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ZnO as a material mostly adapted for the realization of room-temperature polariton lasers

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Wannier-Mott excitons in the wurzite-type semiconductor material ZnO are stable at room temperature, have an extremely large oscillator strength, and emit blue light. This makes ZnO an excellent potential candidate for the fabrication of room-temperature lasers where the coherent light amplification is ruled by the fascinating mechanism of the Bose condensation of the exciton polaritons. We report the direct optical measurement of the exciton oscillator strength f in ZnO. The longitudinal transverse splitting of the exciton resonances $\Gamma_5(B)$ and $\Gamma_1(C)$ are found to achieve record values of 5 and 7 meV, respectively, that, is two orders of magnitude larger than in GaAs. Second, we propose a model ZnO-based microcavity structure that is found to be the most adapted structure for the observation of the polariton laser effect. We thus can compute the phase diagram of the lasing regimes. A record value of the threshold power of 2 mW per device (at power density of 3000 W/cm^2) at room temperature is found for the model laser structure.

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During the last decade planar semiconductor microcavities (MC's) have attracted a lot of attention, due to the possibilities to enhance and control the interaction between light and electronic excitations in these structures. The control of this interaction is a key requirement for realizing ultralow threshold lasers. The observation of the strong coupling of light with excitons in semiconductor MC's has generated much speculation regarding the possibility for low threshold optical devices[1] and optical parametric oscillators.[2]

From the point of view of fundamental physics, MC's are particularly fascinating: they offer interesting possibilities to control the optical properties of excitons via their coupling with photons in resonance with the cavity mode. The polaritonlike coupling between the exciton and the photon mode of the cavities has been shown to produce pronounced Rabi oscillations splitting in the two-dimensional (2D) cavity spectra.[3–5] Contrarily to polaritons in bulk crystals, MC polaritons have a quasi-two-dimensional nature, with a finite energy at zero wave vector $k=0$ and are characterized by an extremely small in-plane effective mass. These unique MC polariton properties offer the possibility to study bosonic effects that cannot be achieved in the excitonic system. In particular, a large occupation number and Bose condensation at the lower polariton (LP) band bottom seem to be accessible at densities well below the onset of exciton bleaching. This paves the way towards the realization of so-called ''polariton lasers.'' A revolutionary feature of these devices is that no population inversion is required to achieve optical amplification in such a system: it starts as soon as the relaxation of excitatons into the ground LP state exceeds its escape time. The experimental observation of this effect is prevented, however, in conventional GaAs-based microcavities. The reason for this is the slow acoustic-phonon-mediated relaxation of photoexcited polaritons into the LP band bottom.[6–8] In addition, as their binding energy amounts to only 4 meV (6–10 meV in quantum wells), excitons are not stable at room temperature in GaAs-based structures. The strong exciton-light coupling regime necessary for polariton lasing does not hold at room temperature in GaAs. Thus, the realization of a commercial polariton laser device requires use of an alternative material. Although cubic II-VI semiconductors have been shown to be better adapted to this purpose than GaAs,[9] the problem of temperature stability of excitons is not entirely resolved to date, and to the best of our knowledge, polariton lasing remains to be seen.

In this work we consider a new wide-band-gap material, ZnO, as a candidate for the realization of polariton lasers operating at room temperature. It fits four major material requirements adapted for observing polariton lasing in the visible spectrum:

1. It emits light in the blue region of the electromagnetic spectrum.
2. It can now be doped p-type.[10]
3. ZnO excitons are stable at room temperature.
4. Light-matter coupling occurs with a record strength, as we will show below.

