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Analysis of the transformation of radiative recombination spectra of *n*-GaN after magnetic field treatments based on the queueing theories concept

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Abstract. Long-term changes in radiative recombination spectra of *n*-GaN after magnetic field treatments have been studied. It has been found out that the intensity of the radiation of donor-acceptor pairs remains unchanged over time, while the intensity of edge photoluminescence significantly decreases. These features have been explained by assuming the formation of additional donor levels and using the concepts of the queueing theory of donor-acceptor recombination.

Keywords: gallium nitride, magnetic field, radiative recombination, queueing theory.

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1. Introduction

Study of influences of physical fields on crystalline solids is an actual scientific problem nowadays. Both destructive [1] and non-destructive techniques [2, 3] are used for such study. It is especially important for analyzing structural perfection of semiconductor materials for microelectronic applications [4] and modern sensor systems based on gallium nitride [5].

In [6–9], evolution of the defect subsystem in III-V semiconductors after exposure to magnetic, electromagnetic (microwave) and radiation (irradiation with an electron flow) fields was studied using the photoluminescence method. Most often, the observed transformations fit into the generally accepted concept of the changes in radiative and nonradiative recombination channels.

However, the initial impurity-defect composition of a material always plays an important role on the after effects. Specific features of radiative recombination spectra may be observed [10–13], which are very difficult to explain by competition of recombination channels. This article analyzes temporal patterns of reorganization of the defect subsystem of semiconductor structures based on *n*-GaN stimulated by action of pulsed magnetic fields.

2. Experimental

GaN thin films (2.0…2.5 μm) were prepared by metal-organic chemical vapor deposition on an Al_2O_3 substrate (430 μm). The epitaxial layers were *n*-type, doped with Si up to $n \sim 1.6\times10^{19}\,\mathrm{cm}^{-3}$. Photoluminescence (PL) spectra were measured at room temperature in the 350…650 nm wavelength range under 315 nm excitation by using a Perkin-Elmer LS55 PL spectrometer. Since weak magnetic field (WMF) treatments at $B = 60$ mT, $f = 10$ Hz, and $\tau = 1.2$ ms were the most efficient for III–V semiconductor materials [6, 14], this regime was chosen for our experiments. The treatment duration was 10 min. The initial sample (*i.e.* not subjected to the WMF treatment) served as a reference. All the measurements were repeated during three months after the WMF treatment to reveal the time-dependent features.

3. Results and discussions

3.1. Study of radiative recombination spectra of *n*-GaN

The PL spectra of the *n*-GaN before the treatment in a pulsed magnetic field and long time after it are shown in Fig. 1.

It can be seen from Fig. 1 that the PL spectra of the initial films contain a band near 3.4 eV corresponding to

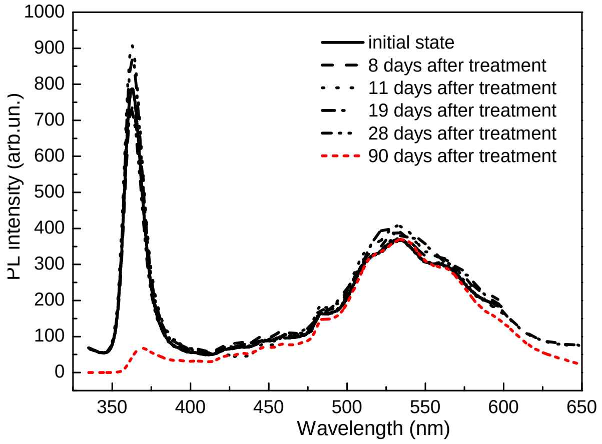

Fig. 1. Transformations of the PL spectra of *n*-GaN with time after treatment in magnetic field. (Color online)

interband transitions in gallium nitride (the GaN band gap is ~3.41 eV at room temperature) [15] as well as a so-called yellow band near 2.2 eV, the origin of which is associated with presence of defects [16, 17]. After treatment in a pulsed magnetic field, long-term nonmonotonic changes in the intensity of the PL bands are observed.

It is shown in [18, 19] that the observed aftereffects, which may be called magnetic memory effects, are possible to explain based on the idea of reorganization of dislocations and defect complexes in the *n*-GaN near-surface region. A characteristic feature of these spectra is that after the treatments, the radiation intensity of the impurity photoluminescence practically does not change, while the intensity of the edge (band-to-band) photoluminescence significantly decreases.

