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The influence of acoustic-dislocation interaction on intensity of the bound exciton recombination in initial and irradiated GaAsP LEDs structures

O.V. Konoreva [a, *], Ya. M. Olikh [b], M.B. Pinkovska [a], O.I. Radkevych [c], V.P. Tartachnyk [a], V.V. Shlapatska [d]

[a] Institute for Nuclear Research, NAS of Ukraine, 47, Pr. Nauky, 03028 Kyiv, Ukraine
[b] V. Lashkaryov Institute of Semiconductor Physics, NAS of Ukraine, 41, Pr. Nauky, 03028 Kyiv, Ukraine
[c] SE "RI of Microdevices" STC "Institute for Single Cristals" NAS of Ukraine, 3, Pivnichno-Syretska Str., 04136 Kyiv, Ukraine
[d] L.V. Pisarzhevski Physical Chemistry Institute NAS of Ukraine, 31, Pr. Nauky, 03028 Kyiv, Ukraine

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ABSTRACT

Acoustic-excitant interaction of GaAsP light emitting diodes (initial and irradiated by 2 MeV electrons) was studied. Structure based on $GaAs_{1-x}P_x$ solid solutions, grown by epitaxy from the vapor phase, were the object of the research. It was observed that ultrasonic treatment (UST) results in the drop of the emitting intensity of structures, which relaxes to the previous values after ultrasound termination. The possible reason of observed changes concerning nonequilibrium dislocation clusters were discussed. Electron irradiation leads to the exponential drop of emitting intensity, which restores after UST much slower than initial one. Radiation degradation parameters τ_0/K_τ of yellow and orange LEDs were found.

1. Introduction

Light emitting diodes (LEDs) based on gallium arsenide-phosphide solid solutions have several advantages over traditional GaP *p-n*-structure, such as high quantum efficiency and the possibility of a smooth change of the spectral color - from greenish to red - by adjusting phosphorus and arsenic concentration. On the other hand in GaAsP solution, as well as in binary GaP, an exciton bound at nitrogen isoelectronic impurity is formed, and its recombination ensures the existence of emission with the high quantum yield. Thus the samples which possess the exciton emission as the main component of the spectrum are useful to study an interaction between ultrasound wave and exciton luminescence centers. In turn, due to the high quantum efficiency it is possible to record more accurately changes caused by ultrasound. That is why GaAsP LEDs are useful object of the research. Note also the higher sensitivity of this recombination mechanism to crystal defects, including radiation ones [1–3]. Irradiation of crystals by quick particles makes it possible to change purposefully the physical characteristics of both materials and devices, to create in their volume nano-size regions with special physical and chemical properties, to build barriers of required profile and so on [4].

* Corresponding author.
E-mail address: okskon@meta.ua (O.V. Konoreva).

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Ultrasound (US) as well can change dramatically the parameters of materials and devices [5–8]. Passing through the crystal an ultrasound wave causes oscillatory processes within different types of dislocations that are ideal sinks for moving atoms [7,8].

Moving deformation fields serve as unlimited capacity absorbers for free excitons. Concerning bound excitons, recombination of which provides the operating of optoelectronic emitters, their interaction with dislocations, moving by the ultrasonic wave, is not currently clear. Therefore, the main purpose of the given work is to clarify the ultrasound influence on the bound exciton luminescence and to compare the effect of two different factors - acoustic and radiation. Appropriating data of degradation parameters of irradiated diodes are also given.

2. The samples and experiment

LEDs, based on solid solutions $GaAs_{1-x}P_x$ ($x = 0.45$) were made by the standard epitaxial technology from the vapor phase. Diodes were irradiated by 2 MeV electrons at electron accelerator ILU-6 at room temperature in a pulsed mode. Electroluminescence spectra were measured at temperatures 77–300 K with the simultaneous ultrasonic loading (frequency $\nu \approx 1$ MHz and power $W \approx 1$ Wt/cm^2) and in the absence of ultrasonic. An automated measuring complex was constructed on the basis of monochromator MDR-23, which made it possible carrying out an experiment at wide temperature interval and different levels of injection.

3. Result and discussion

3.1. Ultrasonic treatment

Fig. 1 shows electroluminescence spectra of orange $GaAs_{1-x}P_x$ ($x = 0.45$) LED at different temperatures. The dependence of luminescence maximum on temperature is separately given in the inset. High luminescence efficiency of such structures is provided by the nitrogen isoelectronic acceptor doping, and also by "the effect of band structure" [9].

