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The relaxation of electrophysical properties HgCdTe epitaxial films affected by plasma of high frequency nanosecond volume discharge in atmospheric-pressure air

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ABSTRACT

In this work the results of the experimental investigation of the influence of the high-frequency nanosecond volume discharge in atmospheric pressure air on the electrophysical properties of epitaxial HgCdTe films are presented. Analysis of magnetic-field dependences of the Hall coefficient have shown that in the near-surface region of the material a high-conductivity n-type layer is formed as a result of irradiation. It is shown that relaxation of the values of the electrophysical parameters of the epitaxial films is observed after the irradiation. The supposition is made that the obtained experimental results may be explained by the formation of thin dielectric oxide film at the surface of the irradiated material, containing built-in fixed and mobile positive charge.

1. Introduction

To date, devices operating in the infrared (IR) spectral range are exploited in various spheres of human endeavor. Besides this, there are higher and higher requirements to the detectivity of IR photodetectors. Only photodetectors based on narrow-gap $Hg_{1-x}Cd_xTe$ (HgCdTe, MCT) semiconducting solid solutions can meet these requirements [1]. Thus, the detectivity of existing bolometers and pyroelectric detectors usually does not exceed 10^8 cm·Hz$^{1/2}$ W^{-1}, while the detectivity of even uncooled non-optimized near-IR detectors based on HgCdTe is higher than 10^{10} cm·Hz$^{1/2}$ W^{-1} (maximum values of the detectivity that are reached with cooling are even higher). That is why this material is the basic for creation of highly effective IR photodetectors for the spectral range of 1–14 μm. In the opinion of specialists this unique status of MCT will remain at least two decades. At present, heteroepitaxial MCT films with barrier wide-gap layers grown by the method of molecular beam epitaxy (MBE) are the most promising material for creation of focal plane array IR photodetectors [1]. During creation of highly effective multiple-element photodetectors it is necessary to achieve high level of homogeneity of properties of photosensitive elements of focal plane arrays. The most crucial for homogeneity of operating characteristics of the elements is the distribution of electronic properties over the surface and over the near-surface region of the epitaxial film, which leads to significant changes in their electrophysical characteristics. Despite great success in the technology of creation of the photodetectors based on HgCdTe, search for new and effective methods of controlled management of electronic properties of initial material is still an important task.

Discharges of various types and electron beams are widely used for modification of near-surface layers of different materials. For HgCdTe materials, the mentioned influence is widely applied in the methods of “dry etching”, such as reactive ion etching, inductively coupled plasma (ICP), etching ion-beam milling and others [2,3]. These methods are based on the treatment of the material's surface by low-energy (0.2–2 keV) ions of discharge plasma in noble gases (mostly, Ar) or in the mixture of Ar with chemically active gases (H_2, CH_4). Initially, such treatment was used to create specific surface topology in the technology of photodetectors [3]. In 1981 in the patent [4] it was shown that etching ion-beam milling results in conversion of the conduction type of the initial materials during processing of HgCdTe with p-type of

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conductivity [5]. To date etching ion-beam milling is widely used in the technology of creation of photodetectors based on MCT [5].

In the Institute of High-Current Electronics SB RAS (Tomsk) the sources are developed that allow one to form pulsed nanosecond volume discharge with low (~1 Hz) and high (~1 kHz) repetition frequency in high-pressure gases and to use it for modification of materials properties [6–11]. The unique feature of this discharge is the complex influence of the plasma of nanosecond discharge with the specific power of energy deposition of hundreds of megawatts per cubic centimeter, ultra-short electron beam with wide energy spectrum and optical radiation of various spectral ranges from the discharge plasma. The features of this discharge make it possible to induce discharge in the air at atmospheric pressure that significantly simplifies experimental equipment.

First experiments on the influence of the volume nanosecond discharge on the electrophysical properties of MCT (low-frequency discharge in the air at atmospheric pressure [8–10] and high-frequency discharge in the nitrogen atmosphere [11]) have shown the possibility of using this method for this material. The aim of the presented work was to determine the influence of the high-frequency volume nanosecond discharge in the air at atmospheric pressure on the electrophysical properties of the epitaxial MCT films and to reveal the dynamics of the change of the electrophysical properties of the material in time.

