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INTERACTION OF LASER RADIATION WITH MATTER. LASER PLASMA

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Interrelation of the laser-induced damage characteristics in statistical theory

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Abstract. An analysis is performed of the general relationships of the statistical theory of laser-induced damage in transparent solids caused by absorbing inclusions. It is shown that the structure of the statistical theory equations determines the interrelation of various dependences of the damage thresholds on physical parameters. This results in the equivalence of the spot-size dependence of the damage threshold and the reliability of the transparent solid, as well as in a similarity of the threshold dependences obtained upon single-shot and multishot irradiation. The predictions of the theory are in good agreement with the experimental data.

1. Introduction

The statistical relationships are most general for laser-induced damage (LID) of transparent solids. They are typical for both the intrinsic mechanisms of LID [1, 2] and the mechanisms related to absorbing inclusions [3, 4], and are observed upon single-shot [5] and multishot irradiation [6].

Statistical features of the damage can be caused by different reasons. These can be spatial or temporal fluctuations of the radiation intensity [7], the random nature of the appearance of the seed electron that leads to avalanche ionisation [1], or a random distribution of absorbing inclusions in a transparent solid [3, 4 – 6].

In any case, despite their different nature, statistical features complicate studies of LID in transparent solids, because they tend to blur the dependences observed in experiments. Statistical features significantly increase the amount of the experimental data needed to identify the damage mechanism. Therefore, one usually seeks to prevent statistical features in LID experiments. In particular, the single-mode single-frequency radiation sources are used in these experiments to avoid the influence of laser fluctuations on experimental results.

When the inclusion-related damage mechanism is dominant, the statistical features are unavoidable. They are intrinsic to the nature of the damage caused by the inclusions randomly distributed over the volume of the transparent solid. For these reasons, investigation of various statistical relationships and their interrelation is a fundamental problem of the theory of LID, which is important for both the metrology of LID and identification of its physical mechanisms.

Interrelation of the statistical relationships was studied in Refs [3 – 6]. In particular, Manenkov et al. [6] used the requirement of correspondence between the damage probability and the spot-size dependence to prove that absorbing inclusions play the dominant role in damage of polymethylmetacrylate (PMMA). The approaches used in these works were based on utilisation of the distribution function of inclusions over damage thresholds, which is usually unknown. The attempts to develop the method for experimental determination of this function met with serious difficulties [8, 9]. Therefore, it is particularly interesting to investigate interrelations between different experimentally observed statistical features of LID without making any assumptions about the distribution function. This work is dedicated to the solution of this problem.

2. Basic concepts of the statistical theory of LID

We review basic concepts of the statistical theory of the LID caused by absorbing inclusions [4, 10], which will be used in the subsequent analysis. The basic assumptions of the theory are the following:

(1) the inclusion size is much smaller than the dimensions of the interaction region;

(2) the inclusions are randomly distributed over the volume (or the surface) of the transparent solid;

(3) an ensemble of inclusions $\{c_s\}$ (where $s = 1, 2, \ldots, L$; c_s is the concentration of the inclusions of the s-th type; L is the number of the inclusion types) is characterised by the distribution over the damage thresholds,

(4) the inclusion of the s-th type initiates damage when the intensity of the incident radiation exceeds the threshold $I_{\rm th}^{(s)}(\mu)$, dependent on μ – the physical and geometric characteristics of the inclusion (absorption coefficients, dimensions etc.) – and the parameters of the incident radiation (pulse duration, wavelength, etc.).

For single-mode and single-frequency radiation, taking into account these assumptions, the reliability $Q(I, \upsilon, \mu)$ of the transparent solid (i.e., the probability of no damage) has the form

$$Q(I, \upsilon, \mu) = \exp\left[-\upsilon \sum_{s=1}^{L} c_s K\left(\frac{I}{I_{\rm th}^{(s)}(\mu)}\right)\right], \qquad (1)$$

where I is the maximum intensity in the interaction region;

