THE ROLE OF ABSORBING IMPURITIES IN LASER-INDUCED DAMAGE OF TRANSPARENT DIELECTRICS

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A theory of light-induced thermal damage of transparent dielectrics constaining absorbing impurities is developed. Temperature nonlinearities of thermal constants of the impurities and the matrix and the temperature dependence of the absorption coefficient are taken into account. It is shown that these factors are significant in the damage mechanism and cause an avalanche increase of temperature within the impurities corresponding to a thermal explosion. The damage threshold conditions are defined and threshold dependence on impurity size and laser pulse length is determined. Threshold damage power is computed by way of example for ruby crystals and glass containing metallic impurities and is found in agreement with observed thresholds.

1. INTRODUCTION

In recent years a number of papers appeared dealing with the damage of transparent dielectrics containing absorbing inclusions[1–3]. This problem is studied in particular detail in the case of laser glass with metallic and dielectric inclusions[2,3]. It was found from experiments that platinum inclusions in glass, for example, affect significantly the damage threshold. An analogous effect was also observed in laser ruby crystals containing impurities of the iron-group metals (Ni, Co, Fe, Ti)[4].

The mechanism of damage on absorbing inclusions discussed in[2,3] is based on strong heating by laser radiation that can be accompanied by phase transitions within the inclusions (melting and vaporization) and thermoelastic stresses in the surrounding matrix. The above papers presented a linear theory of thermoelastic damage and utilized static critical stress data to compute damage thresholds. Such an approach can obviously lead to considerable errors for the following reasons. First, this approach leaves critical temperatures and stresses indeterminate because of the dynamic nature of the damage process (especially in the region of short laser pulses). Second, the temperature dependence of thermal, elastic, and optical parameters is significant at the high temperatures ($\sim 10^4$°K and over[3]) generated in the course of the damage process.

The possible role of temperature nonlinearities in the damage process was indicated in[2,5]. An attempt to account for the temperature dependence of the absorption coefficient of the inclusions was made in[6].

We consider a more consistent theory of thermal damage to transparent dielectrics taking into account the above nonlinearities of the absorbing inclusions as well as of the surrounding matrix. As we show below, with such an approach it is no longer necessary to know the specific values of critical temperature and stress causing the damage in order to determine the damage threshold. Consequently we can determine more realistically the threshold of internal damage in transparent dielectrics containing absorbing inclusions.

2. THEORETICAL ANALYSIS OF THRESHOLD DAMAGE CONDITIONS

For the sake of simplicity we consider an absorbing spherical particle with radius a in a transparent dielectric. The heating of such a particle and its surrounding medium by laser emission is described by the heat-conduction equation[2)]:

$$\frac{\partial}{\partial t}(C\rho T)=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2k\frac{\partial T}{\partial r}\right)+Q(I,T), \tag{1}$$

where C, ρ, and k are heat capacity, density, and thermal conductivity coefficient, have generally different values for the inclusion material and for the surrounding matrix and are temperature dependent, while Q(I, T) is the power of heat sources in the particle and depends on the laser emission intensity and instantaneous temperature T.

The temperature nonlinearity of Q can be expressed by various laws, depending on the mechanism of light absorption; for example it can be expressed by the well-known approximation[7]:

$$Q(I,T)=Q(I)\exp\left(\xi\frac{T-T_0}{T_0}\right), \tag{2}$$

where T_0 is the initial specimen temperature and ξ is a parameter of temperature nonlinearity of the impurity absorption coefficient of the inclusion. In general, such a dependence can be valid within a limited temperature interval.

The temperature dependence of the thermal conductivity coefficient is in general different for different materials. We take it in the form

$$K(T)=\alpha/T, \tag{3}$$

where α is the constant of the material. This dependence is normally realized in dielectrics for T above

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[2)]This is valid for particles that are not too small, when the size effects in the phonon spectrum are insignificant.

the Debye temperature θ and for a number of metals (nickel for example at $T > 300°K$)[8].

We seek the solution of (1) for $r < a$ and time $t \gtrsim \tau_x = T_0 C_1 \rho_1 a^2/\alpha_1$ (index 1 refers to the inclusion meterial and 2 to the surrounding matrix) in the form

$$T(r, t) = T_0 \exp\{u(t)(\beta - r^2/3a^2)\}, \tag{4}$$

where $\beta = \frac{1}{3}(1 + 2\alpha_1/\alpha_2)$ and $u(t)$ is some function weakly varying in time τ_x. Such an approximation is valid if $\tau_x dT/dt \ll T - T_0$. Substituting (4) into (1) we obtain the following implicit function for temperature at the center of the inclusion at the time t:

$$t = \int_{x_1}^{x_2} \frac{T_0 \beta e^{\beta x} C_1(T_0 e^{\beta x}) \rho_1(T_0 e^{\beta x})\, dx}{Q(I, T_0 e^{\beta x}) - 2\alpha_1 x/a^2}, \tag{5}$$

