Influence of Internal Electric Fields on the Ground Level Emission of GaN/AlGaN Multi-Quantum Wells

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ABSTRACT

The spectroscopic investigation of GaN/AlGaN quantum wells reveals that the emission energy of such structures is determined by four parameters, namely composition, well-width, strain and charge density. The experimental data obtained by varying these parameters 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.

INTRODUCTION

The physical behavior of GaN/AlGaN quantum wells (QWs) in view of their application to optical devices is at present under investigation both theoretically and experimentally[1-8]. In particular, the relation between optical properties and geometrical and compositional structure of the systems is of great relevance.
By growing identical quantum wells of various dimensions on different buffer layers, it is possible to clarify the interplay between geometry, strain, piezoelectric field, spontaneous polarization field and quantum size effect in determining the ground level energy of the heterostructures and its well-width dependence.
Having addressed this problem in a recent publication [9], in this paper we further investigate the role of the internal field in determining the ground level emission energy of GaN/AlGaN QWs, with particular concern for the role played by the injected charge which accumulates at the interface between quantum wells and barriers.

MATERIALS AND METHODS

Three sets of samples have been produced for our experiments. The first two sets (A and B) were grown by reactive molecular beam epitaxy (MBE) 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 set A and 10 for set B. 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

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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, and 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). Set C was grown by MOCVD according to the structure already described for set B but with quantum well widths of respectively, 1, 2, 3, 6, and 9 nm.

The optical measurements were performed either under cw excitation (325 nm line of a He-Cd laser) or under a pulsed excitation (4^{th} harmonic of a Nd-YAG laser). The samples were kept in a variable temperature closed cycle cryostat. The spectral resolution was always better than 0.2 meV.

DISCUSSION

The systematic analysis of all samples belonging to sets A and B results in the well-width dependence of the n=1 ground level emission displayed in Fig. 1 (symbols). In this figure the experimental data are compared to the theoretical curves obtained by the analytical models discussed in the following.

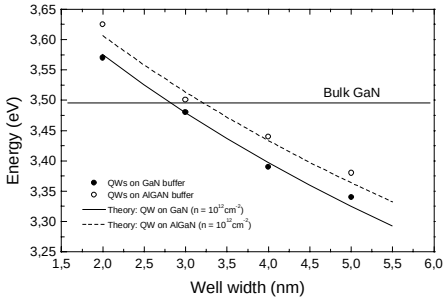

Figure 1. *Well-width dependence of the ground level emission energy of GaN/AlGaN QWs grown on GaN, at 10 K. The curves are calculated by means of Eq. 4 The horizontal line indicates the unstrained bulk energy gap of GaN.*

The main features of the experimental data shown in figure 1 can be summarized as follow:

1) for $L_w \geq 3$ 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;

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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) The emission energy blue-shifts with increasing the photo-generated charge density (data not shown).

The detailed analysis of the results obtained on sets A and B has been already presented in [9].

Figure 2 shows the emission energy of samples belonging to set C. As can be easily noticed, the linear dependence of energy on the quantum well width holds only for the narrower wells (first 3 points) whereas the wider wells exhibit a considerable deviation from such trend. The blue-shift displayed by the emission energy of the samples with larger well widths indicates that the role of the well dimensions must be considered, in this case, with greater attention.

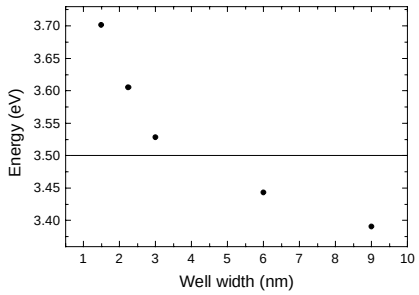

Figure 2. *Well-width dependence of the ground level emission energy of GaN/AlGaN QWs belonging to set C. The horizontal line indicates the unstrained bulk energy gap of GaN.*

The theoretical model presented in [9], describing the well width dependence of the narrow wells (see fig.1) takes into account strain, built-in field and structural parameters of the quantum wells. As far as the strain is concerned, this is evaluated following Ref. [10]. According to this approach, samples with strained quantum wells (set A) 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 GaN layers are assumed to grow pseudomorphically and to undergo a compressive in-plane strain $\sigma_{\parallel}$=-0.37%. We similarly evaluated the strain correction for samples belonging to sets B and C, 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 $\sigma_{\parallel}$=0.37%. The assumption of a strained $Al_{0.15}Ga_{0.85}N$ 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 $Al_{0.15}Ga_{0.85}N$ contained in our 10 periods multiple quantum wells [2].

