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Analysis of Temperature Modes of Hologram Recording on Photothermoplastic Materials

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Abstract

Theoretical study of the process of hologram recording on photothermoplastic media in which a linear relationship between the change in the refractive index and the temperature expansion of the medium is assumed. The results of the study allow us to estimate the required laser radiation power for the information recording as the function of the spatial frequency and the radiation exposure duration.

A solution was found for the heat conduction equation for photothermoplastic materials heated by an interference laser field. The solution obtained allows us to determine the required value of the recording temperature for given spatial frequencies, depending on the thermophysical parameters of the medium, and also on the power and duration of the heating radiation.

The use of the results of the study made it possible to find analytical expressions for the dependence of the diffraction efficiency of thermal gratings on the spatial frequency, pulse duration, and thermophysical parameters of the medium.

It is established that to increase the recording density in these media, materials with a low thermal diffusivity are necessary, while recording should be performed by the short pulses to minimize the length of thermal diffusion.

Keywords: Diffraction efficiency; Hologram; Photothermoplastic materials; Temperature; Heating

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1. Introduction

There is a class of materials in which their optical and spectroscopic properties under irradiation vary depending on the density of the incident energy and the process is accompanied by the formation of a modulated thermal field, leading to a change in the optical characteristics of the medium. The mechanism of changing of the optical parameters responsible for the change in the permittivity of a substance due to the

action of a modulated thermal field can be different [1 - 2]. One of them is associated with the expansion of the medium under the action of heat and, accordingly, a change in its density. The change in the density of the medium leads to a change in its refractive index, which can be used to modulate an external light beam. Depending on whether the imaginary, real or both parts of the substance permittivity change, an amplitude, phase or amplitude-phase lattice, respectively, is formed.

The practical application of such a scheme is connected with the finding of a medium having a sufficiently high expansion coefficient. A number of semiconductor materials, for example, chalcogenide vitreous semiconductors (including As_2Se_3) [2 - 7] and certain types of liquid crystals [8 - 11], in which the action of a heat pulse of a certain magnitude leads to reversible and irreversible changes of optical parameters, have this property. The intensity distribution can be transformed into a temperature distribution by weak absorption and heating in liquid crystals doped with dyes, which make it possible to increase the absorption of the recording light beams and produce heating even at low light intensities [10 - 11].

In these materials a linear relationship between the amplitude of the refractive index changing Δn and the amplitude of the substance temperature changing ΔT exists for sufficiently large temperature ranges [7, 11]

$$\Delta n = \frac{dn}{dT}\Delta T.$$

Here $\frac{dn}{dT}$ is the thermo-optical coefficient of the refractive index. Since the intermediate temperature gratings have a lifetime from fractions of a nanosecond to milliseconds, the resulting optical lattice must be frozen to preserve information for a long time [11] or during the recording the information is rewritten in parallel to another recording medium [1, 2]. In many thermo-recording media, the freezing process occurs as a result of the transition of the medium to the vitrified state [11]. A large amount of experimental data was collected [1 - 11] on the mechanisms of photothermal recording of information in such media. Experiment [7] shows that the thermo-optical gradient remains constant for a sufficiently large temperature interval $\Delta T \approx 300K$. In particular, the thermo-optical index of the refractive index of arsenic selenide As_2Se_3 at a wavelength $\lambda = 0.9\mu m$ is equal to $\frac{dn}{dT} = 2.87 \cdot 10^{-4}K^{-1}$, and when a temperature change is equal to $100K$ then its refractive index changes by $\Delta n = 0.2$ [7] and the hologram can be recorded with a higher resolution.

The main disadvantage of these media is the need to heat the irradiated region of the film to a temperature that is close to its melting point to record data. This circumstance limits the possibilities of improving the technical parameters of recording

media. Clearly, it is necessary to specify the power of the energy pulse with great accuracy, since a relatively small overheating of the film leads to its irreversible structural destruction, for example, to decay into droplets. Or vice versa, underheating to the desired temperature leads to the fact that the information will not be recorded. Such stringent requirements for the energy pulse parameters greatly complicate the use of recording media. Carrying out theoretical studies that allow to calculate the values of the given power of the energy pulse to obtain optimal temperature conditions for the surface of the recording layer as a function of its thermophysical characteristics and taking into account the parameters of the radiation source is one of the solutions to the problem. Publications on this issue are not enough. In the present work, a theoretical study of the features of the formation of flat holograms in absorbing substances changing their refractive index as a result of heat release is considered. Since the coefficient $\frac{dn}{dT}$ and the heat capacity of the substance remain constant and the temperature phase change is linearly related to the temperature change, then the study of the kinetics of the formation of the diffraction maximums of the thermal lattice is reduced to an examination of the thermal mode of the absorbing layer.