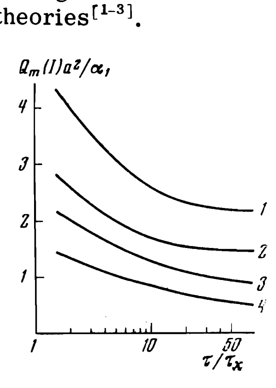
where

$$x_1 = \frac{a^2}{2\alpha_1} Q(I, T_0), \quad x_2 = \frac{1}{\beta} \ln \frac{T}{T_0}.$$

Using relation (2) for $Q(I, T)$, assuming that C_1 and ρ_1 are independent of temperature, and considering the rapid convergence of integral (5), its upper limit can be set at infinity. Then

$$t_{\mathrm{thr}} = \tau_x \int_{x_1}^{\infty} \frac{\beta e^{\beta x} dx}{\alpha_1^{-1} Q_{\mathrm{thr}}(I) a^2 \exp[\xi(e^{\beta x} - 1)] - 2x}. \tag{6}$$

In this case t_{thr} has the meaning of a time interval after which the temperature in the inclusion center begins to rise rapidly. The maximum possible temperature in the particle can be sufficiently high and can be determined either by saturation of the temperature nonlinearity of absorption or by thermal emission energy losses. This temperature determines the maximum stress in the surrounding matrix and consequently the nature, magnitude, and dynamics of matrix damage. However the threshold damage conditions can be found even without knowing these maximum temperatures and stresses. In fact, the rapid avalanche-like increase of inclusion temperature preceding the saturation effect and predicted by (6) means that time t_{thr} can be taken as the onset of damage and the quantity Q_{thr} as the threshold damage power. Here we consider that there is no damage along the linear region of the impurity temperature curve (for $t < t_{\mathrm{thr}}$). This is also indicated by the well-known fact that the damage of transparent dielectrics due to laser emission always has a sharply defined nature and is accompanied by a high-temperature emission (spark) analogous to a laser breakdown in gas. Such a threshold high-temperature luminescence fits well in our model of nonlinear thermal damage and cannot be explained by the existing linear theories[1-3].



Dependence of threshold damage power on laser pulse length. 1–$\beta = 1$, $\xi = 0.1$; 2–$\beta = 2.5$, $\xi = 0.1$; 3–$\beta = 1$, $\xi = 0.2$; 4–$\beta = 2.5$, $\xi = 0.2$.

The damage process in our model thus has the character of a thermal explosion.

Equation (6) determines the threshold damage power as a function of the laser pulse length. In particular, for rectangular pulses with length τ this function has been calculated with a computer for some values of the parameters ξ and β and is shown in the diagram.

In the case when $k(T) = \alpha_1/T$ and the temperature nonlinearity of absorption is weak enough so we can take only the first two terms of the series expansion in (2), we find for the inclusion center temperature:

$$\frac{T(t) - T_0}{T_0} \cong \frac{Q(I) a^2}{\alpha_1 \gamma}\left[1 - (1 - \gamma)\exp\left(-\frac{t\gamma}{\tau_x}\right)\right],$$
$$\gamma = \frac{Qa^2}{\alpha_1}(\xi - 1) - \frac{2}{\beta}\ln \xi. \tag{7}$$

It follows directly from this formula that for

$$\frac{Qa^2}{\alpha_1}|\xi - 1| > \frac{2}{\beta}|\ln \xi|$$

the inclusion temperatures begins to rise exponentially in time, which indeed corresponds to thermal explosion. Hence it follows that for laser pulse lengths $\tau \gg \tau_x$ we have

$$Q_{\mathrm{thr}}(I) = \frac{2\alpha_1}{a^2 \beta} \frac{\ln \xi}{\xi - 1}. \tag{8}$$

To determine threshold intensities of laser emission capable of causing damage we must know the function $Q_{\mathrm{thr}}(I)$, which in general can have a nonlinear character. In the case of ordinary linear (single photon) absorption by small particles ($a \ll \lambda$) the threshold intensity I_{thr} can be written using the expression for absorption cross section[9]:

$$I_{\mathrm{thr}} = \frac{\lambda}{18\pi} \frac{(\varepsilon' + 2)^2 + \varepsilon''^2}{\varepsilon''} Q_{\mathrm{thr}}, \tag{9}$$

where $\epsilon = \epsilon' + i\epsilon''$ is the relative dielectric permittivity of the particle material at the wavelength λ. We note that this formula does not take the magnetic part of absorption into account although it can make a significant contribution in the case of metallic particles with high conductivity.

The above analysis of the damage process concerns "long" laser pulses ($\tau \gtrsim \tau_x$). For short pulses ($\tau \ll \tau_x$) in the case of uniformly absorbing particles (1) gives

$$Q_{\mathrm{thr}} = \frac{C_1 \rho_1 T_0}{\xi \tau}\left\{1 - \exp\left[-\xi\left(\frac{T_c}{T_0} - 1\right)\right]\right\}, \tag{10}$$

where T_c is the critical temperature of a particle at which damage takes place. Since the magnitude of T_c is sufficiently high ($T_c \gg T_0$), it follows from (10) that to find Q_{thr} we also need not know T_c if the nonlinearity parameter is not too small and in the case of short pulses.