2. Samples and experimental methods

Series of samples of epitaxial $Hg_{1-x}Cd_xTe$ films with p-type of conductivity were prepared by the method of molecular beam epitaxy in the Institute of Semiconductor Physics SB RAS (Novosibirsk). The films were grown on GaAs(013) substrates with ZnTe and CdTe buffer layers and varyband layers near the substrate and the surface. The thicknesses of ZnTe and CdTe layers were 0.05 μm and 5.0 μm correspondingly. The composition of varyband layer at the CdTe buffer layer interface was $x = 0.45$.The thickness of varyband layer near the substrate was 1.0 μm, the thickness of base layer was 8.8 μm. The composition of base layer $x = 0.22$. Near-surface varyband layer was etched. After the deposition the films had n-type of conductivity. For conversion into p-type they were annealed in neutral helium atmosphere. For the experiments six series of the epitaxial films were prepared (Table 1).

Specially constructed installation and discharge chamber were used for the irradiation of the samples (Fig. 1). The construction of the generator of diffusive plasma allows us to modify surfaces of flat circular samples with the diameter up to 10 mm. Prepared samples are placed on the anode 1. Point cathode 2 with a small radius of curvature, made of tool steel, was pressed into caprolon insulator 3. To control the operation of generator, quartz windows 5 with the diameter of 20 mm were made in the metal case 4, as well as the voltage at the discharge gap and discharge current were measured with capacitive divider 6 and shunt 7, made from low-inductive chip-resistors. The construction of the setup allows us to form volume discharge in the atmospheres of various gases. For this purpose input 8 and output openings are made in the discharge chamber. Laminar gas flow along the cathode is formed by caprolon conical nozzle 9. Power supply of the generator was performed with high-voltage source of NPG-18/3500N type, forming on matched load voltage pulses of negative polarity with the amplitude of incident wave of 14–52 kV and the duration of 3–5 ns with the frequency up to 3.5 kHz. For registration of the shape of the current pulse TDS 3034 oscillograph (300 MHz, 2.5 GS/s) was used.

Table 1
Electrophysical parameters of the initial samples of epitaxial films.

Sample no.	p, 10^{15} cm^{-3}	μ, cm^{2}V^{-1} s^{-1}	Exposure time t, min
1	7.8	425	1
2	8.1	424	2
3	8.0	430	5
4	7.9	418	10
5	8.0	422	20
6	7.8	415	20

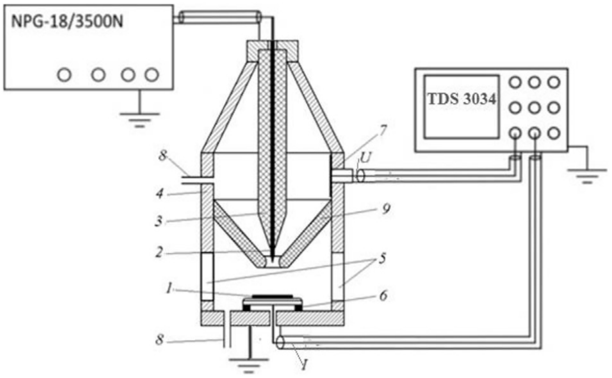

Fig. 1. Experimental setup: (1) – anode, (2) – Point cathode, (3) – caprolon insulator, (4) – metal case, (5) - quartz windows, (6) – shunt, (7) – capacitive divider, (8) – gas inlet and gas outlet, (9) - caprolon conical nozzle.

The MCT samples were irradiated in the air atmosphere at ambient conditions. The irradiation was performed in repetitively-pulsed mode with the pulse repetition frequency 1200 Hz. The amplitude of pulses was 20 kV, the pulse duration at full width at half-maximum (FWHM) was 18 ns, and the pulse rise time was 4 ns. The duration of the exposure was 1, 2, 5, 10 and 20 min.

The electrophysical parameters of the MCT samples before and after the influence of discharge were determined from the Hall measurements by the van der Pauw method. The measurements were conducted at direct current through the sample ($I = 1$ mA) for two directions of the current and two directions of the constant magnetic field. The surface of irradiated samples was investigated with the help of atomic force microscope "Ntegra Prima" (NT-MDT).

