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NONLINEAR OPTICAL PHENOMENA

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Features of photoinduced dynamic gratings in semiconductors

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***Abstract*. The problem of influence of the diffusion process of nonequilibrium carriers on the efficiency of the dynamic grating photoinduced in a semiconductor material is studied theoretically and experimentally. An analytic expression is derived, which allows one to estimate the outlooks for applications of different semiconductor materials as active media for writing dynamic gratings.**

Keywords: *laser, dynamic grating, semiconductor.*

1. Introduction

Amplitude and phase dynamic gratings written in various nonlinear media are used to solve the problem of aberration corrections in observation optics, phase conjugation and control of the parameters of light beams in feedback optical systems [1]. Various semiconductor materials, in which the grating is produced by changing the optical parameters (absorption coefficient or refractive index) due to free-carrier photogeneration can be used along with other nonlinear media [2]. In addition, the results of studies of the formation and development properties of spatially periodic structures photoinduced in the volume of a semiconductor can be used for the elaboration of the optical measurement methods of parameters of charge carriers in semiconductor materials such as their mobility, concentration, and relaxation time.

In this paper, the mechanisms of formation and behaviour of dynamics gratings photoinduced in semiconductors are studied analytically and experimentally.

2. Analysis of formation and estimate of the diffraction efficiency of a dynamic grating

Consider the situation when a grating is written by two interfering short-wavelength beams with the photon energy $\hbar\omega_1$, exceeding the bandgap E_g of a semiconductor, and intersecting in the spatial region occupied by a semiconductor. According to the radiation intensity distribution, pairs of nonequilibrium electron – hole carriers are generated in the semiconductor, which leads to a spatial periodic change in the optical properties of the medium [2, 3]. Long-wavelength radiation, for which the semiconductor material in the initial state is transparent because the photon energy $\hbar\omega_2$ of this radiation is much smaller than E_g, diffracts on the grating produced in this way. Subscripts 1 and 2 hereafter denote physical quantities related to the short-wavelength and long-wavelength radiation, respectively.

The optical parameters of the semiconductor are changed due to the appearance of free carriers and, hence, it is their spatiotemporal distribution that determines the properties of the dynamic grating. Photogeneration and the following redistribution of electrons in the semiconductor volume due to diffusion also determine the properties of the dynamic grating. Therefore, the analysis of the process of grating formation and behaviour is based on the solution of the diffusion equation of photoinduced nonequilibrium carriers. In the case under study ($\hbar\omega_1 \gg E_g$), the absorption coefficient α_1 of radiation writing the grating has a characteristic value of $10^3 - 10^4$ cm^{-1}. The corresponding penetration depth d of writing radiation in the semiconductor does not exceed several micrometers ($d \sim 1/\alpha_1$). This allows one to describe the formation and development processes of a photoinduced grating by the one-dimensional diffusion equation of nonequilibrium carriers

$$\frac{\partial N(x,t)}{\partial t} = D\frac{\partial^2 N(x,t)}{\partial x^2} - \frac{N(x,t)}{\tau} + \frac{\alpha_1 I_1}{\hbar\omega_1}\left(1 + \cos\frac{2\pi x}{\Lambda}\right). \quad (1)$$

Here N, D, and τ are the concentration, ambipolar diffusion coefficient and lifetime of nonequilibrium carriers, respectively; I_1 is the intensity of each of two interfering beams with the wavelength λ_1; Λ is the period of the interference pattern; x is the coordinate in the direction perpendicular to the fringes of the interference pattern.

The solution of Eqn (1) with the initial condition $N(x,t) = 0$ has the form

$$N(x,t) = \frac{\alpha_1 I_1}{\hbar\omega_1}\left\{\tau_r\left[1 - \exp\left(-\frac{t}{\tau_r}\right)\right]\cos\frac{2\pi x}{\Lambda} + \tau\left[1 - \exp\left(-\frac{t}{\tau}\right)\right]\right\}, \quad (2)$$

where τ_r is the time constant of the grating relaxation determined as follows:

$$\tau_r^{-1} = \tau^{-1} + \tau_D^{-1}, \qquad \tau_D = \frac{\Lambda^2}{4\pi^2 D}.$$

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The first term in (2) describes the spatially periodic distribution of carrier concentrations and characterises the temporal change in the dynamic grating contrast. The second term determines the spatially equilibrium concentration distribution of nonequilibrium carriers, which is formed due to their diffusion from the generation regions of an electron–hole plasma.

The spatial distribution of carriers defines variations of both the refractive index and the absorption coefficient of the medium and, in the general case, it is necessary to consider the behaviour of the amplitude–phase grating. However, undesirable effects leading to a decrease in the grating efficiency can be first of all caused by the absorption of light by the carriers distributed uniformly in the volume. Thus, we consider below only the behaviour of the amplitude grating.

