ABOUT NATURE OF LASER-INDUCED OPTICAL BREAKDOWN

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Short comparative analysis of main three concepts of laser-induced optical breakdown (thermal, plasma and direct) are represented. Experimental data for silicon carbide and potassium chloride are selected for modeling. We show that direct (system) concept may be explaining the observing experimental data in full volume. Questions about nature of corresponding transformations light and irradiated matter are discusses too.

Keywords: laser-induced optical breakdown; Relaxed Optics; Cherenkov radiation; interference; diffraction stratification; Zeits energy.

Introduction

Problems of the observation the laser-induced optical breakdown and shock processes in matter as Nonlinear (NLO) and Relaxed (RO) Optical processes are connected with acoustic (thermal) and electromagnetic (plasma and Nonlinear optical) nature [1-3]. These processes may be connected with diffractive stratification of laser beam, self-focusing, self-trapping, generation of super-continuum radiation (ordered – Cherenkov radiation, and disorder – plasma radiation) [2, 3].

The presentation this problem from one point of view for all media – from gases to solid is given in [1-5]. Unfortunately, the first attempt of observation this problem in main details in whole are represented in [5] only.

According to [1, 2], optical breakdown is understood as catastrophic damage caused by strong laser radiation. The cause of optical breakdown is avalanche ionization. This process is differed from heat breakdown, which is result of laser-induced heat of irradiated matter, to direct optical multiphotonic ionization. Roughly speaking the optical breakdown is result of rapid introducing energy to matter with laser help. Optical breakdown determines a limit laser intensity of laser radiation, which irradiated matter can absorb.

In whole this problem [1, 2] is very complex problem. From physical-chemical point of view the optical breakdown is the regime of fool breakage of all chemical bonds in irradiated matter in zone of laser irradiation [2, 3]. In this case we can determine the threshold of breakdown of irradiated matters with help methods of Relaxed Optics (cascade model of excitation the proper chemical bonds in the regime of saturation the excitation) [2, 3]. This regime may be received with help three ways [2]. We represent only direct cascade (system) optical concept [2]. In this case we have direct multiphotonic ionization and transformation of initial radiation and these processes have oriental nature [2].

Here we restrict ourselves to laser-induced optical breakdown in solids (silicon carbide [4, 5] and potassium chloride [6]) and the direct modeling method [2].

Main results and discussions

Typical complex experimental data for silicon carbide, which are included optical breakdown, were received in [4, 5] (Fig. 1).

Sectional area of receiving structures was ~ 22 μm, the depth of ~ 50 μm. As seen from Fig. 1c we have five stages disordered regions, which are located at a distance from 2 to 4 μm apart vertically [4, 5]. Branches themselves in this case have a thickness from 150 to 300 nm. In this case there are lines in the irradiated nanocavity spherical diameter of from 10 nm to 20 nm. In this case irradiated structures have crystallographic symmetry of the initial structure.

Two damages region in a crystal with moderately high density of inclusions were received in [6] for KCl after irradiation by CO_2-laser pulses (wavelength 10.6 μm, duration of pulse 30 ns). These results are presented in Fig. 2 [6].

Fig. 2a shows that spatial inhomogeneities are in fact inclusions [6]. The damage bubbles

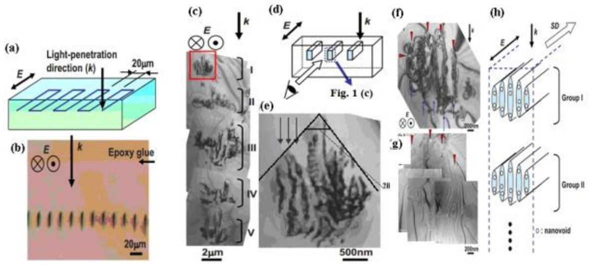

Fig. 1. (a) Schematic illustration of the laser irradiated pattern. The light propagation direction (k) and electric field (E) are shown. (b) Optical micrograph of the mechanically thinned sample to show cross sections of laser-irradiated lines (200 nJ/pulse). (c) Bright-field TEM image of the cross section of a line written with pulse energy of 300 nJ/pulse. (d) Schematic illustration of a geometric relationship between the irradiated line and the cross-sectional micrograph. (e) Magnified image of a rectangular area in (c). Laser-modified layers with a spacing of 150 nm are indicated by arrows. (f) Bright-field TEM image of a portion of the cross section of a line written with a pulse energy of 200 nJ/pulse. (g) Zero-loss image of a same area as in (f) with nanovoids appearing as bright areas. Correspondence with (f) is found by noting the arrowheads in both micrographs. (h) Schematic illustrations of the microstructure of a laser modified line. Light-propagation direction (k), electric field (E), and scan direction (SD) are shown. Only two groups (groups I and II) of the laser-modified microstructure are drawn [4, 5]

occur randomly near, not necessarily in, the tiny focal volume. At a well-defined power threshold, an elongated pointed bubble forms, its vertex falling at the focus (Fig. 2b).

