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Impact of inhomogeneous excitonic broadening on the strong exciton-photon coupling in quantum well nitride microcavities

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GaN/AlGaN and InGaN/GaN quantum wells (QWs) are investigated as the active region for room-temperature strong exciton-photon coupling in high-quality AlInN/(Al)GaN microcavities (MCs). Angular resolved photoluminescence (PL) measurements performed on an AlInN/AlGaN MC with GaN/AlGaN QWs reveal cavity polariton dispersion curves. A vacuum-field Rabi splitting of 30 meV is observed, from which an exciton oscillator strength of 4.8×10^{13} cm^{-2} per QW is deduced, a value ten times larger than for InGaAs QWs. In contrast, the PL spectra of an InGaN/GaN QW MC do not exhibit such an anticrossing behavior as should be expected from the strong-coupling regime (SCR). By modeling cavity absorption and PL spectra at the resonance, the conditions, in terms of inhomogeneous excitonic broadening, for the observation of the SCR in these nitride MCs are determined.

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Since its first demonstration in GaAs-based microcavities (MCs) more than a decade ago,[1] the strong exciton-photon coupling regime (SCR) in solid state systems has generated an intense research effort.[2] In this context, semiconductor MCs have become the structure of choice thanks to the ability to modulate the interaction strength between excitons and cavity photons. Planar MCs containing either quantum wells[1,2] (QWs) or a bulk layer[3,4] (organic or inorganic) as an active region form the prototypical system to investigate the physics of cavity polaritons (CPs), the quasiparticles resulting from the strong interaction between cavity photons and excitons. Recently, CPs underwent a renewed interest owing to their large nonlinear emission properties triggered by their bosonic behavior.[5] In particular, their low density of states[2] could allow for the formation of dynamical Bose-Einstein condensates. Besides fundamental studies, semiconductor MCs operating in the SCR are promising candidates to develop optoelectronic devices based on CP nonlinearities such as low-threshold nonlinear light emitters[6] or ultrafast micro-optical amplifiers.[5,7]

However, the realization of functional devices based on CPs operating at room temperature (RT) is out of reach of present high-quality inorganic semiconductor MCs such as III-V arsenide-based structures. This is primarily due to their reduced exciton binding energy (E_X^B). The larger E_X^B in II-VI compounds allowed for the observation of optical parametric amplification operation up to 220 K in CdTe-based MCs.[7] However, technological applications using wide-bandgap II-VI materials have not been developed so far. By contrast, group-III nitrides exhibit both a large E_X^B ($\sim$26 meV for GaN and $>$40 meV for narrow QWs) (Ref. 6) and a large coupling to the light field as is also the case for organic semiconductors.[3,8] Consequently, nitride-based MCs appear to be promising for the investigation of the SCR at RT. Although the study of CPs in group-III–nitride MCs is still in its infancy, SCR at RT has been reported both in bulk GaN structures[9,10] and in InGaN-based QW MCs.[11,12]

In this Brief Report, we report a RT study of two MCs containing either $In_{0.15}Ga_{0.85}N/GaN$ or $GaN/Al_{0.2}Ga_{0.8}N$ QWs. The latter exhibits an anticrossing behavior with a vacuum Rabi splitting (Ω_{VRS}) of 30 meV whereas the former shows ambiguous photoluminescence (PL) features, which are eventually not ascribed to CPs. Finally, the conditions for achieving SCR in these planar nitride MCs, in which the QW inhomogeneous broadening is expected to play a key role, are determined through a linear dispersion model.

