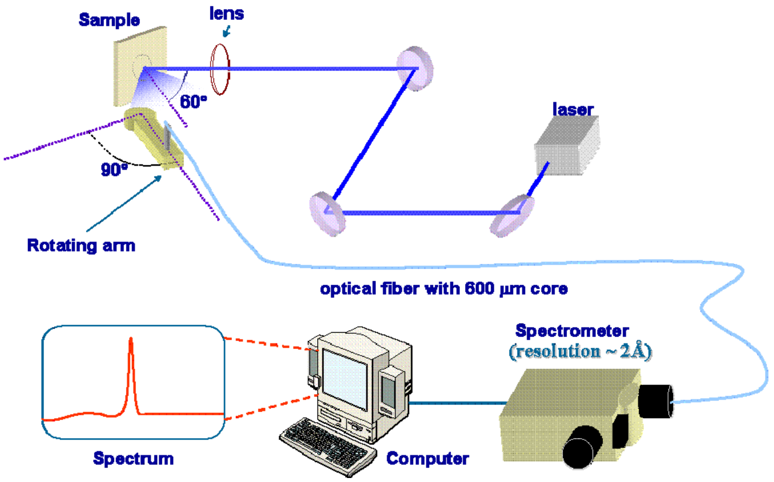

Figure 3. Schematic sketch of ARPL measurement

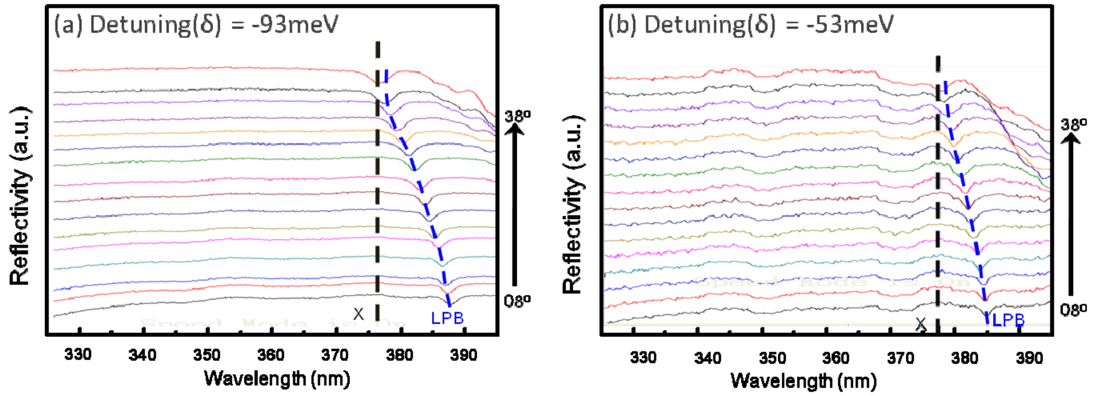

Figure 4. RT angle-resolved reflectivity spectral at (a) δ = -93meV and (b) δ = -53meV.

