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THE INFLUENCE OF LASER PULSE DURATION ON THE KINETICS OF LASER-INDUCED THERMAL EMISSION OF POROUS CARBON MATERIALS

The shape of pulse signals of laser-induced thermal emission of porous carbon material is studied with the duration of laser excitation pulses varied from 20 to 40 ns. It is found that the amplitude and duration of thermal emission pulses depend significantly on the duration of laser pulses. In particular, an increase in the duration of the emission pulses from 70 to 200 ns is recorded, which is caused by changes in the temperature distribution with depth in the surface layer of the irradiated material. Computer modeling of the processes of pulsed laser heating and of the formation of thermal emission signals is carried out. The simulation results show satisfactory agreement with the measurement results.

Keywords: laser-induced thermal emission, kinetics, porous carbon.

1. Introduction

During the laser irradiation of light-absorbing materials, a local increase in the temperature in the area of absorption of the laser radiation can lead to noticeable changes in the pattern of the thermal radiation of the irradiated object, which is the basis of various laser technologies [1–10]. The use of powerful laser pulses of nanosecond duration makes it possible to obtain a laser-induced thermal radiation in the visible light, and this circumstance significantly expands the possibilities for the experimental research and applications. In thermal laser technologies, an important tool is the analysis of kinetics of grow and decay of the laser-induced thermal emission (LITE), since the shape of a laser-induced emission pulse carries the important information about the processes in the object under study [11–15].

This work is a continuation of the cycle of works [16–20] devoted to the study of the kinetics of LITE of surface layers of carbon materials. In previous studies, it was found that the decay of LITE in these materials is quite complex. In particular, at least two components with the characteristic time of the order of 10^{-8} s and 10^{-7} s can be distinguished in the emission decay. The emission decay kinetics depends not only on the properties of the studied material, but also on many other factors, in particular: (i) on the intensity of laser excitation; (ii) on the previous history of laser irradiation of the surface; (iii) on the wavelength of LITE detection; (iv) on the wavelength of the laser excitation; (v) on the surface roughness; (vi) on the presence of ambient air. In this work, another factor affecting the kinetics of LITE of carbon materials is investigated – it is the duration of laser excitation pulse. Experiments show that, when the laser pulse duration increases from 20

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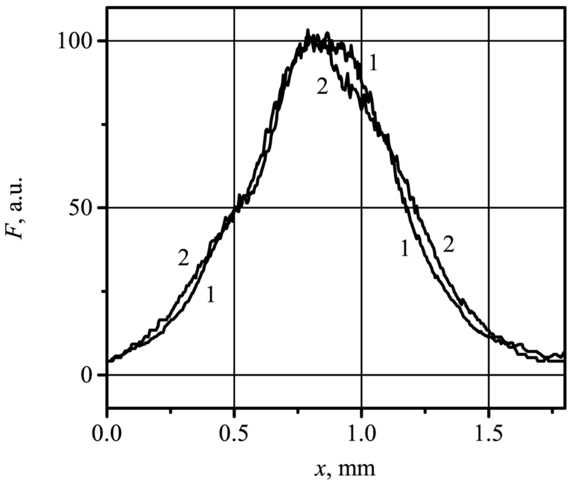

Fig. 1. Dependence of the intensity of laser radiation on the coordinate across the beam in the case of $\tau_i = 20$ ns (curve *1*) and 40 ns (curve *2*)

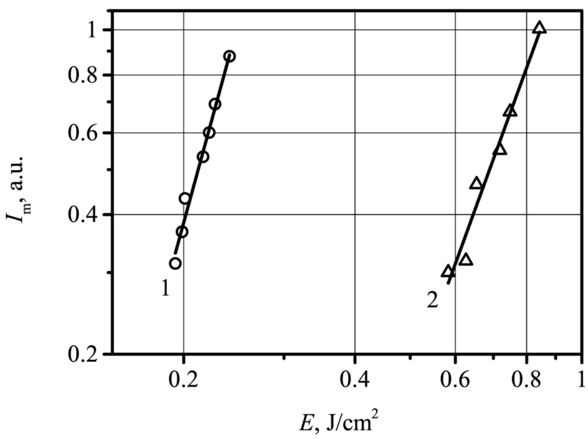

Fig. 2. Amplitude I_m of LITE pulses as a function of surface energy density E of laser excitation pulses with $\tau_i = 20$ (curve *1*) and 40 ns (curve *2*)

to 40 ns, the LITE pulse duration increases several times (for example, from 70 to 200 ns for porous carbon samples).

