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Research Article

Photoacoustic transients generated by laser irradiation of thin films

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ABSTRACT

Irradiation of an optically thin layer immersed in a transparent fluid with pulsed laser radiation can generate photoacoustic waves through two mechanisms. The first of these is the conventional optical heating of the layer followed by thermal expansion, in which the mechanical motion of the expansion launches a pair of oppositely directed sound waves. A second, recently reported mechanism, is operative when heat is conducted to the transparent medium raising its temperature, while at the same time reducing the temperature in the absorbing body. The latter mechanism has been shown to result in compressive transients at the leading edges of the photoacoustic waveforms. Here the photoacoustic effect produced by irradiating thin metal films which undergo negligible thermal expansion under optical irradiation, but which generate sound solely by the heat transfer mechanism is investigated. Solution to the wave equation for the photoacoustic effect from the heat transfer mechanism is given and compared with the results of experiments using nanosecond laser pulses to irradiate thin metal films.

The photoacoustic effect is governed a pair of coupled [1–3] differential equations for the temperature τ and the pressure p given by

$$\begin{aligned}(\nabla^2 - \frac{\gamma}{c^2}\frac{\partial^2}{\partial t^2})p &= -\rho\beta\frac{\partial^2}{\partial t^2}\tau \\ \frac{\partial}{\partial t}(\tau - \frac{\gamma - 1}{\gamma\alpha}p) &= \frac{K}{\rho C_P}\nabla^2\tau + \frac{H}{\rho C_p},\end{aligned} \quad (1)$$

where c is the sound speed, γ is the heat capacity ratio, ρ is the density, β is the thermal expansion coefficient, C_p is the specific heat capacity, K is the thermal conductivity, and H is the energy deposited per unit volume and time by radiation source. Except for extremely small bodies, the time scale for heat diffusion is much longer than that for sound generation, hence, the properties of the photoacoustic pressure are commonly determined by assuming the thermal conductivity to be zero, in which case the coupled equations reduce [4] to a single wave equation

$$(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2})p = -\frac{\beta}{C_p}\frac{\partial H}{\partial t}, \quad (2)$$

obviating solution to the fourth order equation [1,5] that corresponds to Eqs. 1.

It has recently been shown in the case of optically thin fluid bodies immersed in transparent fluids that large amplitude compressive transients are found on the leading edges of the photoacoustic waveforms that are not accounted for by Eq. 2. Despite the fact that production of the photoacoustic effect with pulsed lasers has been intensely investigated [6–8] since the 1970′*s*, the existence of the compressive transients has only been reported recently. It is likely that such transients had not been previously reported owing to the fact that slight misalignment of a plane transducer with respect to a plane absorbing object results in integration of the transient in time, reducing its amplitude so it is not evident from examination of the photoacoustic waveform. In fact, it was shown [9–11] that misalignment of a plane polyvinylidene fluoride (PVDF) transducer by as little as one degree resulted in the complete disappearance of the transient on the wave recorded from a weakly absorbing glass flat. As the disappearance of the transient is caused by its integration over the plane surface of the transducer, recording the wave with a small diameter transducer can be expected to alleviate this difficulty. With spherically or cylindrically symmetric objects, observation of the transients would require the geometry of the transducer to be matched to that of the irradiated object.

In so far as determining the origin of the transients, it was shown in Refs. [12,11,10] that their presence could be accounted for by taking into account heat conduction in the region of the irradiated object nearest the interface between the absorbing object and the transparent fluid. By keeping the heat conduction term in Eqs. 1 and approximating the heat capacity ratio as unity, it was shown that the coupled equations reduce to the wave equation

$$(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2})p = -\rho\beta\chi\frac{\partial}{\partial t}\nabla^2\tau - \frac{\beta}{C_p}\frac{\partial H}{\partial t}, \quad (3)$$

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which contains two source terms, the one found in Eq. 2 and a new term dependent on both the space and time derivatives of the temperature as well as the thermal diffusivity χ. The work reported in Refs. [12,11,10] was restricted to determining the character of the transients when, following short pulse irradiation of an optically thin target, the temperature at the surface of the irradiated object decreased, while that of the liquid in contact with its surface increased. Here we investigate the solution of Eq. 3 for the case of delta function deposition of heat in space and time where only a temperature increase in the fluid surrounding target is considered.

The deposition of heat as a delta function in space can be described by considering a layer whose absorption coefficient tends to infinity while its thickness approaches zero, with the product of the two remaining finite. This product gives a dimensionless quantity denoted $\hat{\alpha}$. The heating function for a laser pulse with a fluence E_0 irradiating a delta function layer can be written

$$H(x,t) = \hat{\alpha} E_0 \delta(x)\delta(t). \tag{4}$$

The heat diffusion equation, which determines τ in Eq. 3, is given by

$$\frac{\partial}{\partial t}\tau = \chi \nabla^2 \tau + \frac{H}{\rho C_p}, \tag{5}$$

which, for the heating function given by Eq. 4, gives the well-known solution [13]

$$\tau(x,t) = \frac{\hat{\alpha} E_0}{2\rho C_p \sqrt{\pi \chi t}} e^{-x^2/4\chi t}. \tag{6}$$

In determining the photoacoustic effect from short pulse excitation, it is convenient to work with the velocity potential φ, which is governed by the wave equation

$$(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2})\varphi = \frac{\beta}{\rho C_P} H + \beta \chi \nabla^2 \tau, \tag{7}$$

where the acoustic pressure [14] is determined from φ through

$$p = -\rho \frac{\partial \varphi}{\partial t}. \tag{8}$$

The solution for the conventional photoacoustic effect, which arises from the first term on the right of Eq. 7, can be found by integrating this term over the Green's function for the one-dimensional wave equation [15], giving the velocity potential as

$$\varphi = -\frac{\hat{\alpha}\beta E_0 c}{2\rho C_P}\int dx' \int dt' \delta(x')\delta(t') \times \tag{9}$$

$$[1 - \mathrm{u}(\frac{x - x'}{c} - (t - t'))], \tag{10}$$

where the factor in brackets containing the Heaviside function u is the Green's function (divided by $2\pi c$) for the one-dimensional wave equation. By differentiating φ with respect t, the factor in brackets becomes $\delta[t' - t - (x - x')/c]$. The integration in Eq. 9 can then be carried out immediately to give

$$p = \frac{\hat{\alpha}\beta E_0 c}{2 C_P}\delta(t - x/c), \tag{11}$$

which describes the right-going photoacoustic wave.

