Nonstationary holographic currents in a $\beta$-Ga$_2$O$_3$ crystal at wavelength $\lambda = 457$ nm

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Abstract. We report the excitation of nonstationary holographic currents in a monoclinic gallium oxide crystal. Although the crystal is almost transparent and insulating for a visible light, the dynamic space-charge gratings are recorded and holographic currents are observed for both the diffusion and drift modes. The anisotropy along the [100] and [010] directions is revealed, namely, there is a small difference in the transport parameters and a pronounced polarization dependence of the signal. The crystal’s photoconductivity, responsivity and diffusion length of electrons are estimated for the light wavelength $\lambda = 457$ nm.

1. Introduction
Monoclinic gallium oxide $\beta$-Ga$_2$O$_3$ is a material, which will probably meet the demands of future electronics on operation in severe conditions. The crystal is highly transparent for visible light and partially for UV due to the large bandgap ($\sim 4.8$ eV). This property is used for development of solar-blind detectors of deep ultraviolet radiation. The material possesses the high breakdown field (6-8 MV/cm), reasonable mobility of carriers and high thermal stability, and this determines its utilization in radio frequency and power field-effect transistors, as well as Schottky rectifiers. The water splitting under UV radiation and gas sensing were also proposed [1,2].

Material characterization is an important stage of the development and production processes. Besides the standard methods for investigation of photoelectric materials [3] there are approaches based on the principles of the dynamic holography [4,5] and some relative techniques including the nonstationary holographic currents. The effect of nonstationary holographic current or, in other terminology, non-steady-state photo-EMF reveals itself as an alternating electric current arising in a semiconductor illuminated by an oscillating interference pattern [6,7]. The electric current appears due to time-varying relative spatial shifts of the space-charge and photoconductivity distributions. Various techniques based on this effect were proposed for semiconductor characterization [8–10] and for detection of phase- and frequency-modulated optical signals [11–14].

Recently we used the non-steady-state photo-EMF technique for characterization of $\beta$-Ga$_2$O$_3$ crystal in the green spectral region [15]. Then we started research with the blue light [16]. In this work we continue the investigation at $\lambda = 457$ nm for the drift mechanism of signal excitation and compare it with the diffusion mode.
2. Experimental arrangement

The experimental setup for investigation of nonstationary holographic currents in $\beta$-Ga$_2$O$_3$ is shown in Fig. 1.a. The light from the solid state diode pumped laser with the wavelength of $\lambda = 457$ nm is expanded and split into two beams, which then create the interference pattern with spatial frequency $K$, contrast $m$ and average intensity $I_0$ on the crystal surface. The electro-optic modulator introduces phase modulation with amplitude $\Delta = 0.51$ and frequency $\omega$ into the signal beam. The polarization plane of the both beams is perpendicular to the incidence plane in the most of experiments. The half-wave plate placed in front of the sample allows the rotation of the polarization plane, when it is necessary. The photocurrent arising in the sample produces a voltage across the load resistor $R_L$, which is amplified and then measured by the lock-in voltmeter. The external high voltage is obtained from the commercial dc power supply. The following parameters differ in the experiments with external fields: $m = 0.38$, $R_L = 100$ k$\Omega$ for the zero field, and $m = 0.20$, $R_L = 10$ k$\Omega$ for the dc one.

We use the same sample of $\beta$-Ga$_2$O$_3$ as in our previous works [15,16]. It has the dimensions of $2.00 \times 2.15 \times 1.35$ mm along the crystallographic directions [100], [010] and direction perpendicular to the plane (001), respectively. The front and back surfaces ($2.00 \times 2.15$ mm) are the (001) crystal’s cleaved facets, no additional treatment was applied to them. The silver paste electrodes are deposited on the corresponding pairs of lateral surfaces in the experiments with the light pattern grating vector $K||[100]$ and $K||[010]$.

3. Experimental results

The presence of the nonstationary holographic current in $\beta$-Ga$_2$O$_3$ crystal at the chosen wavelength is the first result to be reported. The signal amplitude is rather small but allows the reliable detection with the signal-to-noise ratio of $0 - 40$ dB. The phase of the signal corresponds to the electron type of photoconductivity.