2. Methods

An YAG : Nd laser with electro-optical Q-switching was used for the excitation of LITE. To vary the duration of the laser generation pulses, the front of the pulsed electric signal, which opened the electro-optical shutter, was changed. By this method, the laser generation with the controlled pulse duration τ_i from 18 to 40 ns was obtained without changing the geometry of the laser resonator. Typical distributions of the intensity of laser radiation F across the beam are shown in Fig. 1 for two values of the duration of pulses, 20 and 40 ns. As is seen from Fig. 1, in both operation modes, the laser beams have approximately the same cross-beam intensity distributions, which significantly facilitates the comparison of the results of measurements at different values of τ_i.

In the present work, the method of measuring LITE signals is similar to the method used in works [18, 19]. Measurements were carried out in the spectral interval of 560 ± 20 nm.

To demonstrate the effect of the duration of laser excitation pulses on the kinetics of LITE, a porous carbon material – pharmaceutical activated carbon – was chosen. In previous works [18, 19], a computer model was developed for this material, which allows the calculation of the shape of the emission pulse under the pulsed laser excitation. The calculations use the classical heat conduction equation with a spatially inhomogeneous heat source formed in the surface layer of the material due to the absorption of laser radiation. As a result, the transient temperature distribution is calculated, which enables the calculation of the exitance of the surface as a function of time using Planck's formula for black body thermal radiation. The calculations involve the temperature dependence of the coefficient of thermal conductivity and specific heat capacity of the irradiated material and of the surrounding air, as described in [18, 19].

3. Results and Discussion

Figure 2 in a double logarithmic scale shows the dependence of the amplitude I_m of LITE pulses on the surface energy density E of the laser excitation with the pulse durations $\tau_i = 20$ and 40 ns. The results shown in Fig. 2 were obtained on the same part of the sample surface, with a fixed location of the photodetector and of the laser beam, and with a fixed power supply voltage of the photodetector. Under such experimental conditions, it can be assumed that oscillograms with the same amplitude correspond to approximately the same maximal surface temperature T_m, which is reached at the moment of maximum of the emission pulse.

As is seen from Fig. 2, in both cases for $\tau_i = 20$ and 40 ns, the dependence of I_m on E is strongly nonlinear, which is characteristic of this type of secondary radiation under the pulsed laser excitation. As proposed in [21, 22], for characterization of the degree of nonlinearity of LITE, it is convenient to use a dimensionless parameter $\gamma = (\mathrm{d}I_\mathrm{m}/I_\mathrm{m}) / (\mathrm{d}E/E)$, which is easily calculated as the slope of $I_\mathrm{m}(E)$ graph on a log-log scale.

Figure 2 shows $\gamma \approx 4.5$ for $\tau_\mathrm{i} = 20$ ns and $\gamma \approx 3.5$ for $\tau_\mathrm{i} = 40$ ns. The experimentally obtained values of γ are typical of LITE of carbon materials [23], and the closeness of γ values for $\tau_\mathrm{i} = 20$ and 40 ns testifies in favor of the above statement about the closeness of T_m for oscillograms with the same amplitudes I_m.

In Fig. 2, the fact of a significant displacement of curve 2 in relation to curve 1 is especially noticeable. In particular, as it follows from Fig. 2, in order to obtain a pulse of LITE at $\tau_\mathrm{i} = 40$ ns of the same amplitude as with $\tau_\mathrm{i} = 20$ ns, it is necessary to significantly increase the energy density E. This circumstance indicates that the maximal surface temperature is largely influenced by the processes of heat transfer from the surface to the depth of the sample during the duration of the laser pulse.