Thermal and plasma models of laser-induced optical breakdown are represented in [1, 2]. For the modeling experimental data of Fig. 1 – Fig. 2 we used direct (system) method [2]. Next scenario: diffraction stratification of focused laser beam; generation of Cherenkov radiation on each cone of proper diffraction ring; interference of this Cherenkov radiation and optical breakdown in the maximums of interferograms, was used.

The first laser-induced filaments were received in the liquid [2]. Later researches shown that analogous phenomena are generated in solid and gas matter too. Therefore, first models were created for the nonlinear Kerr media and were used for all types of irradiated matter [1, 2]. Strongly speaking, these filaments are sparks of optical breakdown. More universal concept is physical-chemical [2, 3].

It may be physical-chemical method. In this case we must have concentration of proper centers of scattering (absorption) of laser radiation, which are generated proper nonlinear optical phenomenon, and its activation energy. The self-focusing is nonlinear optical process therefore P_{cr} or the critical value of energy may be determined in next way. Volume density of energy of the creation self-focusing process may be determined with help next formula W_{crvol} [2, 3]:

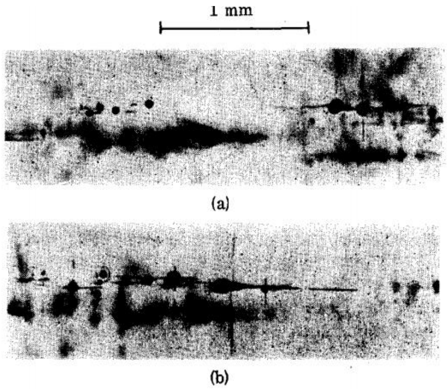

Fig. 2. Two damages regions in a crystal KCl with moderately high density of inclusions. The round black objects are bubbles. The radiation, incident from left to right, was yust at the intrinsic breakdown threshold. In one case (a) there was damage only at the inclusions. In (b), intrinsic breakdown occurred as evidenced by the pointed bubble. The straight lines represent cleavage [6]

$$W_{crvol} = E_a N_{nc}\,, \tag{1}$$

where E_a – energy of activation proper "nonlinear" centers; N_{nc} – their concentration.

Surface density for optical thin may be determined as [3]:

$$W_{crsur}=W_{crvol}/\alpha, \tag{2}$$

where α – absorbance index. Integral value of energy may be determined as [2, 3]:

$$W_{crin} = W_{crsur} \cdot S, \tag{3}$$

where S – the square of irradiation.

In this case [2, 3]

$$P_{cr} = {}^{W_{crin}}\!\big/_{\tau_{ir}}\,, \tag{4}$$

where τ_{ir} is duration of laser irradiation.

Next step of determination the density of energy in our cascade is condition of diffractive stratification. This condition may be determined with help of sizes the diffractive rings. We can estimate density of energy in plane of creation the diffractive stratification for n = 5.

Conic part of filament radiation has continuum spectrum: from ultraviolet to infrared. At first this effect was called superbroadening. Therefore, it may be interpreted as laser-induced Cherenkov radiation [2, 3, 7, 8]. The angle 2θ in the vertex of an angle of Fig. 1 (e) is double Cherenkov angle. In this case we have frozen picture of laser-induced destruction of 4H-*SiC* with help Cherenkov radiation [2, 3].

The Cherenkov radiation is characterized by two peculiarities [2, 3]: 1) creation of heterogeneous shock polarization of matter and, 2) radiation of this polarization. The methods of receiving shock polarization may be various: irradiation by electrons, γ-radiation, ions and excitation with help pulse fields. The stratification of this radiation on other type's radiation (volume, pseudo-Cherenkov i.e.) has relative character and may be represented as laser-induced Cherenkov radiation. Therefore, in future we`ll be represent conical part of filament radiation as Cherenkov [7, 9].

This fact may be certified with macroscopic and microscopic ways.

