

## Electrophysical properties of $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$ epitaxial films irradiated by $\gamma$ -quanta

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**Abstract:** Herein, the effect of  $\gamma$ -quanta on electrophysical and photoelectric properties of p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films obtained from the molecular cluster on the glass substrate by the method of condensation has been investigated. It has been established that the acceptor-type local levels with the ionization energy of 0.14 eV and 0.175 eV are generated, when p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  ( $x = 0.01$ ) epitaxial films are irradiated by  $\gamma$ -quanta at  $D > 10$  kGy doses. The increase in the photoconductivity in the low temperature range 80-180K is due to the discharge of 0.14eV level, but the decrease in the rate of change of photocurrent in the high temperature range is due to the role of local level with 0.175 eV energy as a recombination center.

**Key words:** Epitaxial film, electroconductivity, photoconductivity, volt-ampere characteristics (VAC),  $\gamma$ -quanta

### 1. Introduction

As the development directions of modern micro- and optoelectronics focus on the development of highly efficient multi-component semiconductor structures, it is of particular importance to obtain large-area and homogeneous materials [1–3]. For many years, epitaxial films of  $A^IV B^VI$  - type semiconductor compounds ( $\text{PbS}$ ,  $\text{PbSe}$ ) and their solidsolutions have been widely using in infrared (IR) technology [4]. As their band gap width corresponds to the energy of IR irradiation quanta, these materials play an important role in the development of various optoelectronic devices operating in infrared region [5,6]. From this point of view, the development of elements, structures that are sensitive to infrared rays on the basis of these materials and their use in the different fields of optoelectronics remain as an important issue [7]. The solid solutions and epitaxial films obtained on their basis are widely used in various devices and instruments operating in the 3–5  $\mu\text{m}$  and 8–14  $\mu\text{m}$  wavelength ranges of the spectrum in the IR technology. The presence of manganese (Mn) ions in these solid solutions reveals the existence of new properties of semimagnetic semiconductors and creates wide opportunities for the preparation of magnetic field-controlled diodes operating in the 3–5  $\mu\text{m}$  wavelength of the spectrum based on them [8,9]. Thus,  $E_g$  grows sharply in the solid solutions formed by the inclusion of Mn ions into the crystal lattice of lead chalcogenides [10]. On the other hand, the energy spectrum of charge carriers in the magnetic field changes unusually. Therefore, it is possible to prepare the photoacceptors controlled by the magnetic field and temperature, on the basis of such solid solutions, which has a great scientific and practical importance.

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It is of particular importance to use the method of precipitation from molecular cluster, while obtaining multilayered structures on the basis of narrow-band semiconductor materials. This method ensures the homogeneity of the layer and allows obtaining any profile surface. This method is also used widely in obtaining epitaxial films of  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  solid solutions [11,12].

There is some information on obtaining  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films, and the study of their electrophysical properties in the scientific literature. They are mainly devoted to the improvement of the obtain technology of films. As it is known, initial defects play the main role in the control of physical properties in such compound films, and it is possible to control the physical properties by changing the concentration of these defects. There is no information on the control of their concentration by doping and external effects (thermal annealing, the effect of radiation rays) in the scientific literature.

It is known that shallow and deep energy levels are formed in the structure of semiconductor monocrystals as a result of radiation rays. These levels act as a recombination center for charge carriers, and as a result, they decrease their life term, as well as, efficiency and running time of the photodevices formed on their basis. Therefore, the study of radiation effects in the  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films due to the effects of radiation rays may play an important role in determining the ways of increasing the radiation resistance of semiconducting materials, including the devices formed on their basis [13,14]. The certain studies have been conducted in this direction, but the obtained results have not adequately reflected the information on the mechanism of interaction between the defects, initial and formed within radiation and on the nature of the defects [15].

Herein, both initial (before irradiation) and  $\gamma$ -irradiated electrical and photoelectric properties of epitaxial films of  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  compound obtained on the glass substrate have been studied at various temperatures and electrical fields.

