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Oscillator strength of optical transitions in InGaAsN/GaAsN/GaAs quantum wells

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Experimental and theoretical considerations and results on the effect of nitrogen incorporation on the oscillator strength of optical transitions in InGaNAs/GaAs quantum wells (QWs) are presented. Therefore, a set of dilute nitride quantum well structures was grown by molecular beam epitaxy. Optical investigation via spectroscopic methods have been performed at various temperatures for both the as-grown samples, and after rapid thermal annealing. The fundamental transition energy and its oscillator strength vs. the QW composition have been systematically investigated. Additionally, the effect of the bandgap discontinuities on the transitions intensity has also been considered. The experimental data have been confronted with the band structure calculations within the effective mass approximation employing a two level repulsion model for the nitrogen-containing structures. The obtained results are crucial for possible future applications employing the quantum well in cavity structures and bringing the practical exploitation of quantum electrodynamics phenomena to the telecommunication spectral range.

Keywords: dilute nitride, quantum well, oscillator strength.

1. Introduction

Dilute nitride compounds, as GaInNAs, constitute a novel material system proposed and developed early in the 1990's by KONDOW *et al.* [1] and they remain very interesting nowadays because of their unique physical properties and potential device applications. During the epitaxial crystal growth process small amounts of nitrogen are added and substitute group V atoms in III–V semiconductors. The result is a huge bandgap reduction, which is the main driving force of applications of GaAs-based dilute nitride quantum wells (QWs) in near infrared optoelectronics. Their potential has been demon-

strated in both edge emitting and vertical cavity surface emitting lasers, but also in solar cells, semiconductor optical amplifiers, photodetectors and optical modulators [2].

In the following work, we study experimentally some of unreported yet properties of dilute nitride QWs in the aspect of their possible exploration as light emitting media in QW – optical cavity structures and consequences of tailoring the electronic wave functions and hence the QW transition oscillator strength (OS) and we compare the results to theoretical considerations. Microcavity-type semiconductor structures are reaching high interest because of their potential in studying the interaction between light and matter and in exploring it further in improved performance photonic devices based on polariton lasers. So far, there have been realized such structures emitting in the wavelength range below 1 μm [3–5], whereas it is of a great practical importance to extend the emission range to further infrared, *e.g.*, to II or III fiber telecom window. However, the biggest challenge lies in the choice of the proper QW material. InGaAsN QWs on GaAs, which can be easily embedded into well-established AlAs/GaAs distributed Bragg reflector cavity, could be a possible solution to obtain the strong coupling regime between the QW excitons and photonic cavity mode in the range of 1.3 μm. The exciton oscillator strength in this case is one of the main physical parameters to be optimized towards an enhancement of light-matter coupling and large enough values of Rabi splitting. In order to obtain an experimental estimate on the OS, we rely on the measurements of reflectivity coefficient spectra, as it has been shown being an efficient way of determining the transition intensity of QW structures [6].

2. Experiment

The investigated quantum wells of $In_{0.35}GaAs_{1-x}N_x/GaN_{x1}As_{1-x1}/GaAs$ (with GaNAs as a strain-compensating layer) were grown by molecular beam epitaxy on (001) GaAs substrate and GaAs buffer. The QW itself has been repeated 4 times in order to enhance the optical response, where the wells are separated by 9 nm of GaAs barriers. Figure 1 shows the respective layer structure and energy-band diagram. We examined

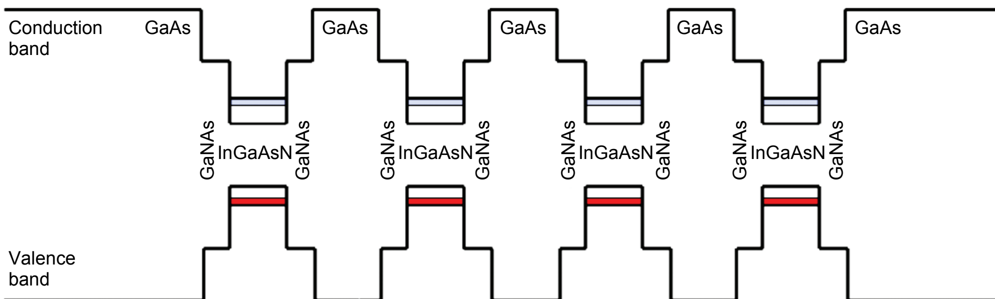

Fig. 1. A scheme of layer structure of the investigated InGaAsN/GaAsN/GaAs MQWs samples and the corresponding alignment of the band edges. The InGaAsN layers are 7 nm thick, the GaAsN ones – 4.5 nm, and the separating GaAs layers – 9 nm.