All the samples are grown by metal-organic vapor phase epitaxy on *c*-plane sapphire substrates after the deposition of a standard GaN buffer layer. The first one, a GaN MC with $In_{0.15}Ga_{0.85}N/GaN$ QWs, consists in a $3\lambda/2$ (λ $\sim$415 nm) cavity layer with two sets of three QWs surrounded by two $Al_{0.83}In_{0.17}N/GaN$ distributed Bragg reflectors (DBRs): a 28-pair bottom one and a 23-pair top one. The second sample, an $Al_{0.2}Ga_{0.8}N$ MC with $GaN/Al_{0.2}Ga_{0.8}N$ QWs, is made of a 35-pair lattice-matched $Al_{0.85}In_{0.15}N/Al_{0.2}Ga_{0.8}N$ DBR, a $3\lambda/2$ (λ $\sim$345 nm) cavity layer with two sets of three QWs, and finally a 13-pair SiO_2/Si_3N_4 dielectric DBR. In both structures, the QWs are inserted at the antinodes of the cavity light field to maximize the light-matter interaction. Details on the specific growth conditions of AlInN-based DBRs are reported elsewhere.[13,14] In addition, single $GaN/Al_{0.2}Ga_{0.8}N$ and $In_{0.15}Ga_{0.85}N/GaN$ QWs were grown in order to assess their optical properties before their insertion in MCs.

Standard and angle-resolved PL measurements are performed in reflection geometry under weak excitation power density (P $\sim$50 W cm^{-2}) using either the UV line (363.8 nm) of a continuous wave (cw) Ar$^+$ laser or the 244 nm line of a cw Ar$^+$ laser frequency-doubling unit. Angle-resolved measurements are performed with the excitation beam focused on the sample at a fixed angle while the PL is collected at various angles by a 600 μm-core UV fiber with an angular resolution below 1° and detected by a UV-enhanced charge-coupled device monochromator combination.

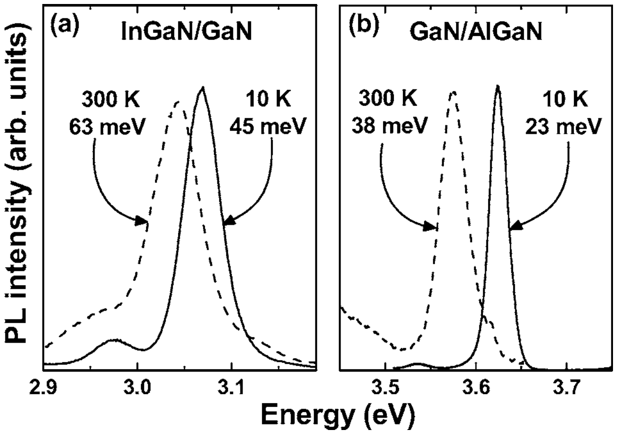

FIG. 1. Low-temperature and RT PL spectra of (a) a single $In_{0.15}Ga_{0.85}N/GaN$ QW and (b) a single $GaN/Al_{0.2}Ga_{0.8}N$ QW.

First, the properties of InGaN and GaN QWs are investigated by standard PL. Usually, InGaN QWs are the heterostructure of choice when dealing with high-efficiency nitride-based light emitters.[15] This is recognized to result from the large degree of carrier localization induced by In composition fluctuations, which inhibits nonradiative recombinations on structural defects such as dislocations.[16] However, these fluctuations lead to an important inhomogeneous broadening[17] which might be detrimental to the SCR.[18] In Fig. 1, we report the PL spectra of extracavity 5-monolayer (ML; 1 ML=0.259 nm) thick InGaN- and GaN-based QWs. The PL linewidths of InGaN QWs at 10 and 300 K are 45 and 63 meV, respectively. They correspond to state of the art values.[19,20] By comparison, the PL linewidths of GaN-based QWs are almost twice smaller and also correspond to the state of the art for GaN QWs grown on *c*-plane sapphire substrates.[21] The low-temperature full width at half maximum (FWHM) being mainly governed by inhomogeneous mechanisms, it can be identified as the inhomogeneous linewidth σ. At RT the homogeneous linewidth γ, essentially due to phonon scattering, cannot be neglected any longer. The measured RT linewidth is thus the convolution of a homogeneous line (Lorentzian) of FWHM γ and an inhomogeneous line (Gaussian) of FWHM σ. As a result, a good fit of the 300 K QW PL spectra gives γ values equal to 29 and 23 meV for InGaN and GaN QWs, respectively.[22]

