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Robust exciton-polariton effect in a ZnO whispering gallery microcavity at high temperature

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A robust exciton-polariton effect in a ZnO whispering gallery microcavity well above room temperature is presented. The lower polariton branches are tuned by current induced thermal effect. The red shift can be as large as ~40 meV. It is found that the strong coupling can be preserved and the polariton-phonon interaction quenching effect remains up to ~550 K, while the Rabi splitting is about 330 meV. The tuning speed is in the order of millisecond, showing its potential in polariton-based optoelectronic device application. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3693378]

The strong coupling of exciton-photon (i.e., exciton-polariton) has been observed in various semiconductor (GaAs, organic, GaN, CdTe, etc.) optical cavities[1–4] and has attracted tremendous attention in the past decades due to its potential in nano-photonics and polaritonics application. Compared with the aforementioned semiconductors, ZnO is an ideal wide band gap semiconductor material for the investigation of exciton-polariton effect at room temperature or even above because of its large exciton binding energy (~60 meV) and oscillator strength.[5] The highest temperature for the strong coupling in ZnO predicted by Chichibu *et al.* is ~ 610 K.[6] So far, the exciton-polariton effect in ZnO whispering gallery (WG) and planar microcavities at room temperature has been demonstrated.[7–9] However, practical polariton light-emitting devices such as polariton laser may require higher tolerable temperature for the strong coupling. Whether the strong coupling in a ZnO WG microcavity can be preserved above room temperature or not is yet to be explored. Besides, it has been proven that polaritons in ZnO WG microcavity can be thermodynamically decoupled from the phonon bath, leading to the preservation of high coherence at room temperature.[8] However, whether this polariton-phonon interaction quenching effect remains at even a higher temperature or not has not yet been investigated experimentally until now, though it is crucial for the realization of high temperature polariton coherence.

For practical device application, tunability of the exciton-photon coupling, namely the detuning between pure optical mode and exciton, is essential. So far, several methods such as electric field, cavity size, temperature, hydrostatic pressure, gas, or stress[7,10–13] have been applied to tune the exciton-photon interaction. However, the exciton energy shift tuned by electrical field is small, while other techniques are either hard to control or lacking in quick response. Thus, a fast and easily controllable tuning method of strong coupling is desirable.

In this Letter, we present a robust exciton-polariton effect in a ZnO WG microcavity at high temperature. The lower polariton branches (LPBs) are tuned by current induced thermal effect. A red shift up to ~40 meV of LPBs is observed. Indeed, it is found that the strong coupling can be preserved and the polariton-phonon interaction quenching effect remains up to a very high temperature (~550 K), showing the opportunity of high temperature polariton condensate, lasing, superfluidity, etc. The Rabi splitting is ~330 meV, and the thermal equilibrium can be established in a very short time (order of millisecond).

The sample used in our experiment is a single-crystalline (wurtzite) ZnO micro-rod grown by carbothermal method.[14] The energy-momentum (E-$\boldsymbol{k}$) dispersion of LPBs with different polarization (TE: electric field $\vec{\varepsilon}$ perpendicular to C-axis and TM: $\vec{\varepsilon}//C$) was measured directly by angle-resolved Fourier space photoluminescence (PL) spectroscopy technique. The luminescence was excited by a 355 nm Nd:YAG picosecond pulsed (repetition rate: 120 MHz) laser, collected by an objective with 0.5 numerical aperture. The normally incident excitation laser beam was focused on the sample to a spot size of ~10 μm^2. The temperature under different current excitation was measured by non-contact Raman scattering spectroscopy method,[15] performed on a micro-PL Raman spectrometer (Labram 800, Jobin Yvon Co., resolution: 1.9 cm^{-1}), and the micro-rod was excited with a continuous Argon ion laser (514.5 nm).

