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INTERACTION OF LASER RADIATION WITH MATTER

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Laser swelling model for polymers irradiated by nanosecond pulses

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Abstract. **Mechanisms of laser swelling of polymers are considered. A theoretical model for one of such mechanisms is constructed and investigated. This mechanism is based on the formation of a thermoelastic wave upon absorption of a laser pulse. Tensile stresses in this wave lead to elastic and plastic deformation of a polymer in the heated region and to the formation of convex structures (humps). The threshold fluence of a laser pulse required for the production of a residual hump under laser irradiation is obtained analytically. A formula for the height of this hump is also derived. The model explains the earlier experimental data from the literature on swelling of a PMMA film irradiated by UV pulses.**

Keywords: *laser swelling, polymer, ultraviolet laser pulses.*

1. Introduction

The interest in laser swelling of polymer materials and biological tissues is due to the application of this effect in modern laser microtechnologies [1 – 5] and in medicine [6]. Swelling of polymer materials was used for obtaining an array of optical microscopic lenses on the polymer surface [1]. The formation of a hump structure and a dent on the polyimide surface irradiated by laser pulses with an energy below the evaporation threshold was studied in [2, 3]. The expansion and contraction dynamics during polymer swelling was studied by the method proposed in [4, 5]. Analysis of swelling dynamics in biological tissues was carried out in [7 – 9]. A model of swelling constructed for a soft biological tissue gave quantitative agreement with experimental data [8, 9]. The main fields of application of laser swelling of polymers are integrated optics, photonics, and micro- and nanopatterning of surfaces. Near-field optical systems can be used for obtaining hump nanostructures.

In spite of a large number of publications on laser swelling of polymers, this effect has been studied insufficiently. The experimental results described in the literature reflect both the ultimate result of swelling (residual hump) and the temporal dynamics of the process of formation and relaxation of the hump structure. Attempts are being made to explain the obtained results and possible swelling mechanisms are considered. However, simulation of this effect has not been carried out. In this study, we consider a simple and original model of polymer swelling, which explains the formation of stationary structures on the surface of the material. This model allows us to calculate the threshold fluence required for the emergence of residual swelling and to estimate the swelling dynamics.

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2. Mechanisms of laser swelling of polymers

Laser swelling is bulging of a medium in the region of radiation absorption and formation of hump structures. As a result of swelling, residual (nonvanishing) humps may be formed, but complete relaxation of the hump structure after irradiation is also possible. The emergence of residual humps during swelling is attributed to the crystal – amorphous phase transition, melting, or plastic flow of the material.

Laser swelling of polymer materials occurs when a laser pulse fluence is below the threshold value for intense removal or destruction of the material. The polymer material is heated by a laser pulse to a temperature above that corresponding to the highly elastic state of the material, so that it can be deformed elastically or plastically by the internal pressure in the medium. Consider a number of swelling mechanisms with taking into account the internal pressure in the medium, which causes deformation of the material.

One of the mechanisms can be described as follows. Under the action of a short (nanosecond) laser pulse absorbed by the medium, a thermoelastic stress wave is formed and propagates to the bulk of the medium. Positive stresses (unloading) in this wave represent a nonstationary internal pressure causing elastoplastic deformation in the heated region of the material. The total (peak) strain in this case is the sum of the strain caused by thermal expansion and elastic and plastic deformation of the medium. An irreversible plastic deformation, remaining after the relaxation of the elastic component of deformation and deformation associated with thermal expansion, determines the final height of the hump. The characteristic time of deformation of the medium and of the formation of a hump with the maximal height corresponds to the time of propagation of a thermoelastic unloading wave over the heated region of the medium.

Another mechanism is associated with the appearance of internal pressure in the medium, which is produced by gases created by a laser pulse. This pressure causes elastoplastic deformation in the medium heated by laser radiation due to the expansion of gases. Gases may be produced in the medium due to the photochemical or thermal decomposition of special components (porophores), depolymerisation of a polymer matrix, and evaporation of the solvent in the bulk of the polymer. An original model of swelling based on this mechanism was developed by us in [8, 9] for biological media.

The necessary condition for these swelling mechanisms in polymers is that a layer of the polymer material must be in a highly elastic state during the time interval of the development of the internal pressure in this layer. In this case, plastic deformation of the medium may occur with maximal strains without mechanical destruction of the material. The most suitable polymer media that can be subjected to the above mechanisms are thermoplastic polymers and composite materials which may contain dyes and porophores and low-boiling solvents.