3. Results and discussion

The investigation of the surface structure for the initial an irradiated epitaxial films with atomic force microscope has shown that surface quality does not change after the exposure. The surface roughness slightly increases from 1.6 to 2.2 nm.

The measurements of electrophysical parameters of the samples of epitaxial MCT films after the irradiation by volume discharge pulses have shown the increase in the conductivity of the material for all the samples (Fig. 2). At the same time with the increase in the exposure time up to 10 min the value of the conductivity increases up to 0.83 Ohm^{-1} cm^{-1} and then does not change, so there is the saturation of the conductivity. Magnetic field dependences of the Hall coefficient

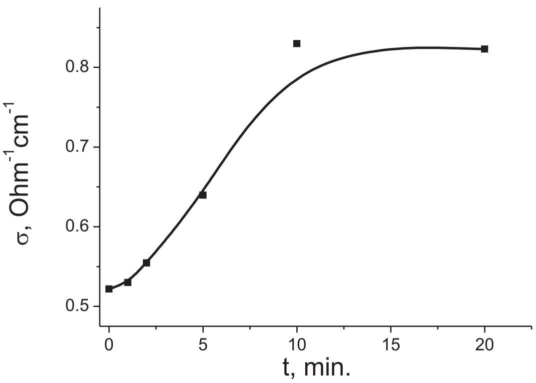

Fig. 2. Dependence of the conductivity σ of the samples of epitaxial MCT films on the time of exposure.

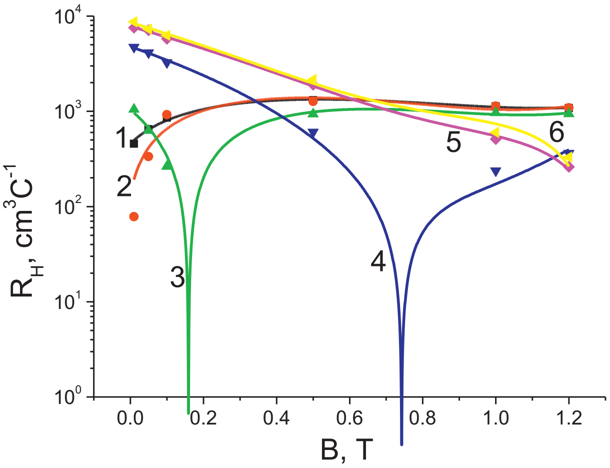

Fig. 3. Field dependence of Hall coefficient $R_H(B)$ of the samples of epitaxial MCT films. Curve 1 – initial sample. Exposure time (min): 2) – 1, 3) – 3, 4) – 5, 5) – 10, 6) – 20.

are also changes with the increase in the exposure time. For the exposure time 1 min there are only small changes of the value of the Hall coefficient in the range of weak magnetic fields (Fig. 3, curve 2), while for the exposure times 3 and 5 min (Fig. 3, curves 3 and 4) the character of $R_H(B)$ dependence changes dramatically and it becomes alternating in sign. For the exposure times 10 and 20 min (Fig. 3, curves 5 and 6) in the whole range of magnetic fields (0.01 ÷ 1.2 T) the character and sign of the Hall coefficient corresponds to the material of n-type of conductivity. Meanwhile, the samples of epitaxial MCT films are characterized by low values of electron mobility $\sim 2{\cdot}10^3$ cm^2 V^{-1} s^{-1}, which is by two orders of magnitude smaller than for corresponding high-quality epitaxial material with n-type of conductivity.