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$K(I/I_{\rm th}^{(s)}(\mu))$ is the dimensionless monotonically increasing function of the intensity, which is determined by the mode structure and the focusing conditions of radiation; and v is the volume of the focal region. The damage probability $P(I, v, \mu)$ is related to the reliability by the normalisation condition

$$P(I, v, \mu) + Q(I, v, \mu) = 1. \quad (2)$$

Note that the dependence $I_{\rm th}^{(s)}(\mu)$ cannot be observed because of spatial variation in the laser damage resistance. Experimentally one investigates the dependence of the maximum radiation intensity $I_\beta(v, \mu)$ inside the interaction region on the testing conditions, i.e., the parameters v and μ (where μ is, for example, the laser pulse duration) that results in the damage for a given reliability β. This dependence can be determined from the condition

$$Q(I, v, \mu) = \beta. \quad (3)$$

Taking into account Eqns (1) and (3), we obtain the main equation of the statistical theory of LID

$$\sum_{s=1}^{L} c_s K\left(\frac{I}{I_{\rm th}^{(s)}(\mu)}\right) = \frac{\ln \beta^{-1}}{v}, \quad (4)$$

The solution of this equation allows one to calculate $I_\beta(v, \mu)$ for given values of $\{c_s\}$, $I_{\rm th}^{(s)}(\mu)$, and a given geometry of the interaction region. A specific feature of the solutions of this equation is that the dependences $I_\beta(v, \mu)$, corresponding to different β and v coincide if β and v satisfy the compensation relationship

$$\frac{\ln \beta}{v} = \text{const}, \quad (5)$$

which means that a variation in the damage threshold caused by the spot-size effect is compensated by a corresponding variation in the reliability.

Although $I_{\rm th}^{(s)}(\mu)$ does not depend on the focal volume size, the observable dependence $I_\beta(v, \mu)$ appreciably depends on v, as follows from (4). This so-called spot-size dependence of the damage threshold is a fundamental consequence of the random distribution of inclusions over the volume of the transparent solid. Many authors investigated it in detail for various materials (see Refs [3, 5]).

A detailed analysis of the general laws of LID in transparent solids (e.g., of the dependences $Q(I, v, \mu)$ and $I_\beta(v, \mu)$, requires a wealth of experimental data. In practice, the studies of statistical relationships are usually limited to investigation of either the spot-size dependence of the damage threshold for a given reliability β_0 or the dependence of the reliability (or the damage probability) of the transparent solid for a given focal volume v_0.

The statistical relationships are fully determined by the distribution of the inclusions over the damage thresholds, i.e., by the set $\{c_s\}$. Knowing this set $\{c_s\}$, one can calculate all the statistical features of LID, in particular, find $Q(I, v, \mu)$ and $I_\beta(v, \mu)$ for any experimental conditions. The importance of the set $\{c_s\}$ for the study of the damage mechanism has led to many attempts to infer it from the statistical properties of the damage (the dependence of the reliability on the intensity [8] or the spot-size dependence [9]) by solving the so-called inverse problem. However, this problem is unstable and therefore very difficult to solve. For these reason, the studies of the statistical relationships that were based on evaluation of $\{c_s\}$ did not provide an efficient method for solving the statistical problems of LID.

3. Interrelation of the reliability and the spot-size dependence of the LID threshold

The dependence of $Q(I, v, \mu)$ on the focal volume results in variation of the absolute value of damage threshold with variation of v and in the modification of the dependence of the LID threshold on other parameters [10]. These modifications hinder a comparison of the experimental data obtained under different conditions. Nevertheless, the structure of the equations of the statistical theory of LID makes it possible in some cases to adequately compare the results of investigations of these dependences. Let us introduce the function

$$C(I, \mu) = \frac{\ln Q^{-1}(I, v, \mu)}{v}, \quad (6)$$

which has the physical meaning of the effective concentration of the inclusions involved in the damage. An important property of $C(I, \mu)$ is that, unlike $Q(I, v, \mu)$ and $I_\beta(v, \mu)$, it is independent of v. Instead, it can be simply calculated from given reliability $Q(I, v, \mu)$, using Eqn. (6), and from the spot-size dependence of the damage threshold $I_\beta(v, \mu)$ using the relationship $C(I, \mu) = (\ln \beta^{-1})/v_\beta(I, \mu)$, where $v_\beta(I, \mu)$ is the inverse function to $I_\beta(v, \mu)$. The function $v_\beta(I, \mu)$ is the volume of the focal region whose irradiation leads to damage with the probability $1 - \beta$. This function is properly defined in the entire intensity range since $I_\beta(v, \mu)$ monotonically decreases with increasing v. The above properties allow one to find the relation between of $Q(I, v, \mu)$ and $I_\beta(v, \mu)$.

