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Effective time of thermal effect of ultrashort laser pulses on dielectrics

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Abstract. **It is shown that for ultrashort laser pulses (10 fs–1 ps) the total time of the thermal effect on a solid depends on the thermal properties of the medium, size and structure of the region under irradiation rather than on the pulse duration. The time of the thermal effect of ultrashort laser pulses amounts to 0.1–10 ns regardless of the pulse duration, which is comparable with the time of the effect of the nanosecond pulses. This fact should be taken into account in a variety of applications, and particularly, when the nonablation regimes are used. Presented are the analytical expressions for calculating the duration of the thermal effect of ultrashort laser pulses on dielectrics both in cases of the surface and bulk absorption.**

Keywords: *ultrashort laser pulses, time of thermal effect.*

1. Introduction

The two-temperature model of heating a condensed medium by means of laser pulses [1] implies that the electron–phonon interaction (the characteristic time of which is 10–11 s [2]) can be ignored if the duration of the pulses does not exceed 1 ns. In this case, the time of the the thermal effect is determined by the laser pulse duration. The physical processes incorporated into this model quite satisfactorily describe the effect of the laser pulses possessing a moderate power density and a duration more than 1 ns.

The situation has changed considerably with the advent of picosecond and femtosecond lasers, the pulse duration of which is by the order or less compared to that of the electron–phonon interaction. This range of durations has required more careful attention of researchers. In particular, the phenomena in the area of high-density flows, with the medium damage threshold exceeded, have been most comprehensively studied. In this regard, the two-temperature model has been significantly improved. These studies have in some degree obscured the area of not so high radiation flux densities being below the medium destruction threshold and even below the melting temperature of the medium. At the same time, provided that the ultrashort effects have the duration of the order of the electron–phonon interaction and less, this area now becomes particularly important in a variety of applications – from information recording and fabrication of diffractive optical elements to ophthalmology and other medical branches.

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With the laser pulse duration of about 10 fs, the phase changes in amorphous semiconductors (chalcogenide glasses [3]) which laid the foundation for a new generation of optical storage devices [4], thermochemical processes in thin metal films [5] which promoted the creation of new types of diffractive optical elements [6], the structural changes in the biological tissue [7], etc., proved out experimentally observable.

For all these applications, the main factors are time and size of the thermal effect of ultrashort laser pulses. It is these parameters that determine the possibility of using ultrashort laser pulses in the regimes with the energy density below the medium damage threshold.

In this regard, the aim of this paper is the calculations of the time of the thermal effect and the size of the region of thermal influence of ultrashort laser pulses, mainly, on dielectrics. When semiconductors and dielectrics are irradiated by ultrashort laser pulses, the generation of free electrons, with the concentration almost comparable to the concentration of those in metals, occurs already on the pulse front edge [8]. Therefore, the radiation absorption in such media may be regarded as the absorption by free electrons. The thickness of the layer in which the radiation energy is absorbed can be estimated as $\delta \sim 1/\alpha$ (α is the absorption coefficient). In case that the pulse duration is less than the time δ^2/a (a is the thermal diffusivity), which for dielectrics is about 1 ns, we may assume that, starting just from this instant, it is necessary to consider the impact of thermal conductivity on the temperature distribution in the area of influence. In the frame of this approach, the temperature distribution $T(x, t_\mathrm{m})$ along the depth at the beginning of the medium cooling can be described by the expression

$$T(x, t_\mathrm{m}) \approx \frac{A\alpha Q_0}{c} \exp(-\alpha x), \tag{1}$$

where A is the absorbability; Q_0 is the energy density; and c is the specific heat.

According to the two-temperature model [3], the electron gas temperature reaches the maximum at the end of the laser pulse action, whilst the atomic lattice continues to warm up until the heat flow from the electron gas into the lattice becomes equal to the flow discharged from it by conduction. It can be shown that that this time is determined by the expression

$$t_\mathrm{m} \approx \frac{c_\mathrm{e}}{\beta} \ln\left(1 + \frac{\beta c_\mathrm{i}}{k\alpha^2 c_\mathrm{e}}\right),$$

where β is the coefficient of heat transfer between the electron and lattice subsystems; c_i, c_e are the heat capacities of the crys-

tal lattice and electron gas; and k is the lattice thermal conductivity.