2. Heating of photothermal materials in an interference field created by laser

Typically, the recording medium is a kind of a layered dielectric-semiconductor structure that is formed from a recording layer enclosed between two transparent coatings, one of which is a substrate and the other is a thermally insulating film. Let two plane coherent waves of wavelength λ fall onto such a recording layer at angles $\pm\frac{\theta}{2}$ to the normal to the surface. Due to interference, the density of the incident energy varies periodically along the y axis. As a result of light absorption in the layer, an appropriate temperature field is induced in the form of a certain thermal lattice. When certain amplitude of the temperature field is reached, the periodic structure of the radiation is registered by means of a corresponding change in the optical parameters, i.e. the refractive index of the layer material. Thus, under the influence of radiation, the properties of the material change. These properties in the general case depend not only on the coordinate *y*, but also on the coordinate *z* and the time *t*.

The distribution of spatially inhomogeneous surface heat sources in a thin semiconductor layer is determined by the nature of this interference pattern formed by

two coherent beams with intensities described by the expression for the case of a rectangular pulse

$$q(y,z) = q_0(z)[1 + m\cos(ky)], \tag{1}$$

where $q_0(z)$ is the heat output averaged over the volume; $k = 2\pi/\Lambda$- spatial frequency; Λ and m - period and depth of modulation of the light energy density in a semiconductor layer; ($\Lambda = \lambda/(2\cos\theta)$- period of the spatial thermal lattice, λ- wavelength of laser radiation, θ- angle of incidence of a light beam on a layer).

In the general case, $q_0(z)$ is a function of the coordinate z, and the distribution of the temperature field $T(y,z,t)$ in the layer is determined by the solution of the thermal conductivity equation (2) with the heat source (1)

$$\frac{\partial T}{\partial t} = a\left(\frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right) + \frac{a}{\eta}q(y,z). \tag{2}$$

Equation (2) is solved under the following conditions: the boundary z = 0 will be considered thermally insulated, that is

$$\frac{\partial T(y,0,t)}{\partial z} = 0, \tag{3}$$

and the absorbing semiconductor layer d is sufficiently thick ($d \to \infty$), so that the influence of the substrate can be neglected, i.e.

$$T(y,z,t) = 0. \tag{4}$$

The initial condition for the induced value of the temperature field $T(y,z,t)$is given

$$T(y,z,0) = T_0, \tag{5}$$

where T_0 is the temperature of an environment.

Here, a and η, are the coefficients of thermal diffusivity and thermal conductivity of the material, respectively.

Let us consider, depending on the conditions of the problem, various approximate solutions of the two-dimensional heat equation (2).

1. Consider the case when the recording of information is carried out in a pulsed mode and at a high radiation energy density. In this case conditions $\sqrt{at} < \Lambda$ and $\sqrt{at} < d$ with good accuracy are satisfied, i.e. the diffusion length is less than the period of the thermal lattice and the thickness of the film. In this case, neglecting the second derivatives with respect to the two coordinates y and z in (2), we easily find the temperature changes in the antinodes of the interference fringes along the y axis on the surface (z = 0) of the semiconductor layer.

$$T(y,t) - T_0 = \Delta T(y,t) = \frac{a}{\eta}q_0 t + \frac{a}{\eta}mq_0 t\cos(ky) = \frac{a}{\eta}q_0(z)[1 + m\cos(ky)]. \tag{6}$$

