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A. Bonfiglio
University of Lecce

M. Lomascolo
University of Lecce

G. Traetta
University of Lecce

See next page for additional authors

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A. Bonfiglio,[a] M. Lomascolo,[b] G. Traetta, and R. Cingolani
INFM-Dipartimento di Ingegneria dell'Innovazione, University of Lecce, 73100 Lecce, Italy

A. Di Carlo, F. Della Sala, and P. Lugli
Dipartimento di Ingegneria Elettronica, University of Roma ''TorVergata,'' 00133 Roma, Italy

A. Botchkarev and H. Morkoc
Electronic Engineering, Virginia Commonwealth University, Richmond, Virginia 23173

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We have performed a systematic investigation of GaN/AlGaN quantum wells grown on different buffer layers (either GaN or AlGaN) in order to clarify the role of strain, structural parameters, and built-in field in determining the well-width dependence of the ground level emission energy. We find that identical quantum wells grown on different buffer layers exhibit strong variation of the ground level energy but similar well-width dependence. The data are quantitatively explained by an analytic model based on the envelope function formalism which accounts for screening and built-in field, and by a full self-consistent tight binding model. © *2000 American Institute of Physics.* [S0021-8979(00)09105-2]

I. INTRODUCTION

In recent years, many research efforts have been made in order to develop the knowledge basis for the application of GaN/AlGaN quantum wells (QWs) to optical devices.[1–8] However, the application of such materials still needs a full comprehension with particular concern to the relation between optical properties and detailed structure of the systems under consideration. For instance, in addition to the well known effect of strain, the built-in electric field has been recently shown[1–7] to affect the ground level energy of GaN/AlGaN quantum wells. Namely, a well-width dependent Stark shift of the emission energy of the QWs has been observed in photoluminescence (PL) and reflectivity experiments. Such a field originates from the superposition of spontaneous polarization charge and piezoelectric charge in the heterostructure, and it can be exploited to tailor the emission energy of a QW-based optical device. In addition, the strength of the built-in field is sensitive to the injected charge density, due to the screening effect.[5,7] In this article, we present a comprehensive spectroscopic investigation of GaN/AlGaN QWs characterized by different strain distribution and structural parameters. By growing identical quantum wells on GaN or AlGaN substrates, we clarify the interplay between strain, piezoelectric field, spontaneous polarization field, and quantum size effect in determining the ground level energy of the heterostructures and its well-width dependence.

The two series of samples are characterized by the same value of the built-in field and by a different strain distribution, namely, strained wells (samples grown on GaN) or strained barriers (samples on AlGaN). Despite the identical quantum wells, the two sets of samples exhibit a strong difference in the ground level emission energy. The comparison to a suitably developed theoretical model provides a good quantitative understanding of the experimental data.

II. MATERIALS AND METHODS

The samples were grown by reactive molecular beam epitaxy on sapphire substrates. Following a chemical *in situ* cleaning of *c*-plane sapphire substrates, a thin AlN buffer layer was grown at 850 °C with ammonia as the active nitrogen source. The AlN buffer layer was followed by the growth of a 1 μm thick GaN buffer layer grown at 800 °C. Then, an $Al_{0.15}Ga_{0.85}N$ layer was grown with two different thickness values for each set of samples: 100 nm for the first and 10 for the second. Finally, the quantum well region was grown. The strain in the QW was varied by changing the buffer layer of the structures: samples grown on the 10 nm thick $Al_{0.15}Ga_{0.85}N$ buffer layer are pseudomorphic to the GaN substrate, so that the GaN QWs are unstrained. Conversely, samples grown on the 100 nm thick $Al_{0.15}Ga_{0.85}N$ buffer layer (which is completely relaxed) result in strained QWs. Each sample consisted of 10 GaN quantum wells. In each set, four samples of well width $L_w = 2,3,4,5$ nm (measured by double-crystal x-ray diffraction) were grown and analyzed. The barrier width and composition were kept constant in all samples ($Al_{0.15}Ga_{0.85}N$, barriers of thickness $L_B = 10$ nm).

The optical measurements were performed either under cw excitation (325 nm line of a He–Cd laser) or under pulsed excitation (fourth harmonic of Nd-Yag laser). The samples were kept in a variable temperature closed-cycle cryostat. The spectral resolution was always better than 0.2 meV.

