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PROGRESS REPORT

PHOTOCHEMICAL GENERATION OF THE OPTOACOUSTIC EFFECT

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ABSTRACT

Research during the last granting period has been conducted in four areas concerned with the photoacoustic effect: proof of six theorems concerning the time profiles of photoacoustic waves, generation of photoacoustic waves from a layered source, production of acoustic waves by laser irradiation of optically thin cylinders and layers, and generation of the photoacoustic effect by excitation of a transient optical grating in viscous fluids.

In the first of these areas we have shown how the time profile of a photoacoustic wave can be found as a mapping of spatial deposition of heat in space when the exciting light pulse is short. Three different mappings have been derived depending on whether there is symmetry in one, two, or three dimensions. We have also found that for long exciting light pulses, the time profile of the photoacoustic wave is proportional to the zero'th, one-half'th, or first time derivative of the intensity profile of exciting light for waves in one, two, three dimensions, respectively. In addition to providing insight into the production of the photoacoustic effect, the theorems permit determination of the shape of photoacoustic waves without resort to tedious solution of differential equations.

We have investigated both experimentally and theoretically a new photoacoustic generator of ultrasound where a layered structure consisting of a series of absorbing and transparent layers is excited by optical radiation to produce a train of pulses. Theoretical methods have been developed for calculation of the properties of the emitted acoustic radiation. When excited with a finite diameter laser beam, the source produces a highly directional acoustic beam depending on the number of layers in the structure and the diameter of the light beam. The characteristics of the photoacoustic effect generated by cylinders and layers have been investigated experimentally and theoretically.

In the case of the cylinder, the mathematical expressions describing the photoacoustic effect are cumbersome; even the limiting cases of long laser pulses, low density, or equal sound speed and density do not yield simple mathematical expressions for the time profiles

of the waves. However, a rigorous description of the photoacoustic effect has been given and compared with experiment.

We have also conducted experiments to investigate the effects of viscosity on the evolution of a picosecond transient grating. Experiments were done with ethylene glycol at a series of different temperatures to produce different viscous damping in the solution. A theoretical description of the photoacoustic effect was given so that sound speed, viscosity, and viscous relaxation times could be extracted from the data.

I. MAPPINGS FOR DETERMINATION OF PHOTOACOUSTIC TIME PROFILES

A. Introduction

The time profile of the sonic wave generated when laser radiation is absorbed by a body is a fundamental property of the photoacoustic effect¹⁻⁸. Over the past several years the determination of the information content of photoacoustic waveforms produced by laser irradiation of various bodies has been a primary goal of this research. Broadly speaking, the present research has been directed toward a rigorous and precise determination of the exact shapes of photoacoustic waveforms with two objectives in mind. The first has been to investigate the properties of the photoacoustic effect itself giving as precise a description of all facets of the effect as possible. The second goal, which relies wholly on achievement of the first, has been to introduce an analytical technique whereby information contained in the time profile of photoacoustic waves can be used to determine properties of an irradiated body. In this part of the progress report, a facet of the work concerned with description of the fundamental properties of the photoacoustic effect is given, namely, a way of determining acoustic waveforms for the extreme cases of very short and very long laser pulses.

Section IB gives derivations of three theorems that describe how a knowledge of the spatial deposition of heat by the optical exciting beam can be used to determine the time profile of photoacoustic waves for bodies with symmetry in one, two, and three dimensions. Section IC gives a derivation of the time profile of photoacoustic waves produced when the time duration of the exciting radiation is long compared with the transit time of sound across the irradiated body. Experimental results are presented in Section ID. The Discussion Section gives brief descriptions of other results and shows the relationship between the present derivations and some previously determined properties of photoacoustic waveforms. Full details of the research presented in this section can be found in *Phys. Rev. Lett.* **67**, 3384 (1991), and "Properties of Photoacoustic Waves in One, Two, and Three Dimensions" (submitted to *Acustica*)

B. Short Optical Pulse Limit

When heat conduction takes place on a time scale long compared with duration of the acoustic wave, the coupled differential equations for the pressure and temperature reduce to a wave equation for pressure. The time profile of a photoacoustic wave is then determined by the wave equation for pressure⁹, which can be written in terms of a velocity potential ϕ as

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \phi = \frac{\beta}{\rho_f C_P} H, \quad (1)$$

where p is determined from $p = -\rho_f \frac{\partial \phi}{\partial t}$, c is the sound speed, ρ_f is the fluid density, β is the isobaric volume expansion coefficient, C_P is the heat capacity per unit mass, t is the time, and H is the heating function defined as the thermal energy per time and volume deposited by the optical beam. In order to derive mappings that determine the spatial profile of the photoacoustic wave we first find a formal solution to the wave equation using a Green's function¹⁰ for the wave equation for the velocity potential,

$$\phi(\mathbf{x}, t) = \frac{-\beta}{4 \pi \rho_f C_P} \int_0^t dt' \int d\mathbf{x}' g(\mathbf{x}, t | \mathbf{x}', t') H(\mathbf{x}', t'), \quad (2)$$

where $g(\mathbf{x}, t | \mathbf{x}', t')$ is the Green's function for an infinite medium.

In one dimension, the appropriate Green's function (for $z > z'$) is

$$g(\mathbf{x}, t | \mathbf{x}, t') = 2\pi c u[(t - t') - (z - z')/c]$$

where $u(\xi)$ is the Heaviside function. Substitution of the one-dimensional Green's function into Eq. 2 followed by differentiation of the resulting velocity potential gives the photoacoustic pressure as the space integral

$$p(z, t) = \frac{\beta c}{2C_P} \int dz' H\left(z', t - \frac{z - z'}{c}\right). \quad (3)$$

Now we take a heating function of the form

$$H(\hat{\xi}, t) = \bar{\alpha} E_0 \hat{h}(\hat{\xi}) \delta(t),$$

where $\bar{\alpha}$ is the optical absorption coefficient, E_0 is the energy fluence in the light beam, and $\hat{h}(\hat{\xi})$ is a spatial heating function written as a function of a generalized dimensionless space coordinate $\hat{\xi} = \xi/\xi_0$ (where ξ is z in one dimension, ρ in two dimensions, or r in three dimensions), and ξ_0 is the length parameter for absorption. On substitution of the above heating function into Eq. 3 and evaluation of the resulting integral, we find the photoacoustic wave at a field point located along the positive z axis for $t > 0$ to be the mapping,

$$p(\hat{\tau}_0) = \frac{\kappa}{2} \hat{h}(\hat{\tau}_0) , \quad (4)$$

where the dimensionless retarded time from the origin $\hat{\tau}_0$ is given by $\hat{\tau}_0 = (c/\xi_0)(t - \xi/c)$ and where an amplitude factor κ is given by $\kappa = \bar{\alpha}\beta E_0 c^2 / C_P$. *Eq. 4 shows that the spatial deposition of heat in the fluid by the laser beam is mapped directly into the temporal profile of the photoacoustic wave.*

We now consider the analogous problem of determining the photoacoustic wave for a spherically symmetric deposition of heat. The wave equation for the velocity potential, which contains only a single radial space derivative, reduces to a one-dimensional wave equation (with H replaced by rH) on introduction of the variable $\phi^\dagger = r \phi$. The one-dimensional Green's function given above can be used for a solution, but must be modified to include the effect of an inwardly propagating spherical wave, which is launched simultaneously with the outwardly travelling wave, and which is reflected at the origin, reaching the field point with an inverted amplitude. The Green's function for Eq. 2 must therefore be the sum of two one-dimensional Green's functions

$$g(r, t | r', t') - g(r, t | -r', t') ,$$

which gives the photoacoustic pressure as

$$p(r, t) = \frac{\beta c}{2 C_P r} \int dr' r' \left[H\left(r', t - \frac{r-r'}{c}\right) - H\left(r', t - \frac{r+r'}{c}\right) \right] . \quad (5)$$

For a delta function heating pulse in time, we thus find the photoacoustic pressure for $t > 0$ to be given by the mapping

$$p(\hat{r}_0) = \frac{\kappa}{2\hat{r}} \hat{r}_0 [\hat{h}(-\hat{r}_0) + \hat{h}(\hat{r}_0)] . \quad (6)$$

In three dimensions the time profile of the photoacoustic wave is determined by a sum of two mappings of the spatial deposition of heat: the first mapping describes a radially expanding wave, the second takes into account reflection of and inwardly propagating wave spherical wave at the origin.