Another point to consider when dealing with nitride heterostructures is the quantum confined Stark effect (QCSE) arising from both spontaneous and piezoelectric polarizations, which strongly reduces the QW oscillator strength (f_{ex}) in wide wells.[21,23] f_{ex} is indeed a key parameter to achieve SCR as Ω_{VRS} is directly linked to this value.[2] For both types of QWs the square of the overlap integral of the electron and hole wave functions[24] (which is proportional to f_{ex}) has been calculated in the envelope function approximation, including the electric field. It is found that an important decrease of f_{ex} with increasing QW thickness occurs. This is why we have considered very thin (5 ML thick) QWs to preserve a large f_{ex} while keeping a small FWHM σ. We also observe that for this QW thickness, f_{ex} is similar for InGaN/GaN and GaN/AlGaN QWs.

RT angular-resolved PL measurements are then performed on the MCs. Spectra measured from 0 to 40° on the InGaN QW MC are reported on Fig. 2(a). Two peaks with a splitting of about 14 meV are clearly seen. These PL spectra are similar to those observed in previous reports of SCR in InGaN QW MC.[11,12] These peaks follow the near-parabolic angular dispersion of a Fabry-Perot resonator and cross the QW energy (dashed line, E_X=2.97 eV at RT). In other words, no anticrossing behavior, characteristic of the SCR, is observed. Consequently, this sample does not exhibit strong exciton-photon interaction despite the observation of two peaks. The origin of this peculiar PL feature will be discussed hereafter. Similar measurements performed on the GaN QW MC are reported on Fig. 2(b). In this case, only one peak is observed at low angle and a second one appears at large angles. Furthermore, the angular dispersion is different: there is an asymptotic approach toward a fixed energy which corresponds to the uncoupled QW exciton energy (E_X=3.625 eV at RT). The high-energy peak appearing at high angles is better resolved in semilogarithmic scale [Fig. 3(a)] and its dispersion can clearly be measured from 28 to 40°. In addition, it is seen that this dispersion curve cannot be the extension of the low-angle dispersion curve of the low-energy peak, indicating that there is no crossing between those two branches. Such dispersion curves are typical of the anticrossing behavior observed between a cavity mode and an exciton in a planar MC,[1,2] and do correspond to the so called lower and upper polariton branches. It is worth pointing out that the weak separation between the peaks close to the anticrossing angle indicates that the system is at the limit of the SCR. The energy positions of these two peaks are reported versus in-plane wave vector[25] ($\boldsymbol{k}_{\parallel}$) on Fig. 3(b) allowing the extraction of a 30 meV Rabi splitting value.

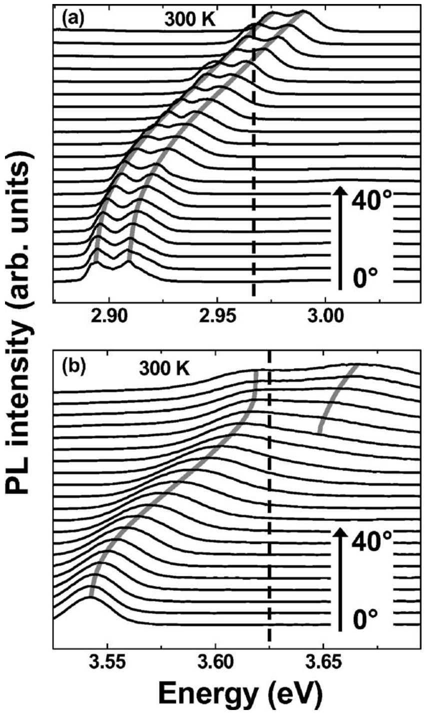

FIG. 2. RT angular-resolved PL spectra ranging from 0 to 40°, measured each 2° and upshifted for clarity. Bold lines are a guide for the eye. The dashed lines correspond to QW exciton energies. (a) InGaN-QW MC. (b) GaN-QW MC.