3.2. Theoretical analysis

The observed features of the edge and donor-acceptor photoluminescence may be explained by assuming that treatments in a pulsed magnetic field induce formation of additional donor levels in the band gap of gallium nitride, *i.e.* their number becomes much larger than the number of acceptor levels. In this case, non-equilibrium electrons appear at the donor levels forming a queue that awaits freeing of recombination channels with holes at the acceptor levels.

It is quite clear that these electrons do not contribute to the edge photoluminescence, and its intensity decreases therefore. On the other hand, the number of donor-acceptor pairs does not change (the number of the acceptor levels does not undergo changes), hence, the intensity of the corresponding radiative recombination will not change either.

These considerations are valid provided that the lifetime τ_1 of non-equilibrium charge carriers during interband radiative recombination is greater than the lifetime τ_2 of non-equilibrium charge carriers during donor-acceptor radiative recombination: $\tau_1 > \tau_2$. The lifetime τ_1 for an *n*-type semiconductor obeys the following relation [20]:

$$\tau_1 = \frac{1}{\gamma n_0}, \tag{1}$$

where γ is the bimolecular recombination coefficient (for GaN it is equal to $\gamma = 2.2\cdot 10^{-16}$ m^{-3} [20]), and n_0 is the equilibrium electron concentration ($n_0 = 8\cdot 10^{23}$ m^{-3} in the studied *n*-GaN structures).

The lifetime τ_2 in *n*-GaN is determined by the lifetime of minority equilibrium charge carriers (holes) and can be found from the following expression [20]:

$$\tau_2 = \frac{1}{\sigma_p v_T N_t}, \tag{2}$$

where σ_p is the cross-section of hole capture by an acceptor level, v_T is the thermal movement velocity of holes, and N_t is the concentration of acceptor levels in the band gap, respectively. It follows from the condition $\tau_1 > \tau_2$ and the expressions (1) and (2) that the following inequality must be satisfied:

$$N_t > \frac{\gamma n_0}{\sigma_p v_T}. \tag{3}$$

We estimate σ_p based on the following considerations. When a hole-acceptor level bound state appears, the space region where it is localized has a radius close to the radius a of a first orbit of the hydrogen-like model in a solid with dielectric constant ε_r. In turn, the radius a is equal to [21]

$$a = a_0\left(\frac{m_0}{m_p}\right)\varepsilon_r, \tag{4}$$

where $a_0 = 0.52917720859\cdot 10^{-10}$ m is the Bohr radius, m_0 is the mass of a free electron, and m_p is the hole mass, respectively. Consequently, the cross-section of hole capture by an acceptor level is of the order of

$$\sigma_p = \pi a^2 = \pi a_0^2\left(\frac{m_0}{m_p}\right)\varepsilon_r^2. \tag{5}$$

The value of the root mean square thermal movement velocity of holes is calculated as $v_T = \sqrt{3kT/m_p}$, where k is the Boltzmann constant and T is the absolute temperature.

Taking into account the following GaN parameters [22]: $m_p = 1.4m_0$ and $\varepsilon_r = 8.9$, calculation by expressions (3) and (5) provides $N_t > 5\cdot 10^{21}$ m^{-3}. In what follows we assume $N_t = 10^{22}$ m^{-3} in the analysis of the effect of formation of a queue of non-equilibrium electrons at the donor levels in the band gap.

Let us consider an ensemble of donor and acceptor levels in the band gap of a semiconductor. Recombination of non-equilibrium charge carriers over these levels produces radiation. The number of the donor levels d is greater than the number of the acceptor levels, $d > a$. On the one hand, recombination of non-equilibrium charge carriers is a random event [23]. This event is complex, since it consists of random elementary events of capture of a non-equilibrium electron to an energy level and its recombination itself. On the other hand, the considered donor-acceptor radiative recombination is characterized by formation of queues of non-equilibrium electrons at the donor levels, “waiting” for freeing a recombination channel with a hole at the acceptor levels. From the viewpoint of the probability theory, this ensemble of energy levels in the band gap may be considered as a multichannel queueing system of donor-acceptor radiative recombination.

