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Effect of annealing on phase separation in ternary III–N alloys

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Major developments in wide band gap III–N semiconductors have led to commercial production of high brightness light-emitting diodes and to demonstration of room temperature laser light emission under pulsed and continuous-wave operation in violet [1], and, recently, in blue spectral region [2]. However, the internal structure of InGaN and AlGaN alloys indispensable for fabrication of high-quality light emitting devices has not been properly investigated. There exist many reports on optical properties of thick ternary alloys and GaN/InGaN and GaN/AlGaN heterostructures. Most of papers are deduced to determination of compositional dependence of band gap of "bulk" AlGaN and InGaN alloys. Values reported for bowing parameters of AlGaN range from $b = 0$ [3] to $b = 1.0$ eV and $b = 1.3$ eV [4, 5]. For InGaN alloys reported range of bowing parameter is much wider: from $b = 1$ eV to $b = 4.8$ eV [6, 7]. Nearly all authors use approximation of strained, but homogenious layer determining In composition from X-ray diffractometry. However, it was shown recently that at least for InGaN ternary alloys compositional fluctuations (phase separation) can play a very important role [8] and must be taken into account calculating In composition from optical transitions energy.

In this paper we report investigation of phase separation in ternary III–N alloys (AlGaN and InGaN) and influence of annealing on optical and structural properties of these samples.

The samples studied in this work were grown by low pressure metalorganic chemical vapor deposition (MOCVD) employing an AlGaN nucleation layer deposited at 530°C on (0001) sapphire substrates. Ammonia, trimethylindium (TMI), trimethylgallium (TMG) and thrimethylaluminum (TMA) were applied as component precursors. Purified hydrogen and/or argon [10] were used as carrier gases. Three types of samples were grown: thick (2–3 μm) AlGaN layers grown directly on sapphire substrate (sample A); structures with thin (50 nm) InGaN layer sandwiched between 3 μm GaN buffer layer and 100 nm thick GaN cap layer (sample B); structures with multiple ultrathin (3–5 nm) InGaN insertions in GaN matrix sandwiched between thick GaN buffer layer and 100 nm GaN cap layer (sample C). The details of growth were reported elsewere [9, 10].

The photoluminescence (PL) study was performed in the temperature range 4–300 K using a continious wave He-Cd laser (excitation density 25 W/cm^2) or a pulsed excimer laser for excitation.

Temperature dependencies of photoluminiscence peak position for sample A and the same sample after rapid thermal annealing (RTA) are shown in Fig. 1. Temperature-dependent PL study of as-grown sample revealed so-called "S-shaped" temperature-dependent emission shift. This feature indicates presence of some kind of localization centers in the layer. After RTA at 1100°C for 120 sec this feature practically disappears and the near-band-edge PL peak position shifts to the high-energy side more than 20 meV. This high-energy

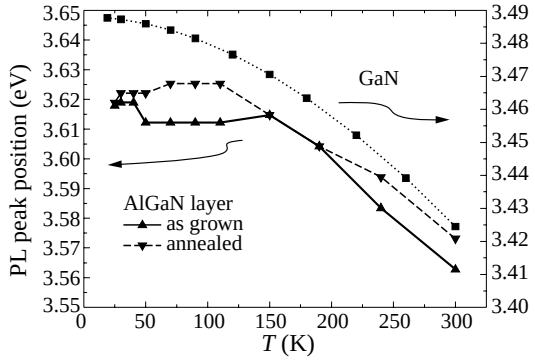

Fig. 1. Photoluminescence peak position *vs* temperature for as grown and annealed AlGaN layer. GaN PL peak position is shown for reference.

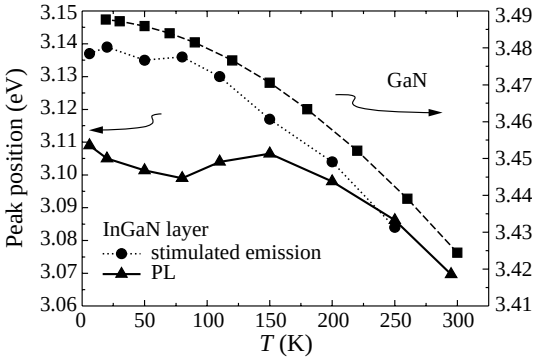

Fig. 2. Photoluminescence and stimulated emission (at threshold) peak position *vs* temperature for InGaN layer. GaN PL peak position is shown for reference.

shift of near-band-edge emission after RTA was also observed in PL spectra of thin AlGaN layers grown on GaN epilayers. RTA also strongly affects electronic properties of the structure. Room temperature Hall mobility after the RTA increased to 200 $cm^2V^{-1}s^{-1}$ from 30–40 $cm^2V^{-1}s^{-1}$ in the as-grown structure indicating efficient decrease in compositional fluctuations for annealed structure.