The first term describes the temperature of the surface under uniform illumination (k = o), and the second term - the contribution from the modulation of the interference of light waves, which depends on the parameter k. In the process of forming the thermal lattice, the spatial distribution of the phase of the temperature wave on the surface does not change with time, and remains proportional to $\cos(ky)$, i.e. has a harmonic character. The surface temperature depends directly on the amount of heat released $Q = q_0 t$ inside the semiconductor layer during the exposure time of the laser radiation and makes it possible to evaluate the temperature information recording modes. Therefore, in the case under consideration the process of forming the thermal lattice has a cumulative character within the framework of applying the above approximation. However, the recording of information occurs when the temperature field changes, which in this case is determined by the modulation of the intensity of the object and reference beams (I_1, I_2) in the layer (7):

$$M(k,t) = m = \frac{2\sqrt{I_1 I_2}}{I_1 + I_2}. \tag{7}$$

The temperature at the antinodes of the interference fringes will be higher by an amount m than the background temperature and $m < 1$, if $I_1 \neq I_2$.

As stated above, the temperature change in the phase of the heat wave $\Delta\Phi$ in the absorbing layer depends on the property of matter and is determined from the expression [2]:

$$\Delta\Phi = kd\Delta n = kd\frac{dn}{dT}\Delta T. \tag{8}$$

As is known, the values $\frac{dn}{dT}$ for the same substance at constant pressure $\left(\frac{dn}{dT}\right)_P$ and at a constant volume $\left(\frac{dn}{dT}\right)_V$ can differ substantially. When a pulsed action on a substance, pressure waves arise that propagate at the speed of sound. If t_s is the time for equalizing the pressure in the substance, and t is the observation time, then $\frac{dn}{dT} = \left(\frac{dn}{dT}\right)_P$ for $t > t_s$, and $\frac{dn}{dT} = \left(\frac{dn}{dT}\right)_V$ for $t < t_s$. The substitution of (6) in (8) leads to the following expression for the phase:

$$\Delta\Phi = kd\frac{dn}{dT}\frac{a}{\eta}q_0 t[1 + mcos(ky)]. \tag{9}$$

The diffractive properties of light-induced gratings will be described using the concept of diffraction efficiency (DE). According to (9), the complex transmission of the thermal lattice is defined as

$$t_r = e^{i\Delta\Phi} = \exp\left(ikd\frac{dn}{dT}\frac{a}{\eta}q_0 t\right)\exp\left[imkd\frac{dn}{dT}\frac{a}{\eta}q_0 t\cos(ky)\right]. \tag{10}$$

Taking into account the formula $e^{iz\cos\phi} = \sum_{m=-\infty}^{\infty} i^m J_m(z) e^{im\phi}$ the final expression for the diffraction efficiency (DE) of a first-order thin phase hologram according to the relation [2] can be represented in next form:

$$\eta_1(t) = SJ_1^2\left(kd\frac{dn}{dT}\frac{a}{\eta}mQ(t)\right), \tag{11}$$

Where $J_1(z)$ is the first order Bessel function of the first kind which describes the amplitude of the diffracted waves, S is the transmission of a layer of thickness d at the wavelength λ of the diffracted radiation, $Q(t) = q_0 t$ is the amount of energy absorbed by the medium. As can be seen, the brightness of the diffraction maxima depends on the amount of heat inside the semiconductor material released by the time t. For small arguments of the Bessel function one can use the approximate formula

$$\eta_1(t) = S\left(kd\frac{dn}{dT}\frac{a}{\eta}mQ(t)\right)^2. \tag{12}$$

It can be seen that the DE depends quadratically on the density of the absorbed energy. As an illustration, Figure 1 shows the dependence η_1 on $Q(t) = q_0 t(\frac{J}{cm^3})$ for the As_2Se_3, and for $\eta = 0.5 - 0.75\frac{W}{m\cdot K}$,

$a = 2\cdot 10^{-6}\frac{m^2}{s}$, $\frac{a}{\eta} = 4\frac{cm^3\cdot K}{J}$, $\frac{2\pi d}{\lambda\cos\theta} = kd = 1000$, $\frac{dn}{dT} = 2.87\cdot 10^{-4}K^{-1}$ and $m = 1$.The maximum value of $\frac{\eta_1(t)}{S}$ equal to 33.9% is achieved at$Q(t) = 1.61\frac{J}{cm^3}$.The increase $Q(t)$ to $3.36\frac{J}{cm^3}$ is accompanied by a drop of η_1 to zero. We consider the solution of (2) for the case when conditions (1), (3) - (5) are satisfied, and the absorption of radiation obeys the Bouguer-Lambert law$q_0(z) = I_0 Ae^{-\alpha z} = q_0 e^{-\alpha z}$. Here $I_0$is the intensity of light at the entrance to the substance, $A$is the absorption coefficient of the substance.

