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Common approach to the problem of defect nucleation in solids under 'pre-threshold' laser irradiation

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Abstract. A universal mechanism for initial defect nucleation in metals and transparent dielectrics, as the sample is irradiated by 'pre-threshold' laser pulses, is proposed. The theoretical model uses the newly described effect of local accumulation of elastic energy due to anharmonic parametric resonance between vibrations localised in an inhomogeneity of the crystal lattice and laser-produced acoustic waves in the solid. Thermal density fluctuation is considered as a universal inhomogeneity of solid matter. The elastic modulus plays the role of an excited parameter, and the coupling constant between this parameter and the elastic wave is the Grüneisen coefficient. Metals and transparent dielectrics have different mechanisms of acoustic wave generation due to laser beam interaction with matter. The thermoelastic effect arising due to pulsed heating of the irradiated region is a source of acoustic waves in metals. Hyperacoustic waves are generated due to stimulated Brillouin scattering in the case of transparent dielectrics. The model explains the main characteristics of laser-induced damage in solids subjected to 'pre-threshold' irradiation. Quantitative estimates have shown that the critical values of the acoustic wave amplitude, under which the proposed mechanism is effective, are within the error bars of the experimental values for both transparent dielectrics (α-Al_2O_3 crystals, amorphous SiO_2) and a typical metal (Al). Several theoretical predictions are compared with original, previously published, results.

1. Introduction

Investigations of laser-induced damage (LID) have been carried out for almost 30 years. Such investigations have always been stimulated by the need for more perfect optical materials, coatings, mirrors, etc. Despite the fact that much of the research has applied character, LID studies have been useful in a number of fundamental works in which the solid-state theory is developed [1–4]. At the same time, no universal conception of the LID phenomenon of condensed matter exists. The mechanisms of failure in metals, crystals, glasses and polymers proposed earlier usually take into account specific features of the named materials. The LID of both metals and dielectrics, nevertheless, have much in common. Note, first of all, that there is a cumulative effect, i.e. damage occurs due to repeated target irradiation by light pulses, each of which, separately, is 'safe' for the sample. The cumulative effect of optical irradiation is treated in some papers as the main feature of LID [5–8]. But the nature of the cumulative effect is still a point for discussion, even among researchers dealing with materials of the same class. As to materials of different classes, the situation is even more complicated. The cumulative effect has been ascribed to the influence of local inhomogeneities of the surface in metals [9], to

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absorbing impurities in glasses and crystals [10], and to thermolysis products in polymers [11]. There are a number of possible explanations for the existence of a cumulative effect invoking intrinsic LID models as well as impurity mechanisms.

In this paper we analyse original work, published previously, dealing with the cumulative effect of laser-induced damage in dielectrics and metals, paying special attention to the coincidence of LID characteristics in these materials when subjected to repeated 'pre-threshold' irradiation. A common approach to the LID problem for the whole class of solids is achieved by way of generalisation of experimental data. The mechanism of nucleation of an initial defect (ID), which is the basis of this approach, has been developed using a new theoretical model of accumulation of elastic energy as the result of laser action on condensed matter. The energy is absorbed by lattice inhomogeneities due to parametric resonance between the intrinsic vibrational modes of an inhomogeneity and a laser-induced acoustic wave. This leads to local heating of the lattice and to deformation of chemical bonds (up to the breaking of bonds). We shall attribute this breaking to nucleation of an incipient defect (a pore or a submicrocrack).

2. Pre-threshold damage

Macroscopic damage is usually manifested as a spark or plasma flare inside a sample or at its surface. The light-pulse power density (Q), or the electric field strength ($\mathscr{E}$), with respect to a spark appearing under the action of a single pulse, is taken to be the 'threshold' for optical resistance (Q_{th} or $\mathscr{E}_{th}$). However, these parameters do not characterise the material unambiguously, since their values depend upon the experimental conditions. The number of pulses prior to macro-damage, N_*, under irradiation with a field $\mathscr{E} < \mathscr{E}_{th}$, shows the resistance of the material to laser action. By definition, $N_* = 1$ when $\mathscr{E} = \mathscr{E}_{th}$, and it increases as $\mathscr{E}$ decreases.

Repeated pulse irradiation of a metal at $\mathscr{E} < \mathscr{E}_{th}$ causes non-plasma surface degradation visualised by electron microscopy [9] or by indirect methods, such as the emission of desorption and evaporation products [12], the generation of an acoustic signal [13], luminescence [14] and others. By using several complementary experimental procedures, one can obtain a complete picture of pre-threshold evolution of a metallic specimen, and relate the erosion of microrelief, under multiple-pulse surface irradiation, to the particular processes of degradation [12].