The literature is well documented with reflectivity measurements performed at liquid-helium temperature on bulk and thin film ZnO epilayers. We refer here to Refs. 11 and 12, although the list is not exhaustive. The spectra show several pronounced but strongly inhomogeneously broadened resonances at the energies of ground state ($1s$) of A,B,C excitons and near their excited states. In order to obtain exciton parameters with good accuracy and despite the inhomogeneous broadening, we have applied an original technique described in Ref. 13 that consists of the simple comparison of not only the frequency-resolved experimental spectra but also of their numerical Fourier transforms with theoretical calculations of the reflectivity and time-resolved reflection, respectively. Results of such a comparison are shown in Fig. 1 for $\Gamma_5(A)$ and $\Gamma_5(B)$ excitons. Analyzing the spectra in the time domain, one can distinguish between homogeneous and inhomogeneous broadening mechanisms and extract the exciton oscillator strength with good accuracy.[14] The excitonic characteristics obtained in this way are summarized in Table I. Note a large, record value of the

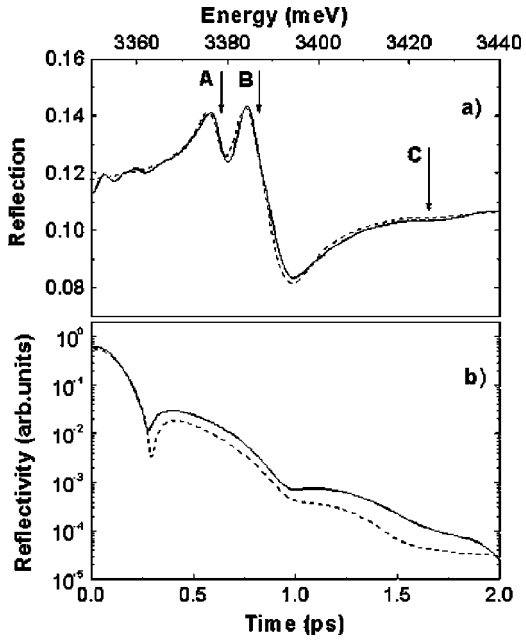

FIG. 1. (a) Reflection spectrum of the thick layer of ZnO grown on a sapphire substrate along the (0001) direction and (b) the time-resolved reflection obtained by the numerical Fourier transform of the frequency resolved spectra. Experimental data are shown by solid lines, theoretical results are shown by dashed lines. Arrows indicate the *A*, *B*, and *C* exciton resonances.

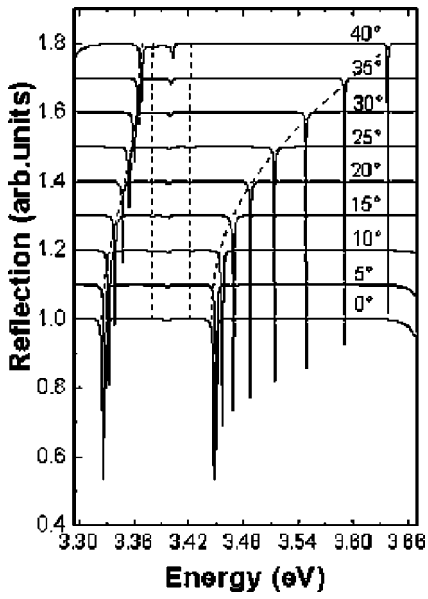

FIG. 2. Calculated spectra of a ZnO-based microcavity for different incidence angles. Dotted lines indicate the cavity eigenmodes.

exciton longitudinal-transverse splitting[15] for $\Gamma_5(B)$ and $\Gamma_1(C)$ resonances (5 and 7 meV, respectively, to be compared with 0.08 meV in GaAs). Note also that the inhomogeneous distribution of the exciton resonance is asymmetric, having a shorter wing at the low-energy side that is a manifestation of the *motional narrowing* effect.[16]

Next, we have examined theoretically the optical properties of a microcavity based on ZnO and grown along the [0001] direction. Zn-Mg-O alloys are available up to a Mg composition of 33%, to the best of our knowledge.[17] The model structure consists of the λ cavity of ZnO sandwiched between $ZnO/Zn_{0.7}Mg_{0.3}O$ Bragg mirrors having 14 and 15 pairs of quarter-wavelength layers in the top and bottom mirrors, respectively. A sapphire substrate has been assumed. The cavity photon mode has been placed at 3385.6 meV (at zero detuning from the *B* exciton resonance).[18] All the excitonic parameters are assumed to be the same as those gathered in Table I for Γ_5 excitons.

Figure 2 shows the calculated reflection spectra of the model microcavity at different incidence angles. A remarkable fact is that the vacuum-field Rabi splitting is as large as 120 meV in our system, which is the highest value recorded for a semiconductor microcavity. Definitely, the strong coupling regime in ZnO microcavities may be held at room temperature, which is one of the most important conditions for realization of the room-temperature polariton lasers.

