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THE PROPAGATION MODE CONTRIBUTION TO THREE-DIMENSIONAL PHOTOACOUSTIC EFFECT

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The theory of three-dimensional photoacoustic effect with a solid sample in a gas-filled chamber is extended to include the propagation mode contribution. The heat transport in a cylindrical photoacoustic cell is analysed by solving a set of linearized hydrodynamic equations and an expression for the photoacoustic signal resulting therefrom explicitly derived. The expression can be used to assess the contribution of the propagation mode as well as that of the thermal diffusion mode to the photoacoustic effect.

О РОЛИ РАСПРОСТРАНЕНИЯ МОДЫ КОЛЕБАНИЙ В ТРЕХМЕРНОМ ФОТОАКУСТИЧЕСКОМ ЭФФЕКТЕ

В работе развита теория трехмерного фотоакустического эффекта в камере, наполненной газом, с помещенным туда образцом твердого тела. Эта теория позволяет учитывать роль распространения моды колебаний в этом эффекте. Проведен анализ переноса тепла в фотоакустическом элементе на основе решения системы линеаризованных гидродинамических уравнений и приведен явный вид полученного отсюда выражения для фотоакустического сигнала. Это выражение можно использовать для оценки роли распространения моды колебаний и термодиффузных колебаний в фотоакустическом эффекте.

I. INTRODUCTION

The photoacoustic effect arises as a result of the heat absorption by a material sample illuminated by an intensity-modulated source and is detected as a sound wave in a gas-filled chamber in which the sample is situated. In general, the intensity and phase of the photoacoustic signal depend in a complicated manner on the physical and geometrical properties of the sample and other materials (the backing substrate and gas in the photoacoustic cell) in a given experimental setup. Insofar as the photoacoustic effect is a useful tool in characterizing the properties of

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the sample, and because it is simpler to do so, much greater attention has so far been directed towards correlating photoacoustic signals with the physical properties of the materials while omitting the geometrical complexities inherent in the experiments. A symptom of this situation is that most considerations in this field of study have been restricted to one-dimensional cases. (For a good review, see Ref. [1].) It is, however, not difficult to appreciate that for many realistic applications the one-dimensional consideration has at best a qualitative usefulness. For example, the incident heat source is typically a laser, which illuminates but a small portion of the sample and therefore the dissipation of heat in the sample and, subsequently, in the gas and indeed the generation of photoacoustic signals itself are essentially three-dimensional effects. That this is the case has of course been recognized in the past. Quimby and Yen [2] have observed that heat transport in the gas transverse to the direction of the incident beam gives the photoacoustic signals characteristics not accountable in a one-dimensional theory. More recently, Murphy and Aamondt [3] have observed and studied the significant signal enhancement due to the three-dimensional effect.

In a previous article [4], hereafter referred to as I, the present author has attempted to develop a fully three-dimensional theory of the photoacoustic effect. Specifically the theory is applicable to cylindrical samples and photoacoustic cells of a finite dimension and to a heat source with a centro-symmetric but otherwise arbitrary beam profile. In I the heat transport in the gas chamber is described as due to a thermal diffusion process. It is well known [5] that in addition to the thermal diffusion mode which decays over a characteristic distance (the diffusion length), a fluid can support another propagating mode of motion which can be sustained indefinitely but for the influence of viscosity and boundaries. The purpose of this note is to study the contribution of this previously neglected propagating mode to the three-dimensional photoacoustic effect. In Sec. II the problem is formulated and an expression for the photoacoustic signal is derived for the same problem as considered in I except that here the temperature variation in the gas, which ultimately produces the photoacoustic signals, is determined by solving a set of linearized hydrodynamic equations (as opposed to the thermal diffusion equation alone) leading to the concomitant presence of the thermal and propagating modes in the signals. In Sec. III a brief discussion of the results of this study is made.