In GaAsP crystals, as in gallium phosphide, nitrogen atom is a short-acting center and by capturing an electron this center localizes strongly its wave function. Reducing of the spatial integral Δx is accompanied by the increased uncertainty of the pulse ΔP and delocalization of the corresponding wave function in the k-space, which removes partially the prohibition on ΔP. The next step after joining the electron is a hole capture by the charged center and appearing of a bound exciton on the nitrogen atom; and exciton recombination generates the near-edge emission intense enough even at room temperature.

The level of nitrogen is bound to the X-bottom of the conduction band, but when due to changes of the solution composition the crystal has direct zones (when X and Γ - minima are equalized at $x = 0.45$), the probability of the emitting intensity increases sharply. Brightness of GaAsP LEDs is limited by the nitrogen solubility ($N_N \approx 10^{20}$ cm^{-3}). An exceeding of this value is accompanied by increased density of dislocations. Dislocations' germination of substrate with a depth of 30–50 μm impacts also negatively on the luminescence intensity [9].

Ultrasonic wave causes the dislocations' oscillation that could end their separation from stoppers and promote moving to other ones, the forming of dislocation networks, the diffusion restructuring of point defects and the emerging of new defects [7–10].

Fig. 2 shows the dependence of maximal emitting intensity of the orange $GaAs_{0.55}P_{0.45}$ diode on the time of UST at 77 K in the current mode ($I = 40$ mA); ultrasonic frequency is ≈ 1 MHz, $W \approx 1$ Wt/cm^2. It is evident that ultrasound leads to a monotonous falling of emitting intensity, which can be approximated by the exponential dependence

$$I = I_0 e^{-t/\tau_1}, \tag{1}$$

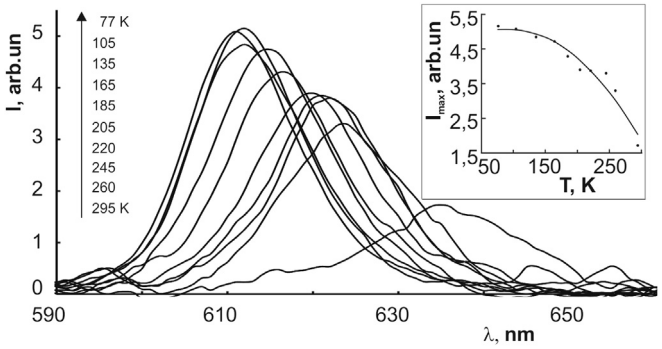

Fig. 1. Electroluminescence spectra of orange $GaAs_{0.55}P_{0.45}$ LED at different temperatures. The dependence of luminescence maximum on temperature is given in the inset.

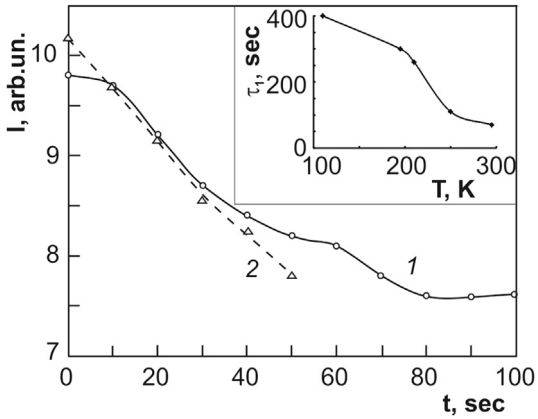

Fig. 2. Dependence of emitting intensity of initial $GaAs_{0.55}P_{0.45}$ LED on time of UST at 77 K (1); calculated value ($I_0 = 10.2$, $\tau_1 = 189$ s) (2). $\tau_1(T)$ for LED with high luminescence intensity is in the inset.

where I_0 is an initial luminescence intensity, t is UST time. τ_1 might be interpreted as an "effective lifetime", necessary for system to reach an equilibrium under external excitation; the loss of ultrasound energy at the main defects is saturated and the absorbed energy is equal to scattered one. τ_1 value for different temperatures is shown in the inset in Fig. 2. Temperature decrease of this parameter can be caused by the increasing mobility of dislocations [11].

