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# Theory of Strong Photothermal Nonlinearity from Sub-Surface Non-Stationary ("Breathing") Cracks in Solids

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**Abstract.** A model for the description of the strong thermal-wave nonlinearity exhibited by a non-stationary ("breathing") crack (delamination) in a solid is proposed. The description of the nonlinear thermal-wave frequency spectrum both in the regime of the laser-induced thermal tansparency and of the laser-induced thermal darkening of the crack is presented. The conditions under which the amplitude of the thermal-wave second harmonic becomes on the order of the fundamental thermal-wave amplitude are derived. It is further demonstrated that the dependence of the photothermal response on the pump-laser intensity provides mechanical information on the crack (delamination). This type of informaton is different from, and additional to, that provided by the traditional measurements of the photothermal-response dependence on laser-beam intensity modulation frequency.

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Growing interest in nonlinear photothermal phenomena has been motivated by several experimental investigations which demonstrated that the thermal-wave second-harmonic detection can provide better contrast both in photothermal microscopy [1, 2] and in photothermal depth-profilometry [3]. Experimentally observed amplitudes of the thermal-wave second harmonic were on the order of one percent of that of the fundamental frequency. This correlates well with our theoretical estimates [4, 5], which take into account the dependence of the material heat capacity $C = C(T)$ and thermal conductivity $K = K(T)$ on temperature $T$ [4, 5], and on the thermal expansion of solid layered structures [5]. The theoretical models presented to-date [4, 5] were based on the method of stepwise successive approximations, the justification of which is based on the assumption of the weak thermal nonlinearity of the system.

Nevertheless, as was first noted in [1], thermal nonlinearity may be caused not only by the nonlinearity of the physical parameters of the material (such as $C$ and $K$) but also by the presence of material defects such as sub-surface cracks and delaminations. In this case, a modulation in the "effective $K$" will be caused by the periodic thermomechanical opening and closing of a crack resulting in a periodic change in the thermal boundary conditions. This effect was first investigated in the regime of weak thermal nonlinearity in [2], where the "breathing" crack was modulated by the nonlinear thermal resistance $R = R(\phi)$, where $\phi$ is the heat flux across the resistance and $\Delta T$ is the temperature change. Under this condition, the temperature change $\Delta T$ can be written [2]

$$\Delta T = R(\phi)\phi \,. \tag{1}$$

In the regime of weak nonlinearity, $R(\phi)$ is described by a linear function of $\phi$ ($\Delta T = R_0\phi + R_1\phi^2$) [2]. Note that the application of the stepwise successive approximation method [1, 2, 4–6] leads directly to linear dependence of the amplitude of the fundamental frequency thermal wave, and to quadratic dependence of its second harmonic, on pump-laser intensity. Thus, there always exists an upper intensity limit for the validity of this approximation.

## 1 Theoretical

In the present work we have theoretically investigated, for the first time, the photothermal response of a non-stationary ("breathing") crack in the regime of strong nonlinearity. We have determined the conditions under which the amplitude of the thermal-wave second (and higher order) harmonics become comparable to that of the fundamental frequency. We have also showed that monitoring the dependence of the amplitudes of the harmonics on the laser-induced heat flux, which is proportional to pump-laser intensity, provides additional information on the crack or delamination.

We start from a simple photothermal model, i.e. we consider a coating ($0 \le z < H$) on a semi-infinite backing

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($z > H$), where $H$ is the thickness of the coating. The modulated light beam induces a heat flux $J_L$ from the irradiated surface ($z = 0$) into the sample:

$$-K\frac{\partial}{\partial z}T(z=0) = J_L\,. \tag{2}$$

Assuming that the crack or delamination is localized at the interface $z = H$ and is thermally thin, we describe it by the thermal resistance $R$ [7, 8]

$$\Delta T = T(z = H - 0) - T(z = H + 0)$$
$$= -RK\frac{\partial}{\partial z}T(z = H - 0) \equiv R\phi\,. \tag{3}$$

