Polariton emission and reflectivity in GaN microcavities as a function of angle and temperature

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We demonstrate strong light-matter coupling at room temperature in a GaN microcavity using simultaneous reflectivity and photoluminescence measurements. At 10 K strong coupling is also observed in both measurements despite the well-known dominance of GaN emission at low temperature by localized neutral donor bound excitons. In addition, the strong light-matter coupling regime is studied as a function of temperature with the tuning of the polariton modes, in this case a result due to the dominant redshift of the excitonlike branch with increasing temperature.

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Recently there has been extensive research in the field of strong light-matter coupling in semiconductor microcavities.[1,2] Strong coupling describes the interaction of excitons and photons with the same energy and momentum to create quasiparticles conventionally termed cavity polaritons.[3] Of particular interest is the very low in-plane effective mass of these particles around $k=0$, which offers the possibility of producing a new generation of parametric optical amplifiers and ultralow threshold lasers[4] based upon stimulated scattering of polaritons.[5]

Strong light-matter coupling was first demonstrated in a semiconductor structure using an Al(Ga)As microcavity;[1] although interesting nonlinear effects were soon demonstrated,[5] these properties have been limited to cryogenic temperatures due to the inherently low exciton binding energy of this system. Recently, strong coupling has been observed at room temperature in the organic materials[6] and polariton scattering has been demonstrated in CdTe microcavities up to 220 K,[4] a direct result of the increased excitonic Rydberg of these systems. Despite this work GaN-based microcavities appear to be the most promising candidates to realize room temperature devices based upon stimulated scattering since the nitrides have large exciton binding energies[7] and high oscillator strengths, are mechanically and environmentally stable, and can be controllably doped *n* and *p* type. Consequently evidence of strong light-matter coupling has now been observed experimentally at room temperature in nitride[8–14] microcavities. Although early work focused on reflectivity measurements,[8–10] it is necessary to observe strong coupling in the luminescence to study interesting nonlinear effects and develop a new generation of optical devices. Evidence of polariton emission in the nitride system was first reported by Tarawa *et al.* in 2004.[11,12] However, these results remain controversial due to the very large emission linewidth (200 meV) with respect to the vacuum Rabi splitting (6 meV). Recently the emission from (Al)GaN microcavities based on (Al,In)N/(Al,Ga)N distributed Bragg reflectors (DBRs)[13,14] was attributed to strong coupling through the analysis of the room temperature photoluminescence spectra. The work in Refs. 13 and 14, however, focuses only on the room temperature emission and does not discuss the evolution of the strong coupling with temperature. This evolution is particularly important since the emission in nitrides is rather complex and can include contributions from free and localized excitons as well as phonon-assisted recombination.[15] Furthermore, we believe it is also necessary to confirm the origin of the polaritons using both luminescence and reflectivity, particularly at 300 K where the resolution of the polariton branches is limited by thermal broadening.

In this paper we present strong light-matter coupling observed in both the reflectivity and photoluminescence (PL) from low to room temperature. The microcavity structure consists of a bulk $\lambda/2$-GaN active region grown by molecular beam epitaxy (RIBER Compact 21) on a ten-period $AlN/Al_{0.2}Ga_{0.8}N$ distributed Bragg reflector (DBR) grown directly on Si (111). During the deposition of the GaN active layer, the substrate rotation was stopped to create a thickness gradient within the active layer. The microcavity was completed by the plasma-assisted chemical vapor deposition of an eight-period SiN/SiO_2 DBR. Angular resolved and position-dependent reflectivity was excited using a standard Xenon bulb and collected using a 50 μm optical fiber mounted on a rotating stage with an angular resolution of $\sim 1°$ and detected with a silicon charge coupled device (CCD). The angle-resolved and position-dependent PL spectra were excited using the frequency-doubled emission of a Rhodamine dye laser at 300 nm, pumped with the frequency-doubled emission of a 76 MHz Nd-yttrium aluminum garnet (YAG) laser.