In [6, 13, 15] surface degradation in metals has been associated with the micromechanical action of heating and cooling cycles as the result of laser pulse action on a sample. By analysing the dependence of the number of pulses, before plasma flare, on the value of Q, Bass *et al* [6, 15] have suggested that the residual deformations that accumulate around the heating region are the reason for damage under repeated irradiation. The authors [6] give a convincing analogy with the cyclic mechanical loading of metals, but they do not consider the mechanism of ID nucleation. The latter is often connected by experimentalists with isolated surface inhomogeneities, impurities, etc [9]. In our opinion, the available experimental data make it possible to propose a general theoretical model for intrinsic ID nucleation. Furthermore, it is well known (see, e.g., [5, 7, 16–20] and elsewhere) that LID of transparent solids under multiple-pulse 'pre-threshold' irradiation is also the result of irreversible processes stored in the material.

As a starting point for our model, we accept that the laser defect formed at 'pre-threshold' field strength is a particular case of mechanical fracture occurring in a dynamic stress field. The elastic (acoustic) waves propagating in the condensed medium due to

laser interaction with matter are the source of mechanical stress. The nature of acoustic wave generation by lasers in metals and transparent dielectrics is different. Therefore, we briefly characterise below the conditions for such generation.

3. Generation of elastic waves on laser interaction with solids

3.1. Metals

The acoustic waves arise under pulsed laser heating of a metal surface [13, 21–24]. The mechanism of wave generation changes slightly as the laser power density overcomes the 'threshold' of plasma flare formation. The temperature of the irradiated region increases sharply at $Q \geqslant Q_{th}$ as a consequence of plasma absorption. As a result, a shock wave of thermal breakdown appears, whose amplitude U_0 is proportional to Q^4 [13, 23]. At $Q < Q_{th}$ a purely thermoelastic effect is observed, for which $U_0 \sim Q$ [13, 21–23]. This is the case in which we are interested. The characteristic values of amplitude and frequency ω of the thermoelastic wave have been determined reliably in experimental work [13, 21, 24]. We shall use the respective data for numerical estimates of the parameters in our theoretical model.

3.2. Transparent dielectrics

The acoustic waves appear in transparent dielectrics due to non-linear scattering of the intense light by thermal density fluctuations, i.e. the effect of stimulated Brillouin scattering (SBS). The experimental observation of SBS made it possible to consider laser damage as a particular case of mechanical fracture by laser-stimulated acoustic waves. Such an approach was first proposed in [1] and studied theoretically in [25]. The idea that SBS is associated with LID was repeatedly mentioned in the literature [26–30]. The experimental data on this problem are quite contradictory (cf. [28–30] and [31–33]), but theoretical estimates show that the pressure of a hyperacoustic wave produced by SBS is probably insufficient to nucleate a defect [25, 26]. Nevertheless, the feasibility of the defect appearing under the action of SBS-produced elastic waves is admitted in theoretical papers [25–27], provided that: (i) there exist some mechanical concentrators of tension (microcracks [25]), or (ii) additional heating of the laser-irradiated dielectric occurs due to electron processes [27].

We shall consider a non-linear effect of acoustic wave interaction with large-scale thermal density fluctuation, which proves to be sufficient for developing the model of ID nucleation.

4. Thermo-fluctuation theory of initial defect nucleation

4.1. Mechanical fracture: previous results

Let us analyse the general physical principles in the theory of ID formation in a stationary mechanical field, in order to clarify the mechanism of LID. There are two main concepts in the theory of fracture: the structural approach and the thermo-fluctuation approach.

According to the first approach, the elementary acts of fracture, or breaking of interactomic bonds, take place in an immovable lattice under the action of an external force concentrated at the structural defects. As an example of such defect concentrators,

we mention the Griffith crack [34] and Cottrell [35] and Stroh [36] dislocation clusters. An elementary act of fracture is considered in [34–36] as a simple mechanical overcoming of near-atomic planes without thermal vibrations of atoms. Here fracture is the result of plastic deformation responsible for the generation of defect concentrators.

According to the other approach, i.e. the Zhurkov kinetic concept of strength [37], one should take into account thermo-fluctuations in order to describe an elementary act of fracture. A loaded solid body is considered, within the framework of the kinetic model, as an essentially non-equilibrium system, in view of both the developing ensemble of defects in the body and the change with time of the phonon mode populations (or filling numbers $n(\bar{k})$, where $\bar{k}$ is the wavevector).

The two approaches are not completely incompatible, as the evolution of defects and the evolution of phonon states are interrelated and should lead to macroscopic fracture because of the intrinsic instability of the ensemble of atoms (at least, in the case of tensile stress). From the theory of non-equilibrium systems it follows [38] that any instability in a system of atoms (including the instability leading to damage) should be developed from large-scale (giant) fluctuations (LSF) having average dimension $L \gg r_0$, where r_0 is the atomic parameter. The LSF can be considered as a nucleus of dissipative structure, which exchanges energy and mass with its environment. A theory of such 'damaging' thermal fluctuations, in the one-dimensional case (a loaded atomic chain), has been proposed in [39]. The idea of the theory is the following. The LSF is the spontaneous dynamic inhomogeneity, i.e. the negative density fluctuation with linear dimension close to the path length of thermal phonons. Since the sound velocity C_l' in the region of the LSF differs from that in the unstretched chain (C_l), the fluctuation exchanges energy with the environment due to the flow of thermal (short-wave) phonons. Under certain conditions energy accumulation is higher than that lost through scattering, which leads, owing to the vibrational anharmonicity of the chain, to local heating of the region involved in fluctuation, and steady stretching of interatomic bonds up to their breaking in the LSF. The local deformation ε and temperature T are the critical parameters of the 'damaging' LSF. They are expressed as [39]:

$$\varepsilon_{cr} = \frac{1}{2g}\left[1 - \left(\frac{f_{fr} - f}{gk_B T}\right)^{-2/3}\right] \tag{1}$$

$$T_{cr} = T\left(\frac{1 - 2g\varepsilon}{1 - 2g\varepsilon_{cr}}\right)^{1/2} \tag{2}$$

where k_B is the Boltzmann constant, f_{fr} is the theoretical strength of the chain, and g is the Grüneisen coefficient. At $\varepsilon < \varepsilon_{cr}$, the LSF relaxes rapidly by releasing energy into the environment. At $\varepsilon \geqslant \varepsilon_{cr}$, the LSF absorbs energy from the environment, which causes chain disintegration (fluctuation instability). Thus, a loaded solid body obtains an initial defect. The concept of 'damaging' LSF described as quasi-particles ('dilatons' [39]) is developed in [39–42] applied to the case of a body loaded by a static mechanical stress.

4.2. Laser-induced damage

In this paper we consider a parametric mechanism for irreversible LSF growth in the field of a laser-produced acoustic wave. The parametric mechanism in this case is different from that described briefly above, as the wavelength of the wave is $\lambda \gg L$ (where L is the dimension of the inhomogeneity), in contrast to the case of static stress, where the wavelength of thermal phonons is much less than L. The mechanism of defect formation,

as a result of the laser-stimulated acoustic waves, suggests that intrinsic vibrations (IV) of the inhomogeneity (here, LSF) should be amplified due to parametric resonance between these vibrations and the long-wave (compared with the LSF dimension) perturbation. The amplified amplitudes of certain modes, $u_n(t)$, grow with time until the absorbed energy causes breaking of bonds in the inhomogeneity due to thermal expansion of the lattice. To obtain the conditions of such a parametric pumping, we make a detailed analysis of the interaction of acoustic waves with a negative density fluctuation. We represent the LSF as some anharmonic inhomogeneity, with dimension $L \ll \lambda$, characterised by tensor $\hat{G}$ of the modulus of elasticity and anharmonicity tensor $\hat{A}$. Suppose that this inhomogeneity involves small IV (say, of thermal origin):

$$\Delta u_n(\bar{r}, t) = \Delta u_n(\bar{r}) \exp(i\omega_n t)$$

where $\Delta u_n(\bar{r}, t)$ is the shift vector in the nth mode, $\bar{r}$ is the spatial coordinate, and ω_n is the frequency of the respective mode. These vibrations arise as a consequence of solving the equation of motion of the inhomogeneity in the harmonic approximation,

$$\rho\ddot{u} = \hat{G} : (\partial \hat{Y}/\partial r) \tag{3}$$

with corresponding boundary conditions,

$$\Delta u_n|_S = 0 \qquad \hat{A} = 0 \tag{4}$$

where ρ is the inhomogeneity density, $\hat{Y}$ is the deformation tensor, S is the inhomogeneity surface and (:) is the designation of scalar dyad. Let us express the potential energy of the inhomogeneity, U, and the stress tensor, $\hat{T}$, by [43]:

$$U = (1/2)\hat{G} :: \hat{Y}\hat{Y} + (1/3!)\hat{A} ::: \hat{Y}\hat{Y}\hat{Y} \tag{5}$$

where the equation of motion of the inhomogeneity in the anharmonic approximation has the form

$$\rho\ddot{u} = (\hat{G} + \hat{A} : \hat{Y}) : (\partial \hat{Y}/\partial r). \tag{6}$$

Let us consider the interaction of the inhomogeneity IV with a longitudinal acoustic plane wave propagating along the x axis, and providing the medium shift $u_x = u_0 \sin(\omega t + kx)$. Here $\omega \ll \omega_n$ due to the condition of the long-wave approximation ($\lambda \gg L$). This wave stimulates a slow deformation of the inhomogeneity, $u'_x = u_0 \sin(\omega' t + k' x)$, compared with the frequency of IV. The shifts u_x and u'_x are matched at the boundary in the long-wave approximation with accuracy up to small terms, which are linear in the parameter L/λ. By substituting the atomic shift, $u = u_x + \Sigma\, \Delta u_n$, into the equation of motion (6), and using the condition $|u'| \gg |u_n|$, we can divide equation (6) into two equations, one for the forward wave and another for the fast IV inside a slowly deforming inhomogeneity:

$$\rho\Delta\ddot{u}_n = [\hat{G} + \hat{A} : \hat{Y} + \hat{A} : (\partial u'/\partial r)] : (\partial^2 \Delta u_n/\partial r^2) \tag{7}$$

where $\partial u'/\partial r$ is the deformation tensor of the acoustic wave. Equation (7) for $\Delta u_n(\bar{r}, t)$ can be rewritten for the time-dependent amplitudes $\Delta u_n(\bar{r}, t) = \Delta u_n(\bar{r}) A_n(t)$ in the form of a Mathieu function describing the parametric resonance:

$$d^2 A_n(t')/dt'^2 + [a_n - 2q_n \cos(2t')] A_n(t') = 0 \tag{8}$$

where

$$a_n = 4\omega_n^2/\omega^2 = 4n^2\lambda^2 C_l'^2/L^2 C_l^2 \tag{9}$$

$$q_n = 8\pi u_0 n^2 \lambda g C_l'/L^2 C_l. \tag{10}$$

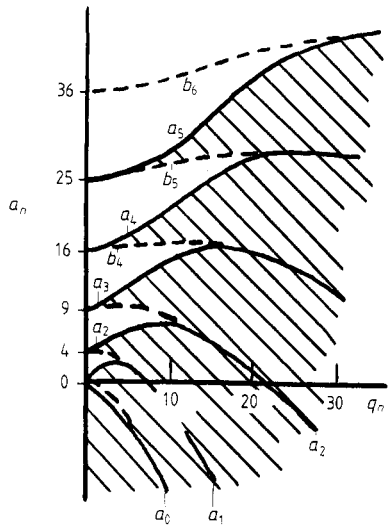

Figure 1. Mathieu equation instability diagram.

Here $t' = \omega t/2$; the right-hand sides of equations (9) and (10) are given for the case of an inhomogeneity in the form of a cube oriented normal to the axis (a model that is easy to calculate). The general solution for equation (9) has the form [44]:

$$A_n(t') = \begin{cases} A\,e^{\mu t'} \sum_{r=-\infty}^{\infty} C_{2r}\,e^{2rit} + B\,e^{-\mu t'} \sum_{r=-\infty}^{\infty} C_{-2r}\,e^{-2rit'} & (11) \\ A\,e^{\mu t'} \sum_{r=-\infty}^{\infty} C_{2r+1}\,e^{(2r+1)it'} + B\,e^{-\mu t'} \sum_{r=-\infty}^{\infty} C_{-(2r+1)}\,e^{-(2r+1)it'} & (12) \end{cases}$$

where A and B are real numbers, r is an integer, C_{2r} and C_{2r+1} are the amplitudes of the Bloch wave, and μ is the characteristic index. Equation (11) is applicable if point $\{a_n, q_n\}$ lies between the curves a_{2n} and b_{2n+1} (or between a_{2n+2} and b_{2n+2}) of the steady-state diagram (figure 1). Equation (12) is valid if point $\{a_n, q_n\}$ lies between the curves a_{2n+1} and b_{2n+2} (or a_{2n+1} and b_{2n+1}). If μ is a real number, the solution of equations (11) and (12) is unstable, i.e. $A_n(t) \to \infty$, which corresponds to the excitation of the nth mode and pumping of energy into the inhomogeneity. The energy pumping occurs in this case with rate $\mu = \{[(a_m - a_n)/(a_n - b_m)]/2m\}^{1/2}$, where m is the number of the resonance. Mathematically, this condition corresponds to the localisation of point $\{a_n, q_n\}$ into the hatched region of figure 1.

Let us use the results of our analysis (equations (9)–(12)) applied to LSF. As LSF parameters we choose the following data:

(i) The dimension of LSF is $L = \Lambda$, where Λ is the average path length of thermal phonons, which can be estimated by the formula [45]

$$\Lambda \simeq MC_l^2 r_0/k_B T g \qquad (13)$$

where M is the atomic mass.

(ii) The substance density in the LSF, with respect to the degree of bond stretching, is close to the mean-square density fluctuation, and has temperature $T' = T$.

Let us assume that all normal modes of vibration in the LSF region are thermally excited, i.e. their amplitudes are determined by the expression [45]:

$$\frac{\Lambda}{r_0} m\left(\frac{\omega_n}{2\pi}\right)^2 u_n^2(0) = \tfrac{1}{2}k_B T. \tag{14}$$

As follows from equations (9) and (10), $a_n \sim \lambda^2$, $q_n \sim \lambda$ and λ lies in quite a wide range. Therefore, the parametric resonance condition can be fulfilled for the mode with number n. This mode proves to be resonant relative to the acoustic wave, and determines the transition of part of the thermoelastic perturbation energy into LSF excitation energy. Since this excitation occurs within a finite time, $t = t^*$, close to the characteristic time of the acoustic wave damping, $t^* = (2/\alpha_p)C_l$. Here $2/\alpha_p$ is the path length of an acoustic phonon. There are two possible situations:

(i) Within time t^* the LSF so heightens the vibration amplitude of the resonant mode, $u_n(t^*)$, that the deformation $\varepsilon_n = u_n(t^*) \simeq \varepsilon_{fr} = 1/2g$ of the bonds, due to the nth mode, proves to be close to rupture. After that, the LSF is broken and a submicrocrack appears, i.e. an initial defect.

(ii) Time t^* proves to be so small that the energy absorbed by the inhomogeneity is insufficient to rupture the bonds ($\varepsilon_n < \varepsilon_{fr}$).