Attention should also be paid to the following circumstance. It is known that aerosol particles of soot under the laser irradiation with the energy density of 0.2–0.5 J/cm^2 reach the sublimation temperature of carbon (about 4000 K) [9], which is an important condition for practical applications. On the other hand, as is seen from Fig. 2, in this work, the experiments were carried out at rather high values of the laser excitation energy density (up to 0.8 J/cm^2), although pyrometric estimates of the surface layer temperature in these experiments did not exceed 3300 K. This circumstance additionally indicates that, during the laser pulse, a significant portion of the laser excitation energy leaves the surface layer as a result of the heat transfer processes.

Figure 3 shows typical oscillograms of LITE pulses with excitation $\tau_\mathrm{i} = 20$ and 40 ns. In Fig. 3, the oscillograms with the same amplitude I_m obtained at different values of the energy density E (according to Fig. 2) are presented. The oscillograms shown in Fig. 3 are shifted horizontally up to their maxima coincide.

It can be seen from Fig. 3 that an increase in the duration of the laser excitation pulse from 20 to 40 ns leads to a significant increase in the duration of leading edge of LITE pulse. If at $\tau_\mathrm{i} = 20$ ns the duration of the leading edge is about 10 ns; then at

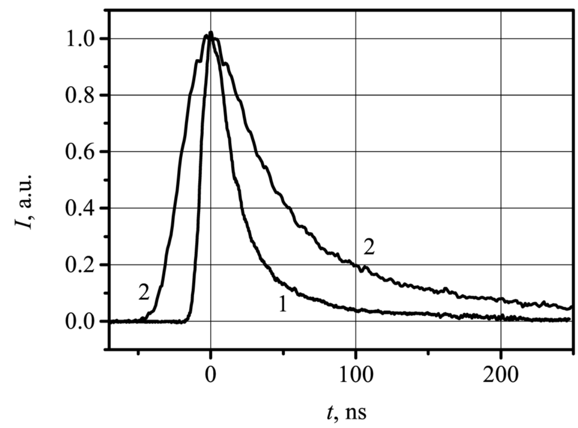

Fig. 3. Oscillograms of LITE pulses at $\tau_\mathrm{i} = 20$ ns (curve *1*) and 40 ns (curve *2*)

$\tau_\mathrm{i} = 40$ ns, this value increases to 25 ns. In both cases, the duration of the leading edge is of the order of half the duration of the corresponding laser excitation pulse. This result, within the accuracy of the measurements, corresponds to the results of computer simulations, which predict the duration of the leading edge of LITE pulse about 9.5 ns at $\tau_\mathrm{i} = 20$ ns and 17 ns at $\tau_\mathrm{i} = 40$ ns. It should also be noted that, according to the results of calculations, the maximum of the emission pulse is reached with the delay of 16 and 29 ns from the maximum of the laser pulse for $\tau_\mathrm{i} = 20$ and 40 ns, respectively.

As can be seen from Fig. 3, the increase in the duration of the laser excitation pulse from 20 to 40 ns leads to the significant lengthening of the trailing edge of a LITE pulse. In view of the fact that a long-term component is observed in the LITE decay, for the further comparison of the experimental and calculated results, it is expedient to introduce a parameter τ_{01} as the duration of a LITE pulse at a level of 0.1 of its maximal value.

The results of τ_{01} measurements are shown in Fig. 4. The pyrometric estimate of the maximal surface temperature T_m is plotted horizontally in Fig. 4. This temperature was calculated using Planck's formula for blackbody radiation based on the results of measurements of the amplitudes of LITE pulses at two wavelengths of 560 and 430 nm.