We compute the threshold values of Q_{thr} and I_{thr} for ruby crystals containing metallic particles of nickel. Such particles (or particles of other metals) can be present in ruby laser crystals[4]. As an example we take a typical particle size of $2a = 3 \times 10^{-6}$ cm and laser pulse length of $\tau = 3 \times 10^{-8}$ sec. In this case $\tau \gg \tau_x$ and we use (6) or (8) to obtain Q_{thr}. For the ξ parameter we take the value $\xi \cong 0.1$ (approximately this value we obtained in direct measurements for Ni and NiO, CoO, and FeO films in the temperature range of 300—1500°K at the wavelength $\lambda = 0.63\ \mu$). Taking the known values for ϵ' and ϵ'' [10] we obtain from (6) and (9) for $Q_{thr} \cong 3.5 \times 10^{14}$ W/cm^3, $I_{thr} \cong 2 \times 10^9$ W/cm^2. Computations according to (8) and (9) for a weaker nonlinearity give $Q_{thr} \cong 3.5 \times 10^{14}$ W/cm^3, $I_{thr} \cong 7 \times 10^9$ W/cm^2. These values of I_{thr} correspond approximately to the observed values of damage threshold in sapphire specimens containing nickel impurities of the above size[4].

Computation of the threshold damage power for glass containing platinum inclusions of $2a = 10^{-5}$ cm exposed to a laser pulse $\tau = 3 \times 10^{-8}$ sec long assuming that $\xi = 0.1$, yields $I_{thr} \cong 8 \times 10^7$ W/cm^2. This value is an order lower than that computed in[3] from linear theory.

3. CONCLUSION

The theory of thermal damage in transparent dielectrics with absorbing inclusions developed here indicates that such damage has the nature of a thermal explosion. This makes it possible to compute the threshold conditions of damage for various types of transparent dielectrics (glass, crystals) without having to analyze the dynamics of thermoelastic stress. The theory shows that the damage threshold depends significantly on the impurity size, thermal and optical constants, and their temperature nonlinearity. Although in our analysis we dealt only with the special cases of temperature dependencies of thermal conductivity and absorption coefficient it is clear that a similar analysis can be made for other types of nonlinearity.

The damage theory discussed here is sufficiently general because it is valid for any mechanism of absorption of light radiation that can cause a quasi-equilibrium phonon heating (the condition for the validity of the instantaneous lattice temperature concent). Such mechanisms can be represented also by multiphoton absorption and absorption by free carriers generated during avalanche ionization of the lattice or impurity centers.

We assume that the above discussion of damage mechanisms can be used for the analysis of the experimental data for damage inside transparent dielectrics in a broad range of laser pulse lengths. In particular it can explain the large dispersion of literature data (see[11] for example) on glass and crystal damage thresholds due to the strong dependence of the threshold on the size of absorbing inclusions and their nature.

[1] E. A. Shefter, Candidate's Dissertation. Institute of Crystallography, USSR Academy of Sciences, 1968.

[2] H. S. Bennett, Proc. of a Symposium on Damage in Laser Materials. Nat. Bur. of Stand. Spec. Publ. N 341, 1970, p. 51.

[3] R. W. Hopper and B. R. Uhlmann, J. Appl. Phys. 41, 4023 (1970).

[4] E. M. Akulenok, Yu. K. Danileĭko, F. M. Zubkova, V. S. Nechitaĭlo, A. A. Manenkov, A. D. Piskun, and V. Ya. Khaimov-Mal'kov, ZhETF Pis. Red. 16, 336 (1972) [JETP Lett. 16, 238 (1972)].

[5] Yu. K. Danileĭko, A. A. Manenkov, A. M. Prokhorov, and V. Ya. Khaimov-Mal'kov, Zh. Eksp. Teor. Fiz. 58, 31 (1970) [Sov. Phys.-JETP 31, 18 (1970)].

[6] B. I. Makshantsev, R. K. Leonov, and P. A. Yampol'skiĭ, ZhETF Pis. Red. 14, 175 (1971) [JETP Lett. 14, 115 (1971)].

[7] W. Franz, Dielectric Breakdown, (Russ. Transl.) IIL, 1970.

[8] C. Werth and R. Thomson, Solid State Physics, (Russ. Transl.) Mir. 1969.

[9] L. D. Landau and E. M. Lifshitz, Elektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Moscow, 1957 [Pergamon, 1959].

[10] M. Born and E. Wolf, Principles of Optics, Pergamon, 1966.

[11] C. R. Giuliano and L. D. Hess, Proc. of a Symposium on Damage in Laser Materials. Nat. Bur. Stand. Spec. Publ. N 341, 1970, p. 76.

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