Consider now a cylindrically symmetric source. We find the photoacoustic wave generated by a delta function heating pulse by expressing Eq. 6 in terms of cylindrical coordinates z and ρ (i.e. $r = \sqrt{\rho^2 + z^2}$) and integrating the result over the z axis. This procedure gives the photoacoustic pressure as

$$p(\hat{\rho}, \hat{t}) = \kappa \int_{-\infty}^{\hat{t}-\hat{\rho}} d\hat{\zeta} \frac{f'(\hat{\zeta}) + f'(-\hat{\zeta})}{[(\hat{t} - \hat{\zeta})^2 - \hat{\rho}^2]^{\frac{1}{2}}} , \quad (7)$$

where $f'(\hat{\zeta})$ is the derivative of a source function, the form of which is not yet specified, and $\hat{t} = ct/\rho_0$. Now immediately after absorption of radiation from a delta function optical pulse in time, the spatial profile of the acoustic pressure in the fluid must equal $\kappa \hat{h}(\hat{\rho})$. Since cylindrically symmetric waves are simultaneously launched inwardly and outwardly from the z axis, the pressure is constrained at $t = 0$ by the initial condition, $\int_{\hat{\rho}}^{\infty} d\hat{\zeta} f'(\pm \hat{\zeta})(\hat{\zeta}^2 - \hat{\rho}^2)^{-\frac{1}{2}} = (1/2) \hat{h}(\hat{\rho})$, which is valid for both the positive and negative signs. To find the source function we transform this expression using a variation of Firsov's inversion¹¹. We determine $f(\hat{\eta})$ as the integral⁷

$$f(\hat{\eta}) = -\frac{\rho_0}{\pi} \int_{\hat{\eta}}^{\infty} d\hat{\rho} \frac{\hat{\rho} \hat{h}(\hat{\rho})}{\sqrt{\hat{\rho}^2 - \hat{\eta}^2}} . \quad (8)$$

Thus in two dimensions the spatial deposition of heat gives a source function that is mapped into the time profile of the photoacoustic wave. As in the case of the three-dimensional theorem, the two-dimensional mapping takes into account the reflection of an inwardly propagating wave.

We now consider an optically thin, infinite fluid layer, a fluid cylinder, and a droplet as examples of the use of the three theorems for short optical pulses given by Eqs. 4, 6, and 7 and 8. The spatial heating function for these bodies is given by $\hat{h}(\hat{\xi}) = \tilde{\Theta}_{0,1}(1 - \hat{\xi})$, where $\tilde{\Theta}_{i,j}(\hat{\xi})$ is a square wave function defined as unity for $\hat{\xi}$ between i and j , and as zero otherwise. Equations 4, 6, and 7 give the photoacoustic pressure for $t > 0$ in one, two (with $\bar{\rho} \gg 1$), and three dimensions as

$$p(\hat{\tau}) = \frac{\kappa}{2} \tilde{\Theta}_{0,1}(\hat{\tau}) \quad (9)$$

$$p(\hat{\tau}) = \begin{cases} \frac{\kappa}{\pi\sqrt{2\bar{\rho}}} \int_{-\hat{\tau}}^0 d\hat{\eta} \frac{(\hat{\eta}+1)}{(\hat{\eta}+\hat{\tau})^{\frac{1}{2}} [1-(\hat{\eta}+1)^2]^{\frac{1}{2}}} & 0 < \hat{\tau} < 2 \\ \frac{\kappa}{\pi\sqrt{2\bar{\rho}}} \int_{-2}^0 d\hat{\eta} \frac{(\hat{\eta}+1)}{(\hat{\eta}+\hat{\tau})^{\frac{1}{2}} [1-(\hat{\eta}+1)^2]^{\frac{1}{2}}} & 2 < \hat{\tau} \end{cases} \quad (10)$$

$$p(\hat{\tau}) = \frac{\kappa}{2\hat{\tau}} (1 - \hat{\tau}) [\tilde{\Theta}_{0,1}(\hat{\tau}) + \tilde{\Theta}_{1,2}(\hat{\tau})], \quad (11)$$

respectively, where $\hat{\tau}$ is the dimensionless retarded time from the edge of the body defined as $\hat{\tau} = (c/\xi_0)[t - (\xi - \xi_0)/c]$, as shown in Fig. 1.

C. Long Optical Pulse Limit

It is possible to derive analytical expressions for the photoacoustic pressure when the exciting optical pulse has a duration far greater than the transit time of sound across the absorption length in the irradiated body. We first solve the problem in the frequency domain by taking a heating function of the form

$$H(\hat{\xi}, t) = \bar{\alpha} I_0 \hat{h}(\hat{\xi}) e^{-i\omega t},$$

In one dimension Eq. 3, gives the acoustic pressure as an space integral of the spatial heating function with the factor $\exp(-i\omega z'/c)$. Since the integration extends only over the source, we approximate the exponential function as unity giving a constant for the space integration. The low frequency response of the distribution and its corresponding Fourier transform thus become

$$p_l = \frac{\bar{\alpha}\beta I_0 \xi_l c}{2C_P} e^{-i\hat{q}_l \hat{\tau}_l} \quad \text{and} \quad p_l(\hat{\tau}_l) = \frac{\kappa}{2} \delta(\hat{\tau}_l), \quad (12)$$

respectively, where $\hat{q}_l = \omega \xi_l / c$ $\hat{r}_l = (c/\xi_l) (t - \xi/c)$ and in n dimensions

$$\xi_l^n = \int_0^\infty d\xi \xi^{n-1} \hat{h}(\xi) .$$

In three dimensions, we find the photoacoustic pressure for a spherically symmetric deposition of heat by substitution of the frequency domain heating function above into Eq. 5. Again, the integration is over a region of space small compared with the acoustic wavelength. Thus we approximate the difference of the two exponential functions as proportional to the radial derivative of $\exp(-i\omega\tau)$. This gives the low frequency response of the spherical distribution and its corresponding Fourier transform as

$$p_l = \frac{\bar{\alpha}\beta I_0 \xi_l c}{C_P \hat{r}_l} \frac{d}{d\hat{r}_l} e^{-i \hat{q}_l \hat{r}_l} \quad \text{and} \quad p_l(\hat{r}_l) = \frac{\kappa}{\hat{r}_l} \frac{d}{d\hat{r}_l} \delta(\hat{r}_l) , \quad (13)$$

respectively, where $\hat{r}_l = r/\xi_l$.

In two dimensions, we determine the frequency domain velocity potential by using a Green's function solution to the Helmholtz equation¹⁰ corresponding to Eq. 1. The Green's function for an infinite medium is $i\pi H_0^{[1]}(k | \boldsymbol{\rho} - \boldsymbol{\rho}' |)$, which for a cylindrically symmetric source reduces to $J_0(k\rho')H_0^{[1]}(k\rho)$, where $H_0^{[1]}$ is the zero'th order Hankel function of the first kind, J_0 is a Bessel function, and k is the wavevector. In the limit of low frequencies, $J_0(k\rho') \cong 1$ which gives the low frequency photoacoustic pressure and its Fourier transform as

$$p_l = \frac{\pi \bar{\alpha}\beta I_0 \xi_l c}{2C_P} \hat{q}_l H_0^{[1]}(\hat{q}_l \hat{\rho}_l) e^{-i \hat{q}_l \hat{t}_l} \quad \text{and,} \quad p_l(\hat{t}_l) = \kappa \frac{d}{d\hat{t}_l} \left[\frac{u(\hat{t}_l - \hat{\rho}_l)}{\sqrt{\hat{t}_l^2 - \hat{\rho}_l^2}} \right] . \quad (14)$$

respectively, where $\hat{\rho}_l = \rho/\xi_l$ and $\hat{t}_l = ct/\xi_l$.

The time profile of the photoacoustic wave generated by a long light pulse is determined by convoluting a normalized light intensity function of the form

$$I(t) = E_0 S(t/\Theta)/\Theta$$

with the long pulse response functions given by Eqs. 12-14 to give the photoacoustic pressure in one, two (for $\hat{\rho}_l \gg 1$), and three dimensions as

$$P(\tilde{\tau}_l) = \frac{\kappa}{2\hat{\Theta}} S(\tilde{\tau}_l) \quad (15)$$

$$P(\tilde{\tau}_l) = \frac{\kappa}{\hat{\Theta}^{\frac{3}{2}} \sqrt{2\hat{\rho}}} \int_{-\infty}^{\tilde{\tau}_l} dT \frac{dS(T)/dT}{\sqrt{\tilde{\tau}_l - T}} \quad (16)$$

$$P(\tilde{\tau}_l) = \frac{\kappa}{\hat{\Theta}^2 \hat{\tau}_l} \frac{d}{d\tilde{\tau}_l} S(\tilde{\tau}_l) \quad (17)$$

respectively, where

$$\hat{\Theta} = c\Theta/\xi_l \text{ and } \tilde{\tau}_l = \tau_l/\hat{\Theta}.$$

According to the definition of the fractional derivative^{13,14}, the photoacoustic pressure in one, two, and three dimensions in the case of long pulse excitation is proportional to the zero'th, the one-half'th, and the first time derivative respectively of the intensity of the exciting optical pulse evaluated at the retarded time from the origin. Figure 2 gives plots of Eqs. 15-17 where S is a Gaussian function of time.