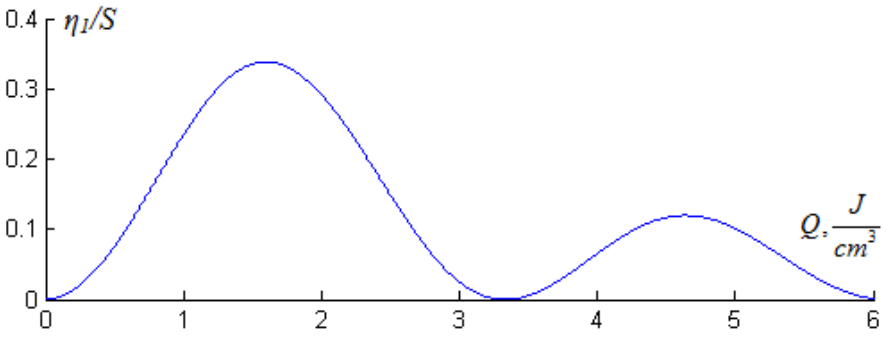

Figure 1: Diffraction efficiency of the thermal phase lattice as a function of the absorbed energy of the recording layer.

The temperature field on the surface can be represented in the form of two terms:

$$T(y,t) = T_1(t) + T_2(y,t). \tag{13}$$

The first of them describes the temperature of the surface of the recording layer when it is uniformly illuminated, i.e. when $k = 0$ (the contribution of the first term in expression (1)). It creates a common background of the temperature field

$$T_1(z,t) = \frac{2I_0A\sqrt{at}}{\eta} i\Phi^*\left(\frac{z}{2\sqrt{at}}\right) + T_0, \tag{14}$$

where $i\Phi^*(s)$ is the integral of the function $\Phi^*(s) = erfc(s) = 1 - erf(s) = 1 - \Phi(s)$, here $\Phi(s)$is the error integral or the Laplace function.

The second term is the contribution of the spatially modulated component, and it has the form:

$$T_2(y,z,t) = \frac{I_0Am}{\eta k}\Phi(k\sqrt{at})\cos(ky)e^{-kz}. \tag{15}$$

Then, according to (13), the total surface temperature of the recording layer $(z = 0)$is determined from the relation

$$\begin{aligned} T(y,t) - T_0 &= \Delta T(y,t) = 2\frac{I_0A\sqrt{at}}{\eta} + \frac{I_0Am}{\eta k}\Phi(k\sqrt{at})\cos(ky) \\ &= 2\frac{I_0A\sqrt{at}}{\eta}[1 + M(k,t)\cos(ky)], \end{aligned} \tag{16}$$

where the coefficient (or amplitude) of the modulation $M(k,t)$ of the temperature field of the interference fringes on the surface along the *y* axis is determined from the relation $M(k,t) = (\Delta T_{max} - \Delta T_{min})/(\Delta T_{\max} + \Delta T_{\min})$ and has the form:

$$M(k,t) = \frac{m}{2k\sqrt{at}}\Phi(k\sqrt{at}). \tag{17}$$

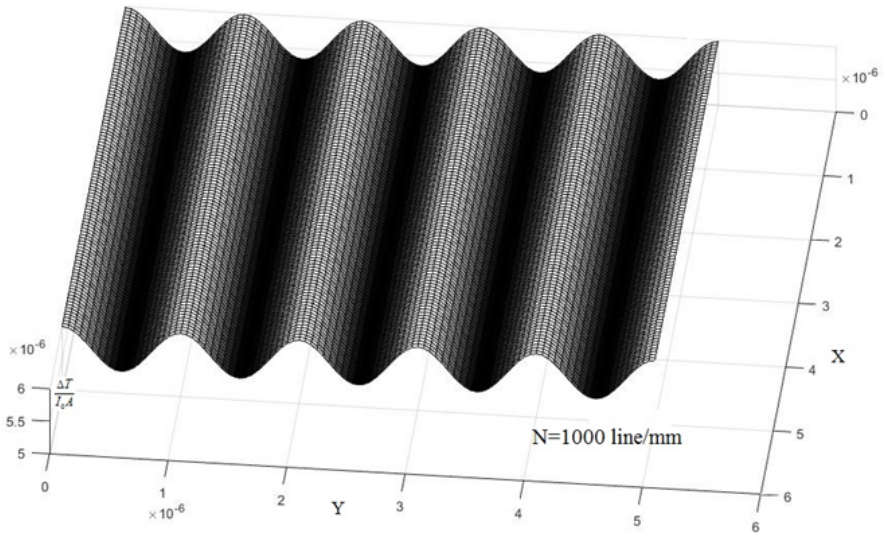

Figure 2: The relief of the temperature field of the medium irradiated by laser radiation.

As can be seen from this expression, the modulation factor depends on the spatial frequency, on the thermal diffusivity of the material, on the duration of the action of

the laser radiation, and on the modulation of the reference and object beams. It does not depend on the power of the heating radiation and on the thermal conductivity of the material. The temperature at the antinodes of the interference fringes, according to (16), is determined by the relation

$$T_2(y,0,t) = \frac{I_0 Am}{\eta k}\Phi(k\sqrt{at})\cos(ky),$$

and it can be considered as a time-dependent Laplace function. Figure 2 shows an example of the distribution of a harmonic temperature field in a medium illuminated by laser radiation. The spatial frequency of the interference pattern is N = 1000 / mm. Depending on the spatial frequency, T_2 grows as rapidly, as the Laplace function, but decreases slowly, as k^{-1}. This circumstance creates a limitation for the recording of information associated with the temperature regime, since for recording purposes the value of the temperature rise at the antinodes of the interference fringes should not exceed the melting point of the test substance and should not be less than the recording temperature, i.e. $T_{rec} < \Delta T(k) < T_{melt}$. The operating temperature range for the recording of information is determined from the experiment, for example, for arsenic selenide $As_2Se_3 T_{rec} \approx 150^0 C$[1], and $T_{melt} = 180^0 C$[2]. Therefore, for each value of the energy of the recording pulse, there is a range of spatial frequencies that will be recorded when the recording temperature falls within the recording temperature range $T_{rec} < \Delta T(k) < T_{melt}$. The limitation of spatial frequencies is related to T_{rec}, since for large *k* the temperature *T* becomes smaller than T_{rec}, and the information will not be recorded. For recording, it is necessary to increase the energy of the radiation pulse, i.e. the power of the radiation source.

Using the properties of the function $\Phi(x)$ such as $\Phi(0) = 0, \Phi(\pm\infty) = \pm 1, \Phi(x > 2.7) \approx 1$ [8], we consider how the modulation coefficient of the thermal field changes as a function of the parameters. Taking into account the condition$\Phi(x > 2.7) \approx 1$, we can write that

$$k\sqrt{at} \approx 2.7. \tag{18}$$

The fulfillment of condition (18) can be achieved in two ways: 1). If the exposure time is large,$t = t_{border} \geq \frac{7.29}{k^2 a}$, and $k = const$. But note that for each spatial frequency there exists a certain value of t_{border}, which decreases with increasing *k*. 2). If the exposure time is fixed, i.e. $t = const$, then condition (18) can be satisfied by changing the given spatial frequency

$$k = k_{border} \geq \frac{2.7}{\sqrt{at}}. \tag{19}$$

In both cases, the modulation factor of the thermal field will tend to

$$M(k,t) \approx \frac{m}{2k\sqrt{at}}, \tag{20}$$

And the corresponding value of the temperature at the antinodes of the interference fringes becomes

$$T_2(y,0,t) = \frac{I_0 Am}{\eta k}\cos(ky). \tag{21}$$

For small values of the argument $k\sqrt{at} << 2.7$, the Laplace function takes the form $\Phi(k\sqrt{at}) \approx \frac{2}{\sqrt{\pi}}k\sqrt{at}$ and for the depth of modulation of the thermal field we obtain expression

$$M(k,t) \approx \frac{m}{\sqrt{\pi}}. \tag{22}$$

The temperature of the interference bands $T_2(y,0,t)$will vary according to the law

$$T_2(y,0,t) = \frac{2m}{\sqrt{\pi}}\frac{I_0 A\sqrt{at}}{\eta}\cos(ky), \tag{23}$$

and its amplitude does not depend on the spatial frequency.