First, macroscopic may be represented according to [7]. The similarity between charge particle and light-induced Cherenkov radiation one can invoke the analogy between Snell's law and Cherenkov radiation [7]. In [7] the point of intersection of a light pulse impinging at an angle φ on a boundary between two media moves with velocity $V=C/n_1\cos\varphi$. This relation with Snell's law, gives the Cherenkov relation [7].

$$\cos\theta = C/n_2(\omega)V \qquad (5)$$

The microscopic mechanism of laser-induced Cherenkov radiation is expansion and application of Niels and Aage Bohrs microscopic theory of Cherenkov radiation as part of deceleration radiation on optical case [8]. For optical case the Bohrs hyperboloid must be changed on Gaussian distribution of light for mode TEM_{00} or distribution for focused light of laser beam [2, 3]. In this case Cherenkov angle may be determined from next formula:

$$\theta_{Ch} + \alpha_{ir} = \pi/2 \text{ or } \theta_{Ch} = \pi/2 - \alpha_{ir}, \qquad (6)$$

where α_{ir} – angle between tangent line and direction of laser beam.

Angle α_{ir} as determined from next formula [2, 3]:

$$\tan\alpha_{ir} = d_b/l_{sf}, \qquad (7)$$

where d_b – diameter of laser beam, l_{sf} – length of self-focusing. In our case α_{ir} is angle of self-focusing.

This formula is approximate for average angle α_{ir}.

The Golub formula (5) was used for the determination product $n_2(\omega)V_{nl\,pol}$ [3, 4]. Self-focusing and Cherenkov angles and product $n_2(\omega)V_{nl\,pol}$ were estimated for LiF, CaF_2, fused silica, water and glass BK-7 in [2, 3].

Thereby microscopic modified Bohrs theory and macroscopic Golub model are mutually complementary methods [2, 3].

The decreasing of Cherenkov angle and product $n_2(\omega)V_{nl\,pol}$ for increasing of laser radiation intensity are corresponded to increasing of nonlinear refractive index and decreasing of velocity of polarization (multiphotonic and multiwave processes) [2].

The estimation of sizes the cascade of volume destructions of Fig. 1c may be explains in next way [2, 3]. The sizes (diameters) of proper stages d_{nir} of cascade are proportionally to corresponding diffraction diameters [2].

The diffraction diameters d_{ndif} may be determined with help condition of diffraction-pattern lobes (modified Rayleygh ratio):

$$d_{ndif} = n\lambda. \qquad (8)$$

The estimations of first five diffraction diameters d_{ndif} for λ = 800 nm were represented in [3, 4]. The distance between diffraction spots and proper moving foci may be determined with help next formula:

$$l_{nf} = \frac{d_{ndif}}{2\tan\varphi/2}. \qquad (9)$$

These distances for $\varphi_1 = 20^o$ and $\varphi_2 = 30^o$ were represented in [3, 4]. In general case the angle φ is depended from homogeneity of irradiated matter or intensity of irradiation.

Qualitative explanation of development of cascade the destructions may be next. The focus of each diffraction zone (spot) is the founder proper shock optical breakdown. But foci with more high number may placed in the "zone" of influence of previous foci. Therefore, only first stage of Fig. 1c is represented pure shock mechanism (Mach cone). Mach cones are characterized the second and third stages of Fig. 1c. But its maximums are displaced from center. It may be result if interaction second and third shock waves with previous shock waves: first – for second wave and first and second for third wave. The chock mechanism of destruction certifies a linear direction of optical breakdown. Thus, basic creator of optical breakdown traces is secondary Cherenkov radiation and shock waves. This radiation is absorbed more effectively as laser radiation and therefore the creation of optical breakdown traces is more effectively as for beginning laser radiation. Cherenkov radiation is laid in self-absorption range of 4H-SiC, but 800 nm radiation – in intrinsic range.

The conclusion about diffractive stratification of focused radiation may be certified by experimental data of Fig. 1c.

We can rough estimate basic peculiarities of energy distribution in Mach cone may be used next formula [3, 4]:

$$E_{1ob} = \frac{\pi^2}{4}\left(\sum_{i=1}^{5} n_{iav}^2 l_{\,iav}\right) r^2 N_{aSiC} E_{Zth}, \qquad (10)$$

where n_{iav}– average visible number of filaments in proper group of cascade, l_{iav}=1000 nm – average length of filaments in proper group of cascade, r = 10 nm – average radius of filament, N_a – atom density of 4H-SiC.