The contribution of the second source term on the right hand side of Eq. 7 can be found by Laplace transformation. The Laplace transform of the time variable in t given in Eq. 6 can be found in mathematical tables [16]. The wave equation for the velocity potential in Laplace space φ is thus given by the Helmholtz equation

$$(\frac{\partial^2}{\partial x^2} - k^2)\varphi = \frac{\hat{\alpha}\beta\chi E_0}{\rho C_p}\frac{\partial^2}{\partial x^2} e^{-\sqrt{sx^2/\chi}}, \tag{12}$$

where $k = s/c$, and s is the Laplace variable. The velocity potentials φ^R and φ^L, which denote potentials to the right and left of the origin, are both governed by Eq. 12 and must obey the acoustic boundary conditions

$$\begin{aligned} \varphi^R|_{x=0} &= \varphi^L|_{x=0} \\ \frac{\partial}{\partial x}\varphi^R|_{x=0} &= \frac{\partial}{\partial x}\varphi^L|_{x=0}. \end{aligned} \tag{13}$$

To simplify solution of the wave equation further, it is convenient to introduce two potentials $\tilde{\Phi}^R$ and $\tilde{\Phi}^L$ defined through $\partial^2\Phi/\partial x^2 = \varphi$, which, on substituion into Eq. 12, gives the Helmholtz equation for both potentials as

$$(\nabla^2 - k^2)\tilde{\Phi} = \frac{\hat{\alpha}\beta\chi E_0}{\rho C_p} e^{-\sqrt{x^2 s/\chi}}. \tag{14}$$

Solutions for the two potentials are found to be

$$\begin{aligned} \tilde{\Phi}^R &= \frac{\hat{\alpha}\beta E_0 c^2 \chi^{3/2} e^{-x\sqrt{s/\chi}}}{2 s^{3/2}(c^2 - s\chi)\rho C_p} + C_R e^{-sx/c} \\ \tilde{\Phi}^L &= \frac{\hat{\alpha}\beta E_0 c^2 \chi^{3/2} e^{x\sqrt{s/\chi}}}{2 s^{3/2}(c^2 - s\chi)\rho C_p} + C_L e^{sx/c}, \end{aligned} \tag{15}$$

where C_R and C_L are constants. The boundary conditions for Φ can be found from Eqs. 13 as

$$\begin{aligned} \frac{\partial^2}{\partial x^2}\tilde{\Phi}^R|_{x=0} &= \frac{\partial^2}{\partial x^2}\tilde{\Phi}^L|_{x=0} \\ \frac{\partial^3}{\partial x^3}\tilde{\Phi}^R|_{x=0} &= \frac{\partial^3}{\partial x^3}\tilde{\Phi}^L|_{x=0}. \end{aligned} \tag{16}$$

Since only the terms in Eqs. 15 containing exponential factors of the form $\exp(\pm sx/c)$ result in travelling waves when transformed back to the time domain, the other terms in Eq. 15 are not carried forward, as they correspond to thermal mode [1] waves that do not propagate. After applying the boundary conditions to determine the constants in Eqs. 15, the velocity potential for the right-going acoustic wave is found to be

$$\begin{aligned} \tilde{\varphi}^R &= -\frac{\hat{\alpha}\beta E_0 c}{2 s\rho C_p} e^{-sx/c} - \frac{\hat{\alpha}\beta E_0 \chi}{2 c\rho C_p} e^{-sx/c} \\ &+ \frac{\hat{\alpha}\beta E_0 \chi^3 s^2}{2 c^3 \rho C_p (s\chi - c^2)} e^{-sx/c}. \end{aligned} \tag{17}$$

The time domain velocity potential is found using the following two inverse Laplace transforms [16] calculated with $x > 0$,

$$\begin{aligned} \mathcal{L}^{-1}[-\frac{\hat{\alpha}\beta E_0 c}{2 s\rho C_p} e^{-sx/c}] &= -\frac{\hat{\alpha}\beta E_0 c}{2\rho C_p}\mathrm{u}(t - x/c) \\ \mathcal{L}^{-1}[-\frac{\hat{\alpha}\beta E_0 \chi}{2 c\rho C_p} e^{-sx/c}] &= -\frac{\hat{\alpha}\beta E_0 \chi}{2 c\rho C_p}\delta(t - x/c). \end{aligned}$$

The pole in complex integration [17] used to determine the inverse Laplace transform of the last term in Eq. 17 lies in the right hand complex plane; hence the value of the integral is taken to be zero. The velocity potential φ^R is thus found to be

$$\varphi^R = \frac{\hat{\alpha}\beta E_0 \sqrt{\chi}}{2\rho C_P}[\sqrt{\frac{c^2}{\chi}}\mathrm{u}(t - x/c) + \sqrt{\frac{\chi}{c^2}}\delta(t - x/c)]. \tag{18}$$