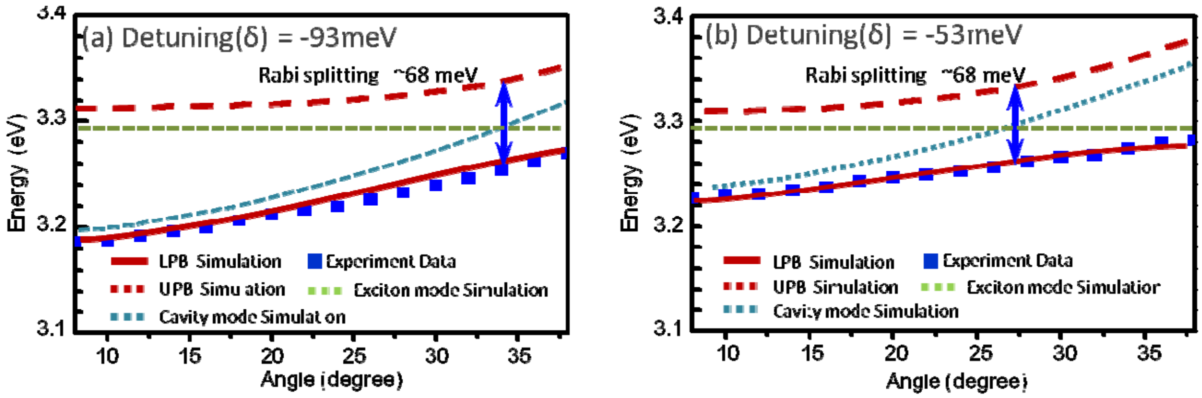

Figure 5. Extracted polariton emission peaks (blue squares) and simulated dispersion curves at (a) δ = -93meV and (b) δ = -53meV.

3. CONCLUSION

In conclusion, The dispersion curves of ZnO MC show obvious characteristics of strong exciton-photon coupling. The large vacuum Rabi splitting value of 68meV at RT reveals that ZnO MC is a promising candidate to achieve the RT polariton laser in the nearly future. Besides, characteristics of the polariton lasing have been systematically measured. The threshold power density, linewidth broadening, blue-shift of emission peak energy and polarization reveal the fundamental difference between polariton lasing and photon lasing.

ACKNOWLEDGMENTS

The authors would like to gratefully acknowledge C. K. Chen, S. W. Chen, Z. Y. Li, and Prof. H. C. Kuo at National Chiao Tung University for technical support. This work has been supported in part by the MOE ATU program and in part by the National Science Council of Taiwan under Contracts NSC99-2221-E-009-035-MY3, NSC99-2120-M-009-007, and NSC98-2923-E-009-001-MY3.

REFERENCES

1. K. J. Vahala, “Optical microcavities”, Nature, vol. 424, pp.839, 2003.
2. Weisbuch, C., Nishioka, M., Ishikawa, A. and Arakawa, Y., “Observation of the coupled exciton-photon mode splitting in a semiconductor quantum microcavity,” Phys. Rev. Lett. 69, 3314–3317 (1992).
3. Kasprzak, J., Richard, M., Kundermann, S., Baas, A., Jeambrun, P., Keeling, J. M. J., Marchetti, F. M., Szymańska, M. H., André, R., Staehli, J. L., Savona, V., Littlewood, P. B., Deveaud, B. and Dang, L. S., “Bose-Einstein condensation of exciton polaritons,” Nature 443, 409– 414 (2006).
4. Deng, H., Weihs, G., Santori, C., Bloch, J. and Yamamoto, Y., “Condensation of semiconductor microcavity exciton polaritons,” Science 298, 199–202 (2002).
5. Deng, H., Weihs, G., Snoke, D., Bloch, J. and Yamamoto, Y., “Polariton lasing vs. photon lasing in a semiconductor microcavity,” Proc. Nat. Acad. Sci. U.S.A. 100, 15318– 15323 (2003).
6. Balili, R., Hartwell, V., Snoke, D., Pfeiffer, L. and West, K., “Bose-Einstein condensation of microcavity polaritons in a trap,” Science 316, 1007–1010 (2007).
7. Kéna-Cohen, S. and Forrest, S. R., “Room-temperature polariton lasing in an organic single-crystal microcavity,” Nat Photonics 4, 371– 375 (2010).
8. Christopoulos, S., Von Högersthal, G. B. H., Grundy, A. J. D., Lagoudakis, P. G., Kavokin, A. V., Baumberg, J. J., Christmann, G., Butté, R., Feltin, E., Carlin, J.-F. and Grandjean, N., “Room-temperature polariton lasing in semiconductor microcavities,” Phys. Rev. Lett. 98, 126405 (2007).
9. Christmann, G., Butté, R., Feltin, E., Carlin, J.-F., and Grandjean, N. “Room temperature polariton lasing in a GaN/AlGaN multiple quantum well microcavity,” App. Phys. Lett. 93, 051102 (2008)
10. Ferrier, L., Pigeon, S., Wertz, E., Bamba, M., Senellart, P., Sagnes, I., Lemaître, A., Ciuti, C., and Bloch, J. “Polariton parametric oscillation in a single micropillar cavity,” Appl. Phys. Lett. 97, 031105 (2010).
11. Christmann, G., Butté, R., Feltin, E., Mouti, A., Stadelmann, P. A., Castiglia, A., Carlin, J.-F., and Grandjean, N., “Large vacuum Rabi splitting in a multiple quantum well GaN-based microcavity in the strong-coupling regime,” Phys. Rev. B 77, 085310 (2008).
12. Chen, J.-R., Lu, T.-C., Wu, Y.-C., Lin, S.-C., Liu, W.-R., Hsieh, W.-F., Kuo, C.-C. and Lee, C.-C., "Large vacuum Rabi splitting in ZnO-based hybrid microcavities observed at room temperature," App. Phys. Lett. 94, 061103 (2009)
13. O□zgür, Ü., Bergmann, M. J., Casey, H. C., Everitt, H. O., Abare, A. C., Keller, S. and DenBaars, S. P., “Ultrafast optical characterization of carrier capture times in $In_xGa_{1-x}N$ multiple quantum wells,” App. Phys. Lett. 77, 109–111 (2000).
14. Malpuech, G., Carlo, A. D., Kavokin, A., Baumberg, J. J., Zamfirescu, M. and Lugli, P., “Room-temperature polariton lasers based on GaN microcavities,” App. Phys. Lett. 81, 412– 414 (2002).
15. Kalusniak, S., Sadofev, S., Halm, S., and Henneberger, F., “Vertical cavity surface emitting laser action of an all monolithic ZnO-based microcavity,” App. Phys. Lett. 98, 011101 (2011).

