Observation of Rabi splitting in a bulk GaN microcavity grown on silicon

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(Received 22 June 2003; published 24 October 2003)

We report the experimental observation of the strong-coupling regime in a nitride-based microcavity. The active layer in the optical cavity consists of a λ/2 GaN layer sandwiched between a dielectric mirror and the silicon substrate, which acts as the bottom mirror. Reflectivity measurements have been performed under various angles of incidence at $T=5$ K, producing evidence of strong-coupling behavior between the exciton and the cavity mode. A Rabi splitting of 31 meV is obtained experimentally. Transfer-matrix simulations have allowed us to account for the exciton-photon interaction. From these calculations, the oscillator strengths of the *A* and *B* excitons are evaluated and these values are in good agreement with those previously determined in bulk GaN.

DOI: 10.1103/PhysRevB.68.153313 PACS number(s): 78.40.Fy, 42.55.Sa, 71.36.+c, 81.15.Hi

Since the first observation of strong exciton-photon coupling, numerous laboratories across the world have achieved microcavities in various material systems in order to study the physics of strong light-matter interaction in semiconductor material.[1] Recent results have highlighted the potential of strongly coupled semiconductor microcavities to achieve a Bose condensate of polaritons in a solid state material.[2–4] This unique entity is of enormous interest from a fundamental point of view and its distinct properties compared to a Bose condensate of atoms make it exceptional for studying the nonclassical states of matter.[3,5] This dynamical condensate has vast potential for a new generation of low threshold coherent light emitters, parametric amplifiers, and switches.[5,6] Although this immense occupation of the lower polariton state has been observed in numerous material systems, these microcavities ultimately demand a material system having a large exciton oscillator strength and binding energy.[4–6] A large exciton oscillator strength permits a large Rabi splitting more resistant to exciton density and temperature broadening while a large exciton binding energy will be more resilient to the dissociation of polaritons or excitons into free carriers.[5] The widely studied GaAs-based microcavity has brought about a significant understanding but unfortunately it does not fit the above requirements. Wide-bandgap semiconductors such as II-VI [ZnSe/ZnMgSe (Ref. 7) and CdTe/CdMnTe (Ref. 8)] and III-N (GaN/GaAlN) based structures provide this dual requirement,[9,10] although, in spite of the remarkable breakthroughs made in the last five years, the quality of crystal growth is still much higher in GaAs-based heterostructures.[11]

To our knowledge the largest observed Rabi splitting in a ZnSe microcavity is 44 meV.[12] In GaN-based microcavities, the larger exciton binding energy of 30 meV for bulk layers[13] and ≈50 meV (Ref. 14) [compared to 40 meV for ZnSe (Ref. 8)] for quantum well structures combined with the larger coupling strength predicted should produce robust polariton states that are more stable at higher temperatures and higher exciton populations. Very large Rabi splittings of 45–90 meV,[9,15] depending on design, have been predicted but up until now there has been no report of strong-coupling observation in a GaN-based microcavity. Due to their exceptional material robustness and their light emitting capabilities, GaN-based optoelectronic devices are having a large impact on industry. Research is now directed towards GaN microcavity-based devices[16–18] and fundamental research devoted to strong coupling is generating much speculation.[6,9,10] It is expected that the strong-coupling regime will have a significant impact in terms of Bose condensation in a solid-state material and can realistically be exploited in GaN-based devices.

Microcavity structures up until now showing strong-coupling behavior consisted of high finesse cavities having mirror reflectances near unity. For III-*N* based structures it is not a trivial issue to grow crack-free, highly reflective distributed Bragg reflectors.[16] Previous simulations have shown that it would be possible to observe strong light-matter interaction in a GaN bulk cavity as soon as the inhomogeneous broadening of the exciton is reduced below 30 meV even if the amplitude of the optical cavity field is not very high.[19] In this work, we present the experimental observation of the strong-coupling regime in an original low finesse GaN microcavity structure grown by molecular-beam epitaxy (MBE) on a silicon substrate.