One more swelling mechanism involves melting of a polymer in the region heated upon absorption of a laser pulse (accompanied by an increase in volume of this region) followed by rapid cooling and solidification of the material over a time shorter than the relaxation time of the free volume in the medium. This leads to a decrease in the density of the material and, accordingly, to an increase in the volume of the irradiated medium as compared to the unexposed medium, leading to the formation of a stationary hump structure.

3. Simulation of laser swelling of polymers

We consider here the first of the above-mentioned mechanisms and construct a model of polymer swelling, in which swelling is associated with the thermoelastic response of the medium to laser radiation and with the emerging elastoplastic deformation of the medium in the region of radiation absorption. For simplicity, we assume that the laser pulse is rectangular. The polymer material has linear absorption at the laser wavelength.

Let a laser pulse be absorbed in the medium, giving rise to a thermoelastic stress wave propagating to the bulk of the medium. When tensile stresses in this wave 'propagate' through the near-surface layer of the medium heated to a temperature above that corresponding to the highly elastic state, elastic and plastic strains appear in this layer. The region of plastic deformation appears at the sites, where tensile stresses exceed the critical stress of plastic flow. In addition to the emergence of elastic and plastic deformations in the material, the volume of the heated region of the medium, which is associated with thermal expansion, increases. Consider the equations of the one-dimensional laser swelling model. The laser radiation intensity distribution in the medium is described by the equation

$$I = I_0 \exp(-\alpha z), \tag{1}$$

where z is the coordinate directed to the bulk of the medium; α is the linear absorption coefficient; and I is the radiation intensity in the medium. The one-dimensional thermal conductivity equation has the form

$$\frac{\partial T}{\partial t} = \chi \frac{\partial^2 T}{\partial z^2} + \frac{\alpha I}{c_p \rho}, \tag{2}$$

where T, ρ, c_p, and χ are the temperature, density, heat capacity, and thermal diffusivity of the polymer, respectively. We will neglect heat conduction in the further analysis of swelling because the duration of this process is longer than that of formation of the hump structure studied here. The hump emerges on the nanosecond time scale. For the absorption coefficient $\alpha \sim 400$ cm^{-1} in the medium, the cooling time for the heated region is a few milliseconds, while this time for 2-μm-thick film samples is $\sim$40 μs. We consider the time dynamics of swelling over shorter times of the order of hundreds of nanoseconds.

In the one-dimensional case under unilateral deformation [10], the stress tensor has the z component only. The z axis is directed to the bulk of the medium. The thermoelasticity equation has the form

$$\frac{\partial^2 \sigma}{\partial t^2} = c_s^2 \frac{\partial^2 \sigma}{\partial z^2} - \frac{\beta_v E_Y}{3(1-2\mu_P)} \frac{\partial^2 T}{\partial t^2}, \quad \sigma(t,0) = 0. \tag{3}$$

Here, σ is the z component of the stress tensor; E_Y is Young's modulus of the material; β_v is the volume expansion coefficient; μ_P is the Poisson coefficient; and c_s is the velocity of sound in the medium. According to [11], the expression for a rectangular laser pulse of duration t_p for nonstationary tensile stress σ_t in a thermoelastic wave for $t \geqslant t_p + z/c_s$ has the form

$$\sigma_t = \frac{\beta_v E_Y I_0}{3(1-\mu_P) c_s c_p \rho} \sinh(\alpha z) \exp(-\alpha c_s t)[\exp(\alpha c_s t_p) - 1]. \tag{4}$$

The maximum tensile stress in the cross section z has the form

$$\sigma_{tm} = \frac{\beta_v E_Y I_0}{6(1-\mu_P) c_s c_p \rho} [1 - \exp(-2\alpha z)][1 - \exp(-\alpha c_s t_p)]. \tag{5}$$