Our task is to evaluate the amplitude of the acoustic wave for the case (i). The amplitude of the resonant mode $u_n(t)$ increases exponentially with time (if point $\{a_n, q_n\}$ is in one of the instability regions of the Mathieu equation), and is expressed by [44]

$$u_n(t) = u_n(0)\, e^{\mu_n t} \tag{15}$$

where the amplification rate is $\mu_n \simeq 10^{-m} q^m (8m)^{-1/2}$. (Here $\{a_m, q_m\}$ are the points of figure 1 corresponding to the intersection of the line $q_n = \text{const}$ with the characteristic steady-state diagrams of the Mathieu equation with the point $\{a_n, q_n\}$ lying between them.) Increasing the amplitude leads to an increase in the maximum of the local deformation in the node of the standing wave with number n:

$$\varepsilon_n = u_n(t) 2\pi n/\Lambda. \tag{16}$$

As soon as $\varepsilon_n = \varepsilon_{fr} = 1/2g$ (i.e. the deformation is close to the rupture deformation, ε_{fr}) the inhomogeneity is broken. From equation (16) it follow that the amplitude of the nth mode, at that moment, is $u_n(t) = 4\pi\Lambda n g$. Taking into account this expression for the amplitude, we get from equation (15) the condition for the inhomogeneity breaking within time $t < t^*$:

$$t^* > \frac{1}{\mu_n} \ln \frac{\Lambda}{4\pi n g u_n(0)}. \tag{17}$$

By substituting $u_n(0)$ from equation (14) and μ_n from equation (15) into this condition, we obtain that damage is possible starting from some critical amplitude u_{cr} expressed by

$$u_{cr} = 10\left(\frac{\lambda\alpha_p m^{1/2}}{2}\right)^{1/m} \frac{\Lambda^2 C_l}{8\pi g n^2 \lambda C_l'} \ln^{1/m}\left[\frac{\Lambda}{4\pi g}\left(\frac{2\Lambda M C_l'^2}{r_0^3 k_B T}\right)^{1/2}\right] \simeq \frac{\Lambda^2 C_l}{\pi g n^2 \lambda C_l'}. \tag{18}$$

By comparing this with experiment we take, for convenience, that $C_l \simeq C_l'$ and $\lambda = 2\pi C_l/\omega$. Then the condition for defect formation becomes

$$u_a > u_{cr} \simeq \frac{\Lambda^2 \omega}{2\pi g n^2 \lambda C_l}. \tag{19}$$

4.3. Numerical estimates

We shall estimate u_{cr} for a metal (Al) and two dielectrics (α-Al_2O_3 and vitreous SiO_2), for which we have found, in the literature, all the necessary parameters for the calculation.

The frequency of the acoustic waves produced under a 'pre-threshold' laser action on metals is 1–10 MHz [22, 24], the path length of thermal phonons in Al is 2×10^{-5} cm [43], the sound velocity is 6×10^5 cm s^{-1} and $g = 2$. By substituting these values into (19) we find that $u_{cr} = 10^{-10}$–10^{-11} cm for the excitation of mode 1–2. This value is much less than the amplitude of laser-stimulated thermoelastic waves found by direct measurements [21] under pre-threshold irradiation of aluminium, $u_a = (1\text{–}10) \times 10^{-8}$ cm. Hence, the condition $u_a > u_{cr}$ can be fulfilled for this metal and the nonlinear interaction of the acoustic wave with the LSF may lead to the formation of a defect.

For dielectrics, Λ is 5×10^{-7} cm [46] and 8×10^{-8} cm [47], and C_l is 1.1×10^6 and 1×10^5 cm s^{-1} for α-Al_2O_3 and SiO_2, respectively. Let us take also that $\omega/2\pi = 2 \times 10^{10}$ Hz [48]. Then, by using equation (19) we find that $u_{cr} \sim 10^{-10}$ cm for α-Al_2O_3 and $u_{cr} \simeq 0.3 \times 10^{-10}$ cm for SiO_2.

The source of acoustic waves on laser interaction with a transparent dielectric is stimulated Brillouin scattering (SBS). We do not know of any work on the direct measurement of the SBS-produced hypersonic amplitudes and, therefore, make a semi-empirical estimate of u_a and compare it with u_{cr} calculated above.

The mean intensity of hypersound, I_a, at SBS can be evaluated by the following formula [47], verified experimentally [48]:

$$I_a = \frac{c^2\gamma^2\mu_0\varepsilon_0}{128}\frac{C_l}{E_0}\left(\frac{2}{\alpha_p C_l}\right)^2\left(\frac{C_l}{\lambda}\right)^2 \mathscr{E}^4. \tag{20}$$

Here $\gamma \leqslant 1$ is the photoelastic constant of the material, ε_0 and μ_0, respectively, are the dielectric and magnetic permeability, c is the light velocity, E_0 is Young's modulus and α_p is the path length of the hypersonic phonon. The acoustic wave intensity is connected with the amplitude by the following relation:

$$I_a = \tfrac{1}{2}C_l\rho\omega^2 u_a. \tag{21}$$

By equating the right-hand sides of equations (20) and (21), and taking $Q = \frac{1}{2}c\mathscr{E}^2$, we find the dependence of u_a on the field in a laser beam:

$$u_a = \frac{c\gamma}{16\pi}\left(\frac{\mu_0\varepsilon_0}{\rho E_0}\right)^{1/2} t^*\mathscr{E}^2. \tag{22}$$