Suppose that we have the an experimentally measured dependence of the reliability of a transparent solid on the laser radiation intensity for a given value of the focal volume v_0. Then, calculating $C(I, \mu)$ from Eqn. (6) for two different values of v, one of them coinciding with v_0, and equating the results, we obtain

$$Q(I, v, \mu) = [Q(I, v_0, \mu)]^{v/v_0}. \quad (7)$$

Expression (7) allows one to calculate $Q(I, v, \mu)$ for any v using the experimentally measured reliability $Q(I, v_0, \mu)$.

The left-hand side of equation (4) is equal to $C(I, \mu)$. For a given reliability $Q(I, v_0, \mu)$, this function can be determined from Eqn. (6). Using Eqns (4) and (6) together, we derive the relationship

$$Q(I, v_0, \mu) = \beta^{v_0/v}, \quad (8)$$

which yields the spot-size dependence of the LID threshold $I_\beta(v, \mu)$.

Thus, if we know the reliability $Q(I, v_0, \mu)$ at some focal volume v_0, we can calculate it for any other v and also determine the spot-size dependence of the LID threshold $I_\beta(v, \mu)$.

On the contrary, if we know the experimental dependence of the LID threshold $I_{\beta_0}(v, \mu)$ for a given reliability β_0, we can calculate $I_\beta(v, \mu)$ for any other value of β and determine $Q(I, v, \mu)$.

Indeed, using Eqn. (4) to calculate $C(I, \mu)$ from $v_\beta(I, \mu)$ for two different values of β (one of them coinciding with β_0) and equating the results, we obtain

$$\upsilon_\beta(I,\mu) = \upsilon_{\beta_0}(I,\mu)\frac{\ln\beta}{\ln\beta_0}. \tag{9}$$

Expression (9) defines the spot-size dependence $I_\beta(\upsilon, \mu)$ as the inverse function to $\upsilon_\beta(I, \mu)$ for any reliability β. Finally, equating the expressions for $C(I, \mu)$ in terms of the functions $Q(I, \upsilon, \mu)$ and $\upsilon_{\beta_0}(I, \mu)$ $[\ln Q(I, \upsilon, \mu)]/\upsilon = (\ln\beta_0)/\upsilon_{\beta_0}(I, \mu)$, we find

$$Q(I,\upsilon,\mu) = \beta_0^{\upsilon/\upsilon_{\beta_0}(I,\mu)}. \tag{10}$$

Expression (10) allows one to calculate $Q(I, \upsilon, \mu)$ for a given value $\upsilon_{\beta_0}(I, \mu)$.

Thus, relationships (7)-(10) demonstrate the interrelation of the reliability and the spot-size dependence of the LID threshold.

4. Similarity of the LID relationships for single- and N-shot irradiation

We introduce notations $I_{\text{th}}^{(s)}(1,\mu)$ and $I_{\text{th}}^{(s)}(N,\mu)$ for damage thresholds of the s-th type inclusions upon single- and N-shot irradiation, respectively. In a similar way we define notations for other characteristics: $Q(I,\upsilon,1,\mu)$ and $Q(I, \upsilon, N,\mu)$, etc. We have shown earlier [11] that Eqns (1) – (4) can be used to describe the statistical features of damage for both single- and N-shot irradiation; of course, $Q(I,\upsilon,1,\mu)$ should then be replaced by $Q(I,\upsilon,N,\mu)$ in these expressions to indicate explicitly the irradiation mode. This means that the reliability and the spot-size dependence are related by Eqns (7) – (10) upon N-shot irradiation as well.

Furthermore, $Q(I,\upsilon,1,\mu)$ and $Q(I, \upsilon, N,\mu)$, as well as $I_\beta(\upsilon,1,\mu)$ and $I_\beta(\upsilon,N,\mu)$ are related to each other by the similarity relationships. This statement is based on the following physical argument. On the one hand, as shown previously [4,12], the temperature T_{th} (1) of the thermal explosion initiation (i.e., the LID upon single-shot irradiation) and the temperature $T_{\text{th}}(N)$ corresponding to the initiation of accumulation of irreversible changes (i.e., the LID upon N-shot irradiation) are determined by the properties of the solid and by the mechanisms of these processes; they are independent of both the properties of the inclusions and the parameters of the radiation pulse.