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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 $P_{tot}= P_{piezo}+P_{spont}$, where P_{piezo} is the piezoelectric charge caused by the lattice mismatch (mis) and by the thermal strain (ts) [$P_{piezo} = P_{mis} + P_{ts}$], whereas P_{spont} represents the spontaneous polarization charge of the GaN/AlGaN interface, as clearly demonstrated by the recent works of Bernardini et al.[11-13]. For an alternating sequence of wells (w) and barriers (b) the total electric field in the well can be calculated as [13]

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

$\varepsilon_{b,w}$ 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_{lm}) induced by the lattice in-plane mismatch ($\sigma_{||}$) can be calculated as

$$P_{lm} = -2\left(e_{33}\frac{C_{11}}{C_{33}} - e_{31}\right)\sigma_{||} \qquad (2)$$

where e_{ij} and C_{ij} are the piezoelectric tensor components and the elastic constants, respectively, as given in ref.[11-13]. Adopting the strain vales quoted above, the piezoelectric polarization charge in our set of samples turns out to be $P^w_{lm}=0$ in the quantum well and $P^b_{lm}= -0.0055$ C/m^2 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 $P^w_{ts}=-3.2 *10^{-4}$ C/m^2. As far as the spontaneous polarization charge is concerned, we take the recent data of ref.[11-13], leading to $P^w_{sp}= -0.029$ C/m^2 and $P^b_{sp}= -0.037$ C/m^2, the latter value being obtained by linear interpolation of the GaN and AlN values ($P_{sp}=-0.08$ 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 0.8 - 1.3 MV/cm depending on the actual well-width.

An 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 [14] resulting from the total built-in field, and ii) the 2D photo injected 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

$$F_w = L_b(\rho + P^b_{tot}- P^w_{tot}) / [\varepsilon_0(L_w \varepsilon_b + L_b \varepsilon_w)] \qquad (3)$$

and the ground level energy

$$E_{1e1h} = E_g - F_w L_w + \left(\frac{9\pi\hbar eF_w}{8\sqrt{2}}\right)^{2/3}\left(\frac{1}{m_e}+\frac{1}{m_h}\right)^{1/3} \qquad (4)$$

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The band gap shift induced by the strain is included in E_g [10]. In order to test the accuracy of the envelope function model we have compared Eqs.(3)-(4) with the results of a full self-consistent tight-binding (TB) model, for several injected charge densities [7,9,16]. 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 photo injected charge density (which is a fitting parameter) 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 17 for details). This method, though intrinsically more accurate than the envelope function model, requires a stronger computational effort.
From Fig.1 we can see that both sets of samples are in good quantitative agreement with the theoretical results. In the case displayed in Fig. 2, the thicker wells exhibit a blue-shift of the emission energy with respect to the ideal trend forecast by Eq. (4). This might be due to the fact that in wide wells the wave functions overlap is decreased by the built-in field (accordingly the oscillation strength becomes very low), resulting in a charge accumulation at the interfaces. Such accumulation induces a further alteration of the electric field (screening), resulting in a blue-shift of the emission energy. Conversely, charge accumulation poorly affects samples with narrower wells because here the spatial extension of the wave functions of electron and holes is comparable to the dimensions of the potential well where these are confined and the probability of recombination is therefore not affected by the field. Experiments are presently under way in our labs to clarify this issue.

CONCLUSIONS

In conclusion, we have completed the analysis of the influence of internal electrical field on the emission energy of GaN/AlGaN MQWs. As demonstrated in a previous paper [9], 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 samples on GaN or AlGaN substrates. The results have been quantitatively confirmed by theoretical calculations based on a self-consistent approach. This demonstrates that GaN QWs are effectively systems in which four parameters (composition, well-width, buffer type and charge density) can be varied to tune the ground level emission and therefore optimizing the characteristic of a possible QW-based optical device.

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