For the samples of MCT epitaxial films No. 6 treated with volume high-frequency discharge for 20 min the detailed investigation of the stability of the electrophysical parameters after the irradiation was carried out. The results of the investigations are presented in Figs. 4 and 5. As it may be seen from Fig. 4 the value of conductivity of the irradiated epitaxial MCT film tends to its initial value. The maximum change in the conductivity is observed in the first day after the exposure, when it decreases from 0.83 to 0.65 Ohm^{-1} cm^{-1}. Then the value of conductivity relaxes gradually to 0.58 Ohm^{-1} cm^{-1}. Moreover, as it is seen from Fig. 5, in the first hour after the treatment of the material by the volume discharge the character of magnetic field dependences of the Hall coefficient of the irradiated samples also changes drastically. Just after the exposure the character of the field dependence and the sign of the Hall coefficient correspond to the materials with n-type of conductivity (Fig. 3, curve 6). After the first hour from the end of the irradiation the $R_H(B)$ dependence becomes alternating in sign. Then, the change in the position of the point of sign inversion for magnetic field dependence of the Hall coefficient is observed, which shifts to the range of weak magnetic fields. Besides that, in the range of strong magnetic fields the value of the Hall coefficient, characterizing the concentration of holes in the bulk of epitaxial material, tends to its initial value.

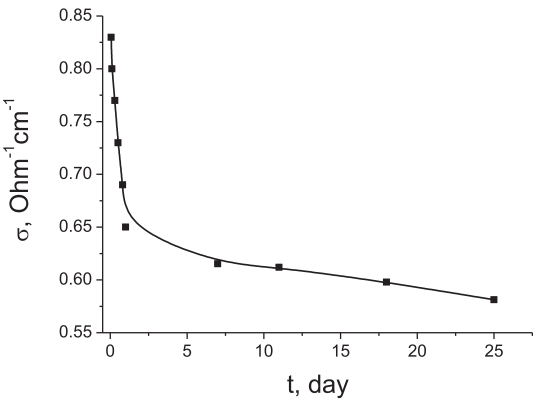

Fig. 4. Time change of the conductivity σ of the sample of the epitaxial MCT film after the irradiation by the high-frequency volume discharge for 20 min.

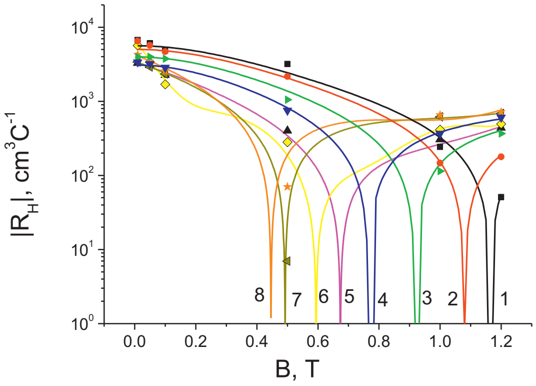

Fig. 5. Field dependence of the Hall coefficient $R_H(B)$ of the sample of the epitaxial MCT film after the irradiation by the high-frequency volume discharge for 20 min. Time after irradiation: 1) – 1 h, 2) – 3 h, 3) – 6 h, 4) – 10 h, 5) - 12 h, 6) 18 h, 7) – 20 h, 8) – 25 days.

The obtained experimental results allows us to suppose that in the process of the treatment of the surface or near-surface area of the samples of epitaxial films by volume discharge the formation of a layer with high concentration of electrons occurs. The conductivity of this layer shunts the bulk of the epitaxial film during the Hall measurements. The reasons of appearing of the layer with high conductivity in epitaxial MCT films treated by the pulses of volume nanosecond discharge still remain controversial. The results may be explained, for example, by the appearance of thin dielectric film on the surface of HgCdTe after the exposure. This may be connected with the formation of an oxide stimulated by the influence of the pulses of the volume nanosecond discharge in the air at atmospheric pressure. It is well known that positive mobile and fixed charge is typical for the anode-oxide film on HgCdTe [12,13] that is usually connected with the presence of oxygen vacancies in the anode oxide.

The presence of built-in positive charge leads to the formation of inversed n-layer with high conductivity at the interface with epitaxial p-MCT film.