Figure 1(a) shows the PL as a function of position at 10 K. At position 1, the photonlike mode (E_{PH}) at 3.463 eV is negatively detuned with respect to the excitoniclike transitions around 3.53 eV. The latter consists of a dominant transition at 3.514 eV (E_{X1}) and a shoulder at 3.538 eV. Polariton modes due to the interaction of the *A* and *B* excitons

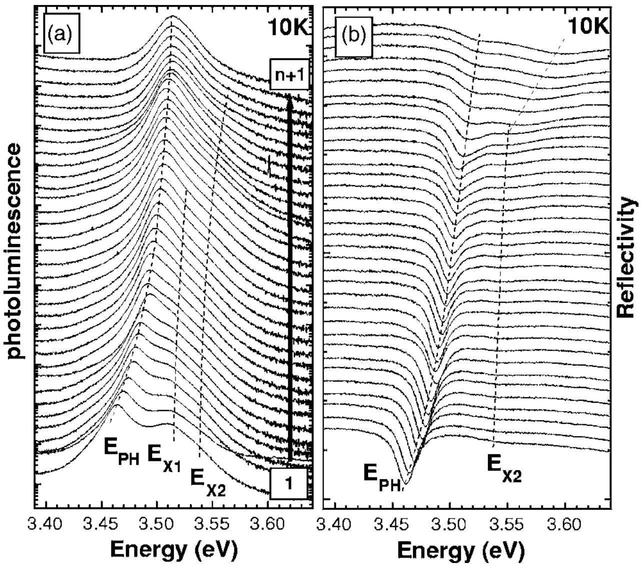

FIG. 1. Position dependent (a) photoluminescence (log) and (b) reflectivity spectra at 10 K respectively. In both cases the photonic mode and excitonic transitions are labeled E_{PH}, E_{X1}, and E_{X2} respectively. The bold line in (a) illustrates the different positions on the sample. The dashed lines are a guide for the eye only.

in GaN microcavities has been observed previously at low temperature, since at $k=0$ the oscillator strength of these two states is comparable.[10] However, in this case the experimental separation of $\sim$24 meV between E_{X1} and E_{X2} is large in comparison to the predicted,[16] and experimentally confirmed,[10] *A* and *B* separation in GaN under high biaxial (0001) compression ($\sim$11–13 meV). At 10 K the low energy excitonic transitions in the PL spectra of GaN are a combination of localized donor and/or disorder bound excitons and *A*-free excitonic emission, we attribute then the broad E_{X1} feature to such a combination. With this is mind, we also tentatively attribute the E_{X2} feature to *B*-free excitons, since its energy is very similar to that previously measured in highly strained GaN grown on $AlN/Al_{0.2}Ga_{0.8}N$ DBRs.[10] As the position is changed, the photonlike branch shifts to higher energy consistent with a decrease in the thickness of the active region and a clear anticrossing of the polariton modes is observed, even if the population of the upper polariton branch is quite small at $T=10$ K.

The position-dependent reflectivity spectra measured at the same points as Fig. 1(a) are shown in Fig. 1(b). At position 1, the photonlike mode ($Q\sim163$) is again negatively detuned with respect to the broad excitonlike transitions around 3.53 eV. The large negative detuning at position 1 results in a relatively weak excitonic mode since the photonic contribution to this polariton mode is low and the optical spectra are rather a probe of the external photons that *leak* from the microcavity. Consequently, as the photonlike mode is tuned towards the excitonic component the resolution of the latter improves. The overall behavior of the polariton modes in reflectivity as a function of the detuning reproduces that of the emission, with an anticrossing of the modes again observed. The demonstration of an anticrossing in both the reflectivity and PL confirms the origin of the features present in the two measurements to that of polariton modes in the strong-coupling regime.

At low temperature the emission from GaN is dominated by localized neutral donor bound excitons.[15] However, due to the huge strain inhomogeneities present in thin GaN layers grown on AlN/(Al,Ga)N DBRs, especially when grown on Si,[10] the resolution of the spectral features is low. As a result it is not possible to resolve the free and bound excitonic states at the E_{X1} energy observed in the PL (Fig. 1). As such, it is not possible to comment on the contribution of the bound exciton levels to the strong light-matter coupling observed. However, since their oscillator strength is expected to be weaker than that of the free excitons (the ratio should be of the order of neutral donor density/molecular density[17]) their contribution is expected to be low.

Increasing the temperature of the structure will result in a significant redshift in the excitonic position. An experimental redshift of the excitonic mode of $\sim$50 meV is observed between 10 K and 300 K for the GaN epilayer prior to the deposition of the upper dielectric DBR, and although the photonic mode is also slightly redshifted with increasing temperature,[18,19] this change is less significant ($\sim$10 meV). As a result, the temperature can be used as an effective way to tune the polariton modes. This method of tuning the polariton branches in the strong coupling regime has been demonstrated previously in the GaAs system.[20–22] Here, a suitable position on the sample wedge was chosen such that the photonic mode (3.505 eV) was negatively detuned with respect to the exciton ($\sim$3.55 eV), but within the range of the temperature-dependent redshift of the excitonic branch. As such, this position is much less negatively detuned than that of position 1 in Figs. 1(a) and 1(b). Figures 2(a) and 2(b) show the temperature-dependent reflectivity and photoluminescence measured in the temperature range between 10 K and 315 K respectively with an anticrossing evident in both cases.