Let us describe the polymer medium using the simplest mechanical–mathematical model. Consider a one-dimensional elastoplastic medium with linear strain-hardening [12]. In the range of tensile stresses, the elastoplastic strain ε of the medium in each section z at instant t is determined by

$$\varepsilon = \begin{cases} \sigma_t / E_Y, & \sigma_t < \sigma_p, \\ \varepsilon_p, & \sigma_t = \sigma_p, \\ \varepsilon_p + (\sigma_t - \sigma_p)/E_p, & \sigma_t > \sigma_p, \quad \dot{\sigma}_t > 0, \\ \varepsilon_m, & \sigma_t = \sigma_{tm}, \quad \dot{\sigma}_t = 0, \\ \varepsilon_m + (\sigma_t - \sigma_{tm})/E_Y, & \dot{\sigma}_t < 0. \end{cases} \tag{6}$$

Here, σ_p is the threshold stress corresponding to plastic flow of the material; ε_p is the strain of the medium at the initial instant of plastic flow; E_p is the plastic modulus; ε_m is the maximal strain attained at point z of the medium under plastic deformation; and $\dot{\sigma}_t = \mathrm{d}\sigma_t/\mathrm{d}t$ is the total derivative of tensile stress with respect to time at point z of the medium. If we know the strain at each instant at each point of the medium, we can easily calculate the height h of the emerging hump by integrating over the region of elastoplastic deformation of the medium:

$$h(t) = \int_0^{z_m} \varepsilon(z,t)\mathrm{d}z. \tag{7}$$

The height of the residual hump h_{pl} of the structure can easily be calculated using the formula

$$h_{pl} = \int_{z_1}^{z_2} \varepsilon_{end}(\sigma_{tm}(z))dz, \quad (8)$$

where ε_{end} is the end plastic strain of the medium at each section z, which is determined by maximal tensile stresses in this cross section. Coordinates z_1 and z_2 determine the boundaries of the plasticity region. The value of ε_{end} can be calculated by the formula

$$\varepsilon_{end} = \sigma_{tm}\left(\frac{1}{E_p} - \frac{1}{E_Y}\right) + \varepsilon_p - \frac{\sigma_p}{E_p}. \quad (9)$$

Plastic strains emerge in the absorption region of the medium, where maximal tensile stresses are attained and where the temperature is higher than the plasticity temperature T_p. Since tensile stresses in a thermoelastic wave and the temperature in the medium are determined by the laser pulse fluence, the plasticity region emerges in a threshold manner as regards the pulse fluence.

Let the region of the medium between coordinates z_1 and z_2 be subjected to plastic deformation. Coordinate z_1 is determined from the condition that the maximal tensile stress at this point is equal to the critical stress σ_p of plastic deformation. Coordinate z_2 is determined from the condition that the temperature decreasing towards the bulk of the medium is equal at this point to the temperature of transition of the material to the highly elastic state. The expressions for z_1 and z_2 have the form

$$z_1 = \frac{1}{2\alpha}\ln\frac{A}{A-\sigma_p},\ z_2 = \frac{1}{\alpha}\ln\frac{\alpha F}{(T_p - T_0)c_p\rho}, \quad (10)$$

where

$$A = \frac{\beta_v E_Y I_0}{6(1-\mu_P)c_s c_p \rho}[1 - \exp(-\alpha c_s t_p)];$$

T_0 is the initial temperature of the medium prior to laser irradiation; and F is the fluence of the laser pulse.

The formation of the plasticity region corresponds to the coincidence of the boundary coordinates ($z_1 = z_2$). For $z_1 < z_2$, the plasticity region exists for a given pulse fluence. For $z_1 > z_2$, no plasticity region is formed since in this case the regions of high temperatures and pressures are separated in space. From the condition of coincidence of the coordinates, we can derive the expression for the threshold fluence F_{th} of the laser pulse, which is required for plastic deformation of the medium and for the emergence of the irreversible swelling effect,

$$F_{th} = \frac{1}{\alpha f(t_p/t_s)}\left\{A_1 + \left[A_1^2 + [f(t_p/t_s)A_2]^2\right]^{1/2}\right\}, \quad (11)$$

where

$$A_1 = \frac{3(1-\mu_P)c_p\rho\sigma_p}{\beta_v E_Y} \text{ and } A_2 = (T_p - T_0)c_p\rho$$

are parameters,

$$f(t_p/t_s) = \frac{1 - \exp(-t_p/t_s)}{t_p/t_s}$$

is the dimensionless function of the laser pulse duration, and $t_s = 1/(\alpha c_s)$ ($t_s = 12$ ns for $\alpha = 400\ \text{cm}^{-1}$) is the propagation time of an acoustic wave over the radiation absorption region. In the case considered here, parameters A_1 and A_2 differ substantially, i.e., $A_2 = 173\ \text{J cm}^{-3} \gg A_1 = 4\ \text{J cm}^{-3}$. Taking this into account, we obtain the following approximations. For $t_p \sim t_s$, it follows from relation (11) that $F_{th} \approx A_2/\alpha$; i.e., the threshold fluence is inversely proportional to the absorption coefficient of the medium and is almost independent of the laser pulse duration. Our calculations were performed precisely under such conditions. A substantial dependence of F_{th} on t_p emerges for $t_p \geqslant (A_2/A_1)t_s$ ($t_p \geqslant 520$ ns).