In this work, in order to demonstrate the major physical features of the phenomena under investigation we further assumed that the backing ($z > H$) plays the role of a heat sink which saturates the temperature

$$T(z = H + 0) \equiv 0\,. \tag{4}$$

This situation is of practical importance, as the backing is typically used in applications not only to support thin films but also for their cooling (e.g. in semiconductor lasers, with silicon-on-sapphire structures etc.). To achieve sufficient cooling it is important to have a thermally thin coating, i.e. the thickness should be less than the thermal wave penetration (thermal diffusion) length $\sqrt{D/2\omega}$, where $D = K/C$ is the thermal diffusivity of the film and $\omega$ is the characteristic angular frequency of the thermal waves.

Under the condition $H^2 \ll D/\omega$, the equation for heat conduction

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{D}\frac{\partial}{\partial t}\right)T(z,t) = 0\,, \tag{5a}$$

with the boundary conditions (2–4) reduces to the ordinary differential equation for the flux $\phi$

$$CH\frac{d}{dt}(R\phi)(t) + \phi(t) \simeq J_L(t)\,, \tag{5b}$$

while the surface temperature usually detected experimentally is determined by

$$T(z = 0; t) \simeq \frac{H}{K}J_L(t) + (R\phi)(t)\,. \tag{6}$$

In the present report we will also assume that the internal thermal resistance of the coating to the heat flux (i.e., $H/K$) is much less than the external thermal resistance

$$H/K \ll R\,. \tag{7}$$

Therefore, we will treat our system as a lumped heat-capacity system [9]. Then, (5b) and (6) provide the following description of the time evolution of the film temperature $T \simeq T(0)$:

$$\frac{\partial}{\partial t}T(0;t) + \frac{1}{\tau_R}T(0;t) = \frac{J_L(t)}{CH}\,, \tag{8}$$

where $\tau_R \equiv CHR$ is the thermal time constant of the film, i.e. the characteristic cooling time of the thin homogeneously heated film, as a result of heat transfer through the thermal resistance.

In order to use (8) for the examination of nonlinear photothermal phenomena it is necessary to describe the dependence of the crack thermal resistance on temperature. We assumed the thermal resistance to be proportional to the thickness $h$ of the gas layer inside the crack or delamination: $R = h/K_g$ [7, 8, 10], where the value of the gas effective thermal conductivity constant $K_g$ depends on the regime of gas heat transfer (i.e. diffusional or ballistic [7, 10]). In most cases the $h$ dependence on temperature may be modelled by $h(T) = h^{(0)} + \gamma T$ (here, $h^{(0)}$ is the gas-layer thickness in the absence of laser action and $\gamma$ is a proportionality constant). For example, if the elastic interaction of the coating and the backing is negligible, then closing of the crack caused by the thermal expansion of the film should take place $\gamma \sim -\beta^* H$ ($\gamma < 0$), where $\beta^* > 0$ is the effective bulk thermal-expansion coefficient of the coating in the one-dimensional geometry [11]. If there exists perfect elastic bonding between the coating and the backing around the delamination not far from the tested area, then additional opening of the crack caused by the predominance of the thermoelastic bending of the thin film may be expected [12–14]:

$$\gamma \sim \beta^* r^2/H \qquad (\gamma > 0)\,, \tag{9}$$

where $r$ is the characteristic linear dimension of the delamination. These possibilities define two different types of non-stationary, or "breathing" cracks.

We can model both situations introducing the constant $\alpha \equiv \gamma/h^{(0)}$ in the description of the temperature dependence of the characteristic time $\tau_R$:

$$\tau_R = \tau_R^{(0)}(1 + \alpha T)\,. \tag{10}$$

Here, $\tau_R^{(0)} \equiv CHR_0 \equiv CHh^{(0)}/K_g$ is the coating time constant in the absence of heating. The description of (8) and (10) encompasses both the regime of laser-induced thermal transparency (i.e. when $\alpha < 0$ and the thermal resistance of the crack decreases with increasing laser intensity) and the regime of laser-induced thermal darkening (i.e. when $\alpha > 0$ and the thermal resistance of the crack rises with rising laser intensity).

