**Laser-Induced Incandescence of Rough Carbon Surfaces**

Kateryna S. Zelenska¹, Serge E. Zelensky², Alexander V. Kopyshinsky², Stanislav G. Rozouvan², Toru Aoki³

¹Educational and Scientific Centre "Institute of Biology", Taras Shevchenko National University of Kyiv, Prosp. Akademika Glushkova 2, Kyiv 03127, Ukraine
²Faculty of Physics, Taras Shevchenko National University of Kyiv, Prosp. Akademika Glushkova 4, Kyiv 03127, Ukraine
³Research Institute of Electronics, Shizuoka University, 3-5-1 Johoku, Naka-ku, Hamamatsu 432-8011, Japan

E-mail: czelenska@gmail.com

(Received October 12, 2015)

Laser-induced incandescence (LII) of rough carbon surfaces was studied under the Q-switched YAG:Nd laser excitation. For the surfaces irradiated by a sequence of laser pulses, the non-monotonic behavior of LII intensity with the increase of number of irradiating laser pulses was observed. Computer simulation of pulsed laser heating of rough carbon surfaces revealed essential non-uniformity of the temperature field on the irradiated surface hence the surface relief is affected by the laser irradiation due to the processes of evaporation of the peaks on the irradiated surface. The intensity of LII was calculated as a function of height of the surface roughness. The results of calculations explain the observed features of LII of carbon surfaces.

1. Introduction

Light-absorbing materials (for example, carbon) can be easily heated up to a temperature of several thousands of Kelvins under pulsed laser irradiation with a moderate level of laser power density (3-30 MW/cm²). As a result, thermal emission with wide spectrum in the visible range is observed, and this emission is usually called laser-induced incandescence (LII). Features and properties of this kind of thermal emission are extensively studied on carbon-based materials and results are presented in numerous papers [1-7]. LII is observed with soot microparticles in flames and engine exhaust gases, in aqueous suspensions of carbon microparticles (carbon black suspensions, CBS), in polymers and borate glass doped by carbon microparticles [1-5]. Besides, LII is emitted by laser-heated surface layers of carbon materials [6,7]. When excited by a sequence of laser pulses, the LII intensity depends on the laser irradiation dose. For example, the intensity of LII of carbon microparticles in CBS decreases with the increase of the dose due to the evaporation of particulate material [3]. For carbon suspensions in polymers (polystyrene, epoxy resin), the intensity of LII can grow with the laser irradiation dose due to the pyrolysis of polymer matrix around the overheated particles [5]. When a rough carbon surface is irradiated by a sequence of laser pulses, the intensity of LII is not a constant, and the increase and further decline of LII intensity is observed with the increase of number of irradiating laser pulses [7]. This feature of LII of rough surfaces requires additional explanation. Therefore, in this work we study the role of laser-induced reduction of the carbon surface roughness on the laser-induced incandescence.
2. Experimental Details

2.1 LII measurements

Samples made of carbon electrode rods with rough and polished surfaces were used. In the experiments, a Q-switched YAG:Nd laser (wavelength of 1064 nm, pulse duration of 20 ns, power density of 2-10 MW/cm$^2$) was employed for LII excitation. To collect the LII signal and to deliver it to a photomultiplier, an optical fiber bundle was used. LII was detected at a fixed wavelength (500 nm). LII pulse duration was determined by oscilloscope measurements with a photomultiplier with 2 ns resolution time. In this paper in the experiments with the irradiation of carbon surfaces by a sequence of laser pulses we denote the integral LII intensity as a time integral of a single LII oscillogram at the above-mentioned fixed wavelength.

2.2 Modeling and Calculation

In the present work, for calculation of the intensity of LII emitted from a rough carbon surface, the surface relief is represented as cylindrical asperities on a flat surface (Fig. 1).