16. Lu, T.-C., Chen, J.-R., Lin, S.-C., Huang, S.-W., Wang, S.-C., and Yamamoto, Y., "Room Temperature Current Injection Polariton Light Emitting Diode with a Hybrid Microcavity," Nano Lett. 11, 2791–2795 (2011)
17. Huang, G.-S., Lu, T.-C., Yao, H.-H., Kuo, H.-C., Wang, S.-C., Lin, C.-W., and Chang, L., "Crack-free GaN/AlN distributed Bragg reflectors incorporated with GaN/AlN superlattices grown by metal-organic chemical vapor deposition," Appl. Phys. Lett. 88, 061904 (2006).
18. Houdré, R., Weisbuch, C., Stanley, R. P., Oesterle, U., Pellandini, P. and Ilegems, M., "Measurement of cavity-polariton dispersion curve from angle-resolved photoluminescence experiments," Phys. Rev. Lett. 73, 2043– 2046 (1994).
19. Sellers, I. R., Semond, F., Leroux, M., Massies, J., Zamfirescu, M., Stokker-Cheregi, F., Gurioli, M., Vinattieri, A., Colocci, M., Tahraoui, A. and Khalifa, A. A., "Polariton emission and reflectivity in GaN microcavities as a function of angle and temperature," Phys. Rev. B 74,193308 (2006).
20. Wang, S.-C., Lu, T.-C., Kao, C.-C., Chu, J.-T., Huang, G.-S., Kuo, H.-C., Chen, S.-W., Kao, T.-T., Chen, J.-R. and Lin, L.-F., "Optically Pumped GaN-based Vertical Cavity Surface Emitting Lasers: Technology and Characteristics," Jpn. J. Appl. Phys. 46, 5397– 5407 (2007).
21. Eliseev, P. G., Perlin, P., Lee, J. and Osiński, M., ""Blue" temperature-induced shift and band-tail emission in InGaN-based light sources," App. Phys. Lett. 71, 569–571 (1997).
22. Tawara, T., Gotoh, H., Akasaka, T., Kobayashi, N. and Saitoh, T., "Cavity polaritons in InGaN microcavities at room temperature," Phys. Rev. Lett. 92, 256402 (2004).