For harmonic modulation of the laser-initiated heat flux

$$J_L(t) = J_0(1 + \cos\omega t)\,. \tag{11}$$

The solution of the problem of (8, 10) by the method of stepwise successive approximations [4, 5, 10] and with the additional separation of the average $T_0$ and oscillating $T_{osc}$ components can be presented in the form:

$$T = T_0 + T_{osc} = T_0 + \sum_{n=1}^{\infty} T_{n\omega}\,, \tag{12}$$

where

$$T_0 \simeq \frac{1}{|\alpha|}\left(\frac{J_0}{J_{cr}}\right), \tag{13}$$

$$T_\omega \simeq \frac{1}{|\alpha|}\left(\frac{J_0}{J_{cr}}\right)\frac{1}{\sqrt{1 + (\omega\tau_R^{(0)})^2}}\cos[\omega t - \tan^{-1}(\omega\tau_R^{(0)})]\,, \tag{14}$$

and

$$T_{2\omega} \simeq \frac{1}{2|\alpha|}\left(\frac{J_0}{J_{cr}}\right)^2 \frac{1}{[1+(\omega\tau_R^{(0)})^2]\sqrt{1+(2\omega\tau_R^{(0)})^2}} \times \cos[2\omega t - 2\tan^{-1}(\omega\tau_R^{(0)}) - \tan^{-1}(2\omega\tau_R^{(0)}) - (1-\operatorname{sgn}\alpha)\pi/2] \,. \tag{15}$$

Here $J_{cr} \equiv 1/|\alpha| R_0$ is the characteristic critical magnitude of the thermal flux. The solution (14) is valid for $J_0 \ll J_{cr}$ at all frequencies. In estimating the nonlinear photothermal effects, a useful parameter is the ratio of the second-harmonic amplitude $A_{2\omega}$ to the square of the fundamental wave amplitude $A_\omega$, because the latter does not depend on laser intensity in the regime of weak pumping $J_0 \ll J_{cr}$ [4]. For the system under consideration we obtain from (14) for $\omega\tau_R^{(0)} \ll 1$:

$$N \equiv A_{2\omega}/(A_\omega)^2 \sim |\alpha| \,. \tag{16}$$

In the case of the laser-induced thermal transparency, (16) leads to $N \sim \beta^*(H/h^{(0)})$. This nonlinearity is significantly stronger than the one associated with the thermal expansion of the system composed of solid layers ($N \sim \beta^*$) [5]. In fact, even under the restriction of lumped heat capacity [condition (7)], i.e. $H/h^{(0)} \ll K/K_g$, one can choose $H/h^{(0)} \sim 10^3$ (for example, for the metal–gas combination $K/K_g \gtrsim 10^4$ is typical). The explanation of this strong nonlinearity is rather self-evident: the thermal expansion of the solid layer predominantly modulates not its own thermal resistance, but the thermal resistance of the trapped gas layer, and the latter process is $\sim (K/K_g)$ times more efficient.

The solution (14) further demonstrates that there exists the parameter $\omega\tau_R^{(0)}$ which, in addition to the parameter $(J_0/J_{cr})$, may force the higher-harmonic amplitudes to decrease with increasing order. This fact allows us to apply the stepwise successive approximation method at high frequencies, too ($\omega\tau_R \gg 1$), assuming that the major thermal resistance changes are controlled by the average temperature field, i.e.