We can now calculate analytically the height of the residual hump in the case when the fluence of the laser pulse acting on the medium exceeds the threshold value F_{th}. By substituting expression (9) into (8) and using formulas (10) for the coordinates of the plasticity region, we obtain the following formula for the residual hump height h_{pl} relative to the unexposed surface:

$$h_{pl} = \left(\varepsilon_p - \frac{\sigma_p}{E_p}\right)(z_2 - z_1) + A\left(\frac{1}{E_p} - \frac{1}{E_Y}\right) \times\left\{(z_2 - z_1) - \frac{1}{2\alpha}[\exp(-2\alpha z_1) - \exp(-2\alpha z_2)]\right\}. \quad (12)$$

Consider the nonstationary component of the hump height h_t, which is determined by thermal expansion of the medium and its elastic strains. Thermal expansion leads to swelling of the medium in the region of heating upon irradiation by a laser pulse. As the thermoelastic stress wave propagates to the bulk of the medium, the latter expands. After the passage of the stress wave through the heated region, the hump structure is formed with a height, which is determined by thermal expansion and which can easily be calculated by integrating the product of the thermal expan-sion coefficient and the temperature increment at a given point as a result of laser heating. In our calculations, we assumed that the thermal expansion coefficient is doubled at the temperature of transition to the highly elastic state [5].

Elastic strains of the medium caused by tensile stresses make a small contribution to the hump height. The elastic component of the hump can be calculated by integrating elastic strains over the heated region of the medium. We assume that the elastic component decreases and vanishes when the thermoelastic wave leaves the region of radiation absorption.

The finite height of the residual hump can be calculated by expression (12) and the time dependence of the hump height can be found using general expression (7). The swelling model is described in greater detail in [13].

In the model described here, we assume that the polymer passes to the highly elastic state instantaneously at temperature T_p; however, the time of this transition obeys an activation dependence. Such a transition in polymers is called the alpha-relaxation transition [14]. As the temperature of the material passes through point T_p, segments in the polymer chain with a length of 3–5 monomer links become mobile, which changes mechanical properties of the material. Let us estimate the time τ of polymer transition to the highly elastic state using the formula [14]

$$\tau = b\exp\frac{E_{ac} - \gamma\sigma}{k_B T_p},$$

where b and γ are the parameters; E_{ac} is the activation energy of the transition; and k_B is the Boltzmann constant. This formula also takes into account the fact that the time of transition of the polymer to the highly elastic state depends on mechanical stresses. We consider induced highly elastic deformation under stresses. For elastomers including PMMA and having parameters $b \sim 5\times\ 10^{-12}$ s^{-1}, $E_{ac} \sim$ 0.31 eV (30 kJ mole^{-1}), $\gamma = 1.7 \times 10^{-27}$ m^3, $k_B T_p = 0.034$ eV, and characteristic stresses $\sigma \sim 20$ MPa ($\gamma\sigma = 0.21$ eV) in a thermoelastic wave, we obtain a transition time of $\tau \approx 0.1$ ns, which is much shorter than the laser pulse duration.

Thus, the swelling mechanism considered here is typical of elastomers, which rapidly pass to the highly elastic state. Such a swelling mechanism can be observed when the medium is heated to the melting point of the polymer (up to 493 K for PMMA).

Below, we will describe the results of calculations of the hump height in the surface of PMMA exposed to nanosecond laser pulses. The parameters of the PMMA sample used in our calculations are borrowed from [5, 15]: $E_Y = 3$ GPa, $\beta_p = 5.6\times10^{-4}$ K^{-1}, $\rho = 1.2$ g cm^{-3}, $c_p = 1.44$ J g^{-1} K^{-1}, $\mu_P = 0.35$, $c_s = 2 \times 10^5$ cm s^{-1}, $\varepsilon_p = 6.7 \times 10^{-4}$, $\sigma_p = 2$ MPa, $E_p = 30$ MPa, $T_p = 393$ K, $T_0 = 293$ K, $\alpha = 400$ cm^{-1}, and $t_p = 30$ ns.