Thus, the time needed to reach the maximum temperature turns out greater than the laser pulse duration, and is associated with the thermo-physical properties of the electron gas and the lattice. Starting from the time t_m, the cooling of the medium begins, governed further by the thermal conductivity. This stage of cooling, being significantly longer than the heating stage, ultimately determines the time of the thermal aftereffect of ultrashort pulses (especially for dielectrics). Consider some practically important cases of cooling of the medium with the specified initial temperature distribution.

2. Cooling of the medium at the surface heating

The problem of cooling of the half-space with the initial temperature distribution (1) has an exact solution for the temperature on the surface (see, e.g., [9])

$$T(0,t) = T(0,0)\exp(\alpha^2 at)\operatorname{erfc}(\alpha\sqrt{at}), \tag{2}$$

where $T(0,0) = T(0,t_m)$ from (1).

For dielectrics, the thermal diffusivity of which is tenfolds less than that of metals, even if the absorption coefficient is comparable with the absorption index of metals (it increases due to the generation of free electrons during the laser pulse action), the temperature in the course of cooling, according to (2), halved relative to the maximum value for the time of a few nanoseconds.

3. Cooling of the medium at the absorption of radiation in the volume

Consider the cooling of the medium in the presence of the internal regions which have a form of a sphere and cylinder uniformly heated up to the temperature T_{max}. Such a configuration is realised when exposing the transparent media, wherein the radiation in the focal region (the region where the energy density is maximal) is absorbed through the nonlinear effects. In the approximation of instantaneous heating, these problems have analytic solutions that can be obtained by the method of Green's functions [9]. In case the heated zone represents of a sphere of radius R, the evolution of temperature at its centre appears as

$$\frac{T}{T_{max}} = \operatorname{erf}\left(\frac{R}{2\sqrt{at}}\right) - \frac{2}{\sqrt{\pi}}\frac{R}{2\sqrt{at}}\exp\left(-\frac{R^2}{4at}\right). \tag{3}$$

When the heated area has a shape of a cylinder of length l and radius R, the evolution of temperature in time in the middle of its axis is

$$\frac{T}{T_{max}} = \operatorname{erf}\left(\frac{l/2}{2\sqrt{at}}\right)\left[1 - \exp\left(-\frac{R^2}{4at}\right)\right]. \tag{4}$$

Figure 1 shows the time dependences of the relative temperature T/T_{max} of the heated area in the form of a glass ball with different radii. The time of cooling the ball center down to a prescribed temperature T_{max} (e.g., to $0.8T_{max}$) depends on the value of $4R^2/a$. Thus, for a glass ball with $2R = 1$ μm, it is close to 30 ns.

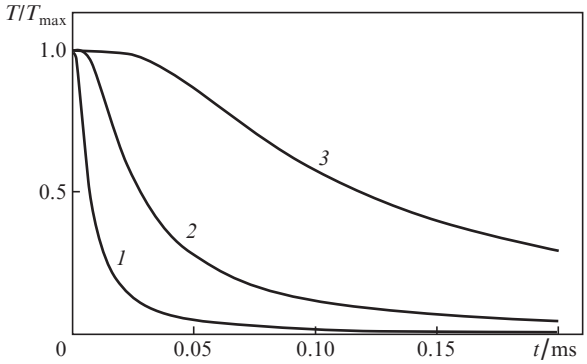

Figure 1. Cooling of the heated region in the form of a sphere of radius R = (*1*) 2, (*2*) 5 and (*3*) 10 μm in K-8 glass.