## 2. Experimental part

Epitaxial films of  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  solid solutions have been obtained by molecular beam condensation (MBC) method in UVN-71P3 vacuum assembly,  $10^{-4}\text{Pa}$  vacuum, ( $T_{subs.} = 663\text{-}673\text{K}$ ) values of temperature of substrates and  $8\div 9\text{\AA}/\text{s}$  values of condensation rate [12]. As a source, it has been used different chemical  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  solid solutions which are primarily synthesized, and as a source of evaporation, the Knudsen-type hole prepared from graphite. The thickness of the films was  $3\mu\text{m}$ . Silver paste has been used as a contact material. The distance between the contacts was 0.6 mm. Silver contacts dried at room temperature for 24 h after applying. It has been determined by the volt-ampere characteristics (VAC) that the contacts are Ohmic. The samples have been irradiated by  $\gamma$ -rays which are irradiated by  $^{60}\text{Co}$  isotope source at 290 K. The energy of  $\gamma$ -rays was 1.17-1.33 MeV.

The value of the lattice constant calculated from the electronogram and the x-ray curve of the obtained films was equal to  $a = 6.11\text{\AA}$ , and the value of crystal excellence calculated from the half-width of the x-ray diffraction curve was in the range of  $\Delta 2\theta = 90\div 100''$ .

The measurement of photoconductivity of the studied samples was carried out in a temperature range of 300K, at the device mounted on the basis of "MS-3504i" monochromator.

## 3. Discussion of experimental section and conclusions

The electroconductivity of thin films of p and n-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  ( $x = 0.01$ ) obtained on  $\text{BaF}_2$  substrates has been investigated in the temperature range of  $T = 80\text{-}450\text{K}$  [16]. The concentrations of charge carriers in

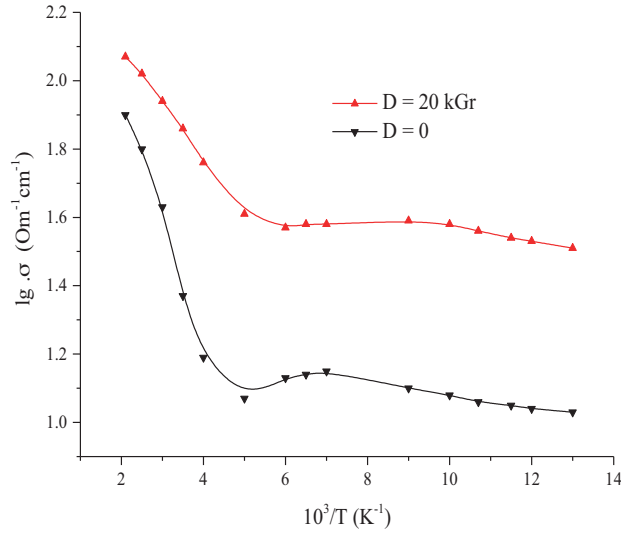
the investigated samples have been determined by measuring the Hall coefficient by the compensation method: the concentration of free electrons in the n-type sample was  $1.1 \cdot 10^{17} \text{ cm}^{-3}$  and in the p-type sample it was  $2.3 \cdot 10^{17} \text{ cm}^{-3}$ . Samples have been irradiated at the same dose under the same conditions, and using the same source- $^{60}\text{Co}$  isotope source during the investigation.

The initial physical properties of the studied p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial film have been studied. The physical properties of the samples have been investigated again after irradiating them with  $\gamma$ -rays of  $^{60}\text{Co}$  isotope at different doses. Figure 1 shows the temperature dependence graphs of specific conductance of p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  ( $x = 0.01$ ) epitaxial films, initial (before irradiation) and after the irradiation with  $\gamma$ -rays ( $D = 20 \text{ kGy}$ ). It is seen from the graphs that the specific electrical conductance increases in the initial samples with p-type conductivity by the increase of temperature in the range of 80-142K, it decreases at the range of 142-200K and increases again at the range of 200-450K (Figure 1). The reason for the decrease in specific electrical conductance in this temperature range is a scattering of charge carriers from thermal motion of ions located in the nodes of crystal lattice. After irradiating with  $\gamma$ -rays, the specific electrical conductance slowly increases compared to the initial sample within the range of 80-140 K, practically does not change in the range of 140-192 K, and increases again in the range of 193-450 K (with a relatively low rate compared to the initial sample).