II. THE PROPAGATION MODE CONTRIBUTION

In this section the contribution of the propagation mode in addition to the thermal diffusion mode to the three-dimensional photoacoustic effect is derived under the following experimental condition. An optically homogeneous cylindrical solid sample with thickness l and radius a is mounted on a backing substrate which is located in the lower portion of a cylindrical photoacoustic cell ($z=-l$ to $z=0$ in the cylindrical coordinates); the remainder of the cell is filled with a gas. An incident light chopped at a frequency $f=\omega/2\pi$ is focused on the centre of the sample and penetrates into it according to the Beer-Lambert law. In I the photoacoustic effect as a result of the thermal diffusional motion of the gas under the same condition has been studied in detail; that study contains several results which will be cited here without derivation in order to avoid repetition.

Central to the determination of the photoacoustic effect is the derivation of the temperature in the gas region, which in turn is affected by the temperatures of the sample and backing material (τ_s and τ_b, respectively). The latter may be found by solving the appropriate thermal diffusion equations:

$$\left(\nabla^2-\frac{1}{\beta_s}\frac{\partial}{\partial t}\right)\tau_s(\varrho, z, t)=I(\varrho)\,e^{-\alpha|z|}\{1+e^{j\omega t}\} \tag{1}$$

$$\left(\nabla^2-\frac{1}{\beta_b}\frac{\partial}{\partial t}\right)\tau_b(\varrho, z, t)=0 \tag{2}$$

where β_i is the thermal diffusivity of the medium i ($i=s$, b for sample and backing material, respectively), α is the reciprocal of the optical penetration length of the sample, and $I(\varrho)$ is the incident beam profile multiplied by $2\kappa_s/\alpha$, with κ_s the sample thermal conductivity.

The temperature in the gas is determined by solving the hydrodynamic equations. In such a treatment the basic variables characterizing a fluid are the density $d_0+\delta$, pressure P_0+p, temperature $T_0+\tau_g$, entropy density S_0+s, and the velocity field $\boldsymbol{u}$. Here the quantities with subscript zero denote the ambient values of the variables. At the level where viscosity may be neglected the linearized hydrodynamic equations are [5] the equation of continuity,

$$\frac{\partial\delta}{\partial t}+d_0\nabla\boldsymbol{u}=0 \tag{3}$$

the equation of motion

$$d_0\frac{\partial\boldsymbol{u}}{\partial t}=-\nabla p \tag{4}$$

and the statement of conservation of energy

$$T_0\frac{\partial s}{\partial t}=\kappa_g\nabla^2\tau_g(\varrho, z, t). \tag{5}$$

These equations are to be supplemented by two thermodynamic relations, the first of which is the equation of state

$$\delta=\left(\frac{\partial d}{\partial P}\right)_T p+\left(\frac{\partial d}{\partial T}\right)_P\tau_g=d_0\left\{\frac{p}{P_0}-\frac{\tau_g}{T_0}\right\} \tag{6}$$

while the second relates the variation of entropy density to the change in pressure and temperature

$$s=\left(\frac{\partial S}{\partial T}\right)_P\tau_g+\left(\frac{\partial S}{\partial P}\right)_T p=d_0C_P\left\{\frac{\tau_g}{T_0}-\frac{\gamma-1}{\gamma}\frac{p}{P_0}\right\} \tag{7}$$

where C_p is the isobaric heat capacity and γ is the specific heat ratio. In both Eqs. (6) and (7), the second equality follows from the assumption that the gas may be regarded as ideal. In addition one may impose the boundary conditions that the gas temperature takes on the ambient value at the cylindrical wall of the cell and that the velocity field vanishes so that there is no motion of the gas at the wall.