On the other hand, the temperature of the inclusion heated by the laser radiation is determined by the properties of the inclusion and the laser pulse, as well as the transparent solid. The critical intensities $I_{\text{th}}^{(s)}(1,\mu)$ and $I_{\text{th}}^{(s)}(N,\mu)$ of the damage initiation are related to the temperatures $T_{\text{th}}(1)$ and $T_{\text{th}}(N)$ by the expressions

$$T_{\text{th}}(1) = k^{(s)}(\mu)I_{\text{th}}^{(s)}(1,\mu),\ T_{\text{th}}(N) = \bar{k}^{(s)}(\mu)I_{\text{th}}^{(s)}(N,\mu),$$

where $k^{(s)}(\mu)$ and $\bar{k}^{(s)}(\mu)$ are the coefficients depending on the material characteristics of the solid and the inclusion (thermal conductivity, absorption coefficients etc.); the bar over k denotes averaging over N laser pulses.

The coefficients $k^{(s)}(\mu)$ and $\bar{k}^{(s)}(\mu)$ can be determined by solving the heat conduction equation; they have the same functional dependence on the heat-transfer properties of the inclusion and the solid. The difference between $k^{(s)}(\mu)$ and $\bar{k}^{(s)}(\mu)$ is due to modification of these properties upon heating. The estimates [13] show that these variations are negligible; therefore, we will neglect the difference between $k^{(s)}(\mu)$ and $\bar{k}^{(s)}(\mu)$ in the following and assume that $k^{(s)}(\mu) = \bar{k}^{(s)}(\mu)$. Taking into account this assumption, the ratio

$$\frac{I_{\text{th}}^{(s)}(1,\mu)}{I_{\text{th}}^{(s)}(N,\mu)} = \frac{T_{\text{th}}(1)}{T_{\text{th}}(N)}$$

is independent of the properties of the inclusion. Introducing the notation $T_{\text{th}}(1)/T_{\text{th}}(N) = \Phi(N)$, for the ratio of the damage thresholds we have

$$\frac{I_{\text{th}}^{(s)}(1,\mu)}{I_{\text{th}}^{(s)}(N,\mu)} = \Phi(N). \tag{11}$$

In accordance with its physical meaning, the function $\Phi(N)$ monotonically increases with increasing N and satisfies the condition $\Phi(1) = 1$.

Therefore, the ratio of the damage thresholds upon single- and N-shot irradiation is independent of the properties of the inclusion and the parameters of the laser pulse. This allows us to establish the relation between $Q(I,\upsilon,1,\mu)$ and $Q(I, \upsilon, N,\mu)$. Using Eqns (1) and (11), we obtain

$$Q(I,\upsilon,1,\mu) = Q(I\Phi(N),\upsilon,N,\mu). \tag{12}$$

In a similar fashion, it follows from Eqns (4) and (11) that the dependence of the damage threshold on any parameter (the pulse duration, the focal region dimensions, etc.) upon single-shot and N-short irradiation are related by the expression

$$I_\beta(I,\upsilon,1,\mu) = \frac{I_\beta(I,\upsilon,N,\mu)}{\Phi(N)}. \tag{13}$$

Thus, the reliabilities $Q(I,\upsilon,1,\mu)$ and $Q(I, \upsilon, N,\mu)$ are related by transformation (12), while the dependences of the LID threshold of the transparent solid upon single- and N-shot irradiation are related by expression (13).

5. Comparison with the experimental data

The developed theory describing the interrelations between the statistical properties of LID is valid for the entire range of the laser pulse durations where the dominant role is played by the damage mechanism related to the absorbing inclusions. It was established earlier that this situation takes place in a wide range of the pulse durations from milliseconds to a few picoseconds and, possibly, femtoseconds [14]. Moreover, it is important for the obtained results to be applicable that the spatial distribution of the laser beam intensity in the interaction volume should not be distorted by any nonlinear effects (self-focusing, self-defocusing etc.).

Comprehensive experimental investigation of statistical relationships of LID in transparent solids demands serious efforts. Therefore, experiments are usually limited to investigation of either the dependence of the reliability on the intensity or the spot-size dependence of the damage threshold. Ref. [6] is perhaps the only work reporting the results of investigation on both the damage probability and the spot-size dependence of the damage threshold upon single- and N-shot ($N = 200$) irradiation.

