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THREE-DIMENSIONAL PHOTOACOUSTIC EFFECT UNDER AN OFF-CENTERED EXCITATION

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A closed form for the three-dimensional photoacoustic effect is obtained as a function of the focus position of the excitation beam. The theory is capable of accounting for the variations of the magnitude and phase of photoacoustic signals with the location of excitation focus.

ТРЕХМЕРНЫЙ ФОТОАКУСТИЧЕСКИЙ ЭФФЕКТ ПОД ДЕЙСТВИЕМ НЕЦЕНТРАЛЬНОГО ВОЗБУЖДЕНИЯ

В работе получено замкнутое выражение для трехмерного фотоакустического эффекта в виде зависимости от расположения фокуса возбуждающего пучка. Теория позволяет объяснить изменения амплитуды и фазы фотоакустических сигналов на основе месторасположения возбуждающего фокуса.

I. INTRODUCTION

Since the inadequacy of one-dimensional theories of the photoacoustic effect in explaining experimental results was first pointed out [1], several experimental and theoretical studies [2—5] which emphasize three-dimensional aspects have been undertaken. The three-dimensional theoretical results obtained previously by the present author are all predicted on the assumption that the heat source employed in generating the photoacoustic effect is focused at the centre of the solid sample. This assumption is made, of course, primarily because it corresponds to many realistic experimental situations but apart from that because the mathematical simplicity which ensues as a result of the guaranteed cylindrical symmetry may be advantageously exploited. However, a variation of such experimental setups is sometimes encountered in that the source is off-centred. For example, in the experiment [1] with the sample excited by a narrowly focused laser beam, the beam spot is actually scanned across the diameter of the sample. In situations such as this, even though the heat source itself is centro-symmetric with respect to the beam spot, the above-mentioned cylindrical symmetry in the overal problem is no longer preserved. The purpose of this note is to generalize the previous theory [3] so as to cover this case. In Sec. II a theory of the three-dimensional photoacoustic effect under an off-centred excitation is presented. In Sec. III the theory is applied to a Corning Glass sample in air, the system investigated in [1], and it will be shown that the agreement between theory and experiment is generally satisfactory.

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II. THEORY

In this section an expression for the photoacoustic signal under an off-centred excitation will be derived. The case under consideration is illustrated in Fig. 1: (a)

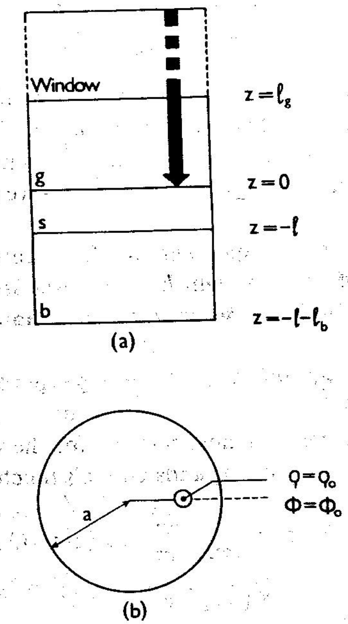

Fig. 1. (a) the cross section of a photoacoustic cell and (b) the top view of the solid sample showing a laser beam focused off-centre at $\varrho=\varrho_0$ and $\Phi=\Phi_0$ in the cylindrical coordinates.

A cylindrical solid sample (s) is mounted on a backing substrate (b) and is contained in a photoacoustic cell filled with gas (g); the sample is excited by an incident beam focused at $\varrho=\varrho_0$ and $\Phi=\Phi_0$ in the cylindrical coordinates with the geometrical centre taken as the origin. Owing to the lack of cylindrical symmetry, the beam profile is not only a function of the variables ϱ and Φ, but it also depends on where the beam is focused and, therefore, it may be labelled as $I(\varrho, \Phi; \varrho_0, \Phi_0)$.

Photoacoustic signals are detected as a sound wave generated as a result of the heat transport in the gas. There are two mechanisms via which the latter is accomplished: heat diffusion and heat wave propagation. Earlier studies [3, 5] indicate that heat diffusion is the more significant of the two for many cases. Therefore, only heat transport via diffusion will be taken into account in what follows. The starting point for a theoretical derivation of the photoacoustic signals is the determination of the departures τ_g, τ_s and τ_b from the ambient values of temperatures in gas, solid and backing regions, respectively, which are governed by the following thermal conduction equations written in ac form:

$$(\nabla^2 - j\omega/\beta_g)\tau_g = 0 \tag{1a}$$

$$(\nabla^2 - j\omega/\beta_s)\tau_s = I(\varrho, \Phi; \varrho_0, \Phi_0)\, e^{-\alpha|z|} \tag{1b}$$

$$(\nabla^2 - j\omega/\beta_b)\tau_b = 0 \tag{1c}$$

where ω is the (angular) modulation frequency of the heat source and β_i $(i = s, g, s)$ are the thermal diffusivities of the media. These equations are subject to the boundary conditions demanded by the vanishing of τ_g, τ_s and τ_b at the cylindrical wall and by the continuity in the temperature and heat flux at each of the two interfaces.