The temperature τ_g can be determined in a manner similar to Kirchhoff's analysis of the effect of heat exchange and viscosity on sound propagation in a narow tube [6]. Eliminating the variable entropy density s from Eqs. (5) and (7), one obtains

$$d_0C_P\left\{\frac{\partial\tau_g}{\partial t}-\frac{(\gamma-1)T_0}{\gamma P_0}\frac{\partial p}{\partial t}\right\}=\kappa_g\nabla^2\tau_g. \tag{8}$$

For the remaining variables τ_g, δ, p, and $\boldsymbol{u}$, one looks for solutions that vary temporally as the modulating incident heat source. Then their spatial dependences (for which the same symbols are used in the interest of simplifying notations) may be obtained by solving the following equations:

$$d_0\nabla\boldsymbol{u}=-j\omega\delta \tag{9}$$

$$\nabla p=-j\omega d_0\boldsymbol{u} \tag{10}$$

$$p=\frac{\gamma P_0}{(\gamma-1)T_0}\{\tau_g+j(\beta_g/\omega)\nabla^2\tau_g\} \tag{11}$$

$$\frac{\tau_g}{T_0}=\frac{p}{P_0}-\frac{\delta}{d_0} \tag{12}$$

where the gas thermal diffusivity $\beta_g=\kappa_g/C_pd_0$ is introduced. Eqs. (9—12) may be used to eliminate p, δ and $\boldsymbol{u}$ in favor of τ_g, yielding

$$j(\beta_g/\omega)\nabla^4\tau_g+\{1+j(\gamma\beta_g\omega/c_0^2)\}\nabla^2\tau_g+(\omega^2/c_0^2)\tau_g=0 \tag{13}$$

where $c_0=(\gamma P_0/d_0)^{1/2}$ is the speed of sound in the medium. The solution to Eq. (13) is given by

$$\tau_g(\varrho, z)=\tau_1(\varrho, z)+\tau_2(\varrho, z). \tag{14}$$

Here τ_1 and τ_2 obey the equations

$$(\nabla^2-\lambda_1)\tau_1(\varrho, z)=0 \tag{15a}$$

$$(\nabla^2-\lambda_2)\tau_2(\varrho, z)=0 \tag{15b}$$

where λ_1 and λ_2 are the roots of the following quadratic equation

$$\mathrm{j}(\beta_g/\omega)\lambda^2+\{1+\mathrm{j}(\gamma\beta_g\omega/c_0^2)\}\lambda+(\omega^2/c_0^2)=0. \tag{16}$$

To the lowest order in $(\omega\beta_g/c_0^2)$, λ_1 and λ_2 are given by

$$\lambda_1=\mathrm{j}(\omega/\beta_g)\{1+\mathrm{j}(\gamma-1)(\omega\beta_g/c_0^2)\} \tag{17a}$$

$$\lambda_2=-(\omega^2/c_0^2)\{1-\mathrm{j}(\gamma-1)(\omega\beta_g/c_0^2)\}. \tag{17b}$$

If the terms in $(\omega\beta_g/c_0^2)$ are neglected in the expressions for λ_1 and λ_2, τ_1 and τ_2 obviously describe two types of temperature propagation, namely, thermal diffusion and acoustic propagation, as may be most clearly seen in the one-dimensional versions of Eqs. (15). In particular, Eq. (15a) for τ_1 with $\lambda_1=\mathrm{j}\omega/\beta_g$ is essentially the starting point of I. It will be noted that the two types of motion are not completely independent of each other with the terms in $(\omega\beta_g/c_0^2)$ providing a coupling between the two.

The solutions to Eqs. (15) that are in accord with the boundary conditions may be developed into a series

$$\tau_1(\varrho,z)=\sum_n G_1(n)J_0(\alpha_n\varrho/a)\,\mathrm{e}^{-\pi_1(n)z} \tag{18a}$$

$$\tau_2(\varrho,z)=\sum_n G_2(n)J_0(\alpha_n\varrho/a)\,\mathrm{e}^{-\pi_2(n)z} \tag{18b}$$

where

$$\pi_1(n)=(\alpha_n^2/a^2+\lambda_1)^{1/2} \tag{19a}$$

$$\pi_2(n)=(\alpha_n^2/a^2+\lambda_2)^{1/2} \tag{19b}$$

and where α_n is the nth zero of the zero-order Bessel function and where λ_1 and λ_2 are related via

$$\{1+\mathrm{j}(\beta_g/\omega)\lambda_1\}G_1(n)+\{1+\mathrm{j}(\beta_g/\omega)\lambda_2\}G_2(n)=0\,, \tag{20}$$

a result which is found to hold to a sufficient degree at typical operating chopping frequencies. In identifying expressions (18) as the solutions to Eqs. (15) an implicit assumption has been made, as in I, that the cylindrical cell is sufficiently long so that the temperature wave reflected from the end wall of the cell (opposite to the sample) may be neglected.