Figure 3 shows the calculated exciton-polariton dispersion in our model microcavity. It shows four branches originating from three exciton resonances and the cavity photon mode. The lowest polariton branch is of the highest interest for the purpose of the polariton Bose condensation and lasing. Note that it is calculated for zero detuning between the photon mode and the exciton *B*. The eigenfunction of the fundamental state (at zero incidence angle) has less than 50% of the photonic component (see the inset of Fig. 2), which is essen-

TABLE I. Parameters for *A*, *B*, and *C* excitons in ZnO that were obtained from the reflection spectra in (0001) and $(10\bar{1}0)$ polarizations. $\hbar\omega_0$ is the exciton energy, $\hbar\omega_{LT}$ is the longitudinal-transverse splitting, Δ_1 (Δ_2) are the parameters of the inhomogeneous distribution of exciton resonances towards lower (higher) energies, and γ is the exciton nonradiative damping due to acoustic phonons. Note that data are given for both Γ_5 and Γ_1 excitons.

	Γ_5 exciton *A*	Γ_5 exciton *B*	Γ_5 exciton *C*	Γ_1 exciton *B*	Γ_1 exciton *C*
$\hbar\omega_0$ (meV)	3377.6	3385.6	3422	3380	3427
$\hbar\omega_{LT}$ (meV)	1.45	5	0.1	0.8	7
Δ_1 (meV)	2.8	3.5	7	4	5
Δ_2 (meV)	2.9	5	7	4	5
Γ(meV)	1	1	1	1	1
Reference	11	11	11	12	12

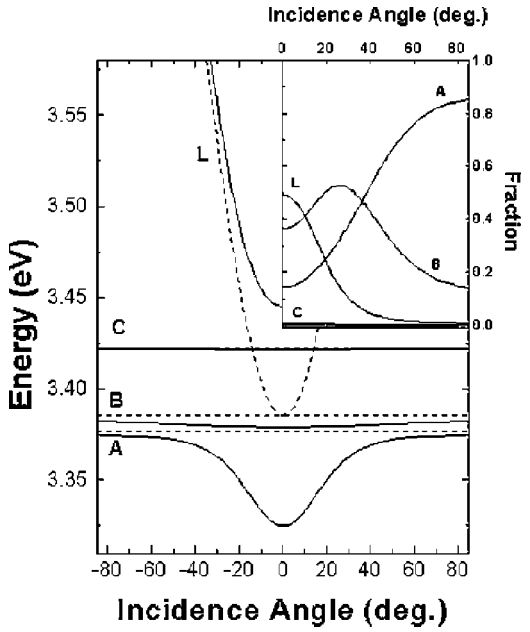

FIG. 3. Eigenenergies of the ZnO cavity modes versus the incidence angle. Dotted lines indicate the positions of the unperturbed modes: L indicates the light mode, and A,B,C indicate the A,B,C exciton resonances in ZnO, respectively. The inset shows the percentage of the excitonic components and the photonic component in the eigenmode of the lowest cavity-polariton state versus the in-plane wave vector.

tial for efficient polariton relaxation toward the ground state via scattering of the excitonic component with acoustic phonons, other excitons, and free carriers eventually introduced in the cavity. We expect no relaxation bottleneck phenomena in ZnO, because at room temperature the relaxation of polaritons via interaction with acoustic phonons is very efficient, especially if a small concentration of free holes is introduced to the cavity by p doping.[19]

Let us now examine the possibility of Bose condensation of exciton polaritons in our structure. The critical density of formation of the condensed phase by an ideal gas of bosons in a finite-size 2D system is given by[20]

$$n_c(T)=\frac{1}{(2\pi)^2}\int_{\vec{k},k>2\pi/R} N_{\vec{k}}(\mu=0)d\vec{k}, \qquad (1)$$

where

$$N_{\vec{k}}=\frac{1}{\exp\{[E(\vec{k})-E(0)-\mu]/k_BT\}-1}. \qquad (2)$$

$E(\vec{k})$ is the energy of the bosonic particle as function of its wave vector $\vec{k}$, μ is the chemical potential, k_b is the Boltzmann constant, T is the temperature, and R is the size of the system. Equations (1) and (2) yield also the critical temperature of Bose condesation for a given density.