The sample was placed on two aluminium (Al) electrodes (the white regions), where a constant current source was applied. The length of ZnO micro-rod is ~190 μm, as can be seen in Fig. 1(a). Fig. 1(b) is the scanning electron microscope (SEM) image of the sample. It is clear that the cross-section of the sample is hexagonal and the facets are optically smooth, which is favorable for an optical microcavity. The quality factor (Q) of the optical modes (WG and quasi-WG) is around 1200. The lateral size is uniform (radius of 2.67 μm) along the C-axis of the sample, forming an ideal one-dimensional (1D) optical system. The insets in Fig. 1(b) are schematic diagrams of WG and quasi-WG modes in the sample, which has already been reported by Sun *et al.*[7,16]

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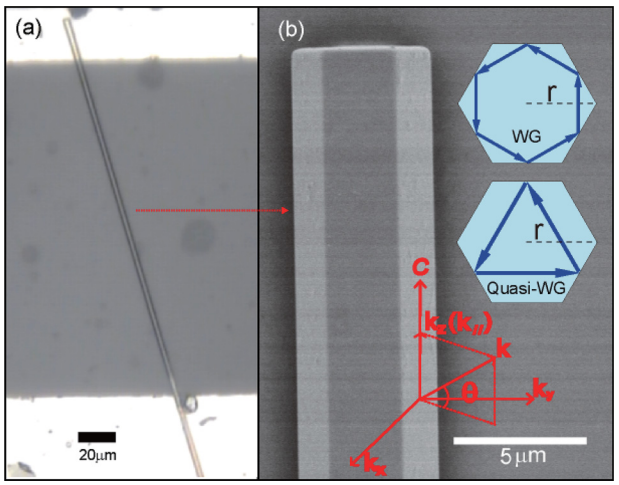

FIG. 1. (Color online) (a) A homogeneous ZnO micro-rod on two Al electrodes (the white regions); (b) SEM image of the ZnO micro-rod, the insets are schematic diagrams of whispering gallery (WG) modes and quasi-WG modes, the external emission angle θ is related to the polariton momentum component parallel to C-axis ($k_{//}$) by $k_{//} = (E/\hbar c)\sin\theta$.

Fig. 2 shows the dispersion of WG and quasi-WG LPBs (TE) at different current: (a) 0 mA, (b) 0.15 mA, (c) 0.25 mA, and (d) 0.28 mA (the behavior of TM LPBs is similar and not shown here). In Fig. 2(a), it can be seen that there are two groups of modes with different curvature and mode spacing. According to the transition selection rules, the two groups of modes with TE polarization are the results of strong coupling between A, B excitons and WG or quasi-WG optical modes.[7] The red and blue dashed lines are simulation results by diagonalizing the exciton-photon Hamiltonian

$$\hat{H}_{pol} = \sum_{\vec{k}}^{i=A,B} E_{Xi}(\vec{k})\hat{b}^{+}_{i,\vec{k}}\hat{b}_{i,\vec{k}} + \sum_{\vec{k}} E_C(\vec{k})\left(\hat{a}^{+}_{\vec{k}}\hat{a}_{\vec{k}} + \frac{1}{2}\right) + \sum_{\vec{k}}^{i=A,B} \hbar g_i(\hat{a}^{+}_{\vec{k}}\hat{b}_{i,\vec{k}} + \hat{a}_{\vec{k}}\hat{b}^{+}_{i,\vec{k}}), \quad (1)$$

where $\hat{H}_{pol}$ is the Hamiltonian of the polariton system, E_{XA} and E_{XB} are energies for A, B exciton, $\hat{b}^{+}_{i,\vec{k}}, \hat{b}_{i,\vec{k}}, \hat{a}^{+}_{\vec{k}}, \hat{a}_{\vec{k}}$ are creation and annihilation operators for excitons and photons, respectively, g_i is the coupling strength between A (or B) exciton and a photon.[17] E_C is the energy of a photon, and the background dielectric constant is chosen based on the reported value of 6.2.[18] Accordingly, the curves are identified as WG and quasi-WG LPBs, respectively. They fit well with the experimental data ($\hbar g_A = 0.034$ eV, $\hbar g_B = 0.174$ eV). The spacings of WG LPBs are smaller than those of the quasi-WG modes. For both WG and quasi-WG LPBs, the closer to A, B exciton (3.309 eV, 3.315 eV),[7] the smaller the curvature and mode interval, showing typical repulsion-like behavior of strong coupling between exciton and pure optical modes. The pure WG cavity optical modes are plotted as green dashed lines in Fig. 2(d), with larger curvature and equal mode interval compared to WG LPBs. When current increases, both WG and quasi-WG LPB modes shift to lower energy (red shift), as shown in Figs. 2(b)–2(d). Take one WG LPB (mode order of 86) for example, when the current increases from 0 mA to 0.28 mA (Fig. 2(d)), the red shift can be as large as ~40 meV. It can be used as a method to tune the energy positions of LPBs continuously. The linewidths of WG (mode order of 86) and quasi-WG (mode order of 76) LPBs hardly change when the current varies from 0 mA to 0.28 mA: about 3–5 meV for WG and 4–6 meV for quasi-WG modes, implying the quenching of interaction between polariton and phonon. This will be discussed later.