The microcavity structure is comprised of a λ/2 GaN active layer grown on a three layer Bragg stack consisting of a 2λ-λ/4 layer of AlN, a 2λ-λ/4 layer of $Al_{0.20}Ga_{0.80}N$, and a λ/4 layer of AlN grown by MBE on a Si(111) substrate. These three layers are the buffer layers necessary to overcome the difficulties encountered during the growth of nitrides on Si substrates.[20] The thickness of each layer is chosen so as to simultaneously satisfy the Bragg condition and the thickness requirement of each buffer layer imposed by the MBE growth. The silicon substrate forms the bottom mirror and produces a reflectivity of 30% in the near-UV range. The upper mirror is a four period SiO_2/Si_3N_4 dielectric mirror producing a reflectivity of 83% in the GaN medium under normal incidence.

The microcavity structure was studied in reflection under various angles of incidence at $T=5$ K. Figure 1 displays the refractive index profile of the microcavity structure together

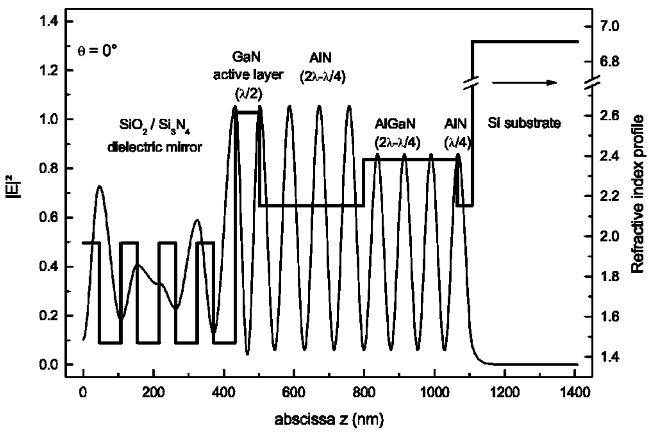

FIG. 1. Electric-field intensity ($|E|^2$) under normal incidence in the investigated bulk GaN microcavity for a photon energy of 3.4 eV corresponding to the resonance of the as-grown cavity. The refractive index profile of the whole structure is also given.

with the electric-field intensity ($|E|^2$) in the growth direction for normal incidence at a photon energy of 3.4 eV. The optical response is measured from reflectivity measurements where the energy of the cavity mode is varied by increasing the angle at which light is incident on the cavity structure.[21] The as-grown structure presents a large negative detuning since the energy of the cavity mode lies at 3.40 eV under normal incidence while the excitonic transition energy is located around 3.5 eV. Angle-resolved reflectivity experiments

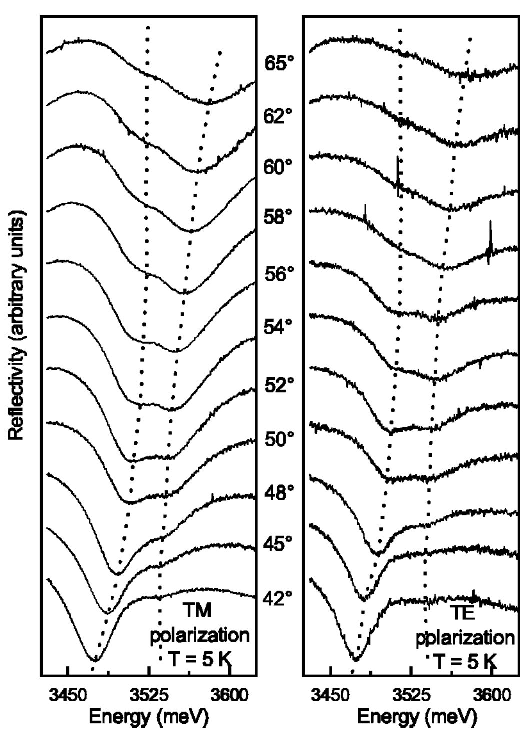

FIG. 2. Angle-resolved reflectivity measurements from the GaN microcavity at $T=5$ K performed for transverse magnetic (TM) and transverse electric (TE) polarizations. The strong light-matter coupling is clearly observed between 52° and 54° for both polarizations.