Now, in accordance with (8), writing down the expression for the phase of the thermal wave as

$$\Delta\Phi = kd\frac{dn}{dT}\frac{2I_0 A\sqrt{at}}{\eta}[1 + M(k,t)\cos(ky)], \tag{24}$$

we have for the diffraction efficiency of the thermal phase lattice, by analogy with (10), the following relation:

$$\eta_1(t) = SJ_1^2[kd\frac{dn}{dT}\frac{2I_0 A\sqrt{at}}{\eta}M(k,t)]. \tag{25}$$

Taking into account (17), relation (25) can be rewritten in the following form:

$$\eta_1(t) = SJ_1^2[m\frac{dn}{dT}\frac{I_0 Ad}{\eta}\Phi(k\sqrt{at})]. \tag{26}$$

When $k\sqrt{at} << 2.7$ expression (26), according to (22), takes the form:

$$\eta_1(t) = SJ_1^2(kd\frac{dn}{dT}\frac{2I_0 A\sqrt{at}}{\eta}\frac{m}{\sqrt{\pi}}). \tag{27}$$

And when $k\sqrt{at} >> 2.7$, according to (20), we have:

$$\eta_2(t) = SJ_1^2(m\frac{dn}{dT}\frac{I_0 Ad}{\eta}). \tag{28}$$

For small arguments $m\frac{dn}{dT}\frac{I_0Ad}{\eta}\Phi(k\sqrt{at}) << 1$ and $m\frac{dn}{dT}\frac{I_0Ad}{\eta} << 1$, then (26) and (28) can be rewritten in the form:

$$\eta_1(t) = S[m\frac{dn}{dT}\frac{I_0Ad}{\eta}\Phi(k\sqrt{at})]^2, \tag{29}$$

$$\eta_2(t) = S(m\frac{dn}{dT}\frac{I_0Ad}{\eta})^2. \tag{30}$$

Then the relation

$$\frac{\eta_1(t)}{\eta_2(t)} = \Phi^2(k\sqrt{at}) \tag{31}$$

behaves as a square of the Laplace function, depending on the argument $k\sqrt{at}$.

We note that the transition (26) to (28) is possible only under the condition that the function $\Phi(k\sqrt{at}) = 1$. If the condition $k\sqrt{at} >> 2.7$ is satisfied for a fixed value $k = k_{fix}$, then the power density of the heat source $q_0 = I_0A$ is chosen for this value $k_{fix}$and not changed during the exposure time in (28). Another situation, if the condition $k\sqrt{at} >> 2.7$ is satisfied for a fixed value $t = const$, then with increasing k, there will be a need to increase the pulse energy in accordance with (21), so that the condition $T_2 - T_0 = \Delta T > T_{record}$ takes place. In other words, the temperature change in the interference fringes must be higher than the recording temperature in order to register the information. Therefore, when calculating the diffraction efficiency from the formula (26), it is necessary to take into account the moment that the value $q_0 = I_0A$should correspond to those values of k when the function $\Phi(k\sqrt{at}) = 1$. Therefore, the stationary values of the diffraction efficiency will take different values depending on the value q_0, which is determined from the relation $q_0 = T_{record}\frac{\eta k}{m}$, i.e. depending on the recorded spatial frequency.

3. Conclusion

Formulas that make it possible to estimate the required laser radiation power for recording information in the material under study, depending on the spatial frequency and the duration of the radiation exposure are obtained. It is shown that the amplitude of the modulation of the temperature field in the interference bands depends on the value of the given spatial frequency, and the recording of high frequencies requires an increase in the energy of the recording pulse. An analytical expression is found for calculating the diffraction efficiency of thermal gratings in the case of recording information due to the mechanism of thermal expansion of matter in photothermal media. It has been found that in order to increase the recording density in these media,

it is necessary to select a material with a low thermal diffusivity and the recording is preferably carried out with short pulses to minimize the length of thermal diffusion.

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