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Physical mechanisms and models of long-term transformations of radiative recombination in n-GaAs due to the magnetic field treatments

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Abstract. Simulation of long-time changes in photoluminescence of *n*-GaAs has been performed, and the mechanism of transformation of the defect structure caused by magnetic field treatments has been represented.

Keywords: magnetic field treatments, photoluminescence, dislocation impurity complex, random variable, cyclotron frequency, electromagnetic radiation.

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1. Introduction

Improving the structural perfection of epitaxial films of semiconductor structures by using non-thermal effects of external fields, including the magnetic one, is a promising direction for modifying their physical characteristics. In its turn, control for changing the microstructure of semiconductor materials is possible when understanding the mechanisms underlying the process of evolution of structural and impurity defects under the influence of a magnetic field. Currently, these mechanisms are not well investigated, and known investigations are mainly related with diamagnetic dielectric (ionic) crystals containing paramagnetic impurities, and their results are interpreted on the basis of the spin dynamics, that is, as a consequence of the singlet–triplet transitions in the radical pair 'defect-stopper', which causes weakening the ability of magnet-sensitive stoppers to decelerate dislocations [1-4].

Therefore, this work to a certain degree is aimed at addressing to remedy this gap and is devoted to the analysis of the mechanisms and time regularities of transformation of defect subsystem of semiconductor structures based on GaAs, stimulated by magnetic field treatment.

2. Experimental

The studied objects were the samples of epitaxial *n*-GaAs doped with tellurium of 300-μm thickness and carrier concentration $5 \cdot 10^{16}$ cm^{-3}. We studied photoluminescence (PL) spectra at 77 K within the spectral range 0.6 to 2.0 eV excited by light with $h\nu \geq 2$ eV. The PL spectra were measured from the side of the epitaxial film for a long period after the magnetic field (MF) treatment (for 60 days). MF treatment was carried out at room temperature with a weak pulsed magnetic field with the parameters: $B = 60$ mT, $f = 10$ Hz, $\tau = 1.2$ ms for 10 min.

3. Experimental results and discussion

The PL spectra of the initial samples showed: the near-edge band with $h\nu_{max} \approx 1.48$ eV and broad bands caused by local states in the forbidden gap with $h\nu_{max} \approx 1.02$ eV and 1.21 eV, which is typical for the samples of this type (Fig. 1). We will associate these bands, as in [5-9] with radiative recombination in the donor-acceptor (DA) pairs formed by gallium vacancies and impurities (Cu).

The intensities of the PL bands after MF treatment generally decrease, but over some relaxation period they return to a previous value or close to it. The observed long-time non-monotonic changes in the intensities of PL bands after MF treatment can be explained being based on the concept of dynamic behavior in near-surface areas of dislocations or complexes of defects.

Thus, in the first scenario, it is assumed that in the initial state, the dislocations are fixed with stoppers, and as a result of the MF treatment and action of the fields of residual mechanical stresses on them, the elastic fields of other defects as well as the force of attraction of dislocations to the crystal surface [10] the dislocations detached from local obstacles and moved from the near-surface layer to the surface (the near-surface area means a thickness of the layer in which the electron-hole pairs are generated, when the sample is exposed to light exciting PL).

In the forbidden gap, the dislocations create dislocation levels, transitions on which preferably have a non-radiative character in GaAs crystals [11, 12], and hence, reduce the intensities of PL bands. Finally, the dislocations cause condensation of defects, in particular gallium vacancies, that are centers of non-radiative recombination [13]. Thus, the dislocations effectively increase the channel of non-radiative recombination.

Therefore, in the analyzed with photoluminescence near-surface area an increase in the dislocation concentration (or other centers of non-radiative recombination) should lead to a decrease in the intensity of all the observed bands – near-edge and impurity ones. Furthermore, the movement of non-radiative centers from the analyzed with photoluminescence layer into the crystal bulk or into drains leads to the increase in intensity of PL bands after reaching a minimum.

Let's consider a second scenario being based on behavior of defect complexes under the influence of MF treatment. It is known that in the doped GaAs crystals containing back-graund copper impurity, the complexes 'donor–acceptor' are formed. The donors are ionized atoms Te^+ and Si^+ in the As sublattice, and acceptor is uncontrolled impurities Cu^- and Cu^{2-} in the Ga sublattice [13]. These associates bind the gallium vacancies, that is they are the place of drain for excess gallium vacancies, which, as already mentioned, are the centers of non-radiative recombination, and this complex $[Cu_{Ga}Te_{As}]$ is responsible for the PL band near 1.3 eV [5].