From the latter equation it is seen that $u_a \sim Q$, as in the case of metals. For SiO_2 and α-Al_2O_3 we have $\varepsilon_0 = 2$, $\rho = 4.0$ and 2.6 g cm^{-3}, $E_0 = 6$ and 4 GPa, and $t^* = 7 \times 10^{-8}$ and 4.5×10^{-7} s [49], respectively. 'Threshold' optical resistances of both dielectrics in the nanosecond pulse region are approximately the same, $\mathscr{E}_{th} = (1\text{–}5) \times 10^5$ V cm^{-1}. By substituting $\mathscr{E} = 10^5$ V cm^{-1} in equation (22) we calculate the maximum possible amplitude of hypersound (without sample damage due to the first light pulse): $u_a \simeq 10^{-9}$ cm for SiO_2 and 2×10^{-9} cm for α-Al_2O_3. The condition $u_a \geqslant u_{cr}$ is fulfilled and, therefore, ID may be formed due to the interaction of elastic waves with LSF for transparent dielectrics.

It is interesting to note that u_{cr} for metals is close to u_{cr} for dielectrics, due to equation (19), where $u_{cr} \sim \Lambda^2$. Although the path length of phonons in metals differs by two orders of magnitude from that in transparent dielectrics, the hypersonic frequency of 10^{10} Hz in dielectrics differs by four order of magnitude from the frequency of

thermoelastic waves in metals ($\sim 10^6$ Hz), and the product $\Lambda\omega^2$ proves to be the same for metals and dielectrics.

5. Thermo-fluctuation nature of inhomogeneities

It is evident that the mechanism of ID formation via a non-linear interaction of the elastic waves with the inhomogeneity of a crystal lattice can also be developed for stable structural or even impurity inhomogeneities. For example, laser breakdown of a liquid may occur due to the interaction of SBS-produced hypersound with intrinsic vibrations of solid impurity particles. Such an approach would, however, deprive the proposed mechanism of its universality, as one would have to suggest the existence of inhomogeneities with admissible parameters, such as dimensions, density, spectrum of vibrations, etc. Having chosen LSF as the inhomogeneity, we have developed an intrinsic mechanism of ID formation. Its efficiency depends solely on the laser intensity and the physical constants of the material (path length of phonons, Grüneisen coefficient, elastic constants, etc). The numerical estimates performed justified the validity of the chosen model of inhomogeneity. Besides, one can verify experimentally some qualitative features of the thermal atomic motion that are typical of the process of ID formation. Such features are the dependences of the critical field of ID formation on the temperature and external stress of the sample. The proposed effects are a consequence of the dependence of thermal phonon path length on temperature T and stress σ. For experimental verification, we get expressions that relate $\mathscr{E}_{cr}$ with T and σ.

Let us use the estimates for the phonon path length in transparent dielectrics from [45]:

$$\Lambda = MC_l^2 r_0 / k_B T g^2 \tag{23}$$

$$C_l = \left(\frac{E_0 r_0}{M}(1 - 2g)\right)^{1/2} \tag{24}$$

$$\varepsilon = (\sigma r_0^3 + g k_B T)/E_0 r_0^3. \tag{25}$$

By equating the right-hand sides of equations (19) and (22) and using the expressions (23)–(25), we get an estimate for the critical field:

$$\mathscr{E}_{cr} = \frac{4E_0 r_0^4}{k_B T g^2}\left(1 - \frac{2g\sigma}{E_0}\right)\frac{(\rho E_0)^{1/4} t^{*-1/2}}{n[c(\mu_0 \varepsilon_0)^{1/2}]^{1/2} g\gamma\lambda}. \tag{26}$$

For metals, $u_a \sim Q$ (pre-threshold irradiation), $u_{cr} \sim \Lambda^2$ (from equation (19)), and taking $Q = \frac{1}{2}c\mathscr{E}^2$, we get $\mathscr{E}_{cr} \sim \Lambda \sim (1 - 2g\sigma/E_0)$. It is seen that this relation is analogous to equation (26) for dielectrics.

5.1. Optical resistance of stressed solids

Supposing that (i) material resistance to the action of repeated laser pulses is determined by the kinetics of microdefect accumulation and (ii) the accumulation rate depends on $\mathscr{E}_{cr}$, we tried to find a relation between $\mathscr{E}_{cr}$ and σ in experiments on multi-pulse irradiation of stressed samples.