Figure 2 shows temperature dependence of PL peak for sample B at low excitation density (25 W/cm^2) (Fig. 2, solid curve) and energies of stimulated emission at edge geometry (Fig. 2, dashed curve). (For stimulated emission energies were taken at threshold excitation dencity). It is clearly seen that stimulated emission follows the band gap of GaN, while PL at low excitation density reveals "S-shaped" emission shift. This behavior points to low density of localized states formed by phase separation of InGaN layer.

In our previous work [10] it was shown that growth of multiple ultrathin (3–5 nm) InGaN insertions in GaN matrix (structure C) leads to formation of dense array of nanoislands with high In composition (quantum dots). The density of these localization centers is so high that at low temperatures these quantum dots (QD) can produce gain necessary even for lasing in vertical direction. Temperature dependencies of photoluminiscence peak position for as-grown sample C and the same sample after rapid thermal annealing (RTA) are shown in Fig. 3. After RTA at 1300°C for 30 sec QD-related PL peak shifts to low-energy side

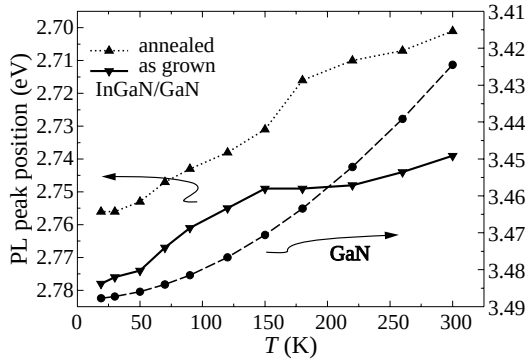

Fig. 3. Photoluminescence peak position *vs* temperature for as grown and annealed InGaN/GaN multilayer samples. GaN PL peak position is shown for reference.

more than 20 meV. This shift is accompanied by increasing width of PL spectra from 100 to 130 meV indicating increase both in maximum depth of localization potential and in its nonuniformity.

In conclusion, we investigated temperature dependence of photoluminiscence for different types of structures with ternary III–N alloys. It is shown that optical properties can be strongly affected by composition fluctuations in AlGaN or InGaN alloys. For AlGaN layers rapid thermal annealing leads to effective decrease in nonuniformity, while for InGaN multilayer structure with dense arrays of quantum dots RTA leads to increase in phase separation.

Acknowledgements

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Light confinement in quantum dots

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Abstract. A concept of light confinement in quantum dots due to diffraction of electromagnetic waves at the dot boundary, is introduced. Possible manifestations of the phenomenon, such as depolarization shift of the exciton frequency, polarization-dependent splitting of the gain band, asymmetry of the absorption and gain spectra, induced magnetization of quantum dots, contribution to radiative lifetime, are discussed both for isolated quantum dots and quantum dot ensembles. We propose that the effect of light confinement should be properly addressed to optimize the design of optoelectronic devices involving quantum dots.

Introduction

A fundamental breakthrough in semiconductor device physics is connected with the recent progress in the synthesis of sheets of nano-scale 3D confined narrow-gap insertions in a host semiconductor, quantum dots (QDs). The large body of recent results on physical properties of QDs and their utilization for the QD laser design has been accumulated in a monograph [1]. The key peculiarity of QDs emerges from the 3D confinement of the charge carriers determined by QD size and shape. However, there exists a class of effects governed by the QD size and shape, which have not received much attention so far. These effects are related to resonant nature of the exciton which provides a dramatic resonant discontinuity of the permittivity at the QD boundary and, consequently, gives rise inhomogeneity of the electromagnetic field both inside and outside QD. By analogy with charge carrier confinement, redistribution of the electromagnetic field energy between the QD interior and exterior under effect of the QD boundary can be referred to as *light confinement*. In many cases the role of light confinement can properly be accounted for the formation in QD of depolarization electromagnetic field, e.g., in dipole approximation of the diffraction theory.

To our knowledge, some physical consequences of the light confinement in an individual QD first time were considered by Schmitt-Rink *et al.* [2]. Manifestation of this phenomenon in relation to the scanning near-field optical microscopy was discussed by Martin *et al.* [3] for geometrically complex mesoscopic systems and by Hanewinkel *et al.* [4] for QDs. An asymmetry of optical absorption and gain spectra in single QD because of depolarization field has been mentioned in Ref. [4]. Recently it has been predicted and experimentally verified that the light confinement in QD arrays constituted by anisotropically shaped QDs manifests itself as polarization splitting of the gain band [5] and, in more general case, as the fine structure of this band [6]. Such a splitting was first experimentally observed by Gammon *et al.* [7], where the depolarization field effect has been proposed as possible

explanation of the splitting. Some new effects related to the light confinement in QDs are considered in Ref. [8].