![Fig. 1. Model of the surface roughness (cylindrical asperity).](image)

For calculation of transient temperature field, $T(r, t)$, the thermal conduction equation was employed with the heat source function $\alpha F(r, t)$, where $\alpha = 10^5$ cm$^{-1}$ is the absorption coefficient of carbon at the laser wavelength [9], and $F(r, t)$ is the local laser intensity. In the calculations, the laser pulse shape was taken as the following Gauss function

$$F = F_0 e^{-4\ln 2 \left(\frac{r}{\tau_i}\right)^2},$$

where $F_0$ is the peak intensity of incident laser beam, $\tau_i = 20$ ns is the laser pulse duration.

The laser intensity decreases along the beam axis according to the following equation

$$\frac{dF}{F} = -\alpha dz.$$ (2)

The integral LII intensity was calculated by integration of Plank’s blackbody emission function [6]

$$i_\lambda = \frac{\text{const} \cdot \lambda^{-5}}{e^{\frac{\lambda}{\Delta r}} - 1}.$$ (3)

over time and over the irradiated sample surface

$$I_{LII} = \iint_S I_5 dS$$ (4)

$$I_5 = \int_{-2\tau_i}^{10\tau_i} i_\lambda dt$$ (5)

$$I_t = \iint_S i_\lambda dS$$ (6)
where \( \xi = \frac{hc}{k_B} = 1.4388 \text{ nm} \), \( h \) is the Planck’s constant, \( c \) is the speed of light, \( k_B \) is the Boltzmann constant.

The penetration depth of the laser radiation into the carbon sample was about 100 nm, and the depth of heat conduction during the 20 ns laser pulse was approximately \( \sqrt{D \tau_i} = 270 \text{ nm} \), where \( D \) is the temperature diffusivity coefficient of carbon.

### 3. Results and Discussion

In this work, the kinetics of LII and transformation of the shape of LII pulses under irradiation by a sequence of laser pulses was studied by oscilloscope measurements. Typical oscillograms are given in Fig.2. Here \( N \) is the number of laser pulse in the sequence, and the incident intensity of laser radiation is \( F_0 = 8 \text{ MW/cm}^2 \). As is seen from Fig.2, after the irradiation by a sequence of laser pulses the LII pulse duration decreases from approx. 30 ns to approx. 20 ns.

![Typical oscillograms of LII of the carbon surface under irradiation by a sequence of laser pulses](image)

**Fig. 2.** Typical oscillograms of LII of the carbon surface under irradiation by a sequence of laser pulses \( N \).

As is known [5,6,7], under irradiation by nanosecond-scale laser pulses with \( F_0 = 3\ldots50 \text{ MW/cm}^2 \), carbon surface layers can be heated up to a temperature close to the temperature of carbon evaporation. Besides, calculations show that peaks and valleys on a rough surface are heated to different temperatures: top areas of asperities are overheated as compared with the valleys around the asperities. In some cases, the mentioned difference in temperature reaches several hundreds of Kelvins. Thus, it seems plausible to expect that irradiation of a rough surface by a sequence of laser pulses can cause the decrease of the surface roughness. STM studies were performed with a sample of polished carbon before irradiation and after the action of the laser pulse. Representative STM scans given in Fig.3 confirm the tendency of decreasing of the surface roughness after the laser irradiation. The above-mentioned shortening of LII pulses (Fig.2) can also be a consequence of faster cooling of the asperities with decreased height after the laser irradiation.

Under the irradiation by a sequence of laser pulses, the laser-induced decrease of the surface roughness can be the cause of a specific non-monotonic behavior of LII intensity with the increase of the number of irradiating laser pulses \( N \). The typical dependence of the integral LII intensity \( I_{\text{LII}} \) on the laser pulse number \( N \) is shown in Fig.4. As is seen from the figure, the increase of \( I_{\text{LII}} \) at \( N = 1\ldots4 \) is followed by the decrease at \( N > 4 \). It should be noted, the maximal value of the integral LII intensity (at \( N = 4 \)) exceeds its start value (at \( N = 1 \)) by a factor of 9.
Fig. 3. STM scans of the polished carbon surface: non-irradiated (curve 1) and irradiated (curves 2, 3) by a single laser pulse with the power density of 2.5 (curve 2) and 6 MW/cm$^2$ (curve 3).