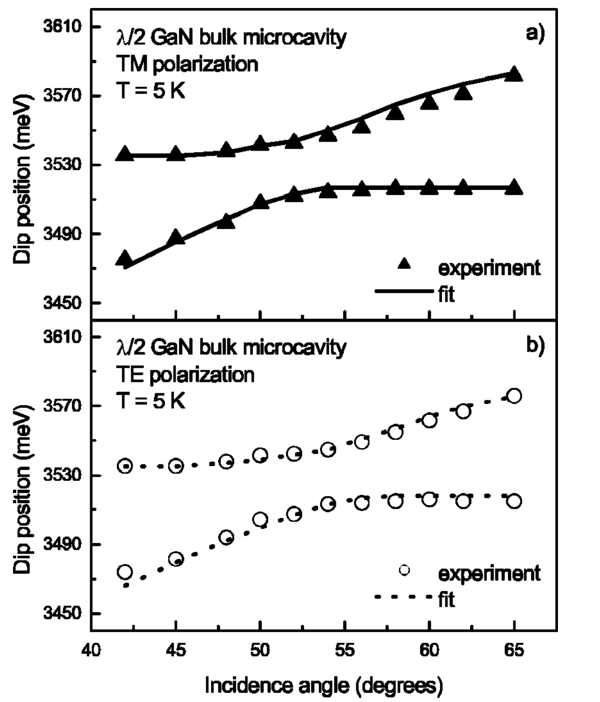

FIG. 3. The energy positions of the reflectivity dips in the bulk GaN microcavity for both TM (a) and TE (b) polarizations as a function of the incidence angle. Solid and dashed lines correspond to simulations.

for both polarizations (TM: transverse magnetic, TE: transverse electric) are reported in Fig. 2 for angle values varying between 42° and 65°. For $\theta=42°$, the uncoupled cavity mode lies at low energy and a weak, rather wide dip, corresponding to the GaN related excitons, is seen at high energy. As the energy of the cavity mode increases, the amplitude of the excitonic dip decreases until its integrated intensity is equal to that of the cavity mode at the resonance for a value of θ lying between 52° and 54° for TM polarization, evidencing strong light-matter coupling.[22] For TE polarization, it occurs also in this range but at a slightly higher angle than that of TM. For larger angles, the coupling progressively disappears and it can be observed that the spectral width of the cavity mode is larger than for smaller values of θ. This is most probably due to the fact that the cavity mode is resonant with the band-to-band continuum states. The lateral inhomogeneity related to layer thicknesses can also be responsible for such a behavior since for large incident angles, the light path in the cavity is increased. The coupling is less pronounced in the TE polarization as previously observed and explained in a GaAs-based cavity.[21] The exciton-photon coupling has also been observed unambiguously at 77 K but at room temperature, due to the additional thermal broadening of excitons ($kT=26$ meV), the splitting is not clearly resolved.

The energy positions of the reflectivity dips are reported in Fig. 3 for TM and TE polarizations. The shift to higher energy of the cavity features is clearly evidenced together with the anticrossing between the exciton and photon mode. This figure shows also a slight difference between the anticrossing positions related to the TE and TM polarizations.

For the angle values lying in the investigated range the TM dip of the cavity mode is observed at higher energy than for TE polarization, the energy difference being maximal for $\theta = 65°$ (6 meV). This is due to the different phase shifts and penetration of the optical modes into the dielectric mirror for the two polarizations. For TM polarization, a minimum exciton-photon splitting of 30.8 meV is achieved for $\theta = 52°$, and for TE polarization this value is 31.5 meV for $\theta = 54°$. Thus an average value of 31 ± 1 meV is measured in both polarizations. The solid and dashed lines are the results of transfer matrix reflectivity simulations as a function of θ.[23] An overall good agreement is found with experimental results for both polarizations. In these calculations, the excitons of the GaN active layer are modeled as Lorentz oscillators without spatial dispersion. Only two oscillators are introduced in order to take into account the *A* and *B* excitons whose oscillator strengths are the most significant. The *C* exciton is not considered here because of its higher energy and its lower oscillator strength.[24] An energy splitting of 5 meV between *A* and *B* is assumed. The agreement between the measured reflectivity spectra and simulation is obtained for oscillator amplitude values proportional to the oscillator strength of 40 000 and 30 000 $(\mathrm{meV})^2$ for the *A* and *B* excitons, respectively; these values agree very well with previous determinations of these parameters.[25] As the linewidth of the excitonic mode is rather large, inhomogeneous broadening is included in the model and a value of 40 meV was deduced for the linewidth of each exciton.[26] Concerning the energy of the *A* and *B* excitons, the values of 3528 and 3533 meV are used in the simulations in order to account for the energy position of the dips. These values could seem to be relatively high but considering the strains imposed on the GaN active layer by the underlying thick AlN layer and the dielectric mirror they are not surprising. Further calculations were also performed in order to evaluate the maximum exciton-photon splitting which could potentially be achieved in such a structure. The structure considered is exactly the same as that investigated in this work, except for the thickness of the GaN active layer. Figure 4 displays the evolution of the exciton-photon splitting at resonance for different thicknesses of the GaN active layer. The Rabi splitting increases with active layer thickness due to the increased superposition between the material and photon wave functions[27] [as a result of the increased ratio value (length of the active region)/(effective cavity length)]. A value of 60 meV appears to be the upper limit achievable with such a structure.