In our paper we introduce sequential concept of light confinement in 3D-confined resonant nanoinsertions and discuss some general consequences of this phenomenon in an isolated QD and in QD arrays. Our consideration is based on classical electrodynamics of inhomogeneous media.

1. Depolarization shift of the exciton resonance

1.1. Polarizability of a single QD

Conventional phenomenological model of the gain in a QD is based on semi-classical theory of two-level systems which gives the equation of motion for the mean polarization $\boldsymbol{\mathcal{P}}$ caused by transitions between the levels:

$$\left(\frac{\partial^2}{\partial t^2}+\frac{2}{\tau}\frac{\partial}{\partial t}+\omega_0^2\right)\boldsymbol{\mathcal{P}}=-\frac{\omega_0}{2\pi\varepsilon_h}\widehat{\mathbf{g}}_0\,\boldsymbol{\mathcal{E}}\,. \tag{1}$$

Here ω_0 is the exciton resonant frequency and τ is the exciton dephasing time in QD. The phenomenological parameter $\widehat{\mathbf{g}}_0$ is proportional to the oscillator strength of the transition. In anisotropically shaped QDs this parameter is tensorial owing to anisotropy of the charge carrier confinement [10]. In an inverted medium $(\widehat{\mathbf{g}}_0)_{ij} > 0$. The field $\boldsymbol{\mathcal{E}}$ stands for the field inside the QD, different from the external acting field $\mathbf{E}$. This difference is determined by the depolarization field which is as follows [9]: $\boldsymbol{\mathcal{E}} = \mathbf{E} - 4\pi\widehat{\mathbf{N}}\,\boldsymbol{\mathcal{P}}$, with $\widehat{\mathbf{N}}$ as the depolarization tensor. This tensor is symmetrical and depends only on the QD shape. If we neglect the contribution of the depolarization field putting $\boldsymbol{\mathcal{E}} = \mathbf{E}$ into Eq. (1), solution of this equation in the vicinity of resonance for time-periodic fields and isotropic $\widehat{\mathbf{g}}_0 = g_0\widehat{\mathbf{I}}$ gives the well-known Lorentz contribution to the medium polarizability: $\alpha(\omega) = (g_0/\varepsilon_h)[\omega-\omega_0+i/\tau]^{-1}$, which is commonly used as phenomenological model of the dispersion and the gain of a single QD: $\varepsilon_d(\omega) = \varepsilon_h[1+\alpha(\omega)]$. Otherwise, taking into account the contribution of the depolarization field, we obtain the tensorial polarizability of QD in the vicinity of resonance:

$$\widehat{\boldsymbol{\alpha}}(\omega)=\frac{1}{\varepsilon_h}\left[\omega\widehat{\mathbf{I}}-\left(\omega_0-\frac{i}{\tau}\right)\left(\widehat{\mathbf{I}}-\frac{1}{\varepsilon_h\omega_0}\widehat{\mathbf{g}}_0\widehat{\mathbf{N}}\right)\right]^{-1}\widehat{\mathbf{g}}_0\,. \tag{2}$$

Thus, the QD's shape reflects itself as fine structure of the resonance which itself is a superposition of three bands with frequencies $\omega_N^{(j)} = \omega_0 - \nu_j$, $i, j = 1, 2, 3$, where ν_j are the eigenvalues of the inner tensorial product $\widehat{\mathbf{g}}_0\widehat{\mathbf{N}}/\varepsilon_h$. For spherical inclusions the tensors $\widehat{\mathbf{N}}$ and $\widehat{\mathbf{g}}_0$ are isotropic and the fine structure manifests itself as a polarization-independent shift of the gain line depicted in Fig. 1. If the energy splittings are much less than the bandwidth, which means the inequality for energy spacings $\max|\Delta\omega_{ij} = \omega_N^{(i)} - \omega_N^{(j)}| \ll 2/\tau$ to be true, the depolarization field will lead to a distortion of the gain band similar to the inhomogeneous broadening. Otherwise, when $|\Delta\omega_{ij}| \sim 2/\tau$, three separate bands will appear in the gain spectrum of a QD array.

1.2. Birefringence in QD arrays

Since the QD linear extension is much smaller than the resonance wavelength, electromagnetic properties of such ensembles – composite materials – can be modeled in the framework