To solve Eqs. (1) it is expedient to first determine Green's function $G(\varrho, \Phi, z|\varrho', \Phi', z')$ of Eq. (1b), which is the response function to a unit source located at (ϱ', Φ', z') and is the solution to the following equation:

$$(\nabla^2 - j\omega/\beta_s)G(\varrho, \Phi, z|\varrho', \Phi', z') = \frac{1}{\varrho}\delta(\varrho - \varrho')\delta(\Phi - \Phi')\delta(z - z'). \tag{2}$$

Taking advantage of the completeness relation for the exponential functions and that for Bessel's functions [6], one expands Green's function into a double series

$$G(\varrho, \Phi, z|\varrho', \Phi', z') = \sum_{\nu=-\infty}^{+\infty}\sum_{n=-\infty}^{+\infty} Z_{\nu n}(z|z')\,Q_{\nu n}J_\nu(\alpha_{\nu n}\varrho/a)J_\nu \times (\alpha_{\nu n}\varrho'/a)\, e^{i\nu(\Phi-\Phi')} \tag{3}$$

where $\alpha_{\nu n}$ is the nth zero of the νth-order Bessel function $J_\nu(x)$ and $Q_{\nu n}$ is a normalization constant given by $Q_{\nu n} = \left\{\frac{1}{2}a^2 J_{\nu+1}(\alpha_{\nu n})\right\}^{-1}$. Upon substituting (3) into Eq. (2), one immediately obtains the equation for the expansion parameter $Z_{\nu n}(z|z')$, namely,

$$\left\{\frac{\partial^2}{\partial z^2} - \left[\left(\frac{\alpha_{\nu n}}{a}\right)^2 + j\left(\frac{\omega}{\beta_s}\right)\right]\right\} Z_{\nu n}(z|z') = \frac{1}{2\pi}\delta(z - z'). \tag{4}$$

Eq. (4) can be solved by finding $Z_{\nu n}(z|z')$ in regions $z > z'$ and $z < z'$ and matching the discontinuity in $\partial Z_{\nu n}/\partial z$ across $z = z'$, yielding $Z_{\nu n}(z|z') = \{4\pi\sigma_s(\nu n)\}^{-1} \exp\{-\sigma_s(\nu n)|z - z'|\}$, with the notation that $\sigma_i(\nu n) = \{(\alpha_{\nu n}/a)^2 + j\omega/\beta_i\}^{1/2}$, $(i = g, s, b)$. The insertion of $Z_{\nu n}$ into Eq. (3) leads to Green's function for Eq. (1b), which on account of the property that $J_{-\nu}(x) = (-1)^\nu J_\nu(x)$, may be simplified as

$$G(\varrho, \Phi, z|\varrho', \Phi', z') = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} \varepsilon_m Q_{mn} J_m(\alpha_{mn}\varrho/a) \times J_m(\alpha_{mn}\varrho'/a)\cos[\nu(\Phi - \Phi')][4\pi\sigma_s(mn)]^{-1}\exp[-\sigma_s(mn)|z - z'|] \tag{5}$$

in which $\varepsilon_m = 1$ if $m = 0$ and $\varepsilon_m = 2$ if $m \geq 1$. With the help of Green's function a particular solution τ_{sp} to Eq. (1b) can be readily constructed:

$$\tau_{sp}(\varrho, \Phi, z; \varrho_0) = \sum_m\sum_n \frac{\varepsilon_m Q_{mn} J_m(\alpha_{mn}\varrho/a)\cos(m\Phi)I_{mn}(\varrho_0)}{4\pi\sigma_s(mn)} \times \int_{-1}^{0} dz'\, e^{-\sigma_s(mn)|z-z'|-\alpha|z'|} \tag{6}$$

where $I_{mn}(\varrho_0)$ is the Fourier-Bessel transform of the beam profile:

$$I_{mn}(\varrho_0) = \frac{1}{2\pi}\int_0^a \varrho\, d\varrho \int_0^{2\pi} d\Phi J_m(\alpha_{mn}\varrho/a)\cos(m\Phi)I(\varrho, \Phi; \varrho_0, 0). \tag{7}$$