This anticrossing is reported in Fig. 2(c) where the energy of the polariton modes for the reflectivity (open squares) and PL (closed circles) are plotted as a function of temperature. The anticrossing in both the reflectivity and PL confirms the origin of the features in the temperature-dependent spectra as that of the upper and lower polariton branches. Also shown in Fig. 2(c) (closed stars) is the temperature-dependent PL of the half microcavity prior to the deposition of the upper dielectric DBR. This PL, which has been blueshifted for clarity, is very different from the dispersion of the polariton branches as a function of temperature, and displays the well-known S-shaped temperature dependence. Such behavior indicates that the emission is dominated by localized states at low temperature.[15] At $T>80$ K the PL energy follows the GaN excitonic band gap[15] (dashed line), indicating the emission is dominated by free excitonic transitions between 80 K and 315 K. Figure 2(c) also shows the results of the numerical analysis (bold lines) in which the variation of the GaN band edge and the cavity mode are included as a function of temperature, coupled with a Rabi-energy of $\sim31\pm4$ meV. This analysis reproduces the variation of the upper and lower branches very well again confirming the strong light-matter coupling regime in both the reflectivity and PL.

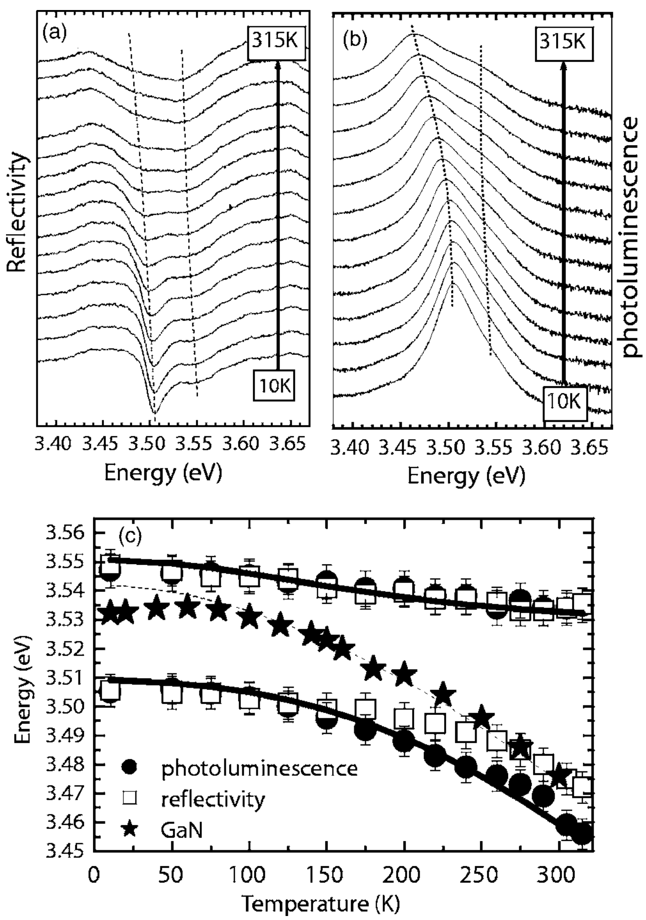

FIG. 2. Temperature dependent (a) reflectivity and (b) photoluminescence (log) spectra between 10 K and 315 K. (c) Variation of the polariton mode energies of the reflectivity (open squares) and photoluminescence (closed circles) as a function of temperature. Also shown is the temperature dependent photoluminescence of the half GaN cavity (closed stars) fitted to the Varshni equation (dotted lines). The bold lines indicate the numerical simulations using transfer matrix analysis of the upper polariton branch and lower polariton branch as a function of temperature.

Angle-resolved measurements offer an alternate method of tuning the polariton modes without changing the position or temperature of the sample. Figures 3(a) and 3(b) show the angle-resolved reflectivity and PL of our sample at 300 K. In the reflectivity spectrum at 5°, the photonic mode is evident at ~3.444 eV, negatively detuned with respect of the excitonic mode at 3.499 eV. As the angle is increased an anticrossing is observed with a resonance at ~35° and a Rabi-energy of ~32±4 meV. The PL spectra display very similar behavior, with both polariton branches evident at all angles.