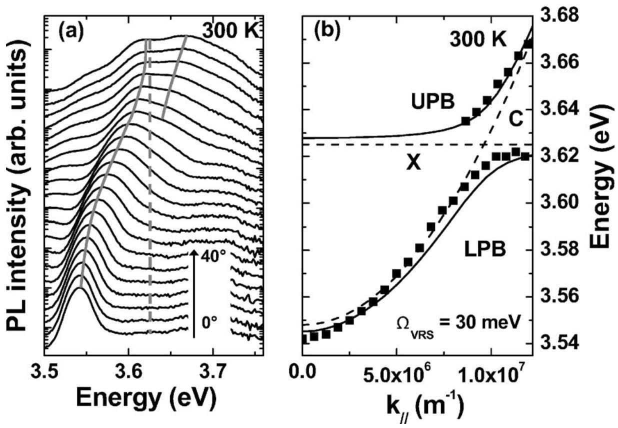

FIG. 3. (a) Angular-resolved PL spectra (semilogarithmic scale) measured on the GaN QW MC ranging from 0 to 40°, measured each 2° and upshifted for clarity. Bold lines are a guide for the eye. The dashed line corresponds to the QW exciton energy. (b) Experimental dispersion curves (black squares) and fits of the lower (LPB) and upper (UPB) polariton branches. The cavity mode (C) and the uncoupled exciton (X) are also reported.

The fitted dispersion curves were deduced from a standard 2×2 matrix Hamiltonian describing the coupling between the uncoupled exciton and the cavity mode and including the homogeneous broadening. From the value of Ω_{VRS}, we deduce an f_{ex} of 4.8×10^{13} cm^{-2} per QW.[18] This value is approximately ten times larger than that measured for InGaAs QWs embedded in a GaAs microcavity[26] and confirms the stronger light-matter interaction in III-V nitrides compared to III-V arsenides. Note that the present GaN QW MC exhibits only regions of large negative detunings (the energy separation between uncoupled excitons and cavity mode at $|k_{\parallel}|=0$).

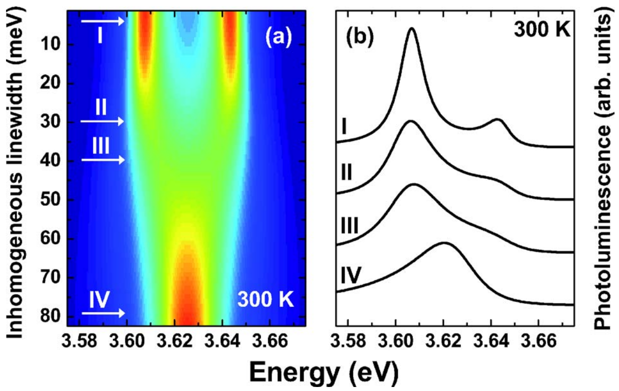

FIG. 4. (Color online) (a) Effect of QW inhomogeneous linewidth on the GaN QW MC absorption spectra at the exciton-photon resonance (i.e., zero detuning). (b) PL spectra corresponding to the arrows I–IV in (a).