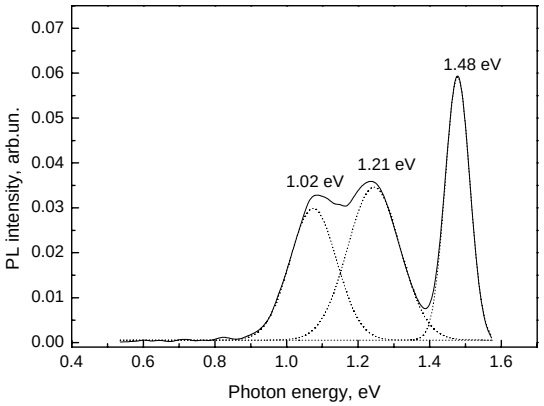

Fig. 1. Typical PL spectrum of the studied samples in the initial state.

Let's assume that under the action of the MF treatment, in the near-surface area these complexes destruct and, consequently, the diffusion of copper occurs principally along the interstices (Cu is a fast diffusing impurity) [13] to the surface under the influence of a concentration gradient. Then, with the lapse of time after the magnetic field treatment as a result of destruction of the donor-acceptor pairs $[Cu_{Ga}Te_{As}]$, the isolated vacancies of gallium could appeared. Because gallium vacancies are non-radiative recombination centers, then with an increase in their number the intensity of all the observed bands decreases. Subsequently, due to, on the one hand, movement of copper released from the impurity complexes to the surface, the concentration of gallium vacancies is recovered and, on the other hand, due to the diffusion of copper into the surface region from the epitaxial layer the impurity tellurium-copper complexes is also recovered. Accordingly, the intensity of the PL bands will tend to its original state, which is observed in the experiment.

In the framework of detailing these mechanisms of change in the intensity of PL bands, it is necessary to solve two problems. The first one concerns the mathematical description of the time regularities of change in PL intensity after MF treatment, while the second one – the mechanism of impact of this treatment on dislocations and impurity complexes.

3.1. Probabilistic-physical modeling of the evolution of the defect semiconductor structure due to magnetic field treatment

To solve the first problem, we use the results of the work [14], which shows that the physical processes are caused by random events, and the corresponding random variables – the times before the events – obey the distribution Weibull–Gnedenko.

We introduce the following random events: a random event – movement of a defect (dislocations, gallium vacancies) from the near-surface region to the

surface (boundary) and a random event – movement of a defect to the near-surface area from the epitaxial layer (source) bordering with the near-surface area and commensurate with the sizes of the latter. Once again, we emphasize that the near-surface area means the thickness of the layer, in which the electron-hole pairs generate, when this layer is exposed to light of photoluminescence excitation. Then, the random variable is time to a random event. Accordingly, $F_1(t)$ is the function of time distribution before movement of the defect from the near-surface area to the boundary (the probability of movement from the near-surface area to the boundary); $F_2(t)$ is the function of time distribution before movement of the defect to the near-surface area from the source (the probability of movement to the near-surface area from the source).

Let's consider a new random event – the absence of a defect in the near-surface area. This event is complex and consists of random event – the movement defect from the near-surface area to the boundary – and a random event – the absence of the movement of a defect to the near-surface area from the source. Let us assume that these events are independent. Then the probability of this complex event is the product of the probabilities of the events constituting:

$$P_{1-2}(t) = F_1(t)\left[1 - F_2(t)\right]. \quad (1)$$

Being based on [14], we use Weibull–Gnedenko distribution as $F_1(t)$ and $F_2(t)$. Then, in a general case $F_1(t)$ and $F_2(t)$ can be expressed as follows [14]:

$$F_1(t) = 1 - e^{-\left(\frac{t}{\tau_1}\right)^{m_1}}, \quad (2)$$

$$F_2(t) = 1 - e^{-\left(\frac{t}{\tau_2}\right)^{m_2}}, \quad (3)$$

where τ_1 and τ_2 are the time constants of random events; m_1 and m_2 – form factors of the time distribution function before corresponding random event. Thus,

$$P_{1-2}(t) = \left[1 - e^{-\left(\frac{t}{\tau_1}\right)^{m_1}}\right] e^{-\left(\frac{t}{\tau_2}\right)^{m_2}}. \quad (4)$$

Accordingly, $I(t)$ is proportional to $P_{1-2}(t)$:

$$I(t) = I_{in} - I_0\left\{\left[1 - e^{-\left(\frac{t}{\tau_1}\right)^{m_1}}\right] e^{-\left(\frac{t}{\tau_2}\right)^{m_2}}\right\}, \quad (5)$$

where I_{in} is the initial value of the intensity photoluminescence of PL band, I_0 – proportionality factor. This function has one extremum (minimum).