As is seen from Fig. 4, the duration of a LITE pulse τ_{01} significantly depends on the duration of the laser excitation pulse τ_i. In particular, with a two-fold increase in τ_i, the value of τ_{01} increases from 70 to

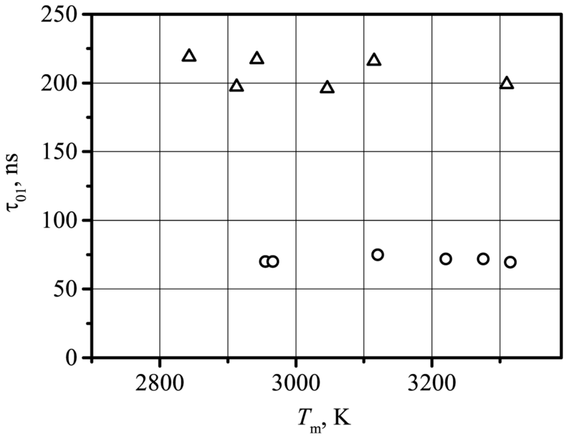

Fig. 4. The duration τ_{01} of LITE pulses at $\tau_i = 20$ ns (circles) and 40 ns (triangles) as a function of maximal surface temperature T_m

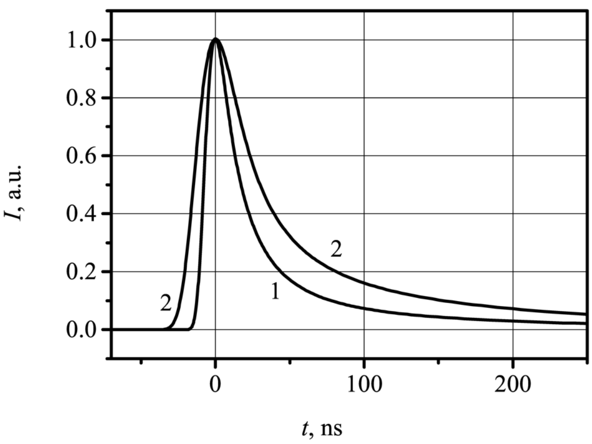

Fig. 5. Calculated LITE oscillograms of the carbon material with cylindrical roughness elements with the height $h = 0.5$ μm for $\tau_i = 20$ and 40 ns (curves *1* and *2*, respectively)

The results of measurements and calculations of the duration of thermal emission pulse τ_{01} at a wavelength of 560 nm

τ_{01}, ns	$\tau_i = 20$ ns	$\tau_i = 40$ ns
Experiment	70	200
Calculations, $h = 0.4$ μm	129	233
Calculations, $h = 0.5$ μm	90	177
Calculations, $h = 0.6$ μm	79	135

200 ns. Such an increase in τ_{01} cannot be explained by an increase in the duration of leading edge and requires a separate analysis.

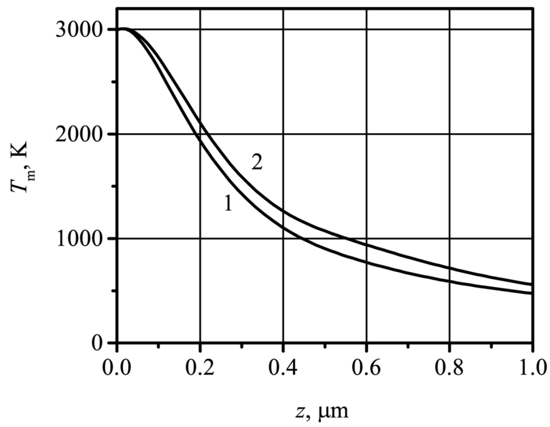

Fig. 6. The calculated maximal temperature T_m as a function of the depth z for $\tau_i = 20$ and 40 ns (curves *1* and *2* respectively) for $h = 0.5$ μm

To find out the reasons for the above-mentioned significant increase in the duration τ_{01} of LITE pulses, when τ_i increases, consider the following results of computer modeling of the processes of laser heating of surface layers of light-absorbing materials.

In previous works [17, 18] it was noted that rough surfaces are heated unevenly under the pulsed laser irradiation; as a result, the surface roughness can significantly affect the characteristics of thermal radiation pulses. Considering this circumstance, it seems appropriate to conduct computer simulations of the formation of LITE pulses by rough surfaces with the variation of duration of laser pulses. The approach described in [18] can be used for calculations. In particular, the rough surface can be represented by uniformly distributed cylindrical protrusions of height h. A surface element with a single protrusion can be used for calculations. In the present work in the calculations, the diameter of the protrusion was set equal to its height. The surface area around the protrusion was equal to the area of the top of the protrusion. The variable parameter h was chosen according to the depth of penetration of laser radiation into the irradiated material. In the calculations, the average intensity of laser radiation was selected for reaching the maximal temperature $T_m = 3000$ K at the top of the protrusion.