[a]Permanent address: INFM-Dipartimento di Ingegneria Elettrica ed Elettronica, University of Cagliari, Italy.
[b]Permanent address: CNR-IME Istituto per lo studio di nuovi Materiali per l'Elettronica, Via per Arnesano, I-73100 Lecce, Italy; electronic mail: mauro.lomascolo@unile.it

III. RESULTS

Figure 1(a) shows the PL spectra of a representative sample (2 nm well width, strained QW) at different temperatures. The $n=1$ level is observed in the high energy wing of the spectra, (around 3.44 eV at 10 K), together with the LO-phonon replicas. With the increasing temperature, the spectra smearout and redshift. The room temperature luminescence is found to drop in intensity by about a factor of 20 with respect to the low temperature emission. The emission energy of the various bands was extracted by a Gaussian deconvolution of the spectra at each temperature, leading to the results displayed in Fig. 1(b). Both the ground level and the phonon replica follow the temperature dependence of the energy gap reported in Ref. 8.

The systematic analysis of all samples results in the well-width dependence of the $n=1$ ground level emission displayed in Fig. 2 (symbols). In this figure, the experimental data are compared to the theoretical curves obtained by the analytical model discussed in the next section.

The main features of the experimental data shown in Fig. 2 can be summarized as follow:

(1) For $L_w \geqslant 3$ nm, the emission energy falls below the bulk energy gap.

(2) The observed well-width dependence differs considerably from the usual square well model (L_w^{-2} dependence).

(3) For a given well width, the ground level emission energy is different in the two sets of samples, i.e., it depends dramatically on the AlGaN buffer thickness.

(4) Despite the difference in the absolute energy value, the well-width dependence of the ground level emission is similar in the two sets of samples, i.e., it does not depend on the thickness of the AlGaN buffer;

(5) In addition, we note that the emission energy blueshifts with increasing the photogenerated carrier density (not shown).

Points (1) and (2) are consistent with other reports, showing that strain and built-in fields affect the emission energy of the GaN-based quantum wells. The redshift of the emission is due to the concomitant effect of the strain and of the quantum confined Stark effect caused by the internal field (given by piezoelectric plus spontaneous polarization fields). In addition, both effects cause a departure from the well-known square-well model prediction, either due to the bending of the well potential or to the nonlinear shift of the bulk band gap edge, respectively.

Points (3) and (4) indicate that the thickness of the AlGaN buffer affects the emission energy but not its well-width dependence. Since the strain distribution is strongly varied by the thickness of the AlGaN buffer (strained well for thin AlGaN buffer or strained barrier for thick AlGaN buffer), whereas the spontaneous polarization field is unchanged, we may infer that the well-width dependence of the emission energy is predominantly determined by the Stark effect associated with the built-in field rather than by the strain distribution in the samples (see next section). Finally, point (5) reveals that with increasing carrier density the internal field is screened and the Stark effect is partly balanced by the screening, resulting in a blueshift of the emission lines.

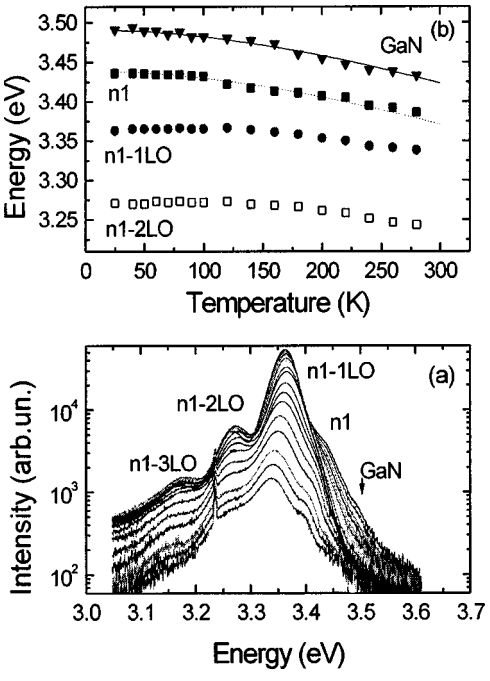

FIG. 1. (a) Temperature-dependent spectra (cw excitation) of the 2 nm thick GaN QWs grown on AlGaN buffer. The spectra are taken every 20 K from 10 to 310 K. (b) Temperature-dependent redshift of the main PL bands obtained by the Gaussian deconvolution of the spectra of (a). The downward triangles are taken from a bulk GaN and the continuous lines are calculated by the Varshni law (see Ref. 8).