Operation of a queueing system includes servicing the flow of requests entering it [24]. In our case, the requests mean non-equilibrium electrons captured by the donor levels. By servicing a request we mean recombination of an electron at a donor level with a hole at an acceptor level. Consequently, service channels are the pairs of the donor-acceptor levels in the semiconductor band gap, over which charge carriers recombine. Moreover, the number of the channels n is equal to the smallest value of the two numbers a and d: $n = \min(a, d)$. In the situation under consideration $n = a$. Non-equilibrium electrons (requests) enter the queueing system, *i.e.* they are captured by the donor levels at random times. Queueing a request (recombination of an electron with a hole) takes a random period of time. After the recombination event, the service channels (donor-acceptor pairs) become vacant and again ready for queueing the next requests (non-equilibrium electrons). Therefore, functioning of the queueing system of donor-acceptor radiative recombination is a random process. The system passes from one state to another at random times. Since the number of the donor levels exceeds the number of the acceptor levels, which is equal to the number of the channels, a non-equilibrium electron at a donor level, which finds all the recombination channels occupied, queues up and waits until some channel becomes free. This queueing system is a multi-channel waiting system. As a limitation imposed on waiting, consider the limit on the number of applications in the queue. In our case, the maximum possible number of non-equilibrium electrons at the donor levels that find all the recombination channels occupied (the maximum possible number of electrons in the queue) is equal to $m = d - a = d - n$. In what follows we will mainly use the terminology of the queueing theory. The approach to consider donor-acceptor recombination from the standpoint of the formalism of the queueing theory is important in that it allows us to establish a relationship between the parameters characterizing the flow of requests as well as the performance of an individual channel and system throughput indicators. The latter mean the probabilities of system idleness, queue formation, denial of service, average number of occupied (idle) channels, average number of requests in the queue and others.

We assume that the flow of requests obeys the Poisson distribution. In this case, the probability that k requests will arrive during the time interval τ is equal to [24]:

$$P_m(\tau) = \frac{(\lambda\tau)^k}{k!}\exp(-\lambda\tau), \tag{6}$$

where λ is the flow density of requests (the average number of requests received per unit of time), λτ is the expected value of the number of requests (the average number of requests per time interval τ).

We assume that the request service time t has an exponential distribution, the probability density $f(t)$ of which has the following form [24]:

$$f(t) = \mu\exp(-\mu t), \tag{7}$$

where μ^{-1} is the expected value (the average time of servicing one request).

The concept of a Poisson flow of requests and an exponential distribution of service time makes it possible to use the apparatus of the Markov random processes in the queueing theory [24]. In accordance with [24], we present here a summary of final expressions for the above-mentioned throughput indicators of an n-channel queueing system, the input of which receives a Poisson flow of requests with the intensity λ. The service intensity of each channel is μ and the maximum possible number of places in the queue is limited by the value m.

The probability P_0 of system idleness (all the system channels are free) has the following form:

$$P_0 = \left\{\sum_{k=0}^{n}\frac{\rho^k}{k!} + \frac{\rho^{n+1}}{n!(n-p)}\left[1 - \left(\frac{\rho}{n}\right)^m\right]\right\}^{-1} \tag{8}$$

and $\rho = \lambda/\mu$.

The probability P_Q of queue formation is equal to

$$P_Q = \frac{\rho^n}{n!} \times \frac{1-(\rho/n)^m}{1-(\rho/n)} P_0. \tag{9}$$

The probability P_R of service denial is

$$P_R = \frac{\rho^{n+m}}{n^m n!} P_0. \tag{10}$$

The average numbers of occupied n_B and idle channels n_I are determined by the following formulas:

$$n_B = \rho(1 - P_R), \tag{11}$$

$$n_I = n - n_B. \tag{12}$$

The average number of requests m_Q in the queue obeys the following relationship:

$$m_Q = \frac{\rho^{n+1}}{nn!} \times \frac{1-(\rho/n)^m(m+1-m\rho/n)}{1-(\rho/n)^2} P_0. \tag{13}$$

The expressions (8)–(10) and (13) include the factorial of the number of servicing channels n equal to the number of acceptor levels. This number takes on large values, which will be estimated as follows. The concentration of the acceptor levels in the band gap of gallium nitride is $10^{22}\,\text{m}^{-3}$, the illumination area of a semiconductor surface during measuring PL spectra is $5\cdot10^{-5}\,\text{m}^2$, and the depth of radiation penetration into the crystal is about $10^{-6}\,\text{m}$, which results in $n = 5\cdot10^{11}$. To approximately calculate the values of the factorials of such large numbers, the Stirling formula is used:

$$n! \approx \sqrt{2\pi n}\left(\frac{n}{e}\right)^n, \tag{14}$$

where e is the natural logarithm base (Euler’s number).

To estimate the throughput indicators of the n-channel queueing system (8)–(13), it is necessary to get an idea of the intensity values λ, μ and queue length m.

As noted above, the recombination process of non-equilibrium charge carriers consists of two stages: capture of carriers at the energy levels in the semiconductor band gap and their recombination itself. Naturally, the intensity of the requests flow exceeds the intensity of servicing by each channel.