Energy, which is necessary for the optical breakdown our nanotubes may be determined in next way. Zeitz threshold energy for 4H-SiC is equaled E_{Zth} ~ 25 eV [2, 3]. Let this value is corresponded to energy of optical breakdown. Therefore, summary energy E_{1ob} is equaled:

$$E_{1ob} = N_{asnt} \cdot E_{Zth} = 23{,}2\, nJ. \qquad (11)$$

This value is equaled of ~ 8% from pulse energy or ~ 30% from the effective absorbed energy of pulse.

We can estimate chain of critical value of energy for the 4H-SiC from physical-chemical point of view too. So, ratio between thresholds of optical breakdown and self-focusing is equaled ~ 10^5. For the modeling sizes and forms of nanovoids the modified Rayleygh model was used [2, 3].

Nanovoids may be represented as results of the laser-induced laser-induce breakdown and creation of cavitations bubbles [2, 3] too.

For the estimations of maximal radius of nanovoids in spherical approximation we must use modified Rayleygh formula [2, 3]:

$$R_{\max} = \frac{2R}{0.915 r}\sqrt{\frac{E_{ir}}{\pi \tau_i c E}}, \qquad (12)$$

where T_c – the time of creation the nanovoid (bubble), τ_i – pulse duration, R is radius of nanovoid, r – radius of irradiated zone, E – Young module, E_{ir} – energy of one pulse [2, 3].

But we have two speeds of sound in elastic body: longitudinal ϑ_{ls} and transversal ϑ_{ts} [2, 3]. The ratio between of these two speeds is equaled:

$$\alpha = \frac{\vartheta_{ts}}{\vartheta_{ls}} = \sqrt{\frac{(1-2\nu)}{2(1-\nu)}}. \qquad (13)$$

But this ratio must be true for shock waves too. Therefore, for silicon carbide for ν = 0.45 [4], α = 0.33. Roughly speaking last ratio is determined the step of ellipsoidal forms of our nanovoids (Fig. 2c).

This method was used for the modeling experimental data for KCl (Fig. 2). Density of atoms of KCl is equaled $3{,}1 \cdot 10^{22}\ cm^{-3}$. Zeits energy for KCl has value ~ 30 eV [2].

We used next approximations. Photography of Fig. 2 gives a blurry image compared to the bright-field TEM image of Fig. 1. Therefore, we can't see the microstructure of optical breakdown. Therefore, we use rough average approximations for diameter $d_{average}$ and length l of cascade laser-induced optical breakdown of Fig. 1. Volume of cascade was determined as cylinder volume [2].

Fig. 2 is similar to Fig 1c. But regimes of irradiation of Fig. 2 are similar to mode TEM_{01}. Therefore, we have two channels of generation the cascade of laser-induced optical breakdown.

The distances between bubbles of Fig. 3b are larger as between regions of destruction of Fig. 1c. But conditions of focusing the radiation in these both cases are equivalence. Therefore, the distances between neighboring bubbles l_2 of Fig. 2b and neighboring regions of destruction l_1 of Fig. 1c are connected by next formula [2]

$$l_2 = \frac{d_{ndif\,2} \tan\left(\varphi_1/2\right)}{d_{ndif\,2} \tan\left(\varphi_2/2\right)} l_1 = \frac{\lambda_2 \tan\left(\varphi_1/2\right)}{\lambda_2 \tan\left(\varphi_2/2\right)} l_1. \quad (14)$$

Energy characteristics of irradiation weren't represented in [6]. Therefore, we select value 2 J/pulse [3]. In this case we have effective using energy. Methods of estimations of energy characteristics for KCl are rougher as for 4H-SiC. But we must suppose that focused laser irradiation has diffraction stratification, generation of Cherenkov radiation and interference of this Cherenkov radiation. On Fig 2b 5-7 steps of cascade optical breakdown we see.

Effective transformation the energy of laser radiation to cascade of laser-induced breakdown for KCl is 11.6-17.4 percents [2].

These models may be used for the case, when kinetic processes of transformations light and matter, according represented scenario, and have a higher rate than the rate of formation of plasma and thermal processes [2]. The establishment of a hierarchy of processes that are described both by the methods of physical optics and by the methods of physical chemistry and radiation physics of solids makes it possible to more adequately describe the presented experimental results.

Conclusions

Experimental data of laser-induced optical breakdown in silicon carbide and potassium chloride are analyzed. Basic peculiarities of nature the laser-induced optical breakdown is shown on the example of direction cascade (system) model.

We show, that this concept describes successfully the represented experimental data.

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