To determine the gas temperature completely one makes use of the equations describing the continuity in the temperature and heat flow at the boundaries that separate any two of the three regions of gas, sample and backing substrate:

$$\tau_g(\varrho,0)=\tau_s(\varrho,0) \tag{21a}$$

$$\tau_s(\varrho,-1)=\tau_b(\varrho,-1) \tag{21b}$$

$$K_g\frac{\partial\tau_g(\varrho,0)}{\partial z}=K_s\frac{\partial\tau_s(\varrho,0)}{\partial z} \tag{21c}$$

$$K_s\frac{\partial\tau_s(\varrho,-1)}{\partial z}=K_b\frac{\partial\tau_b(\varrho,-1)}{\partial z}. \tag{21b}$$

The temperature of the sample τ_s and backing substrate τ_b can be determined via a Green's function method and they are given by (see I)

$$\tau_s(\varrho,z)=\sum_n J_0(\alpha_n\varrho/a)\Big\{S_1(n)\,\mathrm{e}^{\sigma_s(n)z}+S_2(n)\,\mathrm{e}^{-\sigma_s(n)z}+\frac{Q_nI(n)}{2\sigma_s(n)}\int_{-1}^{0}\mathrm{e}^{-\sigma_s(n)|z-z'|-\alpha|z'|}\,\mathrm{d}z'\Big\} \tag{22a}$$

$$\tau_b(\varrho,z)=\sum_n B(n)J_0(\alpha_n\varrho/a)\,\mathrm{e}^{\sigma_b(n)(z+1)} \tag{22b}$$

in which $Q_n=2\{a^2J_1^2(\alpha_n)\}^{-1}$, $J_1(x)$ is a first order Bessel function, $\sigma_i(n)=\{(\alpha_n^2/a^2)+\mathrm{j}(\omega/\beta_i)\}^{1/2}$, $i=s,b$, and $I(n)$ is the Bessel decomposition of the beam profile:

$$I(n)=\int_0^a I(\varrho)J_0(\alpha_n\varrho/a)\,\varrho\,\mathrm{d}\varrho\,. \tag{23}$$

Eqs. (20) and (21) constitute five simultaneous equations which can be solved for the expansion coefficients $G_1(n)$, $G_2(n)$, $S_1(n)$, $S_2(n)$ and $B(n)$. For the gas region with which the subsequent calculations will be concerned, one has

$$G_1(n)=\frac{1+\mathrm{j}(\beta_g/\omega)\lambda_2}{\mathrm{j}(\beta_g/\omega)(\lambda_2-\lambda_1)}\,\tilde G(n) \tag{24a}$$

$$G_2(n)=\frac{1+\mathrm{j}(\beta_g/\omega)\lambda_1}{\mathrm{j}(\beta_g/\omega)(\lambda_1-\lambda_2)}\,\tilde G(n) \tag{24b}$$

in which

$$\tilde G(n)=Q_nI(n)\frac{\{1+b(n)\}\,a_+(n)+\{1-b(n)\}\,a_-(n)}{\{1+b(n)\}\{1+\tilde g(n)\}\,\mathrm{e}^{\sigma_s(n)l}-\{1-b(n)\}\{1-\tilde g(n)\}\,\mathrm{e}^{-\sigma_s(n)l}} \tag{25}$$

$$\tilde g(n)=\frac{K_g}{K_s\sigma_s(n)}[\{\pi_1(n)-\pi_2(n)\}+\mathrm{j}(\beta_g/\omega)\{\pi_1(n)\lambda_2-\pi_2(n)\lambda_1\}] \tag{26}$$