The mechanism of interaction of mobile dislocation with exciton captured by the isoelectronic center can be considered similar to its interaction with an impurity atom [12]. When the dislocation is close to the center, the electron is captured by its core, leading to exciton destruction. Electron transfer into C-zone is a combined process containing thermoactivating and tunnel components.

As shown in Fig. 2, an equilibrium during UST is established for ~90 s, time which is characteristic of large-scale defects, dislocations in our case, unlike to point defects with small relaxation times.

The authors [7] believe that for used UST mode the dislocation average deviation from the equilibrium is close to 100 Å. So, if not even consider its climb due to ultrasound, oscillations of such large defects could lead to the decay of bound excitons in relatively significant part of the crystal.

After termination of ultrasound the effect of luminescence intensity recovery is observed (Fig. 3) in accordance with the equation $I = I_{\max}(1 - e^{-t/\tau_2})$, caused by the reverse transition of nonequilibrium electrons from the conduction band to isoelectronic centers (nitrogen atoms).

Relaxation time is also long, apparently due to the partial carrier capture by shallow traps and immobile dislocations or dislocation grids ("dark spots defects"). Time growth (τ_2) with the temperature of the sample might be caused by the influence of a competing effect - thermal ionization of excitons.

Comparing the change in the luminescence intensity of diode due to US and of the temperature dependence of emitting maximum for the initial sample (inset in the Fig. 1), one can estimate the temperature effect caused by ultrasound. For example, if the ultrasound is performed at 77 K, the luminescence intensity of the diode decreases by 1.27 times (Fig. 2); while it is heated to ~180 K.

Thus, the effect of ultrasound on the exciton luminescence can be regarded as the result of the influence of two factors: the destruction of the excitons by moving dislocations and heat, which contributes to bound exciton ionization.

It is clear that each of them affects on their own, but the existence of long-term relaxation process testifies about the overwhelming influence of dislocation clusters - large-scale structural defects, which have prolonged effects of recovery.

3.2. Irradiation effects

Electron irradiation leads to the exponential drop in radiation intensity of LED (Fig. 4):

$$L = L_0 e^{-K\Phi}, \tag{2}$$

where L_0 is initial luminescence intensity of diode; K is a coefficient that determines the relative rate of change of luminescence intensity.

The current density through p-n-transition is inversely proportional to the minority carrier lifetime as $\tau^{1/2}$ and luminescence intensity is proportional to τ [9];

$$L \sim \tau^{-1/2} \exp\left(\frac{qU}{\beta kT}\right), \tag{3}$$

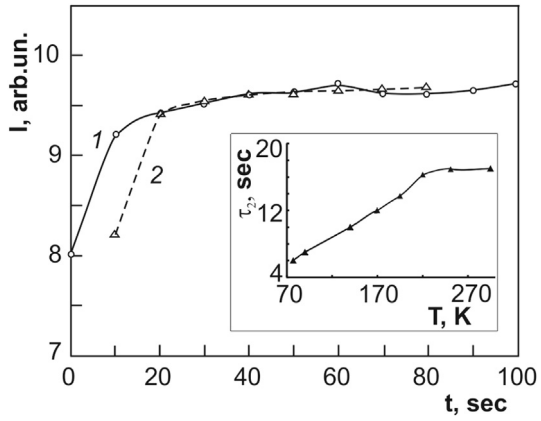

Fig. 3. Dependence of emitting intensity of initial $GaAs_{0.55}P_{0.45}$ LED on time after UST at 77 K (1); calculated value ($I_0 = 9.6$, $\tau_2 = 5.18$ s) (2). $\tau_2(T)$ for LED with high luminescence intensity is in the inset.

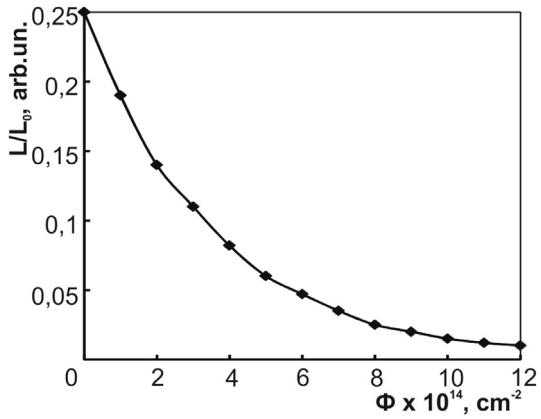

Fig. 4. Dose dependence of emitting intensity of $GaAs_{0.55}P_{0.45}$ LED ($E = 2$ MeV).

$$L \sim \tau \exp\left(\frac{qU}{\beta kT}\right). \tag{4}$$

Therefore, while combining two expressions and considering that $\beta = 1$, we get $L \sim \tau^{3/2} I$.