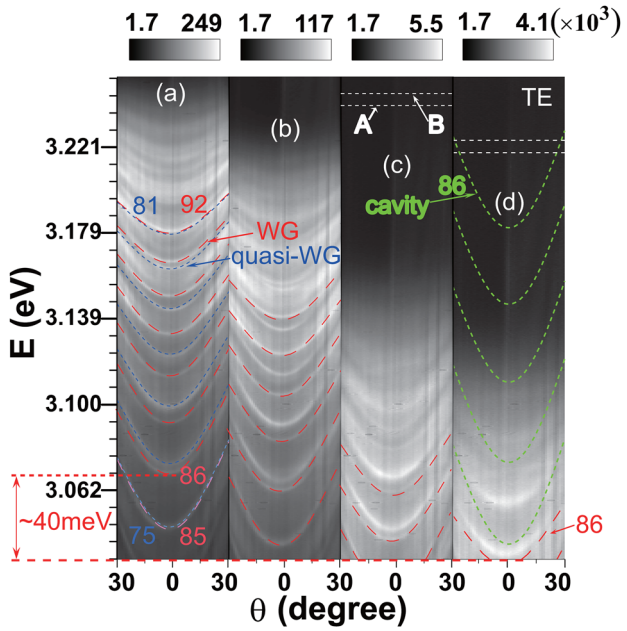

FIG. 2. (Color online) The dispersion of WG LPBs and quasi-WG LPBs (TE) at different currents: (a) 0 mA, (b) 0.15 mA, (c) 0.25 mA, and (d) 0.28 mA with linear gray scale (WG, quasi-WG lower polariton branch, and pure cavity mode are denoted as WG, quasi-WG, and cavity, respectively.).

The red shifts of WG and quasi-WG LPBs may come from current induced thermal effect. The temperature under different current was measured by non-contact Raman scattering spectroscopy according to[15]

$$T = h\nu_i \Big/ \left\{ k \ln\left[\frac{I_S}{I_{AS}} \left(\frac{\nu + \nu_i}{\nu - \nu_i} \right)^4 \right] \right\}, \quad (2)$$

where h is the Plank constant, ν is the frequency of the excitation source, ν_i is the Raman frequency shift, k is the Boltzmann constant, I_S and I_{AS} are the intensities of Stokes and anti-Stokes signal, respectively. The Raman-active phonon mode (E_2^{high}: 438 cm^{-1}) with the strongest Stokes and anti-Stokes peaks in $x(yy)\,\bar{x}$ scattering geometry was chosen to calculate the temperature, as shown in Fig. 3(a).[19] When current increases, the intensity of Stokes peak decreases and blue shift occurs, contrary to the behavior of anti-Stokes peak. By performing the Lorenz fit for each peak, the temperature at different current can be obtained (error smaller than ± 7 K), with the highest value of about 550 K under the current of 0.28 mA, as illustrated in Fig. 3(b). It is noted that the thermal effect caused by the incident laser spot is small, within the error range.[20] There is another concern that the Stark effect may also contribute to the red shift of LPBs. Given the maximum current $I = 0.28$ mA, the electric field strength across the sample is about 0.23 kV/cm, leading to a negligible shift of $\sim 10^{-5}$ meV for exciton.[21–23] Therefore, it can be concluded that the large red shift of LPBs is exclusively ascribed to thermal effect induced by the current.