Fig. 2 (a, b, c) shows the results of approximation by the method of least squares with the expression (5) of change in the intensity of integrated PL for the edge and impurity bands of studied *n*-GaAs with the parameters listed in table, respectively.

Table. Fitted parameters used for simulation with the expression (5)

Fitted parameter	Edge emission	1.02 eV	1.21 eV
I_{in}	1	1	1
I_0	1.02	1.1	1.1
τ_1 (days)	2.47	4.11	1.69
τ_2 (days)	99.58	99.75	99.82
m_1	0.27	0.64	0.46
m_2	10.01	2.51	1.83

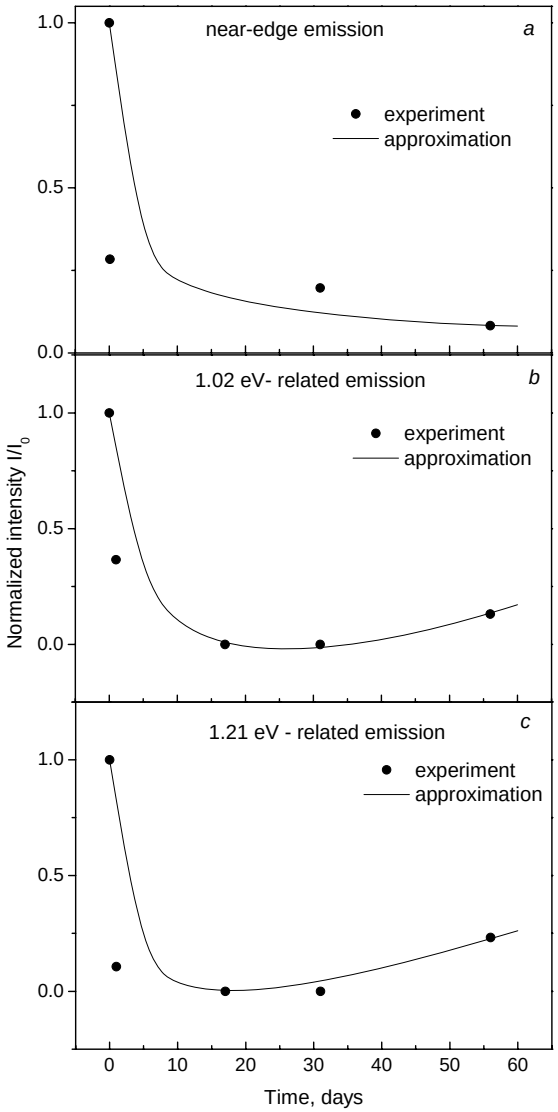

Fig. 2. Variation of the PL intensity of observed bands as a function of time after weak magnetic field treatment (dots – experiment, line – averaging).

One can note the good agreement between the experimental and theoretical results, as well as the fact that the parameters of the approximation (I_{in}, I_0, τ_2, m_1) are almost identical both for impurity and for the edge PL bands. Here, the τ_1 and m_2 parameters are close, but still different from each other. The latter circumstance may indicate that at least two factors are responsible for the change in the intensity of the PL bands: the first factor is common to all the observed bands, and the second one characterizes each band separately. In the first approximation, we have changes in the channels of radiative and non-radiative recombination. The changes of the latter are displayed equally on all the observed PL bands, and those of the first one – only on the certain peak of PL.

Let's estimate the parameters characterizing transformation of defect structure. If the movement of defects is diffusive, in accordance with [14], $\tau_1 = d^2/D$, where D is the effective diffusion coefficient, and d – thickness of the layer in which the electron-hole pairs are generated under exposing to light exciting PL. In our experiments, for the latter d is about $10^{-4} - 10^{-5}$ cm. Then, D is within the ranges $4.7 \cdot 10^{-14} - 4.7 \cdot 10^{-16}$, $2.8 \cdot 10^{-14} - 2.8 \cdot 10^{-16}$ and $6.9 \cdot 10^{-14} - 6.9 \cdot 10^{-16}$ cm^2/s for τ_1 parameters corresponding to the edge and impurity radiations, respectively. The obtained value agrees enough well with similar estimations for the diffusion coefficients of migrating impurities caused by weak magnetic field treatment of Si-SiO_2 and GaN-Al_2O_3 structures [15, 16]. The latter circumstance may indicate some generality of non-thermal mechanisms of interaction of submicrometer waves and weak magnetic fields with semiconductor structures.