two sets of dilute nitride samples grown on GaAs undoped substrates and differing in the In content. First set (series A) consisted of $In_{0.35}GaAs_{1-x}N_x/GaN_{x1}As_{1-x1}$ QWs. In the second set of samples (series B) QW layers were formed of $In_{0.26}GaAs_{1-x}N_x/GaN_{x1}As_{1-x1}$. The width of the InGaAsN layers was 7 nm and the GaAsN ones – 4.5 nm in all the cases. The entire structure has been capped by 100 nm of GaAs. The x_1 mole fraction of nitrogen in GaAsN layers with respect to the x mole fraction of nitrogen in InGaAsN was approximately $x_1 = 1.5x$, when just basing on the growth conditions (estimation based on difference in the growth rates between the layers with and without In). The compositions of the layers are determined based on growth calibration samples or reference samples without nitrogen to estimate the In concentration, whereas the N contents have been determined from HR XRD measurements. The contents for all the samples are listed in Table 1. The same samples have also been annealed using rapid thermal annealing (RTA) procedure at 700 °C in the atmosphere of nitrogen for 3 minutes.

T a b l e. 1. Indium and nitrogen contents in the investigated samples.

Sample	[In]	[N] (QW)	[N] (barrier)
A1	35%	1.3%	1.8%
A2	35%	1.1%	1.6%
A3	35%	0.9%	1.5%
B1	26%	0%	GaAs barrier
B2	26%	1.4%	1.8%
B3	26%	1.9%	2.5%

The measurements of reflectivity coefficient have been performed in a standard experimental configuration, where the light from a tungsten halogen lamp served as a probe beam, which after reflection has been dispersed by a single grating 0.5 m focal length monochromator and detected by a multichannel linear InGaAs charged coupled device.

3. Theoretical approximations of the oscillator strength of QW transition

We have developed a theoretical model in order to investigate the OS issue in a deeper manner and to compare the results of calculations with the experimental data. The calculations of the related energy states in the QW region are obtained by solving the stationary Schrödinger equation assuming parabolic dispersion curves for the particles. The bandgap is given by the band anticrossing (BAC) model which includes the influence of nitrogen on the band structure and estimates the effective bandgap reduction properly [7].

One can derive two important parameters to study the intensity of optical transitions: the overlap integral between the electron and hole wave functions

(square of which reflects the oscillator strength of a band-to-band transition, *i.e.*, in the condition where the excitonic effects can be neglected), or the oscillator strength with the excitonic correction included. As for instance in Refs. [8, 9] the oscillator strength per unit area is related to the exciton wave function by:

$$f = \frac{2M^2}{m_0 E_{\mathrm{exc}}} \left| \int_{-\infty}^{\infty} \mathrm{d}z \, f_e(z) f_h(z) \right|^2 |\Phi(0)|^2 \tag{1}$$

where: M – the optical transition matrix element between the valence and conduction bands, f_e and f_h – the envelope functions of particles in the growth direction, and $\int_{-\infty}^{\infty} \mathrm{d}z \, f_e(z) f_h(z)$ is the overlap integral as mentioned above. $\Phi(\mathrm{r})$ is the function of relative motion of the electron and hole in lateral direction. Simplifying Eq. (1) one can write also the oscillator strengths in following forms [10]:

$$f \sim \frac{1}{a_0^2} \left| \int_{+\infty}^{-\infty} \mathrm{d}z \, f_e(z) f_h(z) \right|^2 \propto \mu^2 \left| \int_{+\infty}^{-\infty} \mathrm{d}z \, f_e(z) f_h(z) \right|^2 \tag{2}$$

where: a_0 – the in-plane exciton radius and μ – the in-plane reduced mass of the exciton. Therefore eventually, in all the cases where the Coulomb correlation cannot be neglected (as very often at low temperatures for QWs based on GaAs) the dependence of OS on any parameter of the well is given by a combined effect of the exciton reduced mass change and the effect of the single particle states overlap integral.