D. Experiments

Experiments with short optical pulses were done using the frequency doubled output of a Q-switched Nd:YAG laser. The acoustic waves were detected with a polyvinylidene fluoride film transducer whose output was displayed on an oscilloscope after amplification by a factor of 40.

The theorems pertain to uniform media. The challenge here is to produce an absorbing body of a specified geometry and to have that body surrounded by an acoustically identical medium. We have approached this problem by finding two immiscible fluids with widely varying polarities so that a dye will remain in one of the fluids only. A fluid layer was thus made by confining dyed methanol between two sheets of 13 μm , transparent polyvinylidene chloride film. The dyed layer was then placed in a methanol-filled photoacoustic cell.

Figure 1

Figure 2

Fig. 1: Photoacoustic waveforms from short laser pulses. Left column: photoacoustic pressure P in arbitrary units versus dimensionless time $\hat{\tau}$ for (a) a fluid layer, (b) a cylinder, and (c) a sphere. Right column: experimental waveforms obtained by irradiating (a) a 3 mm thick layer, (b) a 150 μm radius cylinder, and (c) a 500 μm radius droplet. The time and voltage scales on the oscilloscope are (a) 1 $\mu\text{sec/div}$ and 20 mV/div , (b) 200 ns/div and 20 mV/div , (c) 500 ns/div and 50 mV/div

Fig. 2: Photoacoustic waveforms from long laser pulses. Left column: pressure P in arbitrary units versus dimensionless retarded time from the origin for bodies in (a) one dimension, (b) two dimensions, and (c) three dimensions excited by a Gaussian light pulse in time. Right column: experimental waveforms obtained by irradiating (a) a highly absorbing half space, (b) a 100 μm radius cylinder, and (c) a 200 μm radius droplet of benzaldehyde. The oscilloscope time and voltage scales for the three traces are (a) 200 ns/div and 100 mV/div , (b) 200 ns/div and 50 mV/div , and (c) 200 ns/div and 20 mV/div .

A fluid cylinder was formed by pumping dyed benzaldehyde through narrow bore tubing into a water-filled photoacoustic cell. These two fluids have sound speeds and densities within 4% of each other and are thus essentially identical acoustically. Since benzaldehyde and water are immiscible, at the tip of the tubing a fine cylindrical stream of benzaldehyde is formed which extends over 1 cm into the cell (before breaking up). For the experiments with a sphere, a droplet of benzaldehyde was suspended at the end of a fine capillary. Figure 1 shows waveforms generated by a layer, a cylinder, and a sphere.

We were unable to make a flashlamp with sufficient intensity to provide long optical pulses. Our solution for the long pulse experiments was to use $1.06\ \mu\text{m}$ radiation pulses from the Nd:YAG laser with the quarter wave plate associated with the Q-switch removed. The laser generated a train of approximately $1\ \mu\text{s}$ long pulses, which were typically asymmetric, with slightly faster risetimes than fall times. For the experiment in one dimension, a concentrated solution of infrared absorbing dye in benzaldehyde was used to give an exponential deposition of heat in space. The dyed solution was placed in contact with H_2O , as was the case with the cylinder and the droplet. Experimental waveforms for the long pulse experiments are shown in Fig. 2.

E. Discussion and Conclusions

The results given above for short laser pulses are general proofs relating the photoacoustic waveform to the deposition of heat in space; when the theorems apply, photoacoustic waveforms can be determined directly, obviating solution of a differential equation. It is of interest to compare the general results derived here to some solutions that have appeared in the literature for special cases. The one-dimensional mapping given here for delta function light pulses explains the character of the square waves generated by an optically thin layer or the exponential waves generated by Beer's law absorption of radiation, found in Refs. 2, and 15-17 respectively. In addition, the two-dimensional mapping reproduces⁷ the photoacoustic waveform for a Gaussian heat distribution in space calculated in Refs. 4, 18 and 19 and studied experimentally in Ref. 20, as well as that for an optically thin, infinite cylinder discussed in Refs. 2 and 28. The three-dimensional mapping explains the origin of the *N*-shaped wave found in Refs 2, 21, and 22 for photoacoustic waves generated by a droplet, and can be extended to the problem of the "bursting balloon", (which also generates an *N*-shaped wave), found in texts on classical acoustics.^{23,24} We also believe that the delta function response given here would lead to considerable simplification of some previous calculations in three dimensions^{25,26} since the acoustic wave excited by arbitrary light pulses can be found in terms of simple convolution integrals.

The theorems for long light pulses, show that the photoacoustic waveforms from a number of different bodies can be found directly without resort to solution of the wave equation. For example, the solutions to the wave equation derived in Refs. 15 and 17 are given here as a straightforward consequence of the theorem for one-dimensional waves. We note as well, that the authors of Ref. 18 showed that the temporal profiles of photoacoustic waves produced by square, circular, and Gaussian laser beams with a long Gaussian time profile had nearly identical waveforms. The theorem given for two-dimensional waves clearly explains why these sources produce the same acoustic waveform. We find the waveform in Fig. 2b to match closely those found in Ref. 18. In addition, we note that the amplitude dependence of the photoacoustic pressure in two dimensions, as given by Eq 16, is identical with that calculated in Ref 28 using a completely different method. We also find agreement between the waveform predicted from the long pulse theorem for three dimensions and the waveforms calculated in Refs. 22 and 27.

Figure 3

Fig. 3: Diagram of the layered photoacoustic source. The shaded regions indicate the optically thin layers where sound is generated by the absorption of optical radiation. The layers are marked at the top with the number of the layer, in the center with the number of the region, and at the bottom with the coordinates along the X axis at positions of the interfaces.

Both photoacoustic and electromagnetic radiation are governed by similar wave equations. We note however that the properties of photoacoustic waves given here are unique to monopole radiation, which is necessarily generated by the photoacoustic effect, but which requires the production of charge of a single sign in electromagnetic theory.

II. PHOTOACOUSTIC DISTRIBUTED SOURCE

A. Introduction

In this section of the progress report the properties of a new photoacoustic source of ultrasonic radiation are discussed. By juxtaposing a series of optically thin and transparent layers each having the same sonic transit time, it is possible to construct a distributed feedback generator of ultrasonic waves based on the photoacoustic effect. The response of the device to sinusoidally modulated, continuous radiation is determined by summing the plane wave amplitudes from each absorbing layer, taking into account the effects of reflection and transmission at the interfaces between layers. The frequency domain solution shows the effects of constructive and destructive interference with maxima in the photoacoustic amplitude at points where the modulation frequency of the optical radiation is such that the acoustic wavelength equals twice the layer thickness divided by odd integer. The response of the layered structure to laser pulsed is determined by Fourier transformation of the frequency domain solution. Solutions for short pulse excitation of the layers show the emitted photoacoustic wave to be a series of square waves in time. Experimental waveforms generated by a series of layers excited with the 532 *nm* output of a frequency doubled Nd:YAG laser are presented. The relationship between the layered photoacoustic source and similar devices such as the acoustic filter, multilayer dielectric interference filters, superlattices, and materials with photonic band gaps is discussed.

Section IIB, gives the equations describing the properties of the device for modulated continuous wave radiation. A matrix method is developed for determination of the signal from an arbitrary number of layers. The origin of a somewhat unexpected low frequency oscillation in the photoacoustic signal is discussed in terms of a harmonic oscillator model for

the layered structure. The response of the structure to multiple pulses is discussed. Section IIC gives the results of experiments carried out by irradiating a series of layers with the output of a Q-switched Nd:YAG laser. The Discussion Section, Section IID, describes the relation between the properties of the layered source and similar structures such as super lattices, acoustic filters, and photonic structures. Full details of the research given in this section can be found in *Nature* 355, 806 (1992) and "Distributed Source Photoacoustic Generation of Ultrasound" (submitted to *Ultrasonics*).

B. Frequency Domain Solution

The approach taken here for calculation of the acoustic amplitude emanating from a structure of multiple layers is to use linear superposition of the amplitudes from each photoacoustic source layer in the structure with the acoustic amplitude evaluated at the outside edges of the last layers in the assembly. The acoustic waves produced in each absorbing layer are assumed to be plane waves, as would be produced by uniform irradiation of either an infinitely wide, optically thin, fluid or solid layer²⁹. The effects of heat conduction and viscosity are not treated. A schematic diagram of the layered source is shown in Fig. 3 with the optically absorbing layers indicated with shading. The shaded regions in Fig 3 indicate the optically thin layers where sound is generated by the absorption of optical radiation. The layers are marked at the top with the number of the layer, in the center with the number of the region, and at the bottom with the coordinates along the X axis at positions of the interfaces. Media to the left of layer 1 and to the right of layer N are transparent and of infinite extent. The structure consists of a total of N absorbing layers, each of thickness l , separated by transparent layers of thickness s . The densities ρ and sound speeds c of the absorbing and transparent layers are given subscripts a and t , respectively; the magnitude of the acoustic wavevectors k in the absorbing and transparent layers are denoted with the same subscripts. The photoacoustic pressures in the layers P , and pressure amplitudes A and B of the sound waves are labelled with integer subscripts indicating the region to which they correspond.