When no voltage is applied to the sample, the electric field grating arises in the crystal’s volume due to diffusion of free carriers, and this corresponds to the diffusion mode of signal excitation [16]. The dependence of the signal amplitude on the frequency of phase modulation is presented in Fig. 1,b [16]. The dependence consists of the growing part at low frequencies followed by the frequency-independent part. The signal at low $\omega$ is small, since both the space...
Figure 2. Dependencies of the non-steady-state photo-EMF amplitude and cut-off frequency on the spatial frequency (a). The best fits using Eqs. (1), (2) are shown by the solid lines. Dependencies of the non-steady-state photo-EMF on the angle between polarization and incidence planes (b). The approximation by Eq. (3) is shown for \( a_2 = 0 \) (dotted lines) and \( a_2 \neq 0 \) (solid line).

charge field grating and the grating of free electrons (photoconductivity grating) follow the movement of the interference pattern. The spatial shift between them is nearly equal to \( \pi/2 \), which results in small value of the average drift component of the current. The space-charge grating becomes “frozen-in” at higher frequencies, the spatial shifts between gratings increase, and the signal reaches its maximum at the frequency-independent region. The signal is as follows [6,7]:

\[
j(\omega) = \frac{m^2 \Delta}{2} \sigma_0 E_D \frac{-i\omega \tau_M}{1 + i\omega \tau_M (1 + K^2 L_D^2)}.
\]  

Here \( \sigma_0 \) is the average photoconductivity, \( E_D = (k_B T/\epsilon) K \) is the diffusion field [5], \( \tau_M = \epsilon_0 \tau / \sigma_0 \) is the Maxwell relaxation time, \( L_D \) is the diffusion length of electrons. The growing and frequency-independent regions are separated by the cut-off frequency \( \omega_1 \)

\[
\omega_1 = \left[ \tau_M (1 + K^2 L_D^2) \right]^{-1}.
\]

Both the signal amplitude at the plateau and the cut-off frequency linearly depends on the light intensity [16], which indicates the linear photoconductivity of the crystal \( \sigma_0 \propto I_0 \).

The dependence of the signal amplitude on the spatial frequency is another important characteristic of the nonstationary holographic current (Fig. 2, a [16]). The growth of the signal at low \( K \) is due to the increase of the space charge field amplitude [5]. The decay at high \( K \) is resulted from the diffusion “blurring” of the photoconductivity grating. The maximum of the signal is achieved at \( K = L_D^{-1} \), so the electron diffusion length can be estimated from the experimental curves: \( L_D = 200 \) nm for \( K \parallel [100] \) and \( L_D = 230 \) nm for \( K \parallel [010] \). These estimates are very close to each other, which indicates the slight anisotropy of transport parameters containing \( \mu \tau \)-product.

The cut-off frequency also depends on the spatial frequency (Fig. 2, a [16]). Being measured at low \( K \) it equals to the inverse Maxwell relaxation time \( \omega_1 \propto \tau_M^{-1} \), and this is used for determination of the specific conductivity: \( \sigma_0 = 2.3 \times 10^{-9} \) \( \Omega^{-1} \text{cm}^{-1} \) along the [100] axis and \( \sigma_0 = 1.6 \times 10^{-8} \) \( \Omega^{-1} \text{cm}^{-1} \) along the [010] axis. The experimental dependencies are approximated by Eq. (2) with \( L_D = 110 \) nm for both \( K \parallel [100] \) and \( K \parallel [010] \). The dependence \( \omega_1(K) \) suggests a smaller value for the diffusion length than that for the dependence \( J(\omega)(K) \). This may be explained...
Figure 3. Frequency dependencies of the non-steady-state photo-EMF excited in external dc fields (a). Dependencies of the resonant non-steady-state photo-EMF amplitude and resonant frequency on the spatial frequency (b). The best fits using Eqs. (4), (5) are shown by the solid lines.

by the increased light reflection at large incident angles, by the relatively large amplitude of phase modulation $\Delta$, and by the nonuniformity of the average light intensity $I_0$ due to the light absorption along the sample thickness and residual Gauss profile of the light beams.