4. Results of calculations and discussion

We will consider below the results of calculation of the height of a residual hump on the surface of the material for polymer swelling as a function of the pulse fluence. Calculations were made for the polymer PMMA. The chosen interval of laser pulse durations is typical of laser sources such as excimer lasers and Q-switched Ng:YAG lasers. The range of absorption coefficient of the material used in calculations is typical of intrinsic absorption of radiation from excimer lasers in polymers or absorption of radiation from solid-state lasers and its harmonics by polymer materials doped with dyes.

Calculations were made for swelling of PMMA exposed to UV laser pulses with a wavelength of 248 nm. Figure 1 shows the residual hump height relative to the unexposed surface as a function of the pulse fluence. A residual hump is formed after cooling of the medium to the initial temperature and relaxation of reversible deformations. The height of this structure can be calculated if the distribution of plastic strains over the depth of the medium is known. The calculations were made using formula (12) and are represented in Fig. 1 [curves (*1*–*7*)].

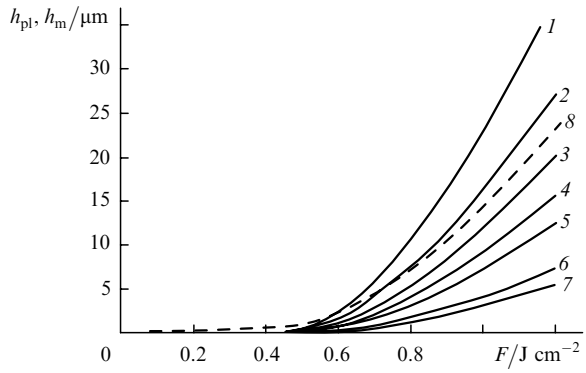

Figure 1. Residual hump height h_{pl} (*1*–*7*) and the maximum hump height h_m (*8*) as functions of the pulse fluence for laser pulse durations of 10 (*1*), 20 (*2*), 30 (*3*, *8*), 40 (*4*), 50 (*5*), 80 (*6*), and 100 ns (*7*).

Figure 1 shows that the dependences are of the threshold type and their behaviour depends on the laser pulse duration t_p. For a fixed pulse fluence, the hump height increases upon a decrease in the laser pulse duration, although the residual swelling thresholds are virtually the same. If the condition $t_p \ll t_s$ is satisfied, analysis of formula (12) taking into account expression (10) shows that the hump height for such short laser pulses will be fixed and will not increase.

Curve (*8*) in Fig. 1 shows the dependence of the maximal hump height attainable in swelling on the pulse fluence. The change in the behaviour of this curve is associated with a transition through the threshold F_{th} and with the emergence of plastic deformations in the medium. Below the threshold, only reversible swelling of the material occurs, which is associated with thermal expansion of the medium and with elastic deformation of the material. The height of reversible hump is proportional to the pulse fluence.

Figure 2a shows the dependence of the residual hump height on two parameters, namely, the pulse fluence and the absorption coefficient of the material. This two-parametric dependence allows us to analyse the ways for swelling optimisation in these parameters. It can be seen that, for