As dielectric samples we used optical fibre of 125 μm thickness. The quartz glass for the fibre is produced by chemical deposition of gaseous $SiCl_4$, i.e. the samples were

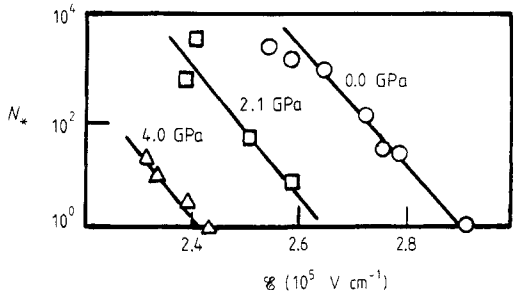

Figure 2. Number of laser pulses, N_*, needed for damage as a function of field strength in the stressed quartz glass fibre.

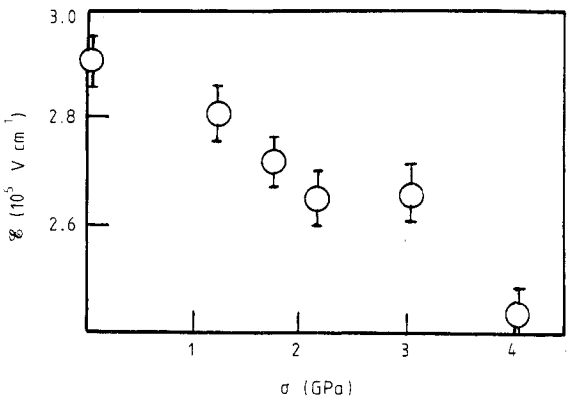

Figure 3. Breakdown field strength, $\mathscr{E}$, versus tensile stress, σ, on the surface of quartz glass.

highly pure and geometrically perfect. The fibres were stressed by means of bending with a fixed radius of curvature; the tensile stress on the surface with greater radius was calculated by the method described in [50]. A neodymium glass laser with 60 ns pulse duration was the source of radiation in these and other experiments. The light beam was focused onto the convex surface of a bent fibre into a 0.07 cm diameter spot.

The influence of tensile stress on the 'waiting time' of damage appears in the dependence of N_* versus $\mathscr{E}$ at σ = const. The respective graph was plotted on semilogarithmic coordinates (figure 2); the intersection of the graph lines with the line $N = 1$ gives the value of $\mathscr{E}_{th}$ at which the sample is damaged in one shot. The sought dependence of $\mathscr{E}_{th}$ versus σ extracted from figure 2 is shown in figure 3. As predicted theoretically, $\mathscr{E}_{cr}$ decreases as σ increases.

Analogous measurements were taken on metal. The samples were commercial gold foils with 99.99% content of pure material. The upper end of a metal strip, of 0.2 mm width and 50 μm thickness, was fastened in a movable gripping device, which provide mutually perpendicular motion of the sample in the focal plane of a lens. To the bottom of the sample was fastened the tensile load. Each point of the sample was irradiated up to the appearance of the damage flash, as in the case of loading of the quartz fibre.

The measurement results are shown in figure 4. Despite a large spread in the experimental points, one can observe the tendency to a reduction of optical resistance with the growth of tensile stress.

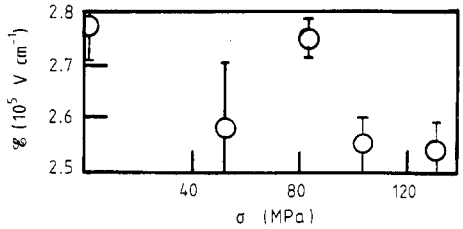

Figure 4. Breakdown field strength, $\mathscr{E}$, versus tensile stress, σ, on a gold foil.

Table 1. Number of laser pulses before plasma flare at various temperatures of metal target.

Metal	Pulse duration, t_p (s)	Field strength, $\mathscr{E}$ (10^5 V cm^{-1})	Temperature, T (°C)		
			20	65	170
Ag	6×10^{-8}	10.0	13560	—	60
Ag	5×10^{-4}	0.5	100	13	3
Au	6×10^{-8}	2.5	210	24	2
Ni	6×10^{-8}	0.2	30	—	1

5.2. Temperature dependence

The temperature dependence of the path length of thermal phonons determines the temperature dependence of LID in our model. The decrease in laser strength with increase in temperature of the sample is a well known effect for transparent dielectrics [16, 51, 52]. The temperature dependence (as well as the dependence on field) should be equally typical of LID not only of dielectrics but of metals as well, in accordance with the thermo-fluctuation model.

The experimental data confirm the theory, e.g. the number of pulses needed to obtain damage decreases noticeably on heating of the metal sample even by several tens of degrees (table 1). Thus, decreasing the threshold of ID formation on heating leads to a faster accumulation of defects, which in turn reduces the 'waiting time' of macroscopic breakdown.