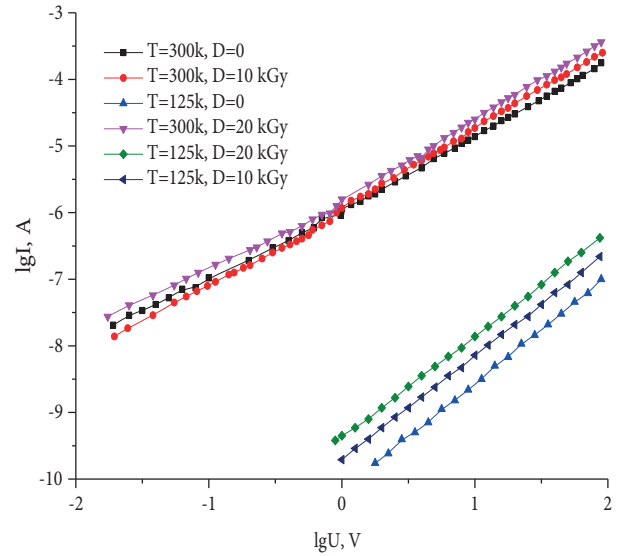
The change in a concentration of free charge carriers and specific electrical conductance in p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films depending on the absorption dose of  $\gamma$ -rays shows that the defects formed in these samples due to the effect of  $\gamma$ -rays are acceptor-types. Therefore, the concentration of charge carriers and accordingly the specific electrical conductance increase in the samples with p-type conductivity, but the concentration of charge carriers and accordingly the specific electrical conductance decrease in the sample with n-type conductivity. It can be said that the generated defects consist of vacancies of Se atoms that replaced their place to intermodal distance as a result of radiation [17–19]. The vacancy of Se atoms leads to the formation of a positive charge state in these compounds. The minimal energy (threshold energy) required to transfer the atoms from the crystal lattice node to the internodal phase was calculated by the Bayerline formula [18]. The increase in conductivity in the p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  samples can be due to the formation of Se vacancies after  $\gamma$ -quanta radiation. As it is known, the formation of halogen vacancies in such compounds leads to an increase in the positive charge state. Therefore, the minimal energy for the transfer of Se atoms to internodal phase in  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  compound has been calculated. The value of this energy (threshold energy) was 14.2 eV and 15.1 eV. As it turns out, the energy given by electrons to the Se atom is enough to pass to the internodal phase, and the point defects consist mainly of Se vacancies.

Thus, it has been determined by the research that although the nature of acceptor-type radiation defects generated while the irradiation of both n-type and p-type  $\text{Pb}_{0.99}\text{Mn}_{0.01}\text{Se}$  samples by the  $\gamma$ -quanta is the same, they can play both the acceptor and donor role for charge carriers. Therefore, the acceptor-type point radiation defects in the p-type samples increase the electrical conductivity, but the electrical conductivity decreases accordingly due to compensation as a result of irradiation in the n-type samples [16, 20].

Herein, the VACs of the initial and irradiated samples in the dark and light and the dependences of electrical conductivity on the temperature have been investigated in order to study the effect of  $\gamma$ -radiation on electrical and photoelectric properties of  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films (Figure 2). It is known that the form of the volt-ampere characteristics (VAC) of semiconductor materials depends on many factors, particularly on the distribution of local levels in the band gap, temperature, etc. and sometimes it has a very complex structure.



**Figure 1.** Temperature dependence of electric conductivity in p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films.



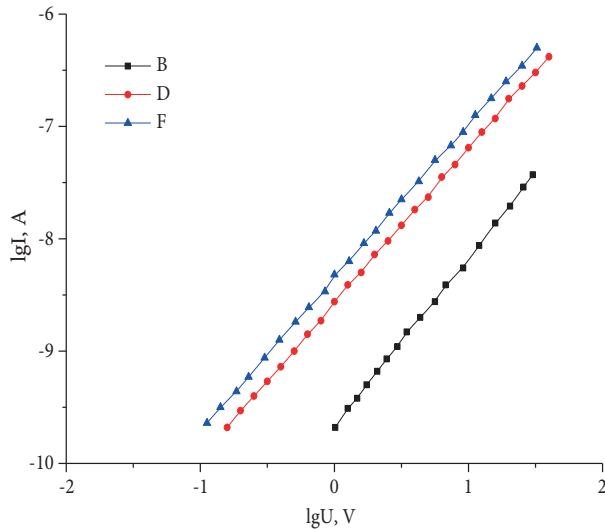
**Figure 2.** Volt-ampere characteristics (VAC) of initial (black and blue) and irradiated by  $\gamma$ -rays (red, purple, pink and yellow)  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films in the dark.