Provided the effects of viscosity and heat conduction can be neglected, the acoustic pressure p produced by absorption of optical radiation is a solution to the wave equation for pressure⁹,

$$\square^2 p = \frac{-\beta}{C_P} \frac{\partial H}{\partial t} , \quad (18)$$

which is equivalent to the wave equation for the velocity potential, Eq 1, given in the previous section. For a continuous laser beam sinusoidally modulated at frequency ω , the alternating component of the heat deposited per unit volume and time H is given by

$$H = \bar{\alpha} I_0 e^{-i\omega t} , \quad (19)$$

where I_0 is the laser intensity and $\bar{\alpha}$ is the optical absorption coefficient of an absorbing layer. For the present problem, which has symmetry in one dimension, the solution to the wave equation in region $2n - 1$ must be a spatially constant term plus the sum of left- and right-going travelling waves; in region $2n$ the solution must be the sum of two travelling waves³⁰. The acoustic pressure in the two regions can therefore be expressed as

$$\begin{aligned} P_{2n-1} &= A_{2n-1} e^{-ik_a x} + B_{2n-1} e^{ik_a x} + \frac{2\kappa_f}{\hat{q}} \\ P_{2n} &= A_{2n} e^{-ik_t x} + B_{2n} e^{ik_t x} , \end{aligned} \quad (20)$$

where an amplitude factor κ_f and a dimensionless frequency $\hat{q}$ have been defined as

$$\kappa_f = i\bar{\alpha}\beta I_0 c_a l / 2C_P \quad \text{and} \quad \hat{q} = \frac{\omega l}{c_a} ,$$

where x is the distance along the X coordinate. If the absorbing layer is an infinitely wide isotropic solid, Eq. 20 gives the correct solution for the acoustic pressure, however, the amplitude factor must be modified slightly²⁹.

Now, the acoustic amplitudes in regions $2n - 1$ and $2n$ arising from light absorption in region $2n-1$ must be related through the acoustic conservation equations at the interface between the two regions. That is, the acoustic pressure and acceleration at the interface approached from either side of the interface must be identical³⁰. These conditions give the

following two equations for the acoustic amplitudes,

$$\begin{aligned} \frac{2\kappa_f}{\hat{q}} + A_{2n-1} e^{-ik_a x_{2n-1}} + B_{2n-1} e^{ik_a x_{2n-1}} &= A_{2n} e^{-ik_t x_{2n-1}} + B_{2n} e^{ik_t x_{2n-1}} \\ - A_{2n-1} e^{-ik_a x_{2n-1}} + B_{2n-1} e^{ik_a x_{2n-1}} &= \hat{Z}[-A_{2n} e^{-ik_t x_{2n-1}} + B_{2n} e^{ik_t x_{2n-1}}] , \end{aligned} \quad (21)$$

where $\hat{Z}$ is a dimensionless impedance factor defined as

$$\hat{Z} = \rho_a c_a / \rho_t c_t .$$

Equations 21, when expressed in matrix form and multiplied on the left by the matrix $\frac{1}{2} \begin{bmatrix} 1 & -1 \\ 1 & 1 \end{bmatrix}$ give the acoustic amplitudes as solutions to

$$\begin{bmatrix} A_{2n-1} e^{-ik_a x_{2n-1}} \\ B_{2n-1} e^{ik_a x_{2n-1}} \end{bmatrix} + \begin{bmatrix} \kappa_f / \hat{q} \\ \kappa_f / \hat{q} \end{bmatrix} = \begin{bmatrix} \alpha \\ \alpha \end{bmatrix} \begin{bmatrix} A_{2n} e^{-ik_t x_{2n}} \\ B_{2n} e^{ik_t x_{2n}} \end{bmatrix} , \quad (22)$$

where the matrix α is given by,

$$\alpha = \frac{1}{2} \begin{bmatrix} e^{ik_t s} (1 + \hat{Z}) & e^{-ik_t s} (1 - \hat{Z}) \\ e^{ik_t s} (1 - \hat{Z}) & e^{-ik_t s} (1 + \hat{Z}) \end{bmatrix}$$

Clearly, if the quantity κ_f is set equal to zero, Eq. 22 expresses the relation between the acoustic amplitudes in regions $2n - 1$ and $2n$ when no source is present. For the acoustic pressure in regions $2n$ and $2n + 1$ arising from a source in region $2n - 1$, an analogous derivation can be carried out, which gives

$$\begin{bmatrix} A_{2n} e^{-ik_t x_{2n}} \\ B_{2n} e^{ik_t x_{2n}} \end{bmatrix} = \begin{bmatrix} \beta \\ \beta \end{bmatrix} \begin{bmatrix} A_{2n+1} e^{-ik_a x_{2n+1}} \\ B_{2n+1} e^{ik_a x_{2n+1}} \end{bmatrix} , \quad (23)$$

where the matrix β is defined as

$$\beta = \frac{1}{2} \begin{bmatrix} e^{ik_a l} (1 + 1/\hat{Z}) & e^{-ik_a l} (1 - 1/\hat{Z}) \\ e^{ik_a l} (1 - 1/\hat{Z}) & e^{-ik_a l} (1 + 1/\hat{Z}) \end{bmatrix} .$$

Consider propagation of waves to the left of an absorbing layer. For waves emanating from source region $2n - 1$ and travelling into the transparent region $2n - 2$, the acoustic amplitudes can be shown to obey the relation,

$$\begin{bmatrix} A_{2n-1} e^{-ik_a x_{2n-2}} \\ B_{2n-1} e^{ik_a x_{2n-2}} \end{bmatrix} + \begin{bmatrix} \kappa_f / q \\ \kappa_f / q \end{bmatrix} = \begin{bmatrix} \alpha^* \\ \alpha^* \end{bmatrix} \begin{bmatrix} A_{2n-2} e^{-ik_t x_{2n-3}} \\ B_{2n-2} e^{ik_t x_{2n-3}} \end{bmatrix} . \quad (24)$$

For the same source, the acoustic amplitudes in regions $2n-3$ and $2n-2$ are related through the equation,

$$\begin{bmatrix} A_{2n-2} e^{-ik_t x_{2n-3}} \\ B_{2n-2} e^{ik_t x_{2n-3}} \end{bmatrix} = \begin{bmatrix} \beta^* \end{bmatrix} \begin{bmatrix} A_{2n-3} e^{-ik_a x_{2n-4}} \\ B_{2n-3} e^{ik_a x_{2n-4}} \end{bmatrix}. \quad (25)$$

Now, the medium surrounding the layered photoacoustic source is assumed to be of infinite extent so that to the right of the source there is only a right-going plane wave. The last absorbing layer is also a source. The amplitude of the right-going travelling wave arising from optical absorption in region $2N-1$ is related to the amplitudes of the standing waves in the last layer by

$$\begin{bmatrix} A_{2N-1} e^{-ik_a x_{2N-1}} \\ B_{2N-1} e^{ik_a x_{2N-1}} \end{bmatrix} + \begin{bmatrix} \kappa_f/\hat{q} \\ \kappa_f/\hat{q} \end{bmatrix} = \begin{bmatrix} \alpha_0 \end{bmatrix} \begin{bmatrix} 0 \\ B_{2N} e^{ik_t x_{2N-1}} \end{bmatrix}. \quad (26)$$

where the matrix α_0 is the matrix α with s set to zero. Similarly, the amplitude of the left-going wave arising from absorption of light in the first absorbing layer and which emanates from the layered source along the negative X axis, is related to the amplitudes of the acoustic waves inside the first absorbing layer through the relation

$$\begin{bmatrix} A_1 e^{-ik_a x_0} \\ B_1 e^{ik_a x_0} \end{bmatrix} + \begin{bmatrix} \kappa_f/\hat{q} \\ \kappa_f/\hat{q} \end{bmatrix} = \begin{bmatrix} \alpha_0^* \end{bmatrix} \begin{bmatrix} A_0 e^{-ik_t x_0} \\ 0 \end{bmatrix}. \quad (27)$$

Again, in calculating the wave amplitudes in the infinite medium outside the layered structure for a source layer other than the end layers, Eqns. 26 and 27 can be used but with κ_f set to zero.