In obtaining (6) we have taken $\Phi_0 = 0$ without any loss of generality, and have insisted that the solution be even with respect to the azimuthal angle (since the heat source is itself centro-symmetric with respect to the focus). Similar considerations can be extended in determining the homogeneous solution of Eqs. (1). One finds then that the temperatures in the gas, solid, and backing material regions can be written as

$$\tau_g(\varrho, \Phi, z; \varrho_0) = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} G(mn) J_m(\alpha_{mn}\varrho/a)\cos(m\Phi)\, e^{-\sigma_g(mn)z} \tag{8a}$$

$$\tau_s(\varrho, \Phi, z; \varrho_0) = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} J_m(\alpha_{mn}\varrho/a)\cos(m\Phi)\Big\{S_1(mn)\, e^{\sigma_s(mn)z} + S_2(mn)\, e^{-\sigma_s(mn)z} + \frac{\varepsilon_m Q_{mn} I_{mn}(\varrho_0)}{2\sigma_s(mn)} \times \int_{-l}^{0} dz'\, e^{-\sigma_s(mn)|z-z'|-\alpha|z'|}\Big\} \tag{8b}$$

$$\tau_b(\varrho, \Phi, z; \varrho_0) = \sum_{m=0}^{\infty}\sum_{n=1}^{\infty} B(mn) J_m(\alpha_{mn}\varrho/a)\cos(m\Phi)\, e^{\sigma_b(mn)(z+l)} \tag{8c}$$

where it has been assumed that the length l_g of the gas column is sufficiently long so that the temperature reflection from the side opposite the sample is negligible. It remains to determine the expansion parameters $G(mn)$, $S_1(mn)$, $S_2(mn)$, and $B(mn)$ by matching the boundary conditions that signify the continuity in the temperature and heat flux at each of the gass-sample and sample-backing material intarfaces:

$$\tau_g(\varrho, \Phi, 0; \varrho_0) = \tau_s(\varrho, \Phi, 0; \varrho_0) \tag{9a}$$

$$\tau_s(\varrho, \Phi, -l; \varrho_0) = \tau_b(\varrho, \Phi, -l; \varrho_0) \tag{9b}$$

$$\kappa_g \frac{\partial \tau_g}{\partial z}(\varrho, \Phi, 0; \varrho_0) = \kappa_s \frac{\partial \tau_s}{\partial z}(\varrho, \Phi, 0; \varrho_0) \tag{9c}$$

$$\kappa_s \frac{\partial \tau_s}{\partial z}(\varrho, \Phi, -l; \varrho_0) = \kappa_b \frac{\partial \tau_b}{\partial z}(\varrho, \Phi, -l; \varrho_0) \tag{9d}$$

in which κ_i is the thermal conductivity of the medium i ($i = g, s, b$). In the subsequent development only the temperature of the gas is of concern and therefore only the expansion coefficient $G(mn)$ will be exhibited. It is given by, from solving Eqs. (9),

$$G(mn) = \varepsilon_m Q_{mn} I_{mn}(\varrho_0) \times \tag{10}$$

$$\times \left\{ \frac{[1 + b(mn)]\,a_+(mn) + [1 - b(mn)]\,a_-(mn)}{[1 + b(mn)][1 + g(mn)]\,e^{\sigma_s(mn)l} + [1 - b(mn)][1 - g(mn)\,e^{-\sigma_s(mn)l}} \right\}.$$

Here

$$b(mn) = \kappa_b \sigma_b(mn)/\kappa_s \sigma_s(mn), \quad g(mn) = \kappa_g \sigma_g(mn)/\kappa_s \sigma_s(mn),$$

and

$$a_+(mn) = \{\exp[\pm\sigma_s(mn)] - \exp[-\alpha l]\}/\sigma_s(mn)[\alpha \pm \sigma_s(mn)].$$

It is necessary next to relate the temperature variation τ_g of the gas to the pressure variation which leads to the photoacoustic signal. To this end, one makes use of the thermal-piston model [7, 3]. In this model a column of gas (whose length is l_p = several thermal diffusion lengths) in the vicinity of the solid sample is envisaged to expand adiabatically as if providing a mechanical piston to drive the gas in the remaining volume of the photoacoustic cell. Treating the gas as ideal and noting that the volume expansion of the piston is out of phase with that of the remaining gas, one finds for the photoacoustic signal $\delta P(\varrho_0)$ when the beam is focused at $\varrho = \varrho_0$,

$$\delta P(\varrho_0) = \frac{l_p \gamma P_0\, e^{j\omega t}}{T_0 \Omega_p} \int_0^{l_p} dz \int_0^a \varrho\, d\varrho \int_0^{2\pi} d\Phi\, \tau_g(\varrho, \Phi, z; \varrho_0) \tag{11}$$

where γ is the specific heat ratio of the gas, P_0 and T_0 are the ambient pressure and temperature, respectively, and $\Omega_p = \pi a^2 l_p$ is the volume of the thermal piston. The result of (11) after the indicated integrations is

$$\delta P(\varrho_0) = \frac{4\gamma P_0\, e^{j\omega t}}{T_0 a^2 l_g} \sum_{n=1}^{\infty} \frac{I_{0n}(\varrho_0)}{\alpha_{0n} \sigma_g(0n) J_1(\alpha_{0n})} \times \tag{12}$$

$$\times \left\{ \frac{[1 + b(0n)]\,a_+(0n) + [1 - b(0n)]\,a_-(0n)}{[1 + b(0n)][1 + g(0n)]\,e^{\sigma_s(0n)l} - [1 - b(0n)][1 - g(0n)]\,e^{-\sigma_s(0n)l}} \right\}.$$