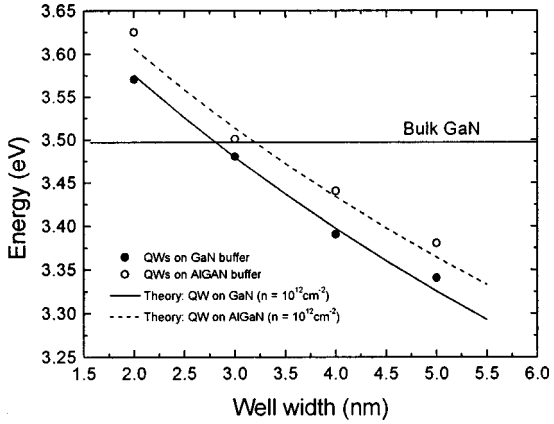

FIG. 2. Well-width dependence of the ground level emission energy of GaN/AlGaN QWs grown on GaN (full dots) or AlGaN (empty dots) buffer layer, at 10 K. The curves are calculated by means of Eqs. (2) and (3). The horizontal line indicates the unstrained bulk energy gap of GaN.

IV. DISCUSSION

In what follows, we attempt a quantitative analysis of our data, taking into account all the possible phenomena affecting the ground level energy of the heterostructures: namely, strain, built-in field and structural parameters of the quantum wells. In order to avoid any confusion, we will refer as ''strain effect'' to the modification of the band structure induced by the strain, while the strain induced polarization will be referred as the piezoelectric component of the built-in field.

As far as the strain is concerned, this is evaluated following Ref. 9. According to this approach, samples with strained quantum wells (those grown on 100 nm thick AlGaN layers) should exhibit a dependence on the in-plane compressive strain of the emission energy. In this case, the barriers take the bulk $Al_{0.15}Ga_{0.85}N$ lattice constant (3.177 Å), whereas the GaN layers are assumed to grow pseudomorphically and to undergo a compressive in-plane strain $\sigma_{\|}=-0.37\%$. We similarly evaluate the strain correction for the samples grown on 10 nm thick AlGaN buffer, i.e., with strained barriers. In this case, the quantum wells take the bulk GaN lattice constant (3.189 Å), whereas $Al_{0.15}Ga_{0.85}N$ layers grow pseudomorphically with a tensile in-plane strain $\sigma_{\|}=0.37\%$. The assumption of a strained AlGaN barrier is consistent with the observation of pseudomorphic growth of AlGaN layers for thicknesses as large as hundreds of nm, which is much thicker than the total amount of AlGaN contained in our 10 periods multiple quantum wells.[2]

The built-in electric field is evaluated by accounting for the accumulation of a polarization charge at the interfaces of GaN/AlGaN heterostructures. The total polarization charge can be written as $P_{\rm tot}=P_{\rm piezo}+P_{\rm spont}$, where $P_{\rm piezo}$ is the piezoelectric charge caused by the lattice mismatch (mis) and by the thermal strain (ts) $[P_{\rm piezo}=P_{\rm mis}+P_{\rm ts}]$, whereas $P_{\rm spont}$ represents the spontaneous polarization charge of the GaN/AlGaN interface, as clearly demonstrated by the recent works of Bernardini *et al.*[10–12] For an alternating sequence of wells (w) and barriers (b), the total electric field in the well can be calculated as[12]

$$F_w=L_b(P^b_{\rm tot}-P^w_{\rm tot})/[\varepsilon_0(L_w\varepsilon_b+L_b\varepsilon_w)]. \tag{1}$$