3.2. Physical bases of influence of magnetic field treatments on the defect structure of semiconductor crystals

When placing the semiconductor crystal in a uniform magnetic field with the constant induction B, conductance electrons in n-GaAs, the effective mass of which is a scalar value (isotropic mass), begin to describe the helical path, that is, helical lines, the axis of which coincides with the line of the magnetic field induction [17]. It means that the particle is involved in two simultaneous motions [17]: under the action of the Lorentz force it is uniformly rotated with the velocity $\upsilon_\perp$, in a circle of the radius $R = m\upsilon_\perp / eB$ where m is the effective mass of electron in a semiconductor crystal, e is the elementary electric charge (electron charge; $\upsilon_\perp$ is a component of the thermal electron velocity that is perpendicular to the magnetic induction vector and moves forward by inertia – uniformly and in a straight line with a constant speed $\upsilon_\parallel$, which is a component of the thermal speed of electron and is parallel to the vector of magnetic induction, and step of helix line is $h = \upsilon_\parallel T$, where $T = 2\pi R / \upsilon_\perp$ is the period of revolution of electron.

The angular velocity of rotation of electron is called a cyclotron (Larmour) frequency. For non-relativistic particles, it is equal to [18, 19]

$$\omega_B = \frac{2\pi}{T} = \frac{eB}{m_n}. \tag{9}$$

Since electron has acceleration when rotating, that is constant in magnitude and directed perpendicularly to the velocity $\upsilon_\perp$, electron is a source of radiation of electromagnetic waves both at the frequency ω_B and at higher harmonics $\omega_B p$, where p is an integer that is greater than unity [20, 21]. Nonrelativistic electrons (for electrons in a semiconductor crystal, the thermal velocity is much less than the speed of light) radiate electromagnetic waves at the fundamental frequency [20, 21].

Furthermore, simplifying the consideration, let's analyze radiation of electron moving in a circle. Nonrelativistic electron radiation power (total power radiated per unit time) at a frequency ω_B is equal to [20, 21]

$$W = \frac{e^2 \omega_B^2 \upsilon_\perp^2}{6\pi\varepsilon_0 \varepsilon c^3}, \tag{10}$$

where ε_0 is the vacuum electric constant, ε is the dielectric constant of the semiconductor crystal.

The radiation power of electromagnetic waves W_n by electrons per unit volume of the semiconductor crystal, in the assumption that all the plurality of electrons $\upsilon_\perp$ has the determined value, is of the form

$$P_n = nP, \tag{11}$$

where n is the concentration of electrons in semiconductor.

However, in the semiconductor crystal, the vectors of the velocity of electrons participating in the thermal motion have various directions relative to magnetic induction vector. In other words, $\upsilon_\perp$ is a random variable, which lies within the range from 0 to υ_T, here υ_T is the mean square thermal velocity of the movement of electrons, which is equal to $\upsilon_T = (3kT/m_n)^{1/2}$, where k is the Boltzmann constant, T – absolute temperature [22]. We assume that this random variable obeys the continuous uniform distribution. Then, the probability density of the random variable is of the form [23]:

$$f(\upsilon_\perp) = \frac{1}{\upsilon_T}. \tag{12}$$

Consequently, for the average value of power of radiation of electromagnetic waves by electrons per unit volume of the semiconductor crystal, we have

$$\overline{W_n} = \int_0^{\upsilon_T} W_n f(\upsilon_\perp) d\upsilon_\perp. \tag{13}$$

Integrating with account of (10), we obtain

$$\overline{W_n} = \frac{ne^2\omega_B^2\upsilon_T^2}{18\pi\varepsilon_0\varepsilon c^3}. \qquad (14)$$

It should be noted that in the semiconductor crystal, electron in motion on a helix line is undergoing the acts of elastic scattering by ions and impurity atoms, dislocations, acoustic and optical phonons [24, 25]. This changes the direction of the velocity of electron motion [24]. As a result of change in the direction of velocity relative the magnetic induction vector, the projections of $\upsilon_\perp$ and $\upsilon_\parallel$ change, too. Respectively, the radius of the circle of rotation *R*, period of revolution of electron *T* and step of helix line *h* change. However, the cyclotron frequency at which the electromagnetic waves radiate remains unchanged. The number of revolutions that electron carries out before scattering, is equal to $\omega_B\tau$, where $\tau = m\mu/e$ is the relaxation time, μ – mobility of electrons in semiconductor [24].