It is clear from the curves obtained at the temperature of 300K that the change in the conductivity in the lower values of intensity occurs mainly due to the thermal ionization of the level. Here,  $n$  is approximately close to one in small rectilinear first part (red, black and purple). This proves that the change in the current in the low intensity region obeys to the Ohmic law. As  $n > 1$  in the next part of the curves (red, black and purple), there is observed deviations from the Ohmic law in the dependence of current on the field. It is known from the Lampert's theory that the observed deviations are due to the local levels with different concentration [21]. Here, the ionization of the levels, a thermal effect may occur, thereby the value of concentration of charge carriers and accordingly the current increases. Only a slight change in the level is seen here. The value of current changes slightly at different temperatures depending on the field. As it is seen in blue curve, current flow mechanism is explained by the formation of charge carriers generated due to injected and thermal ionization. It is seen from the graph that there is an acceptor-type energetic level with the ionization energy of  $\approx 0.08$  eV at low temperatures ( $T < 125\text{K}$ ), and as it has the same nature with the defect formed due to irradiation, only its concentration changes. And this leads to an increase in current, as seen in red and blue curves in Figure 3. In order to reveal the influence of the observed level on conductivity, the sample is illuminated by a white light (Figure 3).

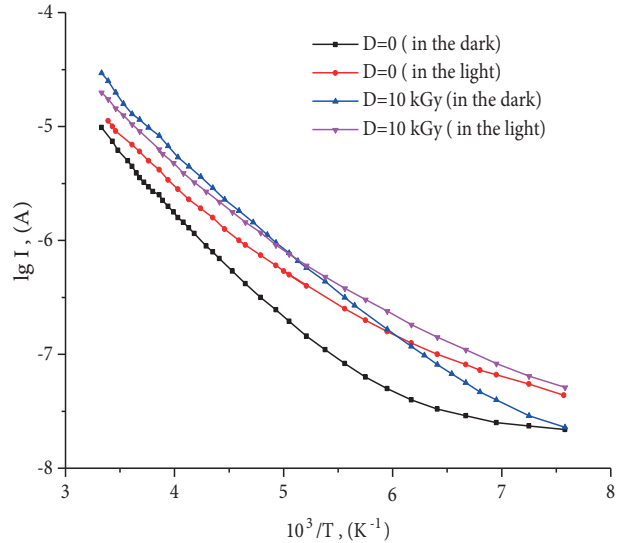
It has been established that the observed level is sensitive to light, and therefore the sample has photosensitivity in the low temperature range. As there isn't observed any level in the high temperature range ( $T > 125\text{K}$ ), the samples do not have photosensitivity. Thus, we can say on the basis of experimental results that there is observed photosensitivity due to the formation of acceptor-type shallow level in  $\text{Pb}_{0.99}\text{Mn}_{0.01}\text{Se}$  epitaxial films as a result of  $\gamma$ -quanta influence.

Figure 4 shows the dependence of the current intensity of the initial and irradiated samples on the temperature (dependence of  $\lg I \sim \frac{10^3}{T}$ ) in the dark and light at the intensity of 10V, but Figure 5 shows at the

intensity of 40V. It is seen from the graphs that the electrical conductivity in dark increases after irradiation compared to initial state at all intensities in the temperature range of 125-300K. The reason for lower value of current intensity in the light than the value of current intensity in the dark after a certain value of temperature in the irradiated sample can be explained as follows.