For the confirmation of this supposition the fundamental system of equations (the Poisson's equation and two continuity equations) was solved with respect to the presence of the positive charge Q(0) on the surface of the epitaxial MCT film:

$$\frac{dE}{dz} = \frac{q}{\varepsilon\varepsilon_0}(p(z) - n(z) + N_d^+ - N_a^-) + Q(0) \tag{1}$$

$$\frac{dp}{dt} = D_p\frac{\partial^2 p(z)}{\partial z^2} - \mu_p E\frac{\partial p}{\partial z} - p(z)\mu_p\frac{\partial E}{\partial z} - R_p = 0 \tag{2}$$

$$\frac{dn}{dt} = D_n\frac{\partial^2 n(z)}{\partial z^2} - \mu_n E\frac{\partial n}{\partial z} - n(z)\mu_n\frac{\partial E}{\partial z} - R_n = 0 \tag{3}$$

where z is the spatial coordinate of the structure, q is the electron charge, ε_0 is the electric constant, ε is the relative permittivity of the semiconductor, n, p, N_a, N_d are the concentrations of free electrons, holes, ionized acceptors and donors, respectively, R_n, R_p are the rates of recombination and generation of electrons (holes), respectively, D_n, D_p are the diffusion coefficients of electrons and holes, μ_n, μ_p are the mobilities of electrons and holes, E is the electric field. The diffusion coefficients of electrons and holes were determined from Einstein relation: $\mu_{n,p} = \frac{q}{kT} D_{n,p}$. It was considered during modeling that carrier recombination is limited by Auger recombination

$$R_{Auger} = C_n(p(z)n^2(z) - n(z)n_i^2(z)) + C_p(n(z)p^2(z) - p(z)n_i^2(z)) \tag{4}$$

where $C_n = 8.3 \cdot 10^{-32}$ m^{6}s^{-1}, $C_p = 3.3 \cdot 10^{-31}$ m^{6}s^{-1} [14] are the coefficients of Auger recombination for electrons and holes, n_i is intrinsic concentration of charge carriers. For the calculations of dependence of the intrinsic concentration of charge carriers on the composition of the material the following expression was used [1]:

$$n_i = (0.585 - 3.82x + 0.001753T - 0.001364xT) \bullet 10^{14} E_g^{3/2} T^{3/2} \exp\left(-\frac{E_g}{2kT}\right) \tag{5}$$

where x is the composition of the material, E_g is the band gap of the material (in eV):

$$E_g = -0.302 + 1.93x - 0.81x^2 + 0.832x^3 + 5.35 \bullet 10^{-4}(1 - 2x)T \tag{6}$$

For the calculations of the electron mobility the following empirical formula was used [14]:

$$\mu_n = 9.0 \bullet 10^8 \left(0.2/_x\right)^{7.5} T^{-2\left(\frac{0.2}{x}\right)^{0.6}} \tag{7}$$

During modeling, the experimental values of the hole mobility determined from the Hall measurements for the initial epitaxial films were chosen (Table 1).

After solving this system of equations, obtained spatial distributions of charge carriers n(z) and p(z) were used for calculations of magnetic field (B) dependencies of the Hall coefficient:

$$R_{Heff}(B) = \frac{\overline{\sigma}_{xy}(B)}{\overline{\sigma}_{xx}^2(B)} \frac{1}{B} \left[1 + \left(\frac{\overline{\sigma}_{xy}(B)}{\overline{\sigma}_{xx}(B)} B\right)^2 B^2\right]^{-1} \tag{8}$$

where $\overline{\sigma}_{xx}$, $\overline{\sigma}_{xy}$ are the mean tensors of conductivity in the directions xx and xy:

$$\overline{\sigma}_{xx} = \frac{1}{d} \int_0^d qn(z) \frac{\mu_h}{1 + \mu_h^2 B^2} + qn(z) \frac{\mu_e}{1 + \mu_e^2 B^2} dz \tag{9}$$

$$\overline{\sigma}_{xy} = \frac{1}{d} \int_0^d qn(z) B \frac{\mu_h^2}{1 + \mu_h^2 B^2} - qn(z) B \frac{\mu_e^2}{1 + \mu_e^2 B^2} dz \tag{10}$$

The value of positive charge on the material's surface was determined by the least squares method:

$$min \sum_i^m |R_{Hexp}(B_i) - R_{heff}(Q, B_i)| \tag{11}$$

The results of the modeling of field dependence of the Hall coefficient for the epitaxial film sample No. 6 are presented in Fig. 6. The calculations show that theoretical curve adequately describes experimental data for the value of the surface positive charge $Q = 2.94 \cdot 10^{11}$ C·cm^{-2} (where C is the electron charge). The conducted modeling of the field dependence of the Hall coefficient presented in Fig. 3 made it possible to obtain the dependence of the formed positive charge Q in the near-surface region of the epitaxial MCT film on the time of the influence of the volume discharge. The results of the modeling are depicted in Fig. 7. It may be seen that the value of Q lays in the range of $(2.3 \div 2.94) \cdot 10^{11}$ C·cm^{-2}.