4. Results and discussion

We have measured reflectivity spectra for all the structures in the conditions of almost normal incidence. An exemplary spectrum for an as-grown sample named A3 (see also Table 1) is shown in Fig. 2**a**. It exhibits a typical feature of a resonant line being

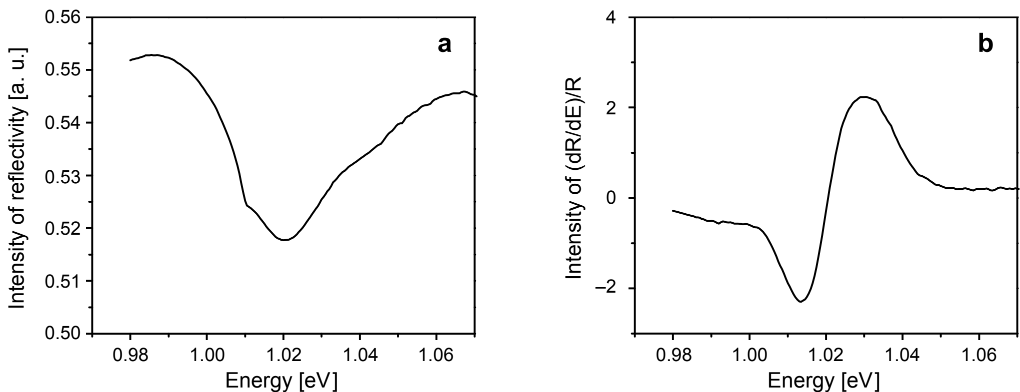

Fig. 2. Reflectance spectra before (**a**) and after (**b**) the procedure of taking the derivative and renormalization.

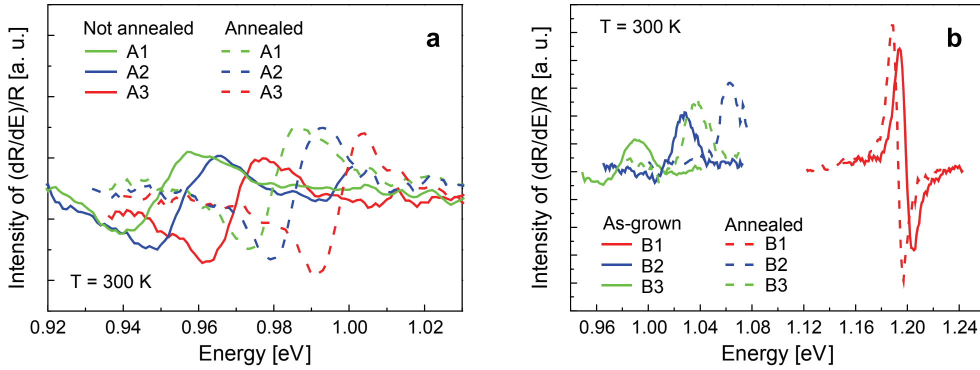

Fig. 3. Room temperature reflectance resonance-type curves for both sets of samples: series with 35% of In (**a**), series with 26% of In (**b**).

an indicator of the optical transition. As the spectra always experience some background effects and the spectrum has rather an indirect connection of the particular lineshape to the transition intensity, then some mathematical processing has been performed: a derivative has been calculated with respect to energy (see Fig. 2**b**), and hence the peak-to-peak intensity can be used as a good estimate of the transition intensity in such a differential spectrum. When it is further multiplied by the linewidth it imitates well the integral transition intensity.

In the case of room temperature measurements the differential reflectivity spectra for all the investigated samples are shown in Fig. 3 in the vicinity of the fundamental optical transition. Beside the normally expected red shift of the transitions with the increase of N concentration, there is also observed a strong blue shift after annealing. The latter is mostly driven by the intermixing effects and the QW interface interdiffusion, which change the well potential profile from a square-like in case of the as-grown structures, into a graded one after annealing. The annealing process is typically used in dilute nitrides to cure some of the defects connected with the interstitial positions of the N atoms and improve the overall sample quality, and has been reported previously [11]. As the annealing modifies the electronic structure and affects the optical material quality, it changes the transition oscillator strength as well, with a general trend to increase after annealing, which is most probably a combined effect of both the mentioned factors.

Figure 4**a** summarizes the values of all the experimentally determined intensities obtained at room temperature for all the samples. It is observed that basically the transition intensity increases with the transition energy. This can, at least in part, be explained by Fig. 4**b** showing the calculated squared overlap integrals for such QW structures of series A (*i.e.*, for 35% In content) in a similar range of energies (similar range of N contents). It is seen that for certain assumptions on the band offset Q_c^* between GaAsN and GaAs (value of which is not very well established in the literature) the transition intensity can indeed increase with energy (*i.e.*, with a decrease of nitrogen concentration).