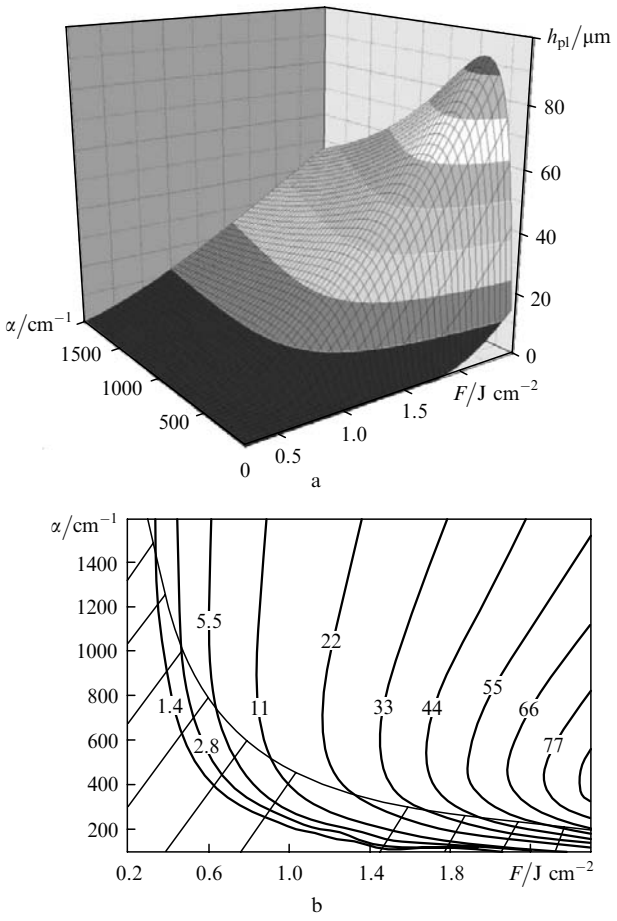

Figure 2. (a) Dependence of the residual hump height on the pulse fluence and on the absorption coefficient of the material; (b) level lines h_{pl} (in μm) corresponding to (a). The range of parameters in which residual swelling takes place is hatched; $t_p = 30$ ns.

a fixed pulse fluence, the dependence of the residual hump height on the absorption coefficient of the material has a peak; i.e., there exists an optimal absorption coefficient for obtaining the maximal effect. Thus, to obtain the maximum swelling effect for a given mode of irradiation, we must prepare a sample with an optimal absorption coefficient. However, in such optimisation, we disregard the range of parameters in which swelling takes place. Fig. 2b shows the level lines corresponding to the dependence depicted in Fig. 2a as well as the range of parameters (hatched) in which the swelling mechanism studied here is realised. The boundary of the region corresponds to heating of the surface of the material to the melting point of PMMA, which is equal to 493 K. Outside this region, swelling may be determined by other mechanisms associated with polymer melting. In the material heated to a temperature above 600 K, effects of destruction and laser ablation of the polymer will emerge and dominate.

Analysis of Fig. 2b shows that the range of parameters in which swelling is realised does not contain the optimal values of the absorption coefficient of the medium upon an increase in the pulse fluence. It can be seen that the swelling effect can be enhanced only by reducing the absorption coefficient of the medium and by increasing the pulse fluence. These conclusions are not universal and were drawn for the PMMA polymer subjected to nanosecond laser pulses. The situation for other polymers may be different.

Figure 3 illustrates the dynamics of hump formation. One can see that a hump structure is formed after the action of a laser pulse, when a thermoelastic wave of tensile stresses is formed and propagates to the bulk of the medium. Plastic deformation of the medium takes place during the propagation of peak tensile stresses of the thermoelastic wave in the heated region of the medium; the deformation time amounts to a few nanoseconds. The time of leading front build-up corresponds to this period. A small change in the height of the hump after the attainment of its peak value corresponds to relaxation of elastic stresses after the departure of the thermoelastic wave from the heated region of the medium. The discontinuity on the axis in Fig. 3 corresponds to the time of cooling of the medium, which amounts to a few milliseconds; after this, the hump height associated with thermal expansion relaxes completely.

Consider the results of calculations of swelling in a 2-μm-thick PMMA polymer film deposited on a quartz glass substrate. We simulated the experiment performed in [5], where a PMMA film was irradiated by 248-nm, 30-ns pulses from an excimer laser. The fluence of this pulse was lower than the laser ablation threshold of this material. In this experiment, the time dependences of the hump height and the dependences of its peak height on the pulse fluence were obtained.

The model of polymer material swelling considered above for a block sample was used for solving the problem of swelling of a PMMA film taking into account the fact that a thermoelastic wave is formed only in absorbing films. Figures 4 and 5 show the results of analysis of swelling of a polymer film and experimental data borrowed from [5], in which an excimer laser pulse ($\lambda = 248$ nm) of duration 30 ns acted on a PMMA film deposited on a quartz substrate. It follows from Fig. 4 that the dependence of the maximum hump height h_m on the pulse fluence successfully describes the experimental data both qualitatively and quantitatively. The characteristic kink on the $h_m(F)$ curve corresponds to the threshold for the emergence of plastic deformations in the film. The dashed line in Fig. 4 shows the theoretical dependence of the residual hump height $h_{pl}(F)$.