The monoclinic $\beta$-Ga$_2$O$_3$ reveals the expected anisotropy demonstrating the larger non-steady-state photo-EMF signal and cut-off frequency for the $K||[100]$ geometry (Figs. 1,b and 2,a [16]). We measure the dependence of the signal amplitude on the angle of the polarization plane (Fig. 2,b) to clear up the origin of such an anisotropy. The experimental dependencies are well approximated by the phenomenological truncated Fourier series:

$$|J_\omega(\Theta)| = a_0 + a_1 \cos 2\Theta + a_2 \cos 4\Theta. \quad (3)$$

As seen from the experimental dependencies, we can confine ourselves with the zeroth component and first harmonic for the case $K||[010]$, while the all three components $a_0, a_1, a_2 \neq 0$ should be taken into account for the case $K||[100]$. The amplitude of the signal in the absolute maxima in these dependencies differs by 5% only. This means that the studied geometries are almost equivalent in their efficiency: the signal amplitude would maintain its maximum value, if we rotated the polarization plane synchronously with the crystal. In our previous work [15] we observed the similar dependencies at $\lambda = 532$ nm, and then we attributed such polarization dependencies to the angular dependence of quantum efficiency: $\beta = \beta_0 + c_\beta \cos 2\Theta$. We think this assumption is valid at $\lambda = 457$ nm as well, at least for the first angular harmonic in Fig. 2,b and Eq. (3).

The increase of the non-steady-state photo-EMF amplitude can be achieved by the application of an external dc voltage [6, 7]. In contrast to the diffusion mode considered above, the drift mode of the signal excitation corresponds to the local response of the photosensitive material, and this results in the different phase relation between incident light oscillations and detected current. The phase and signal enhancement are not the only changes arising in the dc field, the frequency dependence becomes resonant (Fig. 3,a). The appearance of the resonant maximum is due to the excitation of space-charge waves, which are the eigenmodes of space-charge evolution in semiconductors placed in an external dc field [4, 5].

These space-charge waves run along the applied electric field and demonstrate an unusual dispersion law, i.e. dependence $\omega_r(K)$:

$$\omega_r = (\tau_M KL_0)^{-1}. \quad (4)$$
Here $L_0 = \mu \tau E_0$ is the drift length of electrons in the electric field $E_0$, $\mu$ is the mobility of electrons, $\tau$ is their lifetime. This dispersion law is present in our experiments with Ga$_2$O$_3$ (Fig. 3,b).

Relatively high electric fields are necessary for the space-charge wave excitation, so that condition $KL_0 \gg 1 + K^2L_D^2$ is satisfied. The nonstationary holographic current amplitude reaches then [6]

$$J^\omega_r = \frac{m^2 \Delta}{2} \sigma_0 E_0 \frac{KL_0}{1 + K^2 L_D^2}$$

(5)

demonstrating the square-law growth of the signal versus applied field strength and the maximum at $K = L_D^{-1}$ (Fig. 3,b).

The resonant maximum is much wider than its theoretical estimate [6]: the width of $\delta \omega/2\pi = 250$ Hz for the top experimental curve in Fig. 3,a versus $\delta \omega/2\pi = 2(\omega_r/2\pi)(1+K^2 L_D^2)/KL_0 = 23$ Hz calculated for $\omega_r/2\pi = 49$ Hz and $L_D = 200$ nm ($\mu \tau = 1.6 \times 10^{-8}$ cm$^2$/V). This mismatch is probably due to the inhomogeneity of the light intensity $I_0$ and electric field $E_0$ as well as insufficiently low contrast $m$, which is $m \ll 1$ in theory.