We assume that the ratio of the intensity of the requests flow to the intensity of servicing by each channel μ is equal to the number of queuing channels n, *i.e.* $\rho = n$. Moreover, not limiting generality of the consideration, we assume that the number of the donor levels is twice as large as the number of the recombination channels (acceptor levels), $d = 2n$. In this case, the maximum possible number of the places in the queue is $m = n$. Taking into account these assumptions, it is possible to calculate the throughput indicators of the n-channel queueing system of donor-acceptor radiative recombination.

The expression (8) is transformed as follows to determine the probability of system P_0 downtime. Transforming the uncertainty in the second term at $\rho/n = 1$ according to the L'Hopital rule, we obtain:

$$P_0 = \left[\sum_{k=0}^{n} \frac{n^k}{k!} + \frac{n^{n+1}}{n!} \right]^{-1} . \tag{15}$$

It is clear from (15) that even at the maximum value $k = n$ in the first term, the second term is n times larger than the first one. Therefore, we can consider

$$P_0 \approx \frac{n!}{n^{n+1}} . \tag{16}$$

Finally, taking into account (14), we can write $P_0 = \sqrt{2\pi}/e^n\sqrt{n} \approx 0$ for the probability that all the channels in the system are free. Transforming the uncertainty in (9) and taking into account (16), we find that the queue formation probability $P_Q \approx 1$.

According to (10) and (16), the probability of a service denial is equal to

$$P_R \approx \frac{1}{n} \approx 2 \cdot 10^{-12}. \tag{17}$$

In accordance with (11), (12) and (17) as well as taking into account that $1/n << 1$, we have $m_B \approx n \approx 5 \cdot 10^{11}$, $n_I \approx 0$ for the average numbers of the occupied and idle channels, respectively.

Calculation of the uncertainty in (13) and substitution of (16) at $n >> 1$ allows us to obtain the following ratio for the average number of applications in the queue: $m_Q \approx n/2 \approx 2.5 \cdot 10^{11}$. Summarizing the obtained numerical results for the throughput indicators of the queueing system of donor-acceptor radiative recombination, we can state that formation of queues of non-equilibrium charge carriers has a mathematical justification.

Therefore, the observed features of the transformation of PL spectra of n-GaN caused by magnetic field treatments are due to formation of additional donor levels, so that their number becomes much greater than the number of the acceptor levels. As a result, formation of queues of non-equilibrium electrons that await freeing recombination channels with holes at the acceptor levels takes place.

4. Conclusions

After exposure of n-GaN structures to pulsed magnetic fields, long-term nonmonotonic changes in the intensity of the photoluminescence bands are observed. The peculiarity of the transformation of the PL spectra in time is that the radiation intensity of donor-acceptor pairs recombination practically does not change, while the intensity of the edge photoluminescence significantly decreases. The rationale for this phenomenon is based on the assumption that magnetic field treatments stimulate formation of additional donor levels in the band gap of gallium nitride. As a result, the number of the donor levels becomes greater than the number of the acceptor levels. In this case, a queue of non-equilibrium electrons at the donor levels, which await freeing of recombination channels with holes at the acceptor levels, is formed. These electrons do not contribute to the edge photoluminescence, thereby leading to a decrease in its intensity. Since the number of the donor-acceptor pairs remains unchanged, the intensity of the corresponding radiative recombination does not change.

From the viewpoint of the probability theory, an ensemble of donor and acceptor energy levels in the band gap of a semiconductor may be considered as a multichannel queueing system for donor-acceptor radiative recombination. Calculations of the values of the throughput indicators of the queueing system of donor-acceptor radiative recombination indicate that formation of queues of non-equilibrium charge carriers has a mathematical basis.

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Аналіз трансформації спектрів випромінювальної рекомбінації *n*-GaN після обробки магнітним полем на основі концепції теорій масового обслуговування

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Анотація. Досліджено довгострокові зміни спектрів випромінювальної рекомбінації *n*-GaN після обробок магнітним полем. Встановлено, що з часом інтенсивність випромінювання донорно-акцепторних пар практично не змінюється, в той час як інтенсивність крайової фотолюмінесценції суттєво зменшується. Запропоновано обґрунтування цих особливостей трансформації спектрів фотолюмінесценції на основі припущення про утворення додаткових донорних рівнів і концепцій теорії масового обслуговування донорно-акцепторної рекомбінації.

Ключові слова: нітрид галію, магнітне поле, випромінювальна рекомбінація, теорія масового обслуговування.