As mentioned above, in the experiments there are used the epitaxial structures *n*-GaAs doped with Te with the concentration of free electrons in the epitaxial film equal $5\cdot10^{22}\,\mathrm{m}^{-3}$, and the treatment of the researched structures was carried out with weak pulsed magnetic field *B* =60 mT. Moreover, the direction of the magnetic induction vector was normal to the sample surface. Taking into account that for GaAs $m = 0.0063\,m_0$, $\mu = 0.85\,\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$ where m_0 is the free electron mass [26], accordingly to (9) and (14) at *T* = 300 K we obtain: $\omega_B = 1.68\cdot10^{11}\,\mathrm{Hz}$ $(\nu_B = 2{,}67\cdot10^{10}\,\mathrm{Hz})$, $\overline{W_n} = 580\,\mathrm{W}\cdot\mathrm{m}^{-3}$, $\omega_B\tau = 0.051$.

It should be noted, and it is important, that when rotating the investigated GaAs epitaxial structures in the field of a permanent magnet, they move through a region of non-uniform (edge) magnetic fields, in which the value of magnetic induction *B* is by orders of magnitude less than 60 mT. However, this area can be divided into separate sections so small that within each section the field will be approximately constant. [18] Accordingly, in the semiconductor crystal the electromagnetic waves with a wide frequency spectrum $\omega_{em} \le \omega_B = 1.68\cdot10^{11}\,\mathrm{Hz}$ will be generated.

Thus, the task of studying the mechanisms of transformation of the defect structure in *n*-GaAs epitaxial films exposed to pulsed MF treatments is reduced to the task of studying the evolution of the defect structure as a result of exposure to electromagnetic radiation of a wide range of microwave frequencies, which, in turn, was analyzed in detail in [27]. In [27] it is shown that the transformation of defect subsystem under the microwave radiation is caused by detachment and movement of dislocations, as well as the decay of the impurity-defect complexes due to resonance phenomena caused by the coincidence of the frequency of an electromagnetic wave with a frequency of own oscillations of dislocations and ion-plasma oscillations of impurity atoms.

The validity of this view is confirmed by the following facts. First, the relation (11) with account (6) predicts a quadratic dependence of the observed effects on the value of magnetic induction, which was noted in [1, 2]. Second, it is availability of a wide range of oscillation frequencies of electromagnetic waves, as shown in [27], induced by a pulsed magnetic field in this specific case, which is crucial in the evolution of the defect structure of semiconductors.

It is important to treat particularly the pulsed magnetic field, as it was pointed out in the work [28]. Third, since the sensitivity of the defects to the electromagnetic radiation is determined by its frequency as well by the power, *i.e.*, the amplitude of electromagnetic wave electric component [27], which, in its turn, depends on frequency in accordance with (11), then the expression (6) predicts threshold with *B* nature of the induced by magnetic field effects, which was pointed out in [28]. Fourth, taking into account that with *T* decreasing *n* decreases and $\upsilon_\perp$ falls, and respectively $\overline{W_n}$ significantly reduces, it can be expected that at low temperatures the corresponding magnetic-field effects will be also very slightly pronounced or generally absent. This fact was pointed out in [28]. Finally, fifth, if to assume that at the frequencies of electromagnetic radiation above a certain limit frequency, defects become insensitive to it, then as follows from (6), with *B* increasing the saturation effect in changing of the magnetic sensitive parameters must be observed. This fact was marked in the work [28].

The notion of equivalence of action of pulsed magnetic-field treatments and electromagnetic radiation of a wide range of microwave frequencies agrees well with the results of the work [29], in which, in part, there presented the experimental studies of changes in the lattice parameter and structural perfection of the near-surface layer of A^3B^5 semiconductor compounds after action of the pulses of the magnetic field with the amplitude of 0.1 T and duration of 20 s. The observed effects are indicated on generation, transformation and destruction of clusters of Frenkel defects with abnormally low formation energy. And these changes increase in the row GaAs – InAs – InSb. This fact can be attributed to that in the marked row, the effective masses of free charge carriers of both electrons 0.063 – 0.023 – 0.014, and light holes 0.082 – 0.026 - 0.015 decrease [26]. Accordingly, with regard to (6) and (11), we observe the growth of both the frequency ω_B, and thus the range of the frequency spectrum expands, and the power of electromagnetic radiation. Both of these circumstances, according to [27], lead to increasing the changes in the structural perfection of the near-surface layer of semiconductor compounds after exposure to magnetic field pulses.

4. Conclusion

Thus, the probability-physical approach allows to explain the observed long-term changes in the intensity of photoluminescence bands of gallium arsenide as a result of magnetic-field treatments. In turn, the evolution of subsystem of defects is caused by exposure of it to microwave radiation induced by a magnetic field in the semiconductor crystal.

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