The calculation results for a rough surface with $h = 0.5$ μm are shown in Fig. 5 and in Table. As can be seen from Table and from Figs. 3 and 5, the results of calculations correspond to the results of experiments.

Thus, the computer model developed in previous works turned out to be suitable for calculating the parameters of LITE pulses with variation of the duration of laser pulses from $\tau_i = 20$ to 40 ns. The question remains as to the reasons for the observed significant lengthening of the duration of emission pulses.

In previous works [16, 17], for the decay of LITE of various carbon materials, it was noticed that a relatively long-lasting component with the decay time of the order of 10^{-7} s is observed. In [16,17] the presence of this decay component was associated with the fact that, in the studied materials, the penetration depth of laser radiation significantly exceeds the length of thermal diffusion for a time of the order of the duration of laser pulse. Therefore, at the end of the heating laser pulse, an elongated distribution of temperature with the depth is formed in the surface layer of the material. Hence the slow component of the emission decay is observed.

Considering the above mentioned, it seems appropriate to analyze the behavior of the temperature distribution inside the irradiated sample with changes in the duration of laser excitation pulses. The results of the corresponding calculations are shown in Fig. 6 for $h = 0.5$ μm. The z coordinate is counted from the top of the protrusion on the surface. The temperature distributions shown in Fig. 6 correspond to the moments of time, when the temperature at the top of the protrusion reaches its maximum, which roughly corresponds to the moments of the maxima of the emission pulses. It should be noted that the temperature distributions shown in Fig. 6 are initial for the processes of temperature relaxation and for the corresponding decay of the emission.

As can be seen from Fig. 6, when τ_i increases from 20 ns to 40 ns, the distribution $T(z)$ significantly expands. As a result, there is the increase of the duration of the emission decay and, accordingly, the increase of τ_{01}.

4. Conclusions

Summing up, it is worth emphasizing the following circumstances. The experimental and calculated results obtained in this work testify to the significant dependence of the characteristics of LITE pulses of porous carbon materials on the duration of laser excitation pulses. In particular, it is worth paying attention to the experimentally revealed sensitivity of the amplitude of LITE pulses to variations of the duration of laser pulses. In addition, an important circumstance is the significant lengthening of the emission pulses, which correspond to the same maximal surface temperature, but are excited by laser pulses with different durations. The specified properties of LITE are consistent with the existing model conceptions and add additional features to the complete picture of this complex type of laser-induced emission.

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ВПЛИВ ТРИВАЛОСТІ ЛАЗЕРНОГО ІМПУЛЬСУ НА КІНЕТИКУ ІНДУКОВАНОГО ЛАЗЕРОМ ТЕПЛОВОГО ВИПРОМІНЮВАННЯ ПОРУВАТИХ ВУГЛЕЦЕВИХ МАТЕРІАЛІВ

Досліджено форму імпульсних сигналів індукованого лазером теплового випромінювання поруватого вуглецевого матеріалу при варіюванні тривалості імпульсів лазерного збудження від 20 до 40 нс. Виявлено, що амплітуда і тривалість імпульсів теплового випромінювання суттєво залежать від тривалості лазерних імпульсів. Зокрема, зареєстровано збільшення тривалості імпульсів свічення з 70 до 200 нс, що зумовлено змінами розподілу температури в залежності від глибини у поверхневому шарі опромінюваного матеріалу. Проведено комп'ютерне моделювання процесів імпульсного лазерного нагрівання і формування сигналів теплового випромінювання. Результати моделювання показали задовільне узгодження з результатами вимірювань.

Ключові слова: індуковане лазером теплове випромінювання, кінетика, поруватий вуглець.