4. Cooling of thin metal films on dielectric substrates

The time of cooling of a thin metal film positioned on a dielectric substrate with a low thermal conductivity by means of femtosecond laser pulses can be determined by solving a thermo-physical problem [10], comprising a homogeneous heat equation for the film and substrate, supplemented by the conditions of equality of the temperature and heat flux on the boundary of the film and the substrate, at the initial temperature T_{max} of the film defined from heating the film by an ultrashort laser pulse. The solution to this problem is

$$T_1(t) = T_{max}\exp(\gamma^2 t)\operatorname{erfc}(\gamma\sqrt{t}), \tag{5}$$

where $T_1(t)$ is the temperature of the film; $\gamma = k_2 a_1/(k_1\sqrt{at}h)$ (subscript 1 refers to the film, subscript 2 – to the substrate); and h is the film thickness.

The results of calculation, according to formula (5), of the cooling dynamics of a metal film having been fabricated on a glass substrate and preliminary heated by a laser pulse are shown in Fig. 2. It is seen that the time of cooling for a film heated by femtosecond and nanosecond pulses (the difference in the pulse durations is five orders of magnitude), differ from each other only slightly, the time of cooling to a temperature

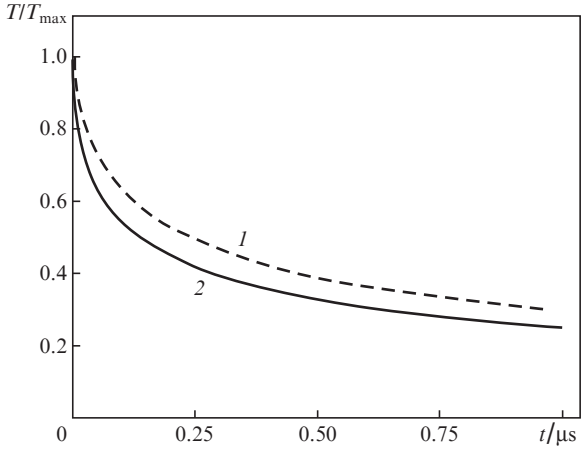

Figure 2. Cooling of the film on a glass substrate heated by (*1*) 10-ns and (*2*) 100-fs laser pulses at $h = 10^{-5}$ cm, $c_1 = 3.3$ J cm^{-3} K^{-1}, $c_2 = 0.28$ J cm^{-3} K^{-1}, $a_2 = 0.01$ cm^2 s^{-1}.

constituting 0.7 of the initial value – about twice, to lower temperatures – even less.

In conclusion, let us briefly discuss the importance of the femtosecond multipulse effects for technological applications. It can be shown from general thermo-physical representations that a sequence of n heating femtosecond pulses, provided that the pulse repetition period $t_r = 1/f$ is much smaller than the exposure time $t = n/f$, can be replaced by a continuous heat radiation with the same average power. In that case, under the condition that $r_0 \gg \sqrt{at}$ (r_0 is the size of the impact area), the temperature at the centre is given by [10]

$$T(0,t) = \frac{2(1-R)q_0\sqrt{at}}{\sqrt{\pi}\,k},$$

and at $r_0 \ll \sqrt{at}$

$$T(0) = \frac{(1-R)q_0 r_0}{k},$$

where q_0 is the power density. Apparently, this regime deserves special consideration for various specific applications.

5. Conclusions

1. The analysis shows that for ultrashort laser pulses (10 fs–1 ps), the total time of their thermal effect on a medium does not depend on the pulse duration, and is mainly determined both by the thermo-physical properties of the medium and the size and structure of the irradiated region.

2. The time of the thermal aftereffect of ultrashort laser pulses constitutes 0.1–10 ns regardless of the pulse duration, which is comparable with the time effect of nanosecond pulses.

3. The difference in the cooling dynamics of a metal film on a glass substrate, when heated either by the pulses of nanosecond duration or by shorter pulses, is negligible.

These results allow one to conclude that the effective time of the thermal effect of ultrashort laser pulses is of the same order of magnitude as that of nanosecond pulses. This fact should be taken into consideration in different applications, at least, when the nonablation regimes are used.

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