The matrix equations given above can be used to determine the acoustic amplitudes at the end of the source produced by layer j of a source having a total of N absorbing layers. For the contribution of the j 'th layer to the acoustic amplitude $B_{2N}^{(j)}$ at the right of the structure and $A_0^{(j)}$ at the left of the structure the following equation obtains,

$$\begin{bmatrix} \alpha\beta \end{bmatrix}^{N-j} \begin{bmatrix} \alpha_0 \end{bmatrix} \begin{bmatrix} 0 \\ B_{2N}^{(j)} e^{ik_t x_{2N-1}} \end{bmatrix} - \begin{bmatrix} \kappa_f/\hat{q} \\ \kappa_f/\hat{q} \end{bmatrix} = \begin{bmatrix} \xi \end{bmatrix} \begin{bmatrix} \alpha^* \beta^* \end{bmatrix}^{j-1} \begin{bmatrix} \alpha_0^* \end{bmatrix} \begin{bmatrix} A_0^{(j)} e^{-ik_t x_a} \\ \end{bmatrix} - \begin{bmatrix} \xi \end{bmatrix} \begin{bmatrix} \kappa_f/\hat{q} \\ \kappa_f/\hat{q} \end{bmatrix}, \quad (28)$$

where the matrix $\alpha\beta$ is defined as the product of α and β which can be written as

$$\begin{bmatrix} \alpha\beta \end{bmatrix} = \begin{bmatrix} \hat{\alpha} & \hat{\beta} \\ \hat{\beta}^* & \hat{\alpha}^* \end{bmatrix}$$

where $\tilde{\alpha}$ and $\tilde{\beta}$ are defined as

$$\begin{aligned}\tilde{\alpha} &= i \frac{(1 + \hat{Z}^2)}{2\hat{Z}} e^{ik_a l} \sin k_t s + e^{ik_a l} \cos k_t s \\ \tilde{\beta} &= -i \frac{(1 - \hat{Z}^2)}{2\hat{Z}} e^{-ik_a l} \sin k_t s ,\end{aligned}$$

where ξ is given by

$$\xi = \begin{bmatrix} e^{-k_a l} & 0 \\ 0 & e^{ik_a l} \end{bmatrix} ,$$

and where the matrix $\alpha^* \beta^*$ is the complex conjugate of $\alpha \beta$. The total photoacoustic amplitude at the ends of the structure is found by solving Eq. 28 for each allowed value of j ($j = 1, 2, 3, \dots, N$) and the contributions of the $A_0^{(j)}$'s to the total wave amplitude for the left-going wave summed, and those of the $B_{2N}^{(j)}$'s for the right-going wave summed.

Figure 4 shows the effect of changing the impedance factor $\hat{Z}$ on the frequency domain response of a layered structure with 5 absorbing layers. For $\hat{Z} = 1$, the response of the structure, as shown in Fig. 4(c), consists of a series of constructive and destructive interferences similar to those found for a single layer²⁵ which appear as rounded peaks, decreasing in amplitude with increasing modulation frequency. The effect of juxtaposing a number of layers with equal sonic transit times however is to sharpen the resonances. Figs. 4(d) and (e) show that an increase in the impedance parameter $\hat{Z}$ has the effect of both sharpening the resonances and suppressing the amplitudes of the side lobes. As shown in Figs. 4(a) and (b), when $\hat{Z}$ is decreased from unity, the decrease in the width of the photoacoustic resonances is accompanied by a sharpening of the cusps in the resonances.

Fig. 4: Left column: photoacoustic pressure versus dimensionless modulation frequency $\hat{q}$ for five absorbing layers with (a) $\hat{Z} = 0.2$, (b) $\hat{Z} = 0.5$, (c) $\hat{Z} = 1$, (d) $\hat{Z} = 2$, (e) $\hat{Z} = 5$. Right column: time domain photoacoustic pressure versus dimensionless time $\hat{\tau}$ for delta function deposition of heat with (f) $\hat{Z} = 0.2$, (g) $\hat{Z} = 0.5$, (h) $\hat{Z} = 1$, (i) $\hat{Z} = 2$, (j) $\hat{Z} = 5$.

The response of the layers in the time domain to a delta function heating pulse is found by Fourier transformation with respect to $\hat{q}$ of the frequency domain response as given by Eq. 28, and replacing κ_f with $i\bar{\alpha}\beta E_0 c_a^2 / 2C_P$ where E_0 is the fluence of the light source. Figures 4(f)-(j) show the results of fast Fourier transformation of the frequency domain curves in Figs. 4(a)-(e). The waveforms are plotted in terms of the retarded time $\hat{t}$ from the edge of the structure where $\hat{t} = (c_a/l)(t - \frac{2N-1}{c_i})$. The train of square waves seen in Fig 4(h) for $\hat{Z} = 1$ follows exactly from the theorem for one-dimensional sources given above, which states that for a delta function heat source in time, the deposition of heat in space is mapped directly into the time profile of the photoacoustic wave. It is noteworthy that the time domain curves in Figs. 4(f) and (g) with $\hat{Z} < 1$ are purely compressive, as are those generated by a single layer. Likewise, waves generated in layers where $\hat{Z} > 1$ consist of alternating compressions and rarefactions.

A somewhat unexpected feature of the time domain waveforms for a structure with two absorbing layers (clearly visible in the experimental waveforms below) is the low frequency sinusoidal wave that appears along with the series of square waves. We have considered a harmonic oscillator model for this response and find the dimensionless oscillation frequency $\hat{\omega}_0$ to be given by

$$\hat{\omega}_0 = \sqrt{\frac{2}{\hat{Z}}} . \quad (29)$$

Note that a number of pulses can be used to excite the layered source in order to produce a train of acoustic pulses. A heating function of the form

$$H(t) = \bar{\alpha} E_0 \sum_{m=1}^M \delta[t - \frac{2l}{c_a}(m-1)] , \quad (30)$$

where M is the total number of pulses describes a train of delta function pulses. The Fourier transform of Eq. 30,

$$H_f(\hat{q}) = \bar{\alpha} I_0 \sum_{m=1}^M \exp i\hat{q}(m-1), \quad (31)$$

Figure 5

Fig. 5: Photoacoustic pressure in arbitrary units versus dimensionless time $\hat{\tau}$ for a layered source with $\hat{Z} = 2$ and $N = 5$ irradiated by (a) 2, (b) 10, and (c) 25 delta function light pulses.

produces a series of square wave acoustic pulses. Another noteworthy property of a series of fluid layers as a generator of ultrasound is that when a finite diameter light beam is used for excitation, the acoustic beam becomes highly directional in space depending on the number of layers. The details of the calculation are not given here for the sake of brevity.

when multiplied with the frequency domain signal from Eq. 28 and Fourier transformed back into the time domain gives the photoacoustic pressure response to a series of M delta function light pulses. The result of exciting the series of layers with multiple laser pulses can be seen in Fig. 5 where M is varied. It can be seen that irradiation of a layered structure with a continuous train of laser pulses, as is commonly produced by a mode-locked, continuous laser,

C. Experiments and Results

Photoacoustic waves were generated by irradiating layered structures with the 532 nm output of a Q-switched Nd:YAG laser. The laser produced pulses that were roughly 8 ns long: the optical pulse duration could thus be considered to be short compared with the transit time of sound across the absorbing layers. In order to approximate a one-dimensional geometry, the laser beam was expanded to a diameter of about 6 cm, which was several times the layer thickness, and the transducer was placed as close to the structure as possible. Experiments were done with two layered sources. The first was constructed of layers of colored polymethylmethacrylate (Lucite) 2.75 mm thick separated by 1.54 mm thick spacers. The assembly was placed in a photoacoustic cell with dimensions 25 cm $\times$ 25 cm $\times$ 25 cm that was filled with water. The cell was equipped with Pyrex windows and a port for an acoustic transducer. The transducer was made from a piece of 52 or 110 μ m thick polyvinylidene fluoride film whose output was fed to a preamplifier with a gain of 40 (Comlinear, Inc. Model E103-1M). The signal was then displayed directly on an oscilloscope whose bandwidth was restricted to 20 MHz in order to provide a signal-to-noise ratio that was acceptable for single shot recording. To avoid a direct hit of the laser beam on the transducer, which is also pyroelectric, the laser was directed either at an angle to the axis of the layered source so as to miss the transducer, or was directed along the source axis and a thin film of metallized plastic or a thin Al foil was interposed between the transducer and the acoustic source. The film eliminated the rather large thermal signal which accompanies the small amount of absorption of the laser beam on the surface of the transducer, but has no discernible effect on the photoacoustic signal. The photoacoustic signals were recorded by irradiation of a series of absorbing Lucite layers in water with $N = 2, 3$, and 5. and compared with the calculated waveforms. The impedance factor for the Lucite-water combination is $\hat{Z} = 2.11$.

Figure 6

Fig. 6: Left column: calculated photoacoustic pressure in arbitrary units versus dimensionless time $\hat{\tau}$, for glass-methanol layers with (a) $N = 2$, (b) $N = 3$, (c) $N = 5$. Right column: Oscilloscope recordings of pressure versus time corresponding to the number of layers in the left column. The time and voltage scales in the experimental traces are $5 \mu\text{s}/\text{div}$ and $20 \text{ mV}/\text{div}$.