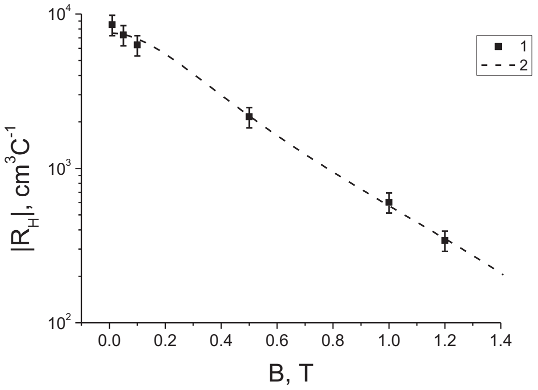

Fig. 6. Field dependence of the Hall coefficient for the sample No. 6 immediately after the irradiation by the volume discharge: 1 – experimental data, 2 – calculated dependence for the value of the positive charge $Q = 2.94 \cdot 10^{11}$C·cm^{-2}.

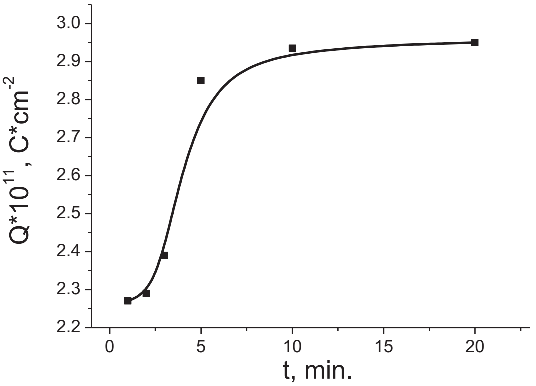

Fig. 7. Dependence of the value of the formed positive charge Q on the time t of the influence of the volume discharge.

As it is seen from Fig. 7, the value of near-surface positive charge changes insignificantly, but the behavior of the corresponding field dependences of the irradiated samples changes drastically. In order to understand the reason of such behavior, we will consider parameters of n-layer: layer thickness d_n and average concentration of electrons n_{av}. The thickness d_n is defined as the point, where the conductivity of near-surface n-layer is equal to the conductivity of p-layer: $\int_0^{d_n} \sigma_n(z)dz = \int_{d_n}^{d} \sigma_p(z)dz$, where $\sigma_n(z) = qn(z)\mu_n$, $\sigma_p(z) = qp(z)\mu_p$, d is the initial film thickness. In this case, average concentration of electrons will be $n_{av} = \frac{1}{d_n}\int_0^{d_n} n(z)dz$. The introduced characteristic n_{av} is convenient for analysis as it simultaneously takes into account both the thickness of high-conductivity n-layer and inhomogeneous distribution of electron concentration over the thickness of irradiated material. Fig. 8 shows the dependence of the average concentration of electrons in n-layer on the value of the positive charge. It is seen that small changes in the Q value result in sharp increase of average electron

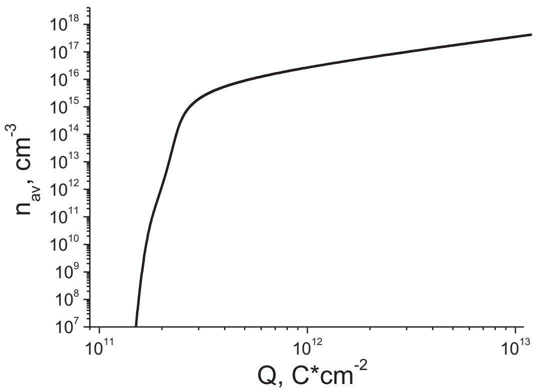

Fig. 8. Dependence of the average electron concentration n_{av} in the inversion layer on the value of the positive charge Q.