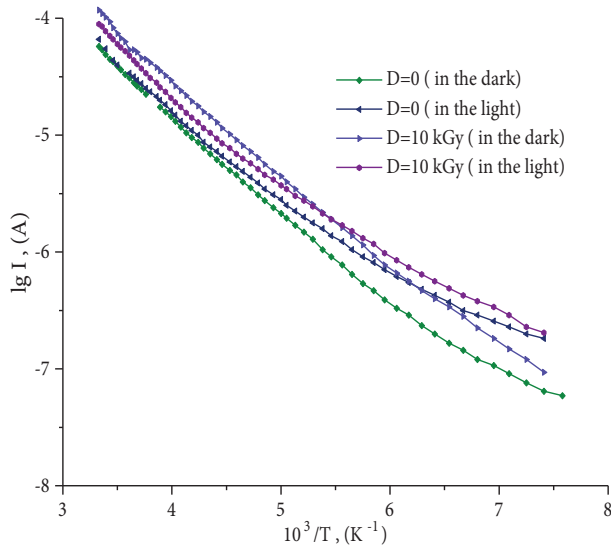
**Figure 3.** Volt-ampere characteristics (VAC) of initial (black) and irradiated by  $\gamma$ -rays (red and blue)  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films in the light.



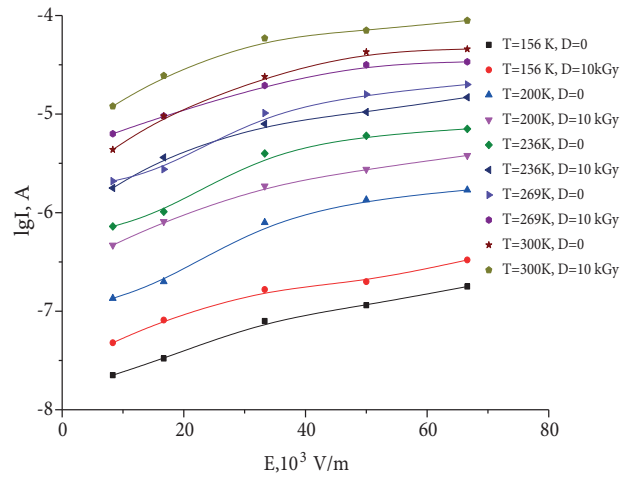
**Figure 4.** The dependence of the current intensity of the initial and irradiated  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films on the temperature (dependence of  $\lg I \sim \frac{10^3}{T}$ ) in the dark and light at the intensity of 10V.

The local levels are generated in the band gap of the  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial film due to the irradiation, so that these levels reduce the concentration of the equilibrium charge carriers by the effects of temperature and light. There is a local level (which means level I) between the Fermi level and permeable zone (located close to the Fermi level), so that electrons pass from the valence zone into this zone by the influence of light. The probability of electrons passing to the permeable zone from this zone through the thermal ionization is very low. Below the Fermi level, there is a local level closer to the valence zone, (which means level II) where the holes are captured by this level. The holes captured by the level II recombine the free equilibrium electrons in the permeable zone, and as a result, reduce concentrations of electrons in the dark. Therefore, the concentration of electrons in the dark decreases under the influence of light, and as a result, the current becomes less than in the dark. In this case, the concentration of holes also increases, but this increase cannot compensate the concentration of electrons [21,22].

In the Figure 6, it has been given the dependence of the logarithmic value of the current intensity on the electrical field intensity at different temperatures for the  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial film, initial and irradiated with  $\gamma$ -rays at 10 kGy dose. It is seen here that a logarithmic value of current intensity has increased at the given temperatures after irradiation with  $\gamma$ -rays, but there was no significant difference in the form of the dependence of the electric field intensity. Herein, may be occurs ionization of levels and thermal effect, which due to this, the value of concentration increases and also occurs the increase of current intensity. This is also explained by the fact that, the mechanism of current conduction occurs not at the expense of the field but at the expense of thermal ionization.



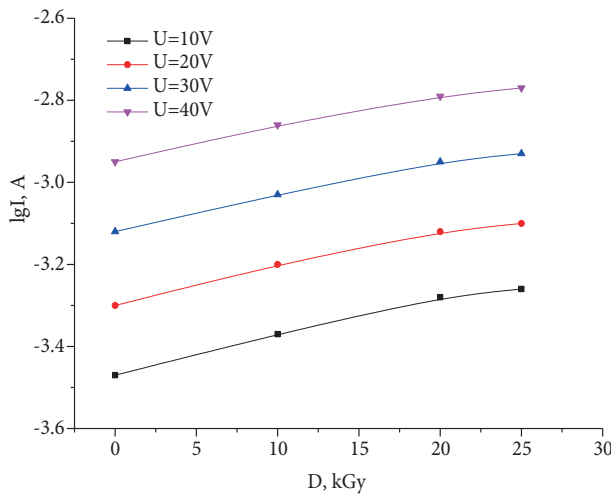
**Figure 5.** The dependence of the current intensity of the initial and irradiated  $Pb_{1-x}Mn_xSe$  epitaxial films on the temperature (dependence of  $lgI \sim \frac{10^3}{T}$ ) in the dark and light at the intensity of 40V.