The radiation lifetime coefficient K_τ is obtain from the equation $\frac{1}{\tau} = \frac{1}{\tau_0} + \frac{D}{K_\tau}$ - where τ_0 is the lifetime of minority carriers of initial sample, D is received dose.

$$\left(\frac{L_0}{L}\right)^{2/3} - 1 = \left(\frac{\tau_0}{K_\tau}\right)\Phi. \tag{5}$$

Than [13].

$$\left(\frac{\tau_0}{K_\tau}\right) = \frac{1}{D}\left(\left(\frac{L_0}{L}\right)^{2/3} - 1\right). \tag{6}$$

In order to obtain D in rad it is necessary to know the path of 2 MeV electrons in GaAsP. Since R (being expressed in g/cm^2) depends on the substance electron density, nearly the same for all elements of the periodic system, one can use the expression for aluminum.

$$R_{Al} = 0.542 \cdot E\,(\text{MeV}) - 0.133 = 0.951\ \text{g/cm}^2$$

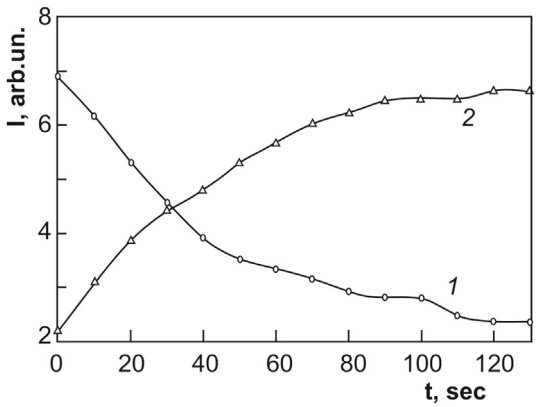

Fig. 5. Dependence of emitting intensity of irradiated $GaAs_{0.55}P_{0.45}$ LED at 220 K on time of UST (1); after UST (2).

The GaAsP density can be estimated as an average between phosphide and gallium arsenide ($\rho_{GaAsP} = 4.72$ g/cm^3). By divided the previous expression on it, we receive $R_{GaAsP} = 0.2$ cm.

For known electron energy and fluence and also the absorption layer thickness R (cm), the dose received in rad is $D_{GaAsP} = 4 \cdot 10^7$ rad.

From the dose dependences of the luminescence intensity of yellow and orange we receive parameters that characterize the rate of radiation degradation of exciton emission:

$$\beta_{1_{yellow}} = \tau_0/K_\tau = 1.89\cdot10^{-7}\text{rad}^{-1} \text{ and } \beta_{2_{orange}} = \tau_0/K_\tau = 2.7\cdot10^{-8}\text{rad}^{-1}$$

Degradation-restoring processes, caused by ultrasound, proceed much more slowly in irradiated samples compared to the initial one (Fig. 5). For example, if in the initial GaAsP sample emission intensity after UST ($T = 220$ K) is reduced by about half, for irradiated sample – arsenic is close to three.

In general, the relationship $I = I(t)$ is still exponential. The deepening of degradation-restoring processes in irradiated diodes may be caused by the existence of complex radiation defects that act as dislocation stoppers which are the extra embryos of dislocation networks during prolonged UST.

4. Conclusions

Ultrasound treatment results in reducing of the emitting intensity of GaAsP LEDs. Long-term brightness recovery occurs due to the impact of large non-equilibrium dislocation clusters. Electron irradiation increases the number of dislocation stoppers, and, accordingly, the density of dislocation networks, deepening the amplitude of degradation-relaxation processes.

Parameters of radiation damage to yellow (β_1) and orange (β_2) diodes are received

$$\beta_1 = \tau_0/K_\tau = 1.89\cdot10^{-7}\text{rad}^{-1} \text{ and } \beta_2 = \tau_0/K_\tau = 2.7\cdot10^{-8}\text{rad}^{-1}.$$

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