4. Discussion

The characteristics of the $\beta$-Ga$_2$O$_3$ crystal can be compared with those for other materials studied in the same spectral range. Since the present and previous experimental data are obtained for different experimental conditions, we should estimate the normalized maximal signal amplitude, i.e. responsivity:

$$R^\omega_m = J^\omega_r/[P_0 m^2 J_0(\Delta) J_1(\Delta)/2],$$

(6)

where $J^\omega_m$ is the amplitude of the nonstationary holographic current measured at $\omega > \omega_1$ and $KL_0 = 1$, $P_0$ is the total light power, which is the sum of powers of the signal $P_s$ and reference $P_r$ beams illuminating the sample, $J_n(x)$ is the Bessel function of the first kind of the $n$-th order. The value provided by Eq. (6) can be used in the interferometric applications, where the contrast and amplitude of phase modulation are small ($m, \Delta \ll 1$), and where it reduces to $R^\omega_m \sim J^\omega_m/(P_0 \Delta)$ being an analog of the photodiode responsivity [6]. After substitution of the data from Fig. 2,a we obtain $R^\omega_m = 3.8 \times 10^{-7}$ A/W. This estimate is lower than those for other wide-bandgap materials studied in the blue spectral range: $R^\omega_m = 1.1 \times 10^{-4}$ A/W for Bi$_{12}$SiO$_{20}$ [7] and $R^\omega_m = 3.3 \times 10^{-5}$ A/W for SnS$_2$ [7]. This difference in responsivities follows from the fact that Ga$_2$O$_3$ crystal is transparent for visible light, while Bi$_{12}$SiO$_{20}$ and SnS$_2$ absorb blue light almost completely. The relatively low diffusion length $L_D$ ($\mu r$-product) is another reason of low responsivity.

If we compare the Ga$_2$O$_3$ parameters obtained for $\lambda = 457$ nm and $\lambda = 532$ nm [15], we can note the slight changes for the most values, which can be considered as an advantage: the sensor based on this material would have rather flat spectral characteristics. Some difference in behavior should be pointed out, however. The second angular harmonic appears in the polarization dependence measured at $\lambda = 457$ nm (Fig. 2,b), but not at $\lambda = 532$ nm. We suppose that this second harmonic arises as product of two angular-dependent functions: $\alpha = \alpha_0 + c_\alpha \cos 2\Theta$ and $\beta = \beta_0 + c_\beta \cos 2\Theta$. Since this peculiarity appears only for $\lambda = 457$ nm and certain sample orientation, coefficient $c_\alpha$ should be a function of the wavelength and some vector, e.g. [100] component of the space charge field: $c_\alpha(\lambda, E_{sc} \cdot \mathbf{a})$. In other words, there can be some effect of electric field induced absorption analogues to Franz-Keldysh effect [17]. This hypothesis surely requires more confirmations with other wavelengths, samples and approaches.

The responsivity defined by Eq. (6) can also be evaluated for the electric-field-enhanced mode: $R^\omega_m = 2.4 \times 10^{-6}$ A/W for the dc field $E_0 = 11$ kV/cm. As seen, the application of the external electric field to the Ga$_2$O$_3$ crystal increases the signal amplitude and responsivity by
several times but not by several orders, as it was in Bi$_{12}$SiO$_{20}$ [6,7]. The reason is the mentioned small $\mu\tau$-product in $\beta$-Ga$_2$O$_3$.

Even demonstrating the rather low responsivity, the studied Ga$_2$O$_3$ crystal has some advantages over other crystals. The frequency dependence of the former is rather flat in the range of 0.1 – 500 kHz, while the signal in Bi$_{12}$SiO$_{20}$ decays over 3 kHz. No influence of shallow traps has been revealed. The crystal has a moderate static dielectric constant $\epsilon \simeq 10$, which is about 5 times lower than that in sillenite crystals, and this allows to achieve the appropriate cut-off frequency at low illumination levels.

5. Conclusion
To conclude, we have studied the nonstationary holographic currents in monoclinic $\beta$-Ga$_2$O$_3$ crystal at $\lambda = 457$ nm. This material allows the signal excitation without any applied voltage as well as the signal enhancement in the presence of dc electric field. The photoconductivity and diffusion length of electrons are estimated for two crystal orientations ($K||[100]$ and $K||[010]$). There is a weak anisotropy of the photo-EMF and electric parameters along the [100] and [010] axes and a pronounced polarization dependence of the quantum efficiency and resulting signal. The noticeable signal level and flatness of the frequency response in $\beta$-Ga$_2$O$_3$ crystal can advance its utilization as a material for adaptive sensors of phase- and frequency-modulated optical signals.

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