D. Discussion

The experimental waveforms differ from those predicted by theory for several reasons. Among these are irradiation of only a bounded area of the structure, a finite response time of the transducer and electronics, viscous damping of the high frequency components of the waveform, imperfect alignment of the layers within the source and with respect to the transducer, and the finite pulsewidth of the laser. The relative importance of these effects is

Experimentally recorded and calculated waveforms for absorbing glass in methanol with $\hat{Z} = 16.0$ are shown in Fig. 6. The extensive ringing in the waveforms in Fig. 6 is a consequence of the high value of $\hat{Z}$ in the glass-methanol source. The low frequency sinusoidal component of the photoacoustic pressure discussed above is clearly visible in the experimental traces in Fig. 6 as well.

still under investigation.

The layered photoacoustic source described here has properties shared with a number of devices in several areas of physics that make use of coherent addition of wave amplitudes. Such devices include interference filters, diffraction gratings, and echelle gratings commonly employed in optical science^{31,32}, layered acoustic filters in the field of classical acoustics³³⁻³⁵, superlattices in solid state physics³⁶⁻³⁸, and photonic crystals in quantum electronics³⁹. *The important difference between the photoacoustic source discussed here and these examples is that the photoacoustic source is an active device.* A second, more subtle difference between passive devices such as an acoustic filter or a dielectric interference filter and the photoacoustic structure is that in an active device, an asymmetry is introduced into the response when only one of the two materials that makes up the structure is the source. The curves shown in Fig. 4, for instance, have different properties for $\hat{Z} < 1$ and $\hat{Z} > 1$; an acoustic filter or an optical interference filter displays no significant difference in its properties when the order of the layers is interchanged. In addition, unlike passive devices, the photoacoustic source, by virtue of the internal generation of waves, displays interference phenomena even when the layers have identical acoustic characteristics as evidenced by the response in Fig. 4(c) where $\hat{Z} = 1$.

A further point of interest is the band structure of a layered acoustic source. It is possible to consider an infinitely long structure of layers with alternating acoustic properties where the acoustic pressure is described by Bloch wavefunctions. Analysis of the wavefunctions gives a dispersion relation for the acoustic waves which shows band gaps directly analogous to that found for electronic wavefunctions as described by the familiar Kronig-Penny model⁴¹. The details of a model describing the properties of the acoustic structure have been given previously^{33-37,42} in conjunction with acoustic filters and with superlattices. Two points of note are, first, that the acoustic resonances shown in Fig. 4 always lie outside a bandgap and thus can always be supported by the structure, and second, that the acoustic bandgaps depend on both the density and sound speed ratios between the layers so that the gap

depends on two parameters as opposed to only one in the Kronig-Penny model.

III. PHOTOACOUSTIC WAVES FROM LAYERS

A. Introduction

In this section the properties of the photoacoustic effect generated by an optically thin fluid as well as an isotropic solid layer in contact with a transparent fluid are derived. We show that an analytical technique is possible where the information contained in the time profile of a photoacoustic wave from short pulse excitation of an infinite layer can be used to determine the thickness and density of the layer, provided the sound speed in the layer and the acoustic properties of the surrounding fluid are known. This work has been done with an objective of determining details of the photoacoustic effect generated by irradiation of an optically thin layer with a view toward distinguishing the characteristics of photoacoustic radiation generated by a fluid layer from those produced by its counterparts in two and three dimensions, namely, an optically thin fluid sphere and cylinder. In Section B, the photoacoustic wave generated by modulated, continuous wave radiation is determined for an absorbing layer surrounded by single transparent fluid whose density and sound speed differ from those of the absorbing layer. In addition, the problem of an absorbing layer in contact with two other fluids, each having different acoustic properties is solved. The frequency domain solutions are used to derive expressions for the acoustic pressure generated by short pulse irradiation of the layers. Section C describes determination of the angular distribution of acoustic radiation in the far field from a fluid layer irradiated by a finite diameter light beam. In Section D, the wave equation for an isotropic solid is used to determine the photoacoustic effect from a solid slab. Experimental results are given in Section E for fluid and solid layers irradiated with modulated continuous radiation from an Ar ion laser and with the pulsed radiation from a Nd:YAG laser. Measurements of the angular distribution of radiation from a continuously irradiated fluid layer are reported as well. The derivations are abbreviated here; full details of the calculations can be found in *J. Acoust. Soc Am.* 93, 1417 (1993).

B. Fluid Layers

1. Absorbing Layer with One Surrounding Fluid

Consider an optically thin fluid layer of thickness l in the Z direction and of infinite extent in the X and Y directions with a density ρ_l and sound speed c_l in contact with a second transparent fluid with density ρ_f and sound speed c_f . By optically thin it is meant that the optical density of the layer is sufficiently low so that absorption of the optical beam produces an essentially uniform heating of the layer. If the layer is irradiated by a light beam with intensity I_0 modulated at a frequency ω , the alternating component of the heating function is described by

$$H = \bar{\alpha} I_0 e^{-i\omega t} \quad (32)$$

where $\bar{\alpha}$ is the optical absorption coefficient of the layer. Since the layer is heated uniformly, the optical beam generates a uniform pressure inside the layer. The spatial derivative in the wave equation is therefore zero, and the solution to the homogeneous wave equation is found by direct integration to be

$$p = \rho_l c_l^2 (i \bar{\alpha} \beta I_0 / \rho_l \omega C_P) e^{-i\omega t} , \quad (33)$$

which has been written so that the factor in parentheses is dimensionless. Since there are discontinuities in the acoustic impedance at the interfaces between the two fluids (taken at the points $z = \pm l/2$), solutions for the pressure inside the layer p , to the right of the layer p_R (along the positive z axis) and to the left of the layer p_L must also include solutions to the inhomogeneous wave equation, which in the case of a one-dimensional geometry are clearly plane waves. The photoacoustic pressures (for unit driving pressure) must be of the form

$$\begin{aligned} p &= [1 + \hat{P}_1 e^{i k_l z} + \hat{P}_2 e^{-i k_l z}] e^{-i\omega t} \\ p_R &= \hat{P}_R e^{i[k_f(z-l/2)-\omega t]} \\ p_L &= \hat{P}_L e^{-i[k_f(z+l/2)+\omega t]} , \end{aligned} \quad (34)$$

where k_i and k_f are the wavevectors inside and outside the layer, respectively, and the $\hat{P}$'s are complex amplitudes.

By equating the acoustic pressure and fluid acceleration for the two waves at the interface of the fluids and through use of identities based on symmetry, the pressure amplitude factors can be found. Solution of the two equations is straightforward and gives the photoacoustic pressures as

$$\begin{aligned} p_R &= i\lambda \left[\frac{\text{sinc } \hat{q}}{\sin \hat{q} + i \hat{\rho} \hat{c} \cos \hat{q}} \right] e^{-i \hat{q} \hat{\tau}} \\ p &= \frac{i\lambda}{\hat{q}} \left[1 - \frac{i \hat{\rho} \hat{c} \cos \hat{q} \hat{z}}{\sin \hat{q} + i \hat{\rho} \hat{c} \cos \hat{q}} \right] e^{-i \hat{q} \hat{t}} \end{aligned} \quad (35)$$

where

$$\hat{\rho} = \rho_i / \rho_f \quad \hat{c} = c_i / c_f$$

and where an amplitude factor λ , a dimensionless frequency parameter $\hat{q}$, a dimensionless retarded time from the right interface of the layer $\hat{\tau}$, and a dimensionless time $\hat{t}$, have been defined as

$$\lambda = \frac{\bar{\alpha} \beta I_0 c_i l}{2 C_P} \quad \hat{q} = \frac{\omega l}{2 c_i} \quad \hat{\tau} = \frac{2 c_i}{l} \left(t - \frac{z - l/2}{c_f} \right) \quad \hat{t} = \frac{2 c_i}{l} t \quad \hat{z} = \frac{2 z}{l}$$

and where the usual definition of the *sinc* function ($\text{sinc } \hat{q} = \sin \hat{q} / \hat{q}$) has been used.

The response of the layer to a delta function heating pulse in time is found by determining a pressure spectral density from Eq. 35 which is then multiplied by the Fourier transform of the intensity of the exciting radiation ($E_0/2\pi$ in the case of a delta function) and integrated over all frequencies ($d\omega/2\pi$). The time domain response of the infinite layer thus becomes

$$p_R(t) = \frac{i\kappa}{2\pi} \int_{-\infty}^{\infty} \frac{\sin \hat{q}}{\hat{q}} \frac{e^{-i \hat{q} \hat{\tau}}}{[\sin \hat{q} + i \hat{\rho} \hat{c} \cos \hat{q}]} d\hat{q} \quad (36)$$

where

$$\kappa = \frac{\bar{\alpha} \beta E_0 c_i^2}{C_P}.$$

It is of note that Eq. 36 gives the acoustic pressure as a function of only two parameters, the retarded time and the product $\hat{\rho} \hat{c}$. Although evaluation of Eq. 36 through the fast Fourier

transform is straightforward, a closed form solution for p_R can be easily found. First, Eq. 36 is rewritten as the double integral, which can be expanded to give,

$$p_R(t) = \frac{\kappa}{(1 + \hat{\rho} \hat{c})} [\tilde{\Theta}_{0,2}(\hat{\tau}) + \xi \tilde{\Theta}_{2,4}(\hat{\tau}) + \xi^2 \tilde{\Theta}_{4,6}(\hat{\tau}) + \dots] \quad (37)$$

where

$$\xi = (1 - \hat{\rho} \hat{c}) / (1 + \hat{\rho} \hat{c}) .$$

and where the square wave function $\tilde{\Theta}_{i,j}(\hat{\tau})$ is defined as unity for $i \leq \hat{\tau} \leq j$ and zero otherwise. The photoacoustic wave described by Eq. 37 is a compressive staircase wave for $\xi \geq 0$ ($\hat{\rho} \hat{c} \leq 1$), a series of alternating compressions and rarefactions for $\xi \leq 0$ ($\hat{\rho} \hat{c} \geq 1$), and a single compressive square wave for $\xi = 0$ ($\hat{\rho} \hat{c} = 1$).