5.3. Time dependence

We examined the dependence of optical resistance on T and σ under multi-pulse irradiation of a sample, and have made only a qualitative verification of equation (26), since this equation determines the value of the critical field for formation of a single ID, while we could record the formation of macroscopic damage. In other words, we assumed implicitly that the 'waiting time' of damage (the summary irradiation time) is determined by evolution of the ID ensemble. Remember that the thermo-fluctuation approach to LID was first considered in studying the damage kinetics under repeated irradiation [7, 8]. The dependence of the number of shots before damage on the field was found to be exponential for transparent dielectrics [7, 8, 16]. Graphically, the dependence of log N_*

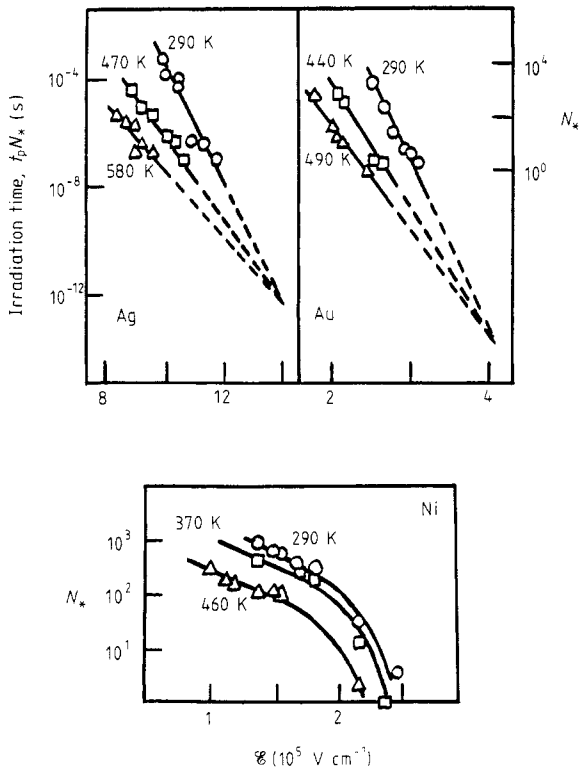

Figure 5. Number of pulses, N_*, before damage versus field strength and temperature for metals.

on $\mathscr{E}$ is plotted in the form of straight lines, and their slope decreases with increase in sample temperature. The experimental dependence of Boltzmann character

$$N_* \sim \exp[U(\mathscr{E})/k_B T] \tag{27}$$

where $U(\mathscr{E})$ is the damage activation energy, made it possible to consider that the dependence of the number of pulses before breakdown on the experimental conditions is governed by the kinetics of submicroscopic defects produced in a thermo-fluctuation manner [8].

The thermo-fluctuation mechanism of LID is equally applicable to both transparent dielectrics and metals, so one might expect that the exponential dependence of N_* on inverse temperature is fulfilled for metals as well. The result of measuring N_*, for different fields in a pulse, is shown in figure 5. When our data for log N_* are plotted as a function of $\mathscr{E}$, they lie on a 'bundle' of straight lines meeting at one point. Hence, equation (27) is also valid for metals.

(Our experiments were performed in air; therefore, we refer the conclusion only to Au and Ag having no oxide films on a newly cleaned surface. For Ni we have observed deviation from straight lines in the region of the first few tens of pulses, which seems to be due to purification of the sample surface from impurities and oxide films during the first few pulses.)

The result obtained for metals justifies a thermo-fluctuation approach to choosing the type of inhomogeneity as the source of the future defect; secondly, it gives an

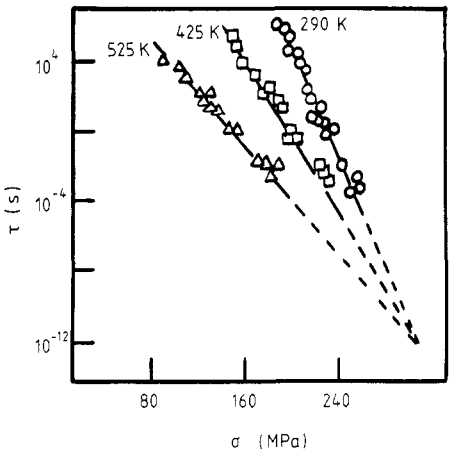

Figure 6. Time, τ, needed for mechanical failure of silver foil versus tensile stress, σ, and temperature.

additional argument for consideration of LID as a specific case of mechanical fracture under dynamic stress at an atomic scale level.

The 'waiting time' of mechanical fracture on static loading depends, analogously, on stress and temperature (figure 6) [53], i.e. at least the temperature–time behaviours of mechanical and laser damage coincide. Note that Bass *et al* [6] also suggested that the 'mechanical' nature of laser defects should be verified through the analogous behaviours of dependences of number of laser pulses needed to produce macro-damage versus Q and number of stress cycles required to cause mechanical failure versus σ.

6. Conclusions

The main results of the work are as follows:

A theoretical model is proposed for the non-linear interaction of elastic waves with a lattice inhomogeneity in solids.

A new approach to the formation of an initial defect under the action of optical irradiation has been developed on the basis of this model, where (i) a thermoelastic effect in metals and stimulated Brillouin scattering in transparent dielectrics were considered as the source of elastic waves under laser irradiation and (ii) a large-scale thermal density fluctuation (with dimension of a thermal phonon path length) was assumed to be a universal inhomogeneity always available in a condensed medium.

The numerical estimates were made on the basis of experimental data, which confirm the possibility of defect formation according to the proposed mechanism. It is shown experimentally that the model is in qualitative agreement with the main trends of laser damage induced by a multi-pulse 'pre-threshold' irradiation.

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