It is known that the electron–hole plasma absorbs photons due to intraband transitions [4]. This process is most strongly pronounced when this plasma interacts with radiation of mid- and far-IR ranges [5, 6]. The absorption coefficient α_2 of the long-wavelength radiation by carriers photoinduced in the semiconductor is related with $N(x,t)$ by a simple relation [6]:

$$\alpha_2 = (\sigma_e + \sigma_h)N(x,t), \tag{3}$$

where σ_e and σ_h are the cross sections of IR absorption by electrons and holes, respectively.

Based on expressions (2), (3), the Bouguer law and by neglecting the reflection from the semiconductor surface, the expression for the coefficient $T_2(x,t)$ of the amplitude transmission of long-wavelength radiation at λ_2 by a photoinduced semiconductor can be written in the from

$$T_2(x,t) = \left\{1 - \frac{\alpha_1 I_1}{\hbar\omega_1}(\sigma_e + \sigma_h)\tau_r d\left[1 - \exp\left(-\frac{t}{\tau_r}\right)\right]\right\}$$
$$\times \exp\left\{-\frac{\alpha I_1}{2\hbar\omega_1}(\sigma_e + \sigma_h)\tau d\left[1 - \exp\left(-\frac{t}{\tau}\right)\right]\right\}. \tag{4}$$

The diffraction efficiency η_2 of a thin amplitude grating is determined by the square of the expansion coefficient of the amplitude transmission function into the Fourier series:

$$\eta_2 = \left\{\frac{\alpha_1 I_1 \tau_r(\sigma_e + \sigma_h)d[1 - \exp(-t/\tau_r)]}{4\hbar\omega_1}\right\}^2$$
$$\times \exp\left\{-\frac{\alpha_1 I_1}{\hbar\omega_1}(\sigma_e + \sigma_h)\tau d\left[1 - \exp\left(-\frac{t}{\tau}\right)\right]\right\}. \tag{5}$$

Based on the analysis of expressions (2) and (5), we can conclude that the maximum efficiency η_2 is realised in semiconductors, which are characterised by a low mobility of carriers and for which $\tau_r \approx \tau$. In this case, the fraction of long-wavelength radiation absorbed by the electron–hole plasma uniformly distributed in the semiconductor will be small. A similar situation can be also observed in the case when the pulse duration τ_2 with the wavelength λ_2 is much shorter than the relaxation time τ_r of the grating. For $\tau_2 \gg \tau \gg \tau_r$ the absorption by uniformly distributed carriers can achieve a substantial quantity. In this case, large energy losses of a long-wavelength radiation diffracted on the grating are inevitable. If a rectangular IR pulse is incident on the grating, the shape of the pulse of the diffracted radiation is determined by the temporal change in the grating efficiency. Thus, Figure 1 shows the transformation of a rectangular light pulse for the grating induced in a germanium single crystal.

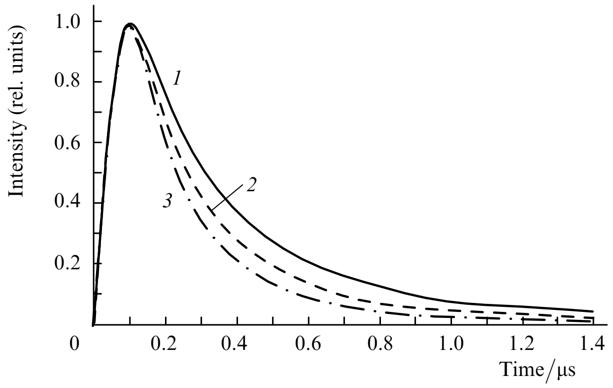


Figure 1. Shape of the rectangular IR pulse diffracted on the grating at writing radiation intensities 800 (*1*), 1000 (*2*) and 1200 W cm^{-2} (*3*).

By supplementing relation (5) with the analytic dependence from paper [5], which describes the IR absorption by photoinduced carriers, one can determine the relaxation time constant of the gratings and, hence, the mobility of nonequilibrium carriers.

3. Experimental results

The dynamic gratings photoinduced in semiconductor materials were studied experimentally at high and low diffusion velocities and, correspondingly, for different ratios of τ_r and τ_1 by the example of crystalline and amorphous germanium.