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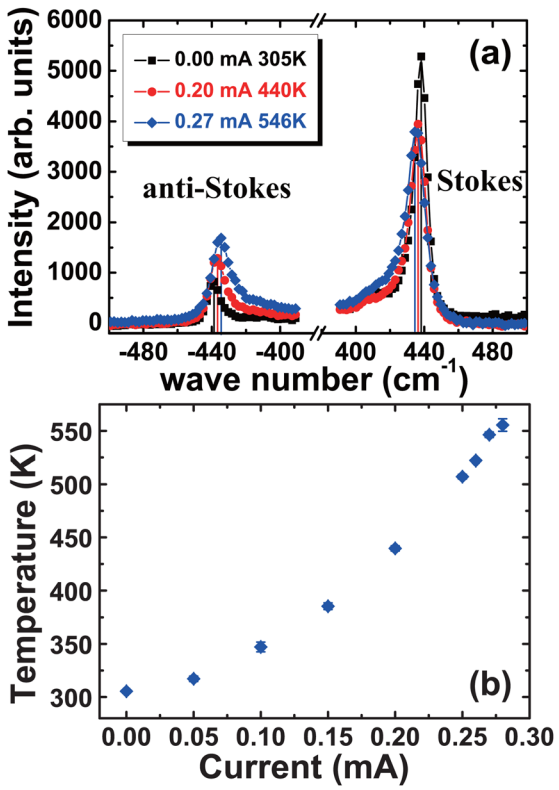

FIG. 3. (Color online) (a) Stokes and anti-Stokes Raman spectra under different currents; (b) temperature of ZnO micro-rod under different current.

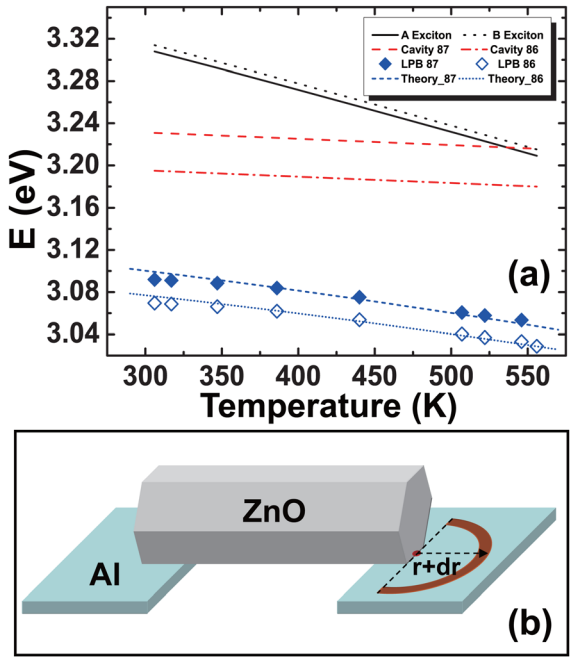

FIG. 4. (Color online) (a) Red shifts of A, B exciton, pure optical WG cavity modes (denoted as cavity 87, 86), experimental and theoretical WG LPBs at $k_{//}=0$ (mode order of 87 and 86, denoted as LPB 87 and 86, Theory_87 and 86, respectively) with temperature; (b) the schematic diagram of heat dissipation model: ZnO micro-rod on two Al electrodes.