When the product $\hat{\rho} \hat{c}$ is much less than one, it can be shown that

$$p_R(t) = \kappa u(\hat{\tau}) e^{-\hat{\rho} \hat{c} \hat{\tau}} \quad (38)$$

where $u(\hat{\tau})$ is the Heaviside function. Clearly, the exponential decay in Eq. 38 is an approximation to the discontinuous staircase wave given in Eq. 37.

In the long pulse limit where the duration of the optical radiation pulse far exceeds the transit time of sound across the layer the photoacoustic pressure can be formally written

$$p_R(\hat{\tau}_0) = \frac{\kappa}{\hat{\rho} \hat{c}} \delta(\hat{\tau}_0) , \quad (39)$$

where $\hat{\tau}_0$ has been substituted for $\hat{\tau}$ in Eq. 39 (where $\hat{\tau}_0$ is the retarded time from the origin) since the two retarded times become indistinguishable in the limit of long exciting pulses.

Equation 39 states simply that the layer responds equally to all frequencies in the low frequency limit. On convolution of Eq. 39 with a function that describes the intensity of a long exciting optical pulse, it is clear that *for long optical pulses the photoacoustic waveform must be proportional to the intensity of the optical radiation pulse evaluated at the retarded time*. This dependence is in agreement with the conclusions given in Section I for the case of identical acoustic media.

2. One Absorbing Layer-Two Surrounding Fluids

In the case of a single layer, it is possible to have the absorbing layer in contact with two different transparent surrounding fluids where the acoustic properties of all three fluids differ. The results of a rather lengthy calculation give the time domain photoacoustic pressure for the right-going wave as

$$p_R(\hat{\tau}_R) = \frac{i \kappa}{4\pi} T_R \int_{-\infty}^{\infty} \left[\frac{1 - \hat{\rho}_L \hat{c}_L T_L e^{2i\hat{q}} - R_L e^{4i\hat{q}}}{\hat{q}(1 - R_L R_R e^{4i\hat{q}})} \right] e^{-i\hat{q}\hat{\tau}_R} d\hat{q} . \quad (40)$$

where the transmission and reflection amplitude coefficients are given as

$$\begin{aligned} T_R &= \frac{2}{1 + \hat{\rho}_R \hat{c}_R} & T_L &= \frac{2}{1 + \hat{\rho}_L \hat{c}_L} \\ R_R &= \frac{1 - \hat{\rho}_R \hat{c}_R}{1 + \hat{\rho}_R \hat{c}_R} & R_L &= \frac{1 - \hat{\rho}_L \hat{c}_L}{1 + \hat{\rho}_L \hat{c}_L} . \end{aligned}$$

and where

$$\hat{\tau}_R = \frac{2 c_l}{l} \left[t - \frac{z - l/2}{c_R} \right] \quad \hat{\tau}_L = \frac{2 c_l}{l} \left[t + \frac{z + l/2}{c_L} \right] .$$

Owing to symmetry it is clear that $p_L(\hat{\tau}_L)$, the pressure in the left-going wave can be obtained from Eq. 40 by interchanging L and R . The photoacoustic pressure can be found in closed form by writing Eq. 40 as a sum of two, two-dimensional integrals, expanding the denominators, and evaluating each term in a manner parallel to the calculation given in Section A, which gives

$$p_R(\hat{\tau}_R) = \frac{\kappa}{2} T_R \sum_{n=0}^{\infty} (R_L R_R)^n [\hat{\Theta}_{4n,4n+2}(\hat{\tau}_R) + R_L \hat{\Theta}_{4n+2,4n+4}(\hat{\tau}_R)] \quad (41)$$

Again, $p_L(\hat{\tau}_L)$ can be found from Eq. 41 by interchanging the R and L subscripts.

C. Fluid Layer Irradiated by Finite Diameter Beam

Consider an absorbing fluid layer of thickness l located symmetrically in the XY plane at the origin, and surrounded by a transparent fluid of infinite extent with identical acoustic properties. Also, let the layer be optically thin so that the heating function is described by Eq. 32, but with the modification that the region of heating extend only over a circular region in the XY plane of radius a . A formal solution to the wave equation, written in terms of the velocity potential ϕ_v , can be written as an integral over the source volume,

$$\phi_v(\mathbf{x}, t) = \frac{-\beta}{4\pi \rho_f C_p} \int \frac{H\left(\mathbf{x}', t - \frac{|\mathbf{x}-\mathbf{x}'|}{c_f}\right)}{|\mathbf{x}-\mathbf{x}'|} d\mathbf{x}' , \quad (42)$$

where the primed quantities refer to the source. Calculation of the photoacoustic pressure is lengthy; only the result

$$p = \frac{-i \lambda}{\hat{r}} \hat{a}^2 \hat{q} \operatorname{sinc}(\hat{q} \cos \vartheta) \left[\frac{J_1(\hat{q} \hat{a} \sin \vartheta)}{\hat{q} \hat{a} \sin \vartheta} \right] e^{-i \hat{q} \hat{\tau}_0} , \quad (43)$$

where $\hat{a} = 2a/l$, $\hat{r} = 2r/l$, and $\hat{\tau}_0$ has been defined in three dimensions as $\hat{\tau}_0 = (2c_f/l)[t-r/c_f]$ is given here.

D. Radiation From a Solid Layer

Consider an infinite isotropic solid slab of thickness l located with its center at the origin and of infinite extent in the XY plane. Let the slab be in contact with a single fluid on both sides. If the slab is optically thin, and the light beam uniformly irradiates the slab then all displacements of the slab must be in the Z direction. The wave equation⁴³ for the displacement vector $\mathbf{u}$ for longitudinal waves in an isotropic solid can be written in terms of a wave equation for the displacement potential Φ

$$\left(\nabla^2 - \frac{1}{c_l^2} \frac{\partial^2}{\partial t^2} \right) \Phi = \frac{K \beta}{\rho_l c_l^2} \theta \quad (44)$$

where the displacement and pressure are determined by

$$p = -\rho_l \frac{\partial^2 \Phi}{\partial t^2} \quad \text{and} \quad \mathbf{u} = \nabla \Phi .$$

and where θ is the temperature increase above ambient caused by absorption of light, K is the bulk modulus, ρ_l is the density, and c_l is the longitudinal sound speed. When the influence of the displacement on the temperature can be neglected⁴³, the equation for θ reduces to

$$\left(\nabla^2 - \frac{1}{\alpha_c} \frac{\partial}{\partial t} \right) \theta = - \frac{H}{\alpha_c \rho_l C_P} \quad (45)$$

where α_c is the thermal diffusivity.

The solution to Eq. 45 is lengthy so only the results will be reproduced here. We find the displacement potentials to be given by

$$\begin{aligned} \Phi_f &= \frac{i \alpha \beta K I_0 (l/2)^3}{\rho_f \rho_l c_l^3 C_P \hat{q}^3} \hat{A}_f e^{-i \hat{q} \hat{r}} \\ \Phi_l &= \frac{i \alpha \beta K I_0 (l/2)^3}{\rho_l^2 c_l^3 C_P \hat{q}^3} (1 + \hat{A}_l \cos \hat{q} \hat{z}) e^{-i \hat{q} \hat{r}} , \end{aligned} \quad (46)$$

which gives the pressure in the fluid as

$$p = \frac{i \alpha \beta K I_0 (l/2)}{\rho_l c_l C_P \hat{q}} \hat{A}_f e^{-i \hat{q} \hat{r}} . \quad (47)$$

It is thus clear from Eqs. 46 and 47, that optically thin, infinite isotropic solid layers and fluid layers act identically insofar as production of photoacoustic waves is concerned, provided the optical beam gives a uniform heating over the entire volume of the layer. On the other hand, for a finite area radiation beam or a solid slab of finite dimensions, several different kinds of acoustic waves can be excited by the light beam; simple displacement in one dimension is not expected.