The results show clear evidence of strong-coupling behavior from an extremely simple GaN microcavity. The cavity and exciton linewidths are broad due to the low cavity finesse and the poor quality of the bulk GaN active layer that is grown directly on a highly defective buffer layer sequence. It is now well known that the quality of this GaN layer improves with thickness and the original structure presented in this paper is just the starting point for a series of rich investigations involving GaN microcavities. Instead of growing a thin highly defective cavity layer, thick GaN films (1–2 μm) can be grown producing state of the art bulk GaN or GaN/AlGaN multiple quantum wells with emission linewidths ≈ 10 meV.[20] The silicon substrate can be easily removed thus opening the way to hybrid structures with highly reflective top and bottom dielectric mirrors. The initial highly defective GaN layer can then be removed by reactive ion etching until the desired cavity length remains with an active layer of much higher material quality. This approach is currently in the process of optimization.[17,28]

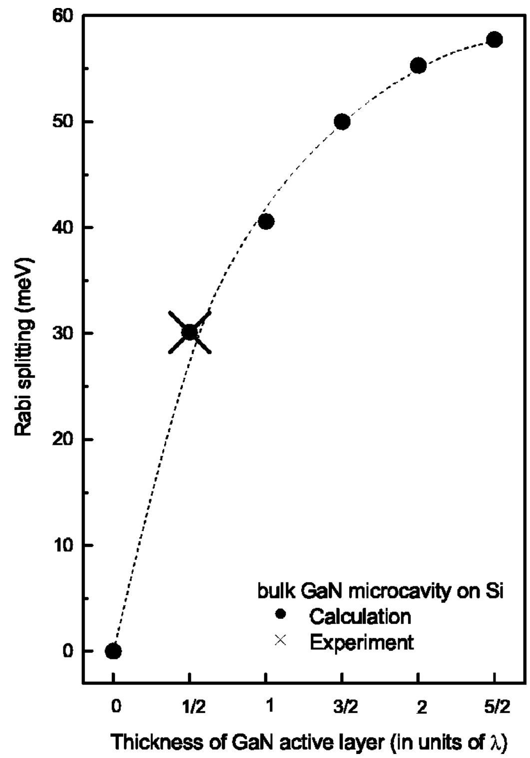

FIG. 4. Evolution of the Rabi splitting at resonance as a function of the thickness of the GaN active layer inserted in a microcavity similar to that investigated in the present work. A maximum coupling strength of 60 meV is predicted in such a structure. The dot-dashed line is a guide for the eye.

In conclusion, strong exciton-photon coupling has been observed experimentally in a bulk GaN microcavity with a Rabi splitting of 31 meV at 5 K well accounted for by transfer matrix simulations. This observation is extremely promising for the study of quantum degeneracy of microcavity polaritons and the realization of future optoelectronic devices based on the polariton Bosonic character.

The authors are indebted to A. Kavokin for stimulating discussions, to S. Walker (University of Sheffield) for dielectric mirror deposition, and to R. Butté and M. S. Skolnick for their interest in this work. This work was supported by the EC-RTN "CLERMONT" program, Contract No. HPRN-CT-1999-00132.

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