$\epsilon_{b,w}$ being the relative dielectric constant of the layers (analogous expression with exchanged indexes holds for the electric field in the barrier). The piezoelectric polarization ($P_{\rm lm}$) induced by the lattice in-plane mismatch (σ_j) can be calculated as $P_{\rm lm}=-2[e_{33}(C_{11}/C_{33})-e_{31}]\sigma_{\|}$ where e_{ij} and C_{ij} are the piezoelectric tensor components and the elastic constants, respectively, as given in Refs. 10–12. Adopting the strain vales quoted above, the piezoelectric polarization charge in the set of samples grown on 10 nm thick AlGaN buffer turns out to be $P^w_{\rm lm}=0$ in the quantum well and $P^b_{\rm lm}=-0.0055\,{\rm C/m^2}$ in the $Al_{0.15}Ga_{0.85}N$ barrier. The opposite holds for the set of samples grown on 100 nm thick AlGaN buffer, where we get $P^w_{\rm lm}=0.0051\,{\rm C/m^2}$ in the quantum well and $P^b_{\rm lm}=0$ in the $Al_{0.15}Ga_{0.85}N$ barrier. The values of the polarization charge either spontaneous or piezoelectric (depending on strain), will be used as input parameters in the modeling of the electronic states discussed later.

In addition, the thermal strain in our experimental conditions amounts to some 0.003%, resulting in an additional polarization charge of the order of $P^w_{\rm ts}=-3.2\times10^{-4}\,{\rm C/m^2}$. As far as the spontaneous polarization charge is concerned, we take the recent data of Refs. 10–12, leading to $P^w_{\rm sp}=-0.029\,{\rm C/m^2}$ and $P^b_{\rm sp}=-0.037\,{\rm C/m^2}$, the latter value being obtained by linear interpolation of the GaN and AlN values ($P_{\rm sp}=-0.08\,{\rm C/m^2}$ in AlN). By using these data and Eq. (1), we can calculate the built-in field in the different samples, which turns out to vary in the range of 0.8–1.3 MV/cm depending on the actual well width.

At first order, neglecting high-field effects and corrections for free-charge screening, we might except that the built-in field causes a redshift of the quantum well gap given by the quadratic Stark effect.[13,14] However, as one can expect, such approximation is too simplistic and actually reproduces only qualitatively the well-width dependence of the redshift.

A more accurate analytical model based on the envelope function formalism can be developed in order to reproduce the effects of the internal field and of the strain by assuming that (i) the rectangular quantum well is replaced by a triangular well[13] resulting from the total built-in field, and (ii) the two-dimensional (2D) photoinjected charge density (ρ) is considered to accumulate at the GaN/AlGaN interface and to screen the built-in field. With these approximations the electric field in the well becomes

$$F_w=L_b(\rho+P^b_{\rm tot}-P^w_{\rm tot})/[\varepsilon_0(L_w\varepsilon_b+L_b\varepsilon_w)] \tag{2}$$

and the ground level energy

$$E_{1e1h}=E_g-F_wL_w+\left(\frac{9\pi\hbar eF_w}{8\sqrt{2}}\right)^{2/3}\left(\frac{1}{m_e}+\frac{1}{m_h}\right)^{1/3}. \tag{3}$$

The band gap shift induced by the strain is included in E_g.[9] In order to test the accuracy of the envelope function model, we have compared Eqs. (2) and (3) with the results of a full self-consistent tight-binding (TB) model, for several injected charge densities.[7,15] The tight-binding model is used to describe the electronic structure in the entire Brillouin zone, up to several eV above the fundamental gap. The self-consistent calculation is performed as follows: the electron and hole Quasi-Fermi levels are calculated for a given 2D photoinjected charge density and the electron and hole charge distributions are obtained and then substituted in the Poisson equation which account also for the spontaneous and piezoelectric polarizations. The obtained potential is inserted in the TB Schroedinger equation which is solved to obtain energy levels and wave functions. Then, the new Quasi-Fermi levels are calculated and the whole procedure reiterated to self-consistency (see Refs. 7 and 16 for details). This method, though intrinsically more accurate than the envelope function model, requires a stronger computational effort. Figure 3 shows the comparison between the analytically calculated emission energies [Eqs. (2) and (3), solid lines], and those obtained by the self-consistent tight-binding model (symbols). The analytical results reproduce quite well the TB results for densities below $4\times10^{12}\,{\rm cm^{-2}}$. At higher densities, the spatial distribution of injected charges should be taken into account properly for a quantitative analysis (indeed under this condition, one cannot assume that charges simply accumulate at the GaN/AlGaN interfaces). From Fig. 3, we also notice that the agreement between Eqs. (2) and (3) and TB results improves as the well width increases. In fact, for narrow wells, the triangular barrier approximation is no longer valid. Such comparison demonstrates that the envelope function approach gives reasonable results provided the injected carrier density is not too high and the screening is