Manenkov et al. [6] studied the damage in PMMA by 20-ns, 1.06-μm laser pulses. The thickness of the investigated samples was $H \ll F$, where F is the focal length of the lens. The spatial distribution of the laser beam intensity was close to the Gaussian one. Under these conditions, $\upsilon = \pi d^2 H/2$, where d is the diameter of the focal spot; therefore,

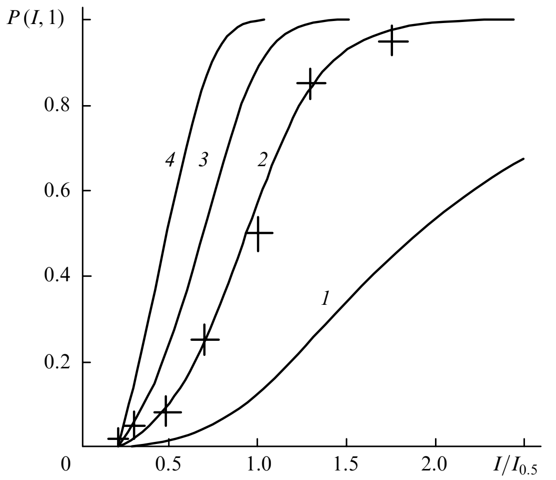

Figure 1. Dependences of the damage probability in PMMA $P(I,\ 1)$ on the maximum intensity of the laser radiation in the focal plane I upon single-shot irradiation for the focal spot diameter $d = 200$ (*1*), 500 (*2*), 800 (*3*), and 1300 μm (*4*), as well as the experimental data of Ref. [6] for $d = 500$ μm (crosses). Here and in Figs 2–4 the intensities $I_{0.5}$ are defined for $d = 500$ μm.

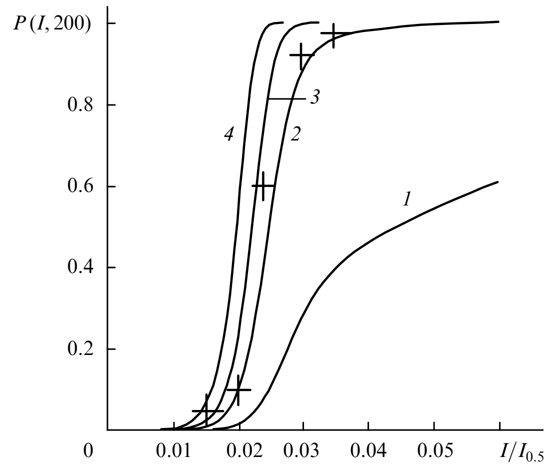

Figure 2. Dependences of the damage probability in PMMA $P(I,\ 200)$ on the maximum intensity of the laser radiation in the focal region I upon 200-shot irradiation for $d = 200$ (*1*), 500 (*2*), 800 (*3*), and 1300 μm (*4*), as well as the experimental data of Ref. [6] for $d = 500$ μm (crosses).

any dependence on υ is equivalent to the dependence on d. Figs 1–3 show the experimental data for $I_{0.5}(d, 1)$, $I_{0.5}(d, 200)$, and $P(I, d, 1)$ and $P(I, d, 200)$ from Ref. [6].

The probability $P(I, d, N)$ of the LID was approximated by the expression

$$P(I,d,N) = \frac{\exp[\gamma_0 + \gamma_1(I/I_{0.5})]}{1 - \exp[\gamma_0 + \gamma_1(I/I_{0.5})]} \times \left\{1 - \exp\left[-\frac{\gamma_2(I - I_0)}{I_{0.5}}\right]\right\}, \quad (14)$$

where γ_0, γ_1, γ_2 and I_0 are the best fit parameters for expression (14) and experimental data; $I_{0.5}$ is the damage threshold of PMMA upon single-shot irradiation for the reliability $\beta = 0.5$.

This function fits the experimental data best upon single-shot irradiation in the region $I > I_0$ (the intensity I_0 is the minimum damage threshold of PMMA) for $d = 500$ μm, if $\gamma_0 = -4.2$, $\gamma_1 = 4.5$, $\gamma_2 = 6$ and $I_0/I_{0.5} = 0.2$ (see Fig. 1).

To find $P(I, d, 1)$ for other values of d, we first calculated the reliability corresponding to expression (14) from normalisation condition (2) and then performed transformation (7). When performing transformation (7), we assumed that $\upsilon/\upsilon_0 = (d/d_0)^2$ in accordance with the condition $H \ll F$ of experiment [6]. We calculated the dependence $Q(I, d, 1)$ for the diameters $d = 200$, 800, and 1300 μm that were used in experiment [6], and then determined the dependence $P(I, d, 1)$ for the same values of d using Eqn. (2). The results of the calculations are shown in Fig. 1. After calculating the family of curves $P(I, d, 1)$, we determined the damage threshold for various values of d (the spot-size dependence) from the relationship $P(I, d, 1) = 0.5$. One can see from Fig. 3 that the results of the calculations for $I_{0.5}(d, 1)$ agree with the experimental data of Ref. [6] to within the experimental error.