Figure 4 shows the phase diagram of the lasing regimes in our model microcavity device. The solid line shows the critical density of polaritons versus temperature calculated from Eq. (1) using the polariton dispersion $E(k)$ from Fig. 3. The

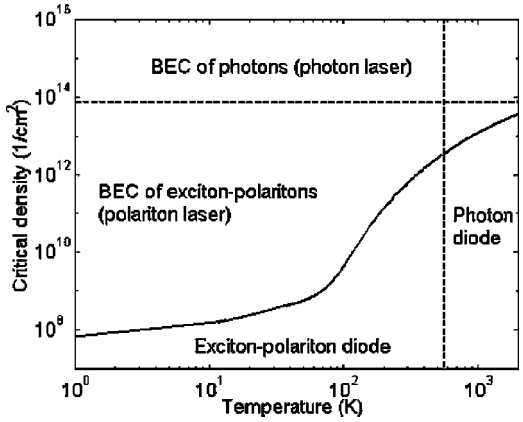

FIG. 4. Exciton-polariton phase diagram in the ZnO microcavity. The solid line shows the polariton critical density versus lattice temperature. The vertical dashed line shows the exciton thermal dissociation limit. The horizontal dashed line shows the Mott transition for excitons.

vertical and horizontal dashed lines show the approximate limits of the strong coupling regime in the microcavity that come from either exciton screening by the photoinduced electron-hole plasma (Mott transition in the excitonic system) or from the temperature-induced broadening of the exciton resonance. One can see that the Bose-condensed phase is formed in a large range of temperatures and pumping powers limited by a critical temperature. The critical temperature $T_c=560$ K is given by the exciton dissociation energy. Such a temperature would be, to the best of our knowledge, the highest critical temperature ever observed for the Bose condensation of a massive particle. Note that the critical temperature for the Wannier-Mott exciton Bose condensation is of the order of hundreds of mK (Ref. 20) and that the critical temperature of polariton lasing in conventional GaAs cavities estimated in the same way is 100 K for the best samples available. Beneath the critical density, the microcavity device operates in the regime of a polariton diode in the strong coupling regime, while in the weak coupling regime the device behaves like a conventional light-emitting diode. Beyond the critical density, in the weak coupling regime, the microcavity acts as a conventional laser. As follows from the phase diagram in Fig. 4, and assuming the exciton nonradiative lifetime of 100 ps (which is a typical value for the VECSEL structures currently used), the lasing threshold power in our microcavity device can be as low as 2 mW at room temperature.[21] This value is lower than the threshold power in any of the currently existing lasers, to our knowledge. The recent achievement of p doping[10] indicates that the realization of the polariton laser with electric pumping that requires simultaneous injection of electrons and holes in the cavity can be envisioned in the near future.

In conclusion, we have shown that a new generation of opto-electronic devices, namely, polariton lasers, can be realized with the use of p-doped microcavities based on ZnO. The model polariton laser we have considered here shows an extremely low threshold power (2 mW) at room temperature.

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[14] J. Lagois *et al.*, Phys. Status Solidi B **72**, 393 (1975), have reported slightly larger values of the exciton oscillator strength in ZnO, while the accuracy of that measurement was not as high because of the strong exciton inhomogeneous broadening.
[15] The longitudinal-transverse splitting is connected to the exciton oscillator strength f by the direct proportionality $\hbar\omega_{\mathrm{LT}} = (2\pi/\epsilon)(\hbar e^2/m_0\omega_0)(f/V)$, where ε is the dielectric constant, m_0 is the free electron mass, V is the volume of the semiconductor, and ω_0 is the exciton resonance frequency.
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[18] Note that in a bulk microcavity a part of the excitons is situated at the nodes of the cavity photon mode. They are weakly coupled to light and manifest themselves as weak dips between cavity-polariton eigenmodes.
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[21] We assume optical pumping by a laser beam having a spatial extension of 10 μm in the cavity plane, which corresponds to the optical power density of 3000 W/cm^2.