$$1 + \alpha T_0 \gg |\alpha T_{osc}| \,. \tag{17}$$

Using the ratio $|\alpha T_{osc}|/(1+\alpha T_0) \ll 1$ as a small parameter we obtained the following solution of (8) subject to (10):

$$T_0 \simeq \frac{1}{|\alpha|}\frac{(J_0/J_{cr})}{[1-\operatorname{sgn}\alpha(J_0/J_{cr})]} \,, \tag{18a}$$

$$T_\omega \simeq \frac{1}{|\alpha|}\left(\frac{J_0}{J_{cr}}\right)\frac{1}{(\omega\tau_R^{(0)})}\cos(\omega t - \pi/2) \,, \tag{18b}$$

$$T_{2\omega} \simeq \frac{1}{4|\alpha|}\left(\frac{J_0}{J_{cr}}\right)^2 \frac{[1-\operatorname{sgn}\alpha(J_0/J_{cr})]^3}{(\omega\tau_R^{(0)})^3} \times \cos[2\omega t - 3\pi/2 - (1-\operatorname{sgn}\alpha)\pi/2] \,. \tag{18c}$$

Given that in the regime of the laser-induced thermal darkening, $\tau_R$ is an increasing function of the thermal flux, the conditions for the validity of (18b) for $\alpha > 0$ are strengthened with increasing pumping:

$$\omega\tau_R^{(0)} \gg 1 - (J_0/J_{cr}) \,; \qquad 0 \le J_0 \le J_{cr} \,. \tag{19}$$

The solution (18a) for the average temperture $T_0$ in the case of thermal darkening becomes unbounded when $J_0$ approaches the critical value. It is apparent that one should take into account other nonlinearities (e.g. the one associated with radiation heat transfer) to avoid this singularity. Furthermore, it is important to remember that the gas layer itself becomes thermally thick for sufficiently large $J_0$ and thus cannot be described by the concept of thermal resistance.

Equation (18b) describes the monotonic increase of the fundamental frequency component $T_\omega$, and (18c) shows the non-monotonic behavior of the second-harmonic amplitude $T_{2\omega}$ with increasing laser-induced thermal flux. The maximum of the second-harmonic amplitude is achieved when $J_0 \simeq (2/5)J_{cr}$. The explanation of the second-harmonic amplitude decrease for fluxes higher than $(2/5)J_{cr}$ can be sought in the relaxational nature of the nonlinearity in the system under consideration: One can readily see from (8) that the nonlinearity is suppressed with increasing $\tau_R$ and this is exactly what takes place with increasing laser pump power in systems with induced thermal darkening. This explanation correlates well with the monotonic decrease of the nonlinear parameter $N$ introduced in (16)

$$N \equiv A_{2\omega}/(A_\omega)^2 \sim |\alpha|(1 - J_0/J_{cr})^3 \,. \tag{19}$$

If one is interested in the relative magnitudes of the fundamental and the second-harmonic amplitudes in (18b, c) then this ratio exhibits a maximum for $J_0 \simeq (1/4)J_{cr}$.

The solution set of (18) shows that in the case of laser-induced thermal transparency ($\alpha < 0$) the average temperature $T_0$ saturates for large pump-induced heat fluxes:

$$T_0 \to 1/|\alpha| \,, \qquad \text{when } J_0 \gg J_{cr} \,,$$

while the thermal resistance approaches the zero value (i.e. a completely closed crack) only asymptotically:

$$R \sim R_0/(J_0/J_{cr}) \text{ when } J \gg J_{cr} \,.$$

Since in this regime the characteristic time $\tau_R$ increases with increasing pump power, the condition for the validity of (18) becomes more stringent under high pumping:

$$\omega\tau_R^{(0)} \gg \max[1, J_0/J_{cr}] \,.$$

The nonlinear parameter $N$ in this regime grows monotonically with increasing laser-induced heat flux in correlation with decreasing $\tau_R$. It should be remembered that the magnitude of the thermal resistance is bounded in the model leading to (8) from below by virtue of the condition (7).