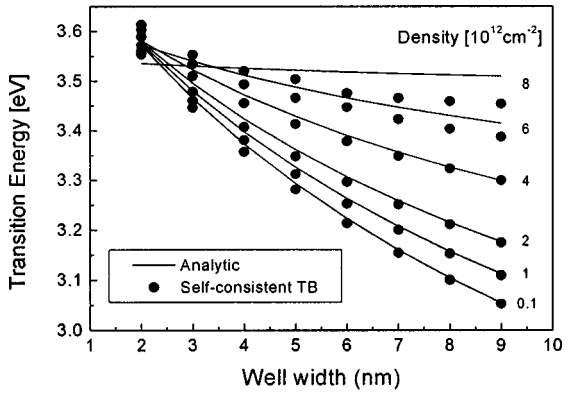

FIG. 3. Comparison between the well-width dependence of the ground level emission calculated by the envelope function model Eqs. (2) and (3) (continuous lines) and by the self-consistent tight-binding model (full dots) for different injected densities. The accuracy of the envelope function model turns out to be very satisfactory for wider wells and for densities up to $4 \times 10^{12}\ \mathrm{cm}^{-2}$.

phenomenologically accounted for. This should be kept into consideration in view of the calculation of the emission energy of Nitride-based laser devices.

The blueshift of the ground level emission versus the excitation intensity is presently under investigation. A precise comparison with the theory requires the exact knowledge of the photogenerated carrier density under stationary conditions. This issue will be investigated in detail in a forthcoming paper.

Nonetheless, under the present experimental conditions, in which the cw photogenerated rate is relatively low, we can confidently compare the experimental data with Eqs. (2) and (3). All the parameters used to evaluate Eq. (2) and Eq. (3) are taken from the literature.[1–12] The AlGaN parameters are obtained by using linear interpolations of the AlN and GaN ones.

From Fig. 2 we can see that the experimental data collected from the QW samples grown on GaN buffer are in good quantitative agreement with the theoretical results for both sets of samples. The well-width dependence turns out to be very similar for both sets of samples, indicating that the Stark effect induced by the internal field prevails on the band shift induced by the strain, and basically determines the ground level energy of nitride QWs. On the other hand, for a given well width, the shift of the energy level induced by the presence of the strain ($\sim$30 meV) is consistent with the theoretical value reported in Ref. 9. We should point out that the measured value of the strain-induced energy shift is not affected by the presence of the polarization field, since polarization fields are nearly the same for the GaN-matched and $Al_{0.15}Ga_{0.85}N$-matched samples. Moreover, within the built-in field correction, the spontaneous polarization term is found to be larger than the strain-induced piezoelectric contribution. The predominant contribution of the Stark effect induced by the built-in field also causes a substantial decrease of the exciton binding energy in these samples. Preliminar calculations based on Green-function models reveal that the exciton binding energy in our structures is less than 10 meV, independent of well width. This supports the use of single particle envelope function model without excitonic contribution for the quantitative analysis of luminescence spectra in GaN/AlGaN quantum wells.

V. CONCLUSIONS

In conclusions, we have studied the interplay between strain and internal electric fields in GaN/AlGaN MQWs. Both the piezoelectric component of the field and the strain induced shift of the gap can be tuned by varying the strain distribution in the heterostructure, i.e., by growing the samples on GaN or AlGaN substrates. The results have been quantitatively confirmed by theoretical calculations based on a self-consistent approach and by a simpler but effective analytical expression. This demonstrates that GaN QWs are effectively systems in which four parameters (composition, well width, buffer type, and charge density) can be varied for tuning the ground level emission and therefore optimizing the characteristic of a possible QW-based optical device.

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