(12) is the final expression for the three-dimensional photoacoustic signal, valid for any centro-symmetric but otherwise arbitrary beam focused off-centre at $\varrho = \varrho_0$. For the special case in which the heat source is focused at the centre of the sample, (12) can easily be shown to take the form of the previously obtained result.¹)

III. APPLICATION AND CONCLUSION

In order to test the validity of the theory presented in Sec. II a detailed numerical calculation has been carried out for the system investigated in [1], namely a Corning Glass sample mounted in a photoacoustic cell filled with air. In this calculation advantage is taken of the narrowness of the excitation beam so that the beam profile function is assumed to be $I(\varrho, \Phi; \varrho_0, \Phi_0) = I_0 \varrho^{-1} \delta(\varrho - \varrho_0)\,\delta(\Phi)$ with the constant I_0 representing the integrated intensity. The Fourier-Bessel transform of the beam profile, Eq. (7), is thus simply $I_0 J_0(\alpha_{0n}\varrho/a)$. Calculations based on this latter function in Eq. (12) are performed for two chopping frequencies $f = 20$ Hz and $f = 250$ Hz, together with the following reported values of parameters: $a = 0.135$ cm, $l = 0.25$ cm, $\beta_s = 6 \times 10^{-3}$ cm²/s, $\beta_g = \beta_{b0} = 0.19$ cm²/s, $\kappa_s = 10.48 \times 10^{-3}$ J/cm s °C, $\kappa_g = \kappa_b = 23.87 \times 10^{-5}$ J/cm s °C, and $\alpha = 12$ cm⁻¹. The theoretical result of the photoacoustic signal magnitude and phase values as well as experimental measurements are plotted in Fig. 2. In conformity with [1] the results are presented as a function of the position (along a diameter) of the excitation focus, the distance x measured from the extreme of the diameter, and they are furthermore normalized to those values obtained when the beam is focused at the centre of the sample. There is a slight asymmetry in the reported measurements with respect to the central position, a fact which is understandable in view of the somewhat asymmetrical photoacoustic chamber used. Otherwise the agreement between theory and experiment is generally satisfactory.

In summary, when a photoacoustic signal is generated under an off-centred excitation, the resultant temperature variations of the sample, gas and backing

¹) Eq. (24) of Ref. [3]. (In that expression a factor of 2π has been inadvertantly left out.)

material no longer enjoy the cylindrical symmetry and a theoretical analysis of the three-dimensional photoacoustic effect such as presented in [3] must be generalized further. This note reports the result of this generalization. While the final expression for the photoacoustic signal derived herein bears much resemblance to that obtained earlier, there exists an important difference. Namely, the

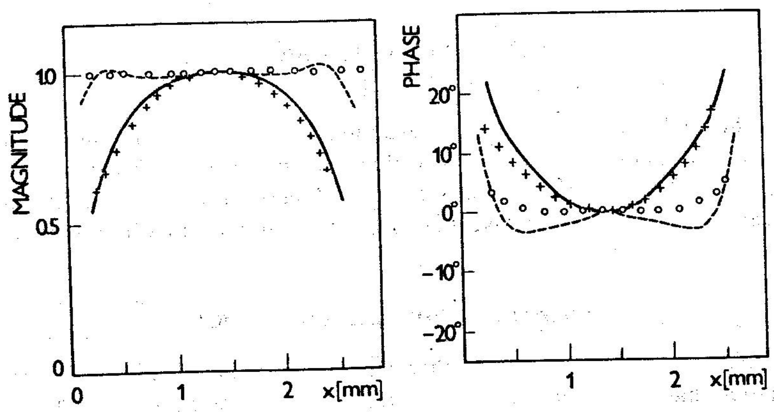

Fig. 2. Comparison between the theoretically calculated values of photoacoustic signal's amplitude and phase with measurements. Solid (dotted) lines are the theoretical results and crosses (circles) are measured values for the frequency $f = 20$ (250) Hz.

signal now explicitly depends upon where the excitation is focused. The variations of the signal's magnitude and phase with the focus position have already been observed experimentally and the present theory seems to be capable of accounting for these observations.

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