An exact analytical description of the entire thermal-wave spectrum can be formulated in the quasi-stationary regime, i.e. at low frequencies $\omega\tau_R \ll 1$:

$$T = \frac{1}{|\alpha|}\left\{\frac{J_N}{\sqrt{1-J_N\operatorname{sgn}\alpha}(\sqrt{1-J_N\operatorname{sgn}\alpha}+1)} + \sum_{n=1}^{\infty}\frac{2(J_N)^n}{\sqrt{1-J_N\operatorname{sgn}\alpha}(\sqrt{1-J_N\operatorname{sgn}\alpha}+1)^{2n}} \times \cos[n\omega_0 t - (1-\operatorname{sgn}\alpha)(\pi/2)(n-1)]\right\} \,. \tag{20}$$

Here $J_N$ is the normalized laser-induced thermal flux $J_N \equiv 2J_0/J_{cr}$. By comparing (18) and (20), one can readily see that in this low-frequency regime the critical value of the flux is twice as low as in the high-frequency limit. This is caused by the fact that in the quasi-stationary solution (20) of (8) and (10) we were able to model the contribution of the oscillating flux components to the average temperature field, as a result of the oscillating nonlinearity introduced by the breathing crack itself.

In the limit of weak pumping (i.e. $J_N \ll 1$) the solution (20) reduces to

$$T \simeq \frac{1}{|\alpha|}\left\{\frac{J_N}{2} + \sum_{n=1}^{\infty} \frac{(J_N)^n}{2^{2n-1}} \times \cos\left[n\omega_0 t - (1 - \operatorname{sgn}\alpha)\frac{\pi}{2}(n-1)\right]\right\}. \quad (21)$$

This expression is related to (14) but, in addition, it very clearly and directly demonstrates the broadening of the thermal wave spectrum as a result of the multiple reflections of the thermal wave from the breathing crack. Note that the amplitude of the $n$-th harmonic grows proportional to $(J_N)^n$.

In systems exhibiting laser-induced thermal darkening ($\alpha > 0$) the solution (20), in the high pumping limit, i.e. for $J_N \to 1$, transforms to:

$$T \simeq \frac{1}{|\alpha|}\left(\frac{1}{2\sqrt{1-J_N}} + \sum_{n=1}^{\infty} \frac{1}{2^{2n-1}\sqrt{1-J_N}} \cos n\omega t\right). \quad (22)$$

This expression describes a divergence in the temperature field components when $J_0$ approaches the critical value $(J_{cr}/2)$. In agreement with (20) the nonlinear parameter decreases with increasing thermal flux:

$$N \sim |\alpha|\sqrt{1-J_N}\,. \quad (23)$$

It also follows from (20) that the ratio of the amplitudes of the second and the first harmonic increases roughly proportional to $J_N$. The solution (20) is valid in a system exhibiting laser-induced darkening for $\omega\tau_R^{(0)} \ll \sqrt{1-J_N}$.

In a system with laser-induced thermal transparency ($\alpha < 0$) the solution (20), in the high pumping limit ($J_N \gg 1$) transforms to:

$$T \simeq \frac{1}{|\alpha|}\left\{1 + \sum_{n=1}^{\infty} \frac{2}{\sqrt{J_N}} \cos\left[n\omega_0 t - \frac{\pi}{2}(n-1)\right]\right\}. \quad (24)$$

This equation describes the saturation of the average temperature field, as well as the decrease of the oscillating components with increasing laser intensity:

$$T_n \sim 1/\sqrt{J_N}\,.$$

The dependences of the normalized partial thermal wave component amplitudes

$$\theta_m = |\alpha| A_m\,; \qquad m = 0, 1, 2, 3 \quad (25)$$

on the normalized heat flux $J_N$ plotted in accordance with the solution (20) are presented in Fig. 1. One can see that the amplitudes of the first and the second harmonic, curves 2 and 3, respectively, become of the same order of magnitude already for $J_N \sim 1$, i.e. for $J_0 \sim J_{cr}$. The nonlinear parameter $N$ is a growing function of $J_N$:

$$N \sim |\alpha|\sqrt{1+J_N}\,. \quad (26)$$

Considering the induced thermal transparency one should take into account the fact that $\tau_R$ decreases with increasing pump power in such systems. Thus, the condition for the validity of the quasi-stationary approximation is strengthened with increasing thermal flux:

$$\omega\tau_R^{(0)} \ll \sqrt{1+J_N}\,.$$

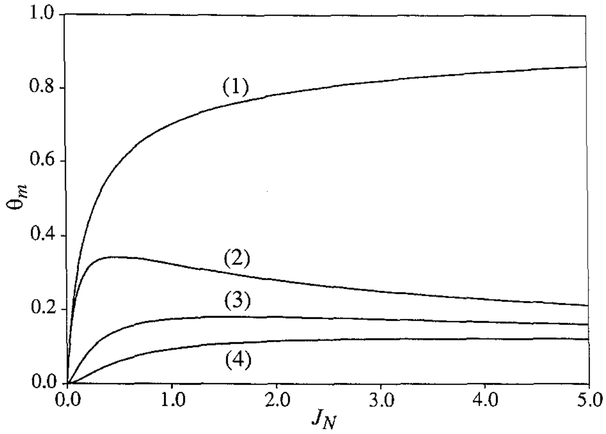


**Fig. 1.** The dependence of the normalized amplitude $\theta_m$ of the average temperature field and the first, second and third harmonics, curves 1–4, respectively, on the normalized laser-induced heat flux $J_N$ in the quasi-stationary regime of thermal transparency [(20), $\alpha < 0$]

## 2 Discussion and Conclusions

In this work, we presented a theoretical model for the description of the laser-induced thermal transparency and thermal darkening of a sub-surface non-stationary crack, or delamination, thermally close to the irradiated surface. The derived asymptotic solutions of the nonlinear thermal-wave problem describe both the regimes of low- and high-laser modulation frequencies, as well as the cases of weak and strong laser-induced thermal fluxes. The presented theory predicts significant changes of the amplitude and phase of the fundamental frequency and of the second-harmonic amplitude and phase with increasing laser pumping. In the weak pumping limit ($J_0 \ll J_{cr}$) the description of both systems (i.e. those with $\alpha < 0$ and $\alpha > 0$) is the same and is given by (14). However, in the strong pumping limit a system with induced thermal transparency can be described in the low-frequency regime by (20) for $J \gg (J_{cr}/2)\,[1 + (\omega\tau_R^{(0)})^2]$. A system with induced thermal darkening can be described in the high-frequency regime by (18) for $(1 - J/J_{cr}) \ll \omega\tau_R^{(0)}$.

It is noteworthy that both amplitude and phase dependences on the modulation frequency in the weak pumping regime $J \ll J_{cr}$, (14), can be used in applications for the determination of the characteristic time $\tau_R$, which contains information about both the layer thickness and the crack thermal resistance, $\tau_R = CHR$. These two parameters can be separated out by absolute temperature measurements at low and high frequencies. For example, in accordance with (14), the amplitude of the fundamental-frequency wave at low frequencies ($\omega\tau_R^{(0)} \ll 1$) depends only on the parameters of the crack:

$$A_\omega \sim J_L R\,,$$

while at high frequencies ($\omega\tau_R^{(0)} \gg 1$) it depends only on the parameters of the coating:

$$A_\omega \sim J_0/CH\omega .$$

The amplitude of the thermal-wave second harmonic in these limiting cases depends both on the parameters of the coating and on the parameters of the crack (14). This may provide better contrast in nonlinear photothermal microscopy and depth profilometry than in conventional photothermal measurements.

Furthermore, the developed theory predicts that additional information on the system under consideration may be obtained from its nonlinear behavior by observing the dependences of the harmonic amplitudes on pump-laser intensity, (18, 20). For example, one can extract the value of $J_{cr} \sim |\alpha| R$ from the saturation of the growth of the oscillating components in the case of induced thermal transparency, (20) and Fig. 1, or from the maximum of the second-harmonic amplitude in the case of the laser-induced thermal darkening (18).

Finally, we wish to point out that there are no difficulties, in principle, in taking into account in the above theory the internal thermal resistance of the overlayer, as well as the possible existence of additional thermally thin layers of other materials between the delamination and the heat-sink. This extension may be important for experimental applications.

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