Fig. 4(a) shows the red shifts of A, B exciton, pure optical WG cavity modes, WG LPBs (order of 86 and 87) with increasing temperature. The temperature dependence of A, B exciton can be described by Manoogian-Wooley (M-W) model: $E=E_0+\mathrm{UT}^s+\mathrm{V}\theta\ [\coth\ (\theta/2\,\mathrm{T})-1]$. Here, the second term is lattice dilatation, the third term is electron-phonon interaction, and the coefficient θ is related to the Debye temperature.[24] The pure optical WG cavity modes show a small red shift originating from the slight change of refractive index when the temperature increases,[10] as can be seen in Fig. 4(a). Therefore, the temperature dependence of WG LPBs at $k_{//}=0$ (mode order of 87 and 86) can be calculated: denoted as Theory_87 and 86, fitting well with the experimental data (LPB 87 and 86). The Rabi splitting is about 330 meV for the WG polariton modes, which is the same order as the reported values for ZnO.[25,26] The fitting of WG LPBs at high temperature in Figs. 2(b)–2(d) are also based on M-W formula. In Figs. 2(c) and 2(d), A, B exciton are illustrated as white dashed lines. The excitonic fraction of the 86th WG LPB at $k_{//}=0$ varies from 33.4% in Fig. 2(a) to 44.1% in Fig. 2(d). As mentioned above, the linewidths of WG and quasi-WG LPBs remain nearly constant while temperature increases from room temperature to ~550 K, which is different from the result of ZnO based distributed Bragg reflector (DBR) structure reported by Sturm *et al.*[10] According to A. Trichet *et al.*'s theory, when temperature increases, the thermal broadening increases greatly only for polariton modes within the energy range $E_{Xi}\sim E_{Xi}-E_{LO}$ $(i=\mathrm{A,B}$, $E_{LO}=72$ meV), while those at lower energy remain virtually unaffected.[8] Here, the 92nd order of the WG LPB at room temperature is about 120 meV lower than the exciton energy $((E_{XA}+E_{XB})/2)$, while the 85th order at 500 K is about 150 meV lower than that at the corresponding temperature, as shown in Figs. 2(a) and 2(c). They are all beyond the strong polariton-LO phonon scattering range and hardly interact with LO phonon, leading to the quenching of polariton-phonon interaction. This is the reason for the existence of LPBs and the nearly unchanged linewidths under such high temperature. As these 1D polaritons can decouple from phonon bath, high coherence may be preserved up to a very high temperature, which is essential for realizing high temperature polariton condensate, lasing, superfluidity, etc.

To estimate the tuning speed of this method, a simple heat dissipation model was considered with the schematic diagram shown in Fig. 4(b). Because the thermal conductivity of pure Al (204 W/m · K) is about 7500 times larger than that of air (0.027 W/m · K),[27] heat will mainly dissipate at the contact Al electrodes. Since Al electrode sheet is very thin (~60 nm), heat can be modelled to dissipate radially from the contact point. The thermal equilibrium equation can then be written as[27]

$$Cm\Delta T=[I^2R-2\pi\kappa d\Delta T/\ln(L/r)]t. \tag{3}$$

Here, C is the specific heat capacity of ZnO (0.1248 kcal/g deg),[28] m is the mass of the sample (density of ZnO: 5.606 g/cm^3),[29] ΔT is the variation of temperature. The two terms on the right hand side of the equation are heat production and dissipation power, where R is the resistance of the ZnO micro-rod (~100 kΩ), obtained by measuring its current-voltage (I-V) curve, κ is the thermal conductivity of Al, d and L (~100 μm) are the thickness and length of Al electrode, respectively, r is the radius of the sample (2.67 μm), t is the time for establishing thermal equilibrium. By solving this equation numerically, it is found that when ΔT is 250 K,

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thermal equilibrium can be established in about one millisecond (ms). This is confirmed experimentally by a pulsed current source (pulse width: 1 ms, period: 2 ms). The redshift within the pulse width is almost the same with that under *CW* current and vanishes quickly in the pulse interval, indicating the efficient temperature tuning by current. The fast tuning speed will be very helpful to control the temperature and consequently the energy positions of excitons and LPBs in ZnO microcavities.

In conclusion, we report a robust exciton-polariton effect in a ZnO WG microcavity at high temperature. The energy positions of WG and quasi-WG LPBs are tuned by current induced thermal effect. The red shift can be as large as ~40 meV. It is also demonstrated that strong coupling in ZnO can be preserved up to ~ 550 K, and the linewidths are nearly temperature independent, which is ascribed to quenching of polariton-phonon interaction. The tuning speed is fast (in the order of millisecond). Our results show opportunity of high temperature polariton condensate, lasing, superfluidity, etc. and polariton-based optoelectronic application such as wavelength-tunable polariton light emitting devices.

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