The energy position of the polariton modes as a function of angle for both the reflectivity (open squares) and PL (closed circles) at 300 K are shown in Fig. 4. Again, a clear anticrossing is evident in both cases. Despite the limited resolution of the features observed in the angle-resolved PL, they are consistent with those observed in the reflectivity and their origin can once again be attributed to the polariton branches of a strongly coupled system. Also shown in Fig. 4 (in bold) are the results of a (3×3) matrix calculation, considering the system as the interaction between A and B excitons of constant energy and the angle-dependent photonic mode. The energy of the A and B excitons used are 3.481 eV and 3.494 eV, respectively. The separation energy of 13±6 meV was confirmed by PL measurements of the GaN layer prior to the deposition of the upper dielectric DBR and is consistent with that of compressively strained GaN.[10,16] The resulting eigenvalues reproduce the experimental data very well with a Rabi-energy of 31±6 meV determined between the upper and lower branches at resonance. The inclusion of both A and B excitons in the analysis, despite the screening of their resolution through inhomogeneous and thermally mediated homogeneous broadening, is justified since the oscillator strength of the A and B excitons in biaxially compressed GaN at near $k=0$ is almost equal.[3,16] The C exciton is not included in this analysis since, at near normal incidence, it is at much higher energy and has considerably

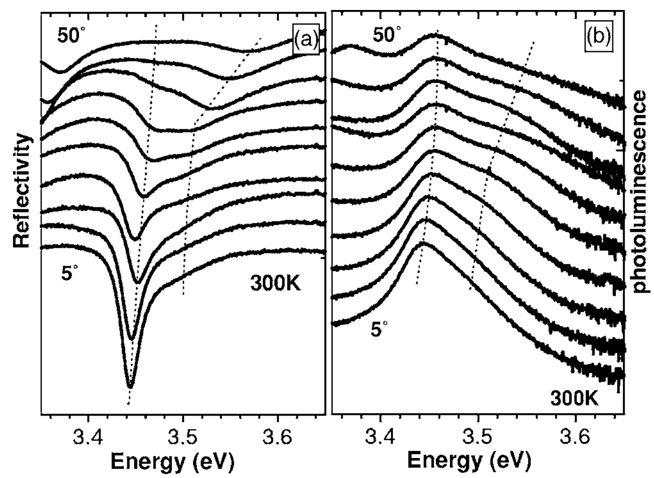

FIG. 3. Angle-resolved (a) reflectivity and (b) photoluminescence (log) spectra at 300 K. The dotted lines are a guide for eye only.

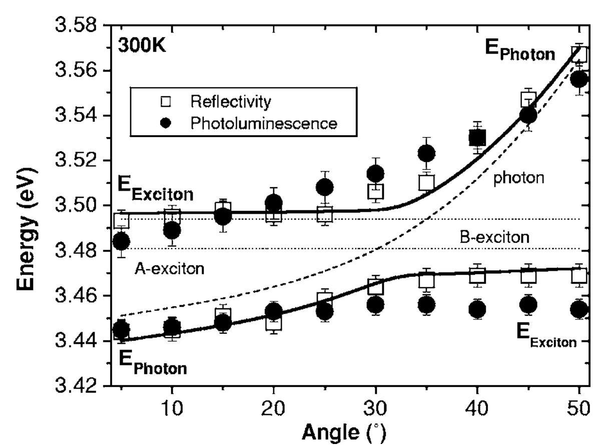

FIG. 4. Variation with the angle of incidence of the polariton mode energies at 300 K for both the reflectivity (open squares) and photoluminescence (closed circles). Also shown as a solid line is the numerical simulation using (3×3) matrix analysis. The uncoupled excitonic (dotted lines) and photonic modes (dashed line) as a function of angle are also included in both cases.

weaker oscillator strength than that of the A and B excitons.[16,19] Finally, we note that in Figs. 2(c) and 4 that the PL transitions at room temperature are slightly Stokes-shifted by ~15 meV relative to reflectivity in the case of large positive detuning. This suggests that though the PL energies follow the excitonic gap for $T > 80$ K [Fig. 2(c)], the active layer quality is still not optimum, with exciton localization persisting up to room temperature. Such analysis is also consistent with recent time-resolved photoluminescence measurements.[23]

In summary we have demonstrated strong light-matter coupling at room temperature in both reflectivity and photoluminescence. Furthermore, we have shown that at 10 K it is possible to observe strong light-matter coupling in the luminescence despite the dominance of localized excitons in GaN at low temperature. Strong light-matter coupling is also presented as a function of temperature in both PL and reflectivity measurements.

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