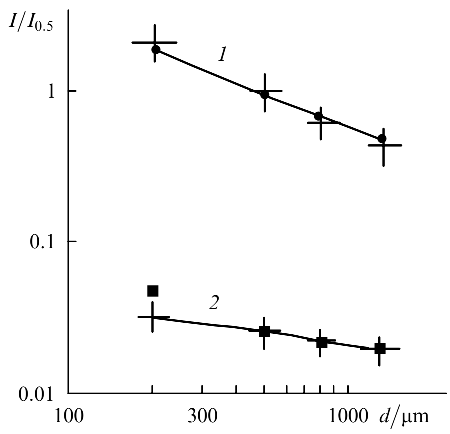

Figure 3. Spot-size dependences of the damage threshold in PMMA upon single-shot (●, *1*) and 200-shot (■, *2*) irradiation, and the experimental data of Ref. [6] (+).

We calculated $P(I, d, 200)$ and $I_{0.5}(d, 200)$ in a similar manner. The best agreement between the function (14) and experimental data for $P(I, d, 200)$ and $d = 200$ μm was observed when $\gamma_0 = -11$, $\gamma_1 = 450$, $\gamma_2 = 100$ and $I_0/I_{0.5} = 0.001$. The curve family $P(I, d, 200)$ was calculated for the same sizes of the focal spot as above. We calculated the spot-size dependence of the damage threshold with the aid of the expression $P(I, d, 200) = 0.5$; the results agreed with the experimental data [6] to within the experimental error (see Fig. 3).

To compare the damage probabilities $P(I,\ d, N)$, measured upon single- and 200-shot irradiaton, one has to perform the transformation (12), which depends on a single parameter $\Phi(200)$. By transforming $P(I, d, 200)$ to the reliability with the aid of expression (2), performing transformation (12) with $\Phi(200) = 40$, and converting the result back to the damage probability, we obtained the curve shown in Fig. 4. One can see that the transformed dependence $P(I, d, 1)$ agrees with the dependence $P(I, d, 200)$ measured in Ref. [6] to within the experimental error. Similarly, the spot-size dependences $I_{0.5}(d, 1)$ and $I_{0.5}(d, 200)$ satisfy relationship (13) to within the experimental error.

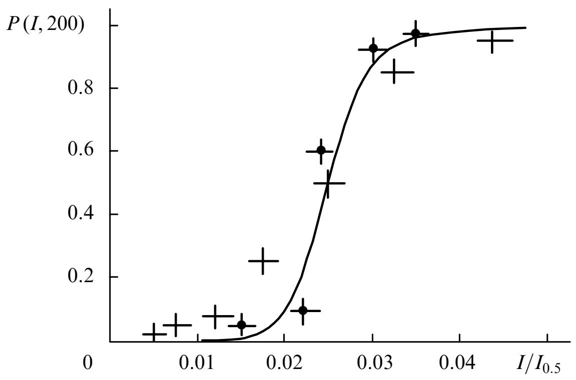

Figure 4. Similarity of the LID probabilities upon single-shot and 200-shot irradiation. The figure shows the experimental data [6] upon the single-shot irradiation that were re-scaled to the 200-shot irradiation (+) and the data for the 200-shot irradiation mode for $d = 500$ μm (•).

Thus, the dependences $I_{0.5}(d, 1)$ and $I_{0.5}(d, 200)$, as well as the dependences $P(I, d, 1)$ and $P(I, d, 200)$ measured in Ref. [6] are interrelated to each other in accordance with expressions (7) – (10) and (12), (13).

6. Conclusions

Our analysis of LID properties under conditions of spatial variation of the laser-induced damage resistance has shown that unique interrelation between the damage probability (reliability) and the spot-size dependence has to be ob-served upon both single-shot and multishot irradiation. The established interrelation is a fundamental property of the statistical theory of LID and is of principal importance for the studies of the LID mechanism.

The analysis, based on the concepts developed, of published experimental data has shown their good agreement with the theory. This indicates that the damage mechanism related to the absorbing inclusions played the dominant role in the investigated sample.

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