with

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

and

$$b(n)=\kappa_b\sigma_b(n)/\kappa_s\sigma_s(n).$$

In relating the gas temperature variation to its pressure which photoacoustic effect measures, a distinction must be made between the manner in which pressure variation is generated by the thermal mode τ_1 (consisting of terms with $G_1(n)$) and propagating mode τ_2 (made up of terms with $G_2(n)$). The contribution $\delta P^{(1)}$ from the thermal mode is envisioned to arise from the compression effect of a small region adjacent to the sample, the "thermal piston" in the sense of Ref. [7], and it is given by (see 1)

$$\delta P^{(1)} = \frac{\gamma P_0 \, e^{j\omega t}}{T_0} \frac{l_p}{l_g} \langle \tau_1(\varrho, z) \rangle_p \tag{27}$$

where l_p is the thickness of the piston (several thermal diffusion lengths), l_g is the length of the gas column, and $\langle \ \rangle_p$ denotes a spatial average over the volume of the piston. The contribution $\delta P^{(2)}$ from the propagation mode, on the other hand, is determined by the hydrodynamic flow of the gas and is given by, upon making use of Eq. (11).

$$\delta P^{(2)} = \frac{\gamma p_0 \, e^{j\omega t}}{(\gamma - 1) T_0} \{1 + j(\beta_g/\omega)\lambda_2\} \langle \tau_2(\varrho, z) \rangle_\Omega \tag{28}$$

where $\langle \ \rangle_\Omega$ indicates a spatial average over the volume Ω_0 of the gas column. It is then seen that the photoacoustic signal δP may be expressed as

$$\delta P = \delta P^{(1)} + \delta P^{(2)} \tag{29}$$

with

$$\delta P^{(1)} = \frac{4\pi\gamma P_0 \, e^{j\omega t}}{T_0 \Omega_0} \frac{\{1 + j(\beta_g/\omega)\lambda_2\}}{j(\beta_g/\omega)(\lambda_2 - \lambda_1)} \sum_n \frac{K(n)}{\pi_1(n)} \tag{30}$$

$$\delta P^{(2)} = \frac{4\pi\gamma P_0 \, e^{j\omega t}}{(\gamma - 1) T_0 \Omega_0} \frac{\{1 + j(\beta_g/\omega)\lambda_2\}\{1 + j(\beta_g/\omega)\lambda_1\}}{j(\beta_g/\omega)(\lambda_1 - \lambda_2)} \sum_n \frac{K(n)}{\pi_2(n)} \tag{31}$$

$$K(n) = \frac{I(n)}{\alpha_n J_1(\alpha_n)} \frac{\{1 + b(n)\} a_+(n) + \{1 - b(n)\} a_-(n)}{\{1 + b(n)\}\{1 + \bar{g}(n)\} \, e^{\sigma_s(n) l} - \{1 - b(n)\}\{1 - \bar{g}(n)\} \, e^{-\sigma_s(n) l}} \tag{32}$$

Eqs. (29—32) constitute the extension of the study of the three-dimensional photoacoustic signal to include the contribution from the propagating mode. It may be readily verified that in the neglect of the latter, in which case $\lambda_1 = j\omega/\beta_g$ and $\lambda_2 = 0$, Eq. (29) is reduced to the previously obtained expression for the photoacoustic effect arising from the thermal diffusion mode alone (namely, Eq. (24) of I, in which, however, a factor of 2π has been inadvertantly left out.)

III. DISCUSSION

The objective of this note is the derivation of the propagation mode contribution to the three-dimensional photoacoustic effect, which has been omitted in I. A motivating factor for the present undertaking has been the observation that while the theory of I agrees rather well with the experimental results using a Corning glass sample in air and in helium [2] over a wide range of modulation frequency, the agreement between theory and experiment is poorer in the low chopping frequency regime and it is suggested [4] that the inclusion of propagation mode contribution be examined. The results of the present study have indeed been applied to the above mentioned systems, but detailed numerical calculations indicate that the propagation mode contribution is much too small to bring about a significant improvement in agreement. On the other hand, since the currently available theoretical acounts [8—10] that do include propagation mode all deal with one-dimensional cases, it is felt that the results of the present three-dimensional treatment may be useful in the future in analysing other systems.

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