The scheme of the experimental setup is shown in Fig. 2. The grating in germanium sample (*3*) was written by $\tau_1 \sim 160$-μs pulses from free-running Nd:YAG laser (*1*). Laser radiation was split by beamsplitter (*2*) into two beams of equal intensities, which later fell onto sample (*3*) at an angle $\Theta \sim 2.5 \times 10^{-3}$ rad with the help of mirrors (*4*) and (*5*). As a result, an interference pattern with a period ~100 μm was formed in the semiconductor. Because of irradiation ($\hbar\omega_1 = 1.17$ eV), nonequilibrium carriers intensely absorbing radiation of the CO_2 laser were excited in germanium ($E_g = 0.67$ eV at $T = 298$ K) [5]. The carrier lifetime $\tau \approx 1$ μs was much shorter than τ_r [6]. Thus, a quasistationary grating of the absorption coefficient at $\lambda_2 = 10.6$ μm was produced in the semiconductor. Sample (*3*) was excited by pulsed LGI-50 CO_2 laser (*6*). The shape of pulses from Nd:YAG and CO_2 lasers and the synchronisation degree of their interaction on the sample was detected with FSG-22-3A (*7*) and LFD-2 (*8*) photodetectors, respectively. Electric signals from photodetectors were fed to the inputs of double-beam S8-14 oscilloscope (*11*). The shape of the radiation pulse diffracted to the first order was detected with FSG-22-3A photoresistor (*12*) whose signal was fed to the input of S8-12 oscilloscope (*13*). Lens (*14*) in the focal plane of which the sample was mounted focused the diffracted beam on photoresistor (*12*).

Experimets on writing gratings in the semiconductor with a high mobility of carriers were performed on single-crystal GMO-3 germanium samples made in the form of plane–parallel plates of thickness 0.5 mm. The diffraction efficiency of the grating at $\lambda_2 = 10.6$ μm was ~10^{-3} in

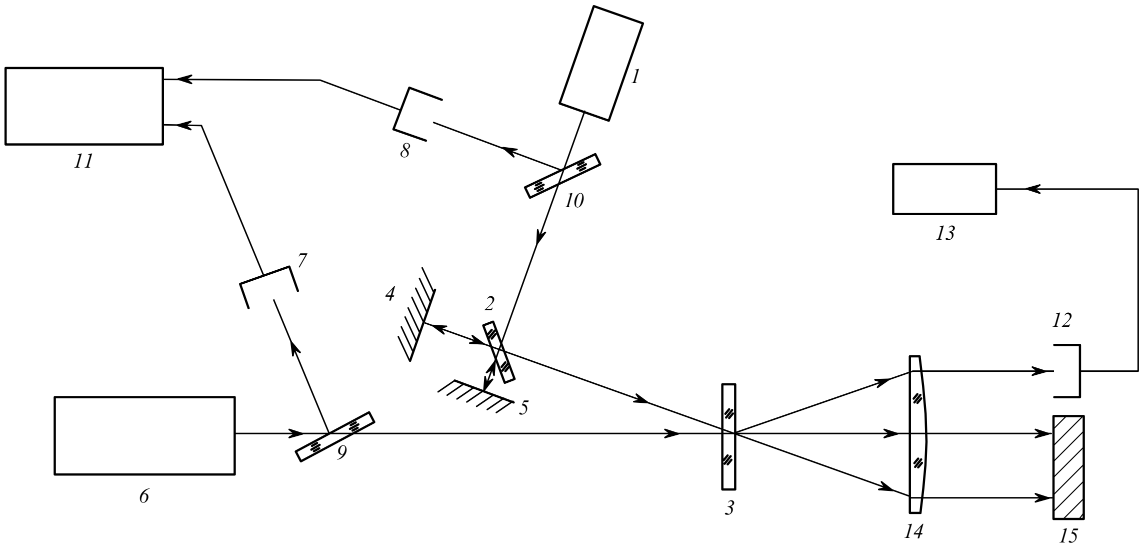


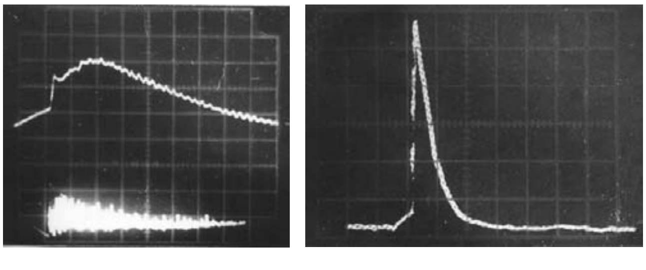
Figure 2. Scheme of the experimental setup: (*1*) Nd : YAG laser; (*2*, *9*, *10*) beamsplitters; (*3*) sample; (*4*, *5*) mirrors; (*6*) CO_2 laser; (*7*, *8*, *12*) photodetectors; (*11*, *13*) oscilloscopes; (*14*) lens; (*15*) absorbing screen.