Fig. 4. The integral LII intensity $I_{LII}$ of the carbon surface as a function of the laser pulse number $N$.

$h \sim (D \tau_i)^{1/2} = 270$ nm

Fig. 5. The calculated integral LII intensity of the carbon surface as a function of the asperity height $h$. The laser power density $F_0 = 10$ MW/cm$^2$. 

To explain the observed increase and further fading of the integral LII intensity, a model is proposed, which accounts (i) for the non-uniform heat distribution inside the asperities and (ii) for the evaporation of the surface material mainly from the top areas of asperities step-by-step with the increase of number of irradiating laser pulses [8]. The results of calculations are presented in Fig.5. The calculations of $I_{\text{LII}}$ were performed for various values of height $h$ of cylindrical asperities with the diameter $d = 300$ nm. As is seen from Fig.5, the integral LII intensity is a non-monotonic function of the asperity height, with the upper limit at $h \approx \sqrt{D\tau_i}$.

To clarify the physical mechanism of the observed behavior of LII intensity with the decrease of the asperity height, consider the following graphs (see Fig.6) of the surface temperature, $T$, and of the local LII intensity, $I_S$, as functions of the distance from the asperity axis measured along the irradiated surface. The graphs in Fig.6 were calculated for $F_0 = 10$ MW/cm$^2$ and for the asperity height of 0.1, 0.25, and 0.5 µm. The graphs 1a, 2a, 3a in Fig.6 correspond to the point of time $t = 10$ ns at which the maximal value of temperature is reached. The graphs 1b, 2b, 3b in Fig.6 are time-integrated LII signals according to expression (5). As is seen from Fig.6, temperature field at the irradiated surface depends significantly on the height of the asperity. When the height exceeds the value of thermal diffusion length, local temperature is decreased at a significant area of the irradiated surface ($x = 300\ldots700$ nm in Fig.6, curve 1a), and the appropriate contribution of this area to the surface-integrated LII signal is small (Fig.6, curve 1b). As a result, the decreased values of surface-integrated LII signals are observed at $N = 1\ldots3$ (see Fig.4). Besides, when the asperity height is of the order of the thermal diffusion length, local temperature at the asperity top area ($x = 0\ldots150$ nm, Fig.6, curve 2a) and the appropriate LII intensity (curve 2b) are increased as compared with graphs 1a, 1b and 3a, 3b, hence the surface-integrated LII signals demonstrate maximum at $N \approx 5$ (see Fig.4).

![Fig. 6. Calculated temperature and $I_S$ distribution on the irradiated surface.](image)

Note should be made, as is seen from Fig.5, the integral LII intensity can increase by a factor of 3, whereas the experiments demonstrate the factor of 9 (see Fig.4). This circumstance shows the limitation of the proposed simplified model. Nevertheless, this model demonstrates the important tendency in the dependence of LII on the surface roughness height.

Another model can be proposed for explanation of the observed non-monotonic behavior of the integral LII intensity under the irradiation by a sequence of laser pulses. This model can be actual for porous materials (or porous surface layers), where the laser irradiation can change the size and depth...
of air-filled undersurface microcavities. The appropriate calculations show that the intensity of LII strongly depends on the cavity size and on the thickness of carbon layer covering the cavity. For ground surfaces of carbon-based materials, the mentioned two models – laser-induced decrease of the surface roughness and transformation of porous structure of the surface layer – are complementary, and their mutual action requires additional study.

4. Conclusion

For carbon surface layers irradiated by nanosecond-scale laser pulses, the observed phenomena of LII increase and degradation with the increase of laser irradiation dose and the results of computer simulations show that the intensity of LII is sensitive to the parameters of surface roughness. Physical mechanisms of this sensitivity include non-uniform heating of the surface asperities and evaporation of carbon.

References