To investigate further the conditions leading to the SCR in nitride-based QW MCs, which exhibit a significant inhomogeneous broadening, the GaN QW MC is modeled using the transfer matrix method.[27] The refractive indices for nitride compounds are taken from the work of Brunner *et al.*[28] (AlGaN alloys) and Carlin *et al.*[13] (AlInN alloys). The optical properties of SiO_2 and Si_3N_4 layers were determined using thick layers grown on sapphire substrates. The quantum wells are modeled by a Lorentz oscillator dispersive dielectric function $\epsilon(\omega)$.[18] To account for the inhomogeneous linewidth of the excitonic transition, a normalized Gaussian distribution $g(\omega)$ of FWHM σ is introduced: $\epsilon_{tot}(\omega)=\int_{-\infty}^{+\infty}\epsilon(\omega-\omega')g(\omega')d\omega'$.[18] From the PL measurements carried out on the extracavity single GaN QW, γ is fixed at 23 meV. Absorption spectra computed for various values of σ are reported on Fig. 4(a). The transition from the strong- to the weak-coupling regime can clearly be seen on this figure, the latter occurring between 35 and 50 meV. Taking the presence of two well-resolved peaks (i.e., there is a minimum between the peaks) in the absorption spectra as the relevant parameter to be in the SCR,[26] we observe with this figure that this condition is satisfied for σ up to 45 meV. This corresponds to a total RT QW linewidth of 60 meV, i.e., a linewidth larger than the Ω_{VRS} value. Such an observation has already been reported for GaAs-based MCs.[29] To compare absorption spectra to PL ones, we assume that the latter have a thermal line shape, i.e., PL spectra are well accounted for by the absorption spectra multiplied by a Boltzmann factor.[30] Calculated PL spectra are reported on Fig. 4(b). It is seen that, when considering an inhomogeneous broadening around 40 meV, the line shape of spectrum III is in good qualitative agreement with the experimental PL spectra measured close to the anticrossing [Fig. 2(b)]. Note that this linewidth is larger than that observed for a single QW in Fig. 1(b). However, it is in good agreement with PL measurements performed on the half GaN QW MC, i.e., before depositing the top dielectric DBR, which revealed an inhomogeneous excitonic linewidth σ about 30 meV (not shown here). This can be explained as two sets of three QWs grown on top of a DBR will inevitably lead to a PL broadening. Note also that the absorption spectra corresponding to arrows II and III on Fig. 4(a) exhibit two well-resolved peaks whereas the corresponding PL spectra show hardly separable peaks.

These simulations also confirm the lower potential of InGaN QWs as their larger σ would be far less compatible with SCR. On the other hand Fig. 4(a) shows that the system should exhibit clearly separated peaks provided that GaN/AlGaN QWs embedded in the cavity have σ values ~20 meV. Note also that PL linewidths in the 5–10 meV range have been reported for GaN QWs grown on *a*-plane bulk GaN substrates as the latter are not subject to a built-in electric field.[31] The QCSE is thus clearly a key issue for the SCR in GaN based MCs.

Finally, we point out that the presence of two peaks on the PL spectra of the InGaN QW MC still remains unclear. One possible explanation might be the presence of cavity thickness fluctuations. This splitting would correspond to (5–6)-ML-thick fluctuations and is kept almost unchanged along the wafer radius. In the previous report of strong exciton-photon interaction in a MC with InGaN/AlGaN QWs (PL linewidth about 200 meV at RT),[11,12] a similar splitting (17 meV) is attributed to the SCR. However, the standard linear dispersion model we used to simulate MCs taking into account the inhomogeneous excitonic broadening,[18] would not reproduce these observations. Indeed, using the parameters quoted in Ref. 11, namely, ten QWs, $f_{ex}=2$

$\times 10^{13}$ cm^{-2} per QW, and a QW inhomogeneous linewidth larger than 150 meV, our modeling would result in a single PL peak.

In conclusion, cavity polariton emission has been demonstrated at RT by means of angular resolved PL measurements in a hybrid AlInN/AlGaN MC containing GaN/AlGaN QWs. An exciton oscillator strength of 4.8×10^{13} cm^{-2} per QW has been estimated. The use of GaN/AlGaN QWs versus InGaN/GaN QWs has been compared; the former appearing more suitable for strong exciton-photon coupling purposes due to their significantly lower inhomogeneous broadening. In this respect, the development of nonpolar nitride-based QW MCs would also be a promising route to observe clearly resolved CP branches as the detrimental impact of the QCSE should be eliminated.

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