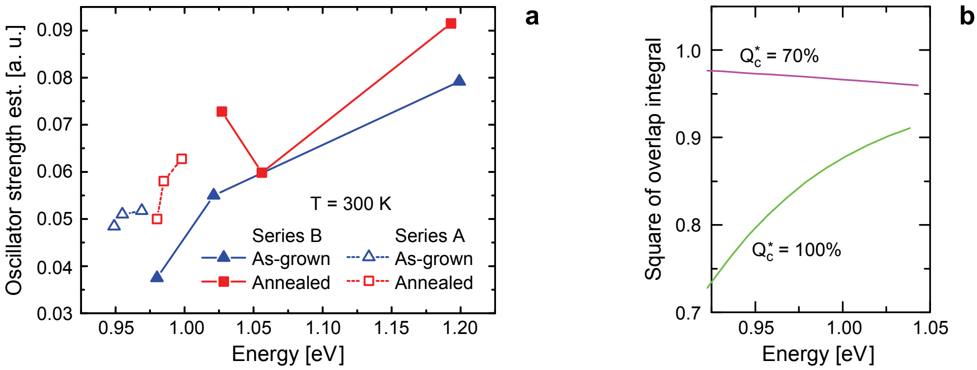

Fig. 4. Results concerning estimation of quantity equivalent to optical transition oscillator strength in room temperature: experimental results (**a**), theoretical predictions (**b**). Q_c^* is the chemical band offset between the GaAsN layer and GaAs.

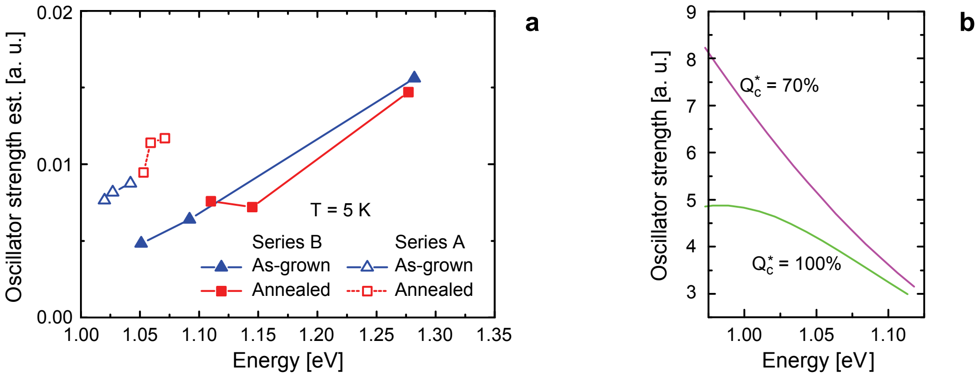

Fig. 5. Results concerning the estimation of quantity equivalent to optical transition oscillator strength: experimental results (**a**), theoretical predictions (**b**).

In the case of low temperature measurements ($T = 5$ K) the experimental results are qualitatively similar (see Fig. 5**a**) and can be expressed as an increase of the transition intensity with its energy. However, now the transitions are to be considered as excitonic and need to be compared to the model including the Coulomb interaction correction (as in Eq. (2)). The latter is shown in Fig. 5**b** for two values of the band offsets Q_c^*. It is seen that the theory predicts a trend which is opposite to the experimental one. The latter is a consequence of an increase of the electron effective mass (and hence also the exciton mass) with the increasing N mole fraction (as a direct outcome of the BAC model). This is not observed in the experiment, most probably due to trapping of carriers at defect-related centers at low temperatures. Therefore, less QW excitons can be formed, which decreases the absorption, and hence also the determined transition intensity, and hence deteriorates the contribution to the intensity coming from the excitonic effect.

What is worth noting, the experimentally determined intensities are always larger for the annealed samples in agreement with the room temperature data, origin of which has been mentioned above.

5. Conclusions

In this article, we have presented an experimental approach towards the estimation of oscillator strength for two different designs of multiple quantum wells that also varied with nitrogen content. We have used reflectance spectroscopy at room and low temperatures and judging by resonance-type curves after some signal processing we could extract the important parameters for the oscillator strength estimations like transition energy, linewidth and relative intensity of absorption mechanism. First of all, there has been observed a significant effect of post-growth annealing for all kinds of samples and based on the OS values, it improves the QW structural quality. Increasing the nitrogen content causes the OS decrease, which is in qualitative agreement with theoretical predictions for room temperature data, but in contrary to the low temperature experimental results, where the expected excitonic oscillator strength should increase with the nitrogen content. The latter is most probably related to trapping states being present and acting effectively at low temperatures in these dilute nitride wells. Decreasing the concentration of these defects seems to be the main route for further material optimization.

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