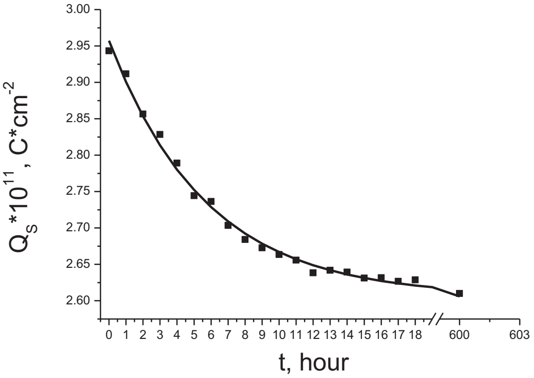

Fig. 9. Dependence of the value of the positive charge Q at the surface of the epitaxial MCT film No. 6 on time t.

concentration. For the value of near-surface positive charge $Q = 2.94 \cdot 10^{11} \text{C} \cdot \text{cm}^{-2}$ the value of average electron concentration in inversion n-layer is $1.9 \cdot 10^{15}$ cm^{-3}, and the thickness of the layer for this conditions is $d_n = 0.27$ μm.

In the frames of the presented model an analysis of the experimental data on the relaxation of the magnetic-field dependences of the Hall coefficient for the sample No. 6 (Fig. 5) was carried out. The result of model calculations of the dependence of the value of positive charge Q at the surface of epitaxial film on time after exposure of volume discharge is given in Fig. 9.

Results of model calculations presented in Fig. 9 are well approximated by the following exponential dependence:

$$Q(t) = Q_F + Q_M \exp\left(-\frac{t}{\tau_M}\right) \tag{12}$$

where $Q_F = 2.6 \cdot 10^{11}$ $\text{C} \cdot \text{cm}^{-2}$, $Q_M = 3.5 \cdot 10^{10}$ $\text{C} \cdot \text{cm}^{-2}$, $\tau_M = 5.6$ h. Within the frames of the presented model of the formation of thin dielectric oxide film on the surface of the material after the irradiation of HgCdTe samples the Q_F value may be linked with the fixed positive charge forming in the oxide film, while Q_M value may be associated with the mobile positive charge. τ_M is the time constant for the process of the relaxation of the value of the mobile positive charge Q_M.

4. Conclusions

The conducted research of the influence of the high-frequency volume discharge on the properties of the epitaxial MCT films have shown that the change in the electrophysical parameters of the investigated samples is observed. The relaxation of the value of the electrophysical parameters with time after the exposure is found. The analysis of the experimental results shows that the formation of n-layer with high concentration of electrons occurs on the surface or in the near-surface region. The conductivity of this layer is such that it shunts the main bulk of the epitaxial film in the process of the Hall measurements. The supposition is proposed that high-conductivity n-layer appears due to the formation of the fixed and mobile positive charge, for example, by virtue of the thin oxide film formation on the surface of the irradiated material. The conducted model calculations in the frames of this supposition have shown that as a result of the irradiation of the epitaxial MCT films by high-frequency volume discharge (in the range of exposure times 1–20 min) surface positive charge with the value of $Q = (2.3 \div 2.94) \cdot 10^{11}$ $\text{C} \cdot \text{cm}^{-2}$ is formed in the near-surface region. The observed relaxation of the electrophysical parameters of the epitaxial films after the exposure may be associated with the change in the concentration of the mobile positive electrical charge. At the same time the nature of this positive charge and the mechanisms of its formation still remain controversial and demand further investigations.

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Author contributions

A.G. Korotaev – Conceptualization, Writing - Review & Editing
D.V. Grigoryev - Investigation, Formal analysis, Writing - Original Draft
A.V. Voitsekhovskii – Project administration, Funding acquisition
K.A. Lozovoy –Writing - Review & Editing
V.F. Tarasenko – Supervision
V.S. Ripenko - Methodology,
M.A. Shulepov - Methodology,
M.V. Erofeev - Methodology
M.V. Yakushev – Supervision
S.A. Dvoretskii – Resources
N.N. Mikhailov – Resources
V.S. Varavin – Methodology

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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