power for the average intensity of the writing radiation 1.4 kW cm^{-2}. Figure 3 presents the oscillograms of CO_2-laser pulses incident on the sample and diffracted into the first order. Under the experimental conditions, $\tau_D = \Lambda^2/(4\pi^2 D) \approx 10^{-8}$ s. Because the duration of the light pulse substantially exceeds the relaxation time, apart from spatially periodic distributions, a uniform distribution of concentrations of electrons and holes also appears in the semiconductor. This process causes a rapid increase in the radiation absorption by free carriers at $\lambda_2 = 10.6$ μm and a corresponding decrease in the transmission coefficient of the semiconductor down to $T_2 = 0$. The oscillograms presented in Fig. 3 show this particular situation. An increasing absorption leads to a significant decrease in the pulse duration of diffracted radiation compared to the initial pulse.

The numeric estimate of the diffraction efficiency by expression (5) for the experimental conditions yields $\sim 0.6 \times 10^{-3}$, which is close to the experimental value. The parameters α_1, σ_e, and σ_h of germanium were borrowed from paper [6].

A completely different situation was observed in experiments with amorphous germanium deposited in the form of a 1.3-μm film on the BaF_2 substrate. Because of a short diffusion length and a low mobility of free carriers, which are typical of amorphous semiconductors [7], a uniformly distributed electron–hole plasma was almost absent in the film. In this case, $\tau_r \approx \tau_e \sim 1$ μs for amorphous germanium. One can see from oscillograms presented in Fig. 4 that no significant changes in the pulse shape and duration of the diffracted radiation compared to the initial pulse were observed, which corresponded to the result following from expression (5). The diffraction efficiency of the grating in these experiments was 3×10^{-4} for the same intensity of writing radiation (1.4 kW cm^{-2}) as in the previous experiment. The comparison of the experimental value η_2 with the numerical estimate have not been performed because data on σ_e and σ_h are absent for the experimental samples.

A low value of η_2 is explained by the fact that due to the small thickness of the film, it absorbed only about 30 % of radiation from the Nd : YAG laser. Note, in addition, that the layer of germanium was deposited on the substrate surface by the method of resistive spraying. It is known from literature [7] that the sample obtained in this way has a large number of structural defects, whose concentration achieves 10^{19} cm^{-3}. These defects are efficient traps for photoinduced



a b

Figure 3. Oscillograms of CO_2-laser (top) and Nd : YAG-laser (bottom) pulses (a) and of the CO_2-laser pulse diffracted on the grating in the crystalline germanium (b). Time sweeps of the oscillograms are 20 (a) and 1 μs div^{-1} (b).

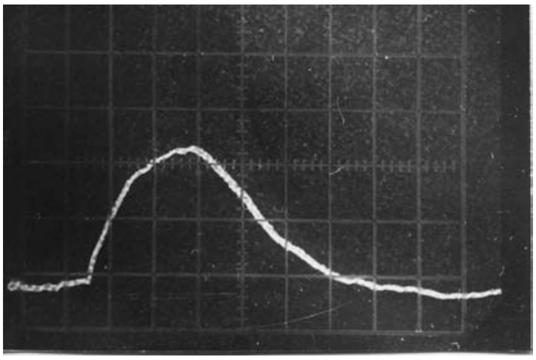


Figure 4. Oscillogram of the CO_2-laser pulse diffracted on the grating in the amorphous germanium. The time sweep of the oscillogram is 20 μs div^{-1}.

carriers, whose concentration in the experiments did not exceed 10^{17} cm^{-3}. In this case, almost all electrons were trapped, which explains the low grating efficiency obtained in the experiment.

To increase the grating efficiency, it is desirable to use hydrogenised amorphous semiconductors whose defects are filled with hydrogen atoms and where the centres of carrier trapping are virtually absent. Because in this case the concentration of carriers in the region of their excitation significantly increases at least by two orders of magnitude, a proportional increase in the efficiency to values larger than 10^{-2} should be expected.

4. Conclusions

The analysis of diffusion processes of nonequilibrium carriers in the volume of the semiconductor under conditions of spatial periodicity of their excitation has shown that to write dynamic gratings, semiconductor materials with a low ambipolar diffusion coefficient and, hence, with a low mobility of the charge carriers are preferable. A comparative estimate of the outlooks for the application of different semiconductor materials as media for writing dynamic gratings can be performed by using relation (5).

By detecting the energy and temporal pulse parameters of radiation diffracted on the grating and by comparing them with the initial ones, the relaxation time constant of the grating and the mobility of nonequilibrium carriers can be determined.

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