Figure 5 shows the dynamics of hump formation. The sharp leading edge on the theoretical time dependence of the hump height is formed due to rapid departure of the

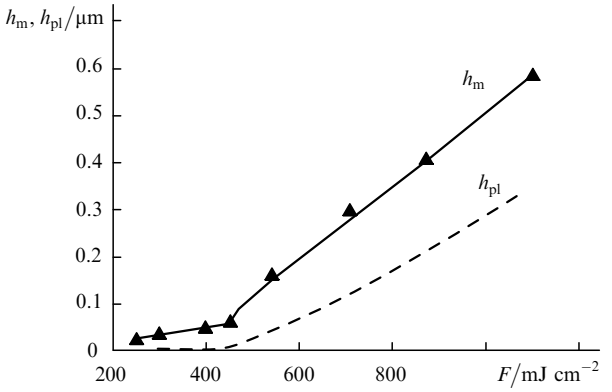

Figure 4. Dependence of the hump height on the pulse fluence. The solid curve corresponds to theoretical analysis of the maximal hump height h_m, the symbols are the corresponding experimental data from [5], and the dashed line is the theoretically calculated height h_{pl} of the residual hump.

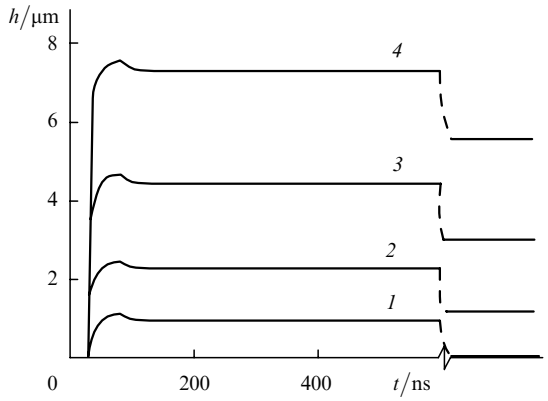

Figure 3. Time dependences of the hump height for the pulse fluence of 0.5 (*1*), 0.6 (*2*), 0.7 (*3*), and 0.8 J cm^{-2} (*4*) for $t_p = 30$ ns. The region behind the discontinuity on the time axis is of the millisecond scale.

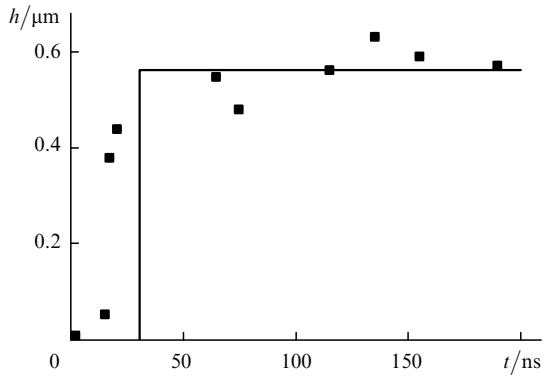

Figure 5. Time dependence of the hump height for $F = 1.1$ J cm^{-2}. The solid line corresponds to theoretical calculations and squares are experimental data from [5].

thermoelastic wave from the heated PMMA film. It can be seen that the theoretical dependence is also in satisfactory agreement with the experiment.

5. Conclusions

We have constructed a model of laser swelling for an absorbing polymer material, in which a thermoelastic stress wave generated by a short (nanosecond) pulse plays the leading role in the formation of a hump structure on the surface of the medium. Within the framework of this model, the threshold laser pulse fluence required for the formation of a plastic deformation region and a residual (stationary) hump structure is obtained analytically. It is shown that this threshold fluence depends on the laser pulse duration and is inversely proportional to the absorption coefficient of the material. The dependence of the residual hump height on the pulse fluence is derived analytically. The dependences of the peak hump height and the height of the residual hump structure on the pulse fluence and the absorption coefficient of the material are obtained for PMMA. The temporal dynamics of swelling of polymer materials is analysed for PMMA block and film samples.

A comparison of the results of theoretical calculations based on this model with the available experimental data on swelling of PMMA film samples reveals good qualitative agreement and satisfactory quantitative agreement.

The results of our calculations can be used for optimising the conditions of polymer swelling in the development of convex structures and for optimising the properties of polymers to be used for swelling.

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