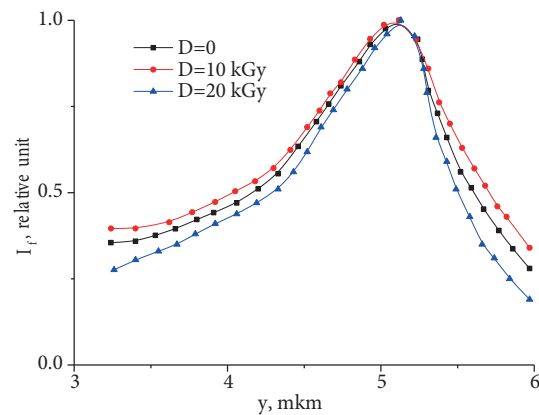
**Figure 6.** The dependence of the logarithmic value of the current intensity on the electrical field intensity at different temperatures of  $Pb_{1-x}Mn_xSe$  epitaxial films.

In the Figure 7, it has been given the absorption dose dependence of current intensity at 300K temperature for the  $Pb_{1-x}Mn_xSe$  (p-type,  $x = 0.01$ ) epitaxial film irradiated with  $\gamma$ -rays. As it is seen, the conductivity changes less by the increase in adsorption dose.

In the Figure 8, it has been given the spectral distribution of photoconductivity for the  $Pb_{0.99}Mn_{0.01}Se$  epitaxial film, initial and irradiated with gamma rays at 10 kGy and 20 kGy doses.



**Figure 7.** Absorption dose dependence of current intensity at 300K temperature for the  $Pb_{1-x}Mn_xSe$  epitaxial film.



**Figure 8.** Spectral distribution of photoconductivity for the  $Pb_{0.99}Mn_{0.01}Se$  epitaxial film.

As it is seen, the maximum of photocurrent practically does not change its place in the spectrum when irradiating the sample with gamma rays at 10 kGy dose, and it remains  $\lambda_{max} = 5 \mu m$  as a initial sample.

But the value of the photocurrent increases in the short- and long-wave region of the spectrum. Even after the irradiation at 20 kGy dose, the maximum of the photocurrent does not change its place practically. The value of photocurrent decreases relative to the value of  $\lambda_{max} = 5 \mu\text{m}$  in the short- and long-wave regions of the spectrum. The decrease in the spectrum width by the increase in radiation dose is likely due to the defects formed on the surface of the  $\text{Pb}_{0,99}\text{Mn}_{0,01}\text{Se}$  epitaxial film and lateral surface areas and they lead to a decrease in photocurrent in the short and long-wave regions of the spectrum.

#### 4. Conclusions

Summarizing to the obtained results, it can be noted that the electrical conductivity of the p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial films which irradiated with gamma rays at a temperature of 300K varies slightly in comparison with the electrical conductivity of initial samples. At this temperature they can be considered as materials resistant to their gamma-radiation.

It has been established that the defects formed as a result of  $\gamma$ -quanta radiation is simple Frenkel defects, which consists mainly of Se atoms that have passed into the internodal phase and their vacancies. After radiation, the physical properties of epitaxial films of  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  at room temperature change slightly, but the difference at low temperatures is relatively large. This is due to the fact that the  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  epitaxial layer has a narrowband semiconductor.

Thus, it has been established that the acceptor-type local levels with the ionization energy of 0.14 eV and 0.175 eV are generated, when p-type  $\text{Pb}_{1-x}\text{Mn}_x\text{Se}$  ( $x=0.01$ ) epitaxial films are irradiated by  $\gamma$ -quanta at  $D > 10$  kGy doses. The increase in the photoconductivity in the low temperature range (80-180K) is due to the discharge of 0.14eV level, but the decrease in the rate of change of photocurrent in the high temperature range is due to the role of local level with 0.175 eV energy as a recombination center.

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