E. Experiments and Results

Experiments with modulated continuous wave radiation were done using the 514.5 nm output of an Ar ion laser operating in the TEM_{00} mode. The laser beam was amplitude modulated with an electrooptic modulator, passed through a beam expander and a circular aperture, and directed into a square photoacoustic cell with dimensions of 30 cm on a side. The aperture selected only the central portion of the laser beam so as to obtain a spatial intensity profile as nearly uniform as possible. The power in the laser beam after passing through the electrooptic modulator and its associated optics was on the order of 1 W. In the experiments where the frequency response of the layers was measured, a PVDF transducer was mounted in the side of the cell; in the experiments where the angular distribution of radiation was measured, the PVDF transducer was mounted so that it could be rotated about a direction perpendicular to the direction of propagation of the laser beam. In order to avoid the spurious photothermal signal from directly hitting the transducer with the laser beam, a piece of PVDF film with a metal coating was placed between the fluid layer and the transducer. The film effectively reflected the optical beam, and introduced a negligible attenuation in the acoustic wave. Below 200 kHz a spurious signal from the film could be detected, thus data below this frequency were not recorded. The output of the transducer was fed to the 1 M Ω input of an oscilloscope (Tektronix Inc. Model 3765) which was used here as a preamplifier (with a gain of 10) owing to its flat frequency response and low noise. The output of the oscilloscope was fed to the Comlinear amplifier and then to a high frequency lock-in amplifier (EGG PAR Inc. Model 5202).

The continuous wave experiments were done by irradiating either an optically thin sheet of polymethylmethacrylate (Lucite brand) or fluid layers containing a small amount of dye. The layers were made by confining the dyed liquid between two 13 μ m thick transparent polyvinylidene chloride films (Dow Chemical Corp. Saran Wrap) mounted on a metal support. The area of the layer illuminated by the laser beam, a circle 3.5 cm in

Figure 7

Fig. 7: Magnitude of the dimensionless photoacoustic pressure p_R versus frequency for a layer irradiated by modulated optical radiation with (A) $\hat{\rho}\hat{c} = 1$, (B) $\hat{\rho}\hat{c} = 2.11$, and (C) $\hat{\rho}\hat{c} = 3.58$. The experimental data were taken with (A) a 1.5 mm dyed methanol layer surrounded by methanol, (B) a 2.75 mm thick layer of absorbing Lucite surrounded by water, and (C) the same absorbing layer of Lucite surrounded by methanol.

diameter, was made as large as possible, and the distance between the layer and transducer was made as small as possible in order to approximate the one dimensional solutions given above. The data shown in Fig 7 were taken at a series discrete frequencies and averaged over five experiments.

The angular distribution of the acoustic radiation was determined by measuring the output voltage on the lock-in amplifier at a constant modulation frequency of 460 kHz as

Figure 8

Fig. 8: Photoacoustic pressure P (in arbitrary units) versus angle from the propagation direction of the laser beam θ (calculated from Eq. 26) with (A) $\hat{q} = \pi/2$ and $\hat{a} = 8.1$ and (B) $\hat{q} = \pi/2$ and $\hat{a} = 14.4$. Both the experimental data as well as the theoretical curves have been normalized. The data were taken with a 1.6 mm thick water layer irradiated by a 1.2 cm diameter laser beam and (B) a 2.3 cm diameter laser beam. The layer had an absorbance of 2.5 cm^{-1} at 514.5 nm.

F. Discussion

In the case of a plane wave striking a fluid interface at perpendicular incidence it is well known^{44,45} that the amplitude reflection R and transmission T coefficients are given by

$$R = \frac{1 - \hat{\rho}\hat{c}}{1 + \hat{\rho}\hat{c}} \quad \text{and} \quad T = \frac{2}{1 + \hat{\rho}\hat{c}} . \quad (48)$$

It is thus possible to write the expression for the pressure generated by the fluid layer, Eq. 48, in terms of T and R as

$$p = \frac{\kappa}{2\pi} T \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} \text{sinc } \hat{q} (e^{2i\hat{q}} R)^n e^{-i\hat{q}(\hat{r}-1)} d\hat{q} . \quad (49)$$

the the position of the PVDF transducer relative to the incident laser beam in the photoacoustic cell was varied. Data are shown in Fig 8. The angle θ was determined with an accuracy of approximately ± 2 degrees.

We have also recorded a number of photoacoustic waves from fluid layers surrounded by two different fluids corresponding to the theory worked out in Section B above. The results of the experiments have been published, but are are not reproduced in this report.

The production of the photoacoustic wave can be viewed as a process where the displacement corresponding to the photoacoustic wave is uniform about the plane $x = 0$. The $n = 0$ term describes the emission of a single compressive wave with the amplitude governed by the *sinc* $\hat{q}$ integral, but altered in amplitude by the transmission coefficient. The $n = 1$ term can be viewed as arising from the original left-going compressive wave that has propagated to the left fluid interface, where it has been altered in amplitude by a reflection, and then transmitted through the right interface. This originally left-going wave appears at a dimensionless retarded time $\hat{t} - 3$, which represents a time delay relative to the $n = 0$ pulse of c/l . Each successive wave emanating from the layer to the right arises from the original left- and right-going waves, but with amplitudes altered by reflections at the left and right faces and arrival times at the field point determined by the traversal time across the layer. This process of reflection and time delay is clearly analogous to that found for photoacoustic waves radiated from a sphere given by Eq. 30 in Ref. 21.

Analogues of the results given in this paper for the photoacoustic effect can be found in other fields as well. For instance, the angular dependence of the far field photoacoustic pressure described by J_1 is seen to be analogous to that for a harmonically vibrating piston in a rigid wall⁴⁵, or the electric field amplitude of a plane wave passing through a circular aperture in the Fraunhofer approximation⁴⁶. This result is a purely geometric property of waves.

IV PHOTOACOUSTIC WAVES FROM CYLINDERS

A. Introduction

In this section, solutions to the wave equation in two dimensions are given for the problem of an infinitely long, optically thin, fluid cylinder in contact with a second, transparent fluid. The two fluids are considered to have different sound speeds and densities. In Section B, a frequency domain solution to the wave equation is found using the acoustic conservation equations at the interface between the two fluids. The solution shows the effects of constructive and destructive interference in the waves emitted by the cylinder. The radial

eigenfrequencies of a cylinder and low frequency response of the cylinder are found from the solution for the photoacoustic pressure. In Section B, an expression for the acoustic pressure as a function of time is given for short pulse excitation of the cylinder. Because of the discontinuities in acoustic properties at the interface between the two fluids, the solution for the pressure shows the reflection and transmission phenomena seen in the case of a droplet and a fluid layer. The response of the cylinder to a long light pulse is found from the time domain expression for the pressure. By taking the low density limit of the integral expression for the photoacoustic pressure, the response of a cylindrical gas bubble is found. Section C gives the results of experiments carried out with fluid cylinders excited by the frequency doubled output of a Nd:YAG laser. The Discussion Section shows how the time dependence of the photoacoustic wave from a cylindrical source can be explained in terms of multiple reflections of a cylindrical wave at a cylindrical interface. The results given in this paper show that an analytical technique is possible whereby an accurate recording of the time profile of a photoacoustic wave generated by laser irradiation of an optically thin cylinder can be used to determine acoustic parameters characteristic of the cylinder. Abbreviated derivations are given here. For further details see "Photoacoustic Waves Generated by Absorption of Laser Radiation in Optically Thin Cylinders" (submitted to *Ultrasonics*).

B. Frequency Domain Solution

Consider an infinitely long, optically thin cylinder with a sound speed c_c and a density ρ_c surrounded by a second, transparent fluid with a sound speed c_f and a density ρ_f . If the cylinder is irradiated with an optical beam with intensity I_0 modulated at a frequency ω , then the alternating component of the heat deposited in the cylinder per unit volume and time can be written as in the case of the layer as

$$H = \bar{\alpha} I_0 e^{-i\omega t} , \quad (50)$$

where $\bar{\alpha}$ is the optical absorption coefficient of the cylinder and t is the time. This is of course the same heating function used as in the case of the infinite layer given above. When

the effects of heat conduction and viscosity can be ignored, the photoacoustic pressure p is given as a solution to the wave equation, Eq. 18. The cylinder is considered to be uniformly irradiated so that the solution to the inhomogeneous wave equation for the driving pressure with the heating function H given by Eq. 50 is

$$p = \rho_c c_c^2 (i \bar{\alpha} \beta I_0 / \rho_c \omega C_P) e^{-i\omega t}, \quad (51)$$

which has been written with a dimensionless factor in parentheses. The solution for the total acoustic pressure inside the cylinder p_c and that in the surrounding fluid p_f for unit driving pressure can be written for unit driving pressure as

$$\begin{aligned} p_c &= [1 + \hat{P}_c J_0(k_c r)] e^{-i\omega t} \\ p_f &= \hat{P}_f H_0^{(1)}(k_f r) e^{-i\omega t}, \end{aligned} \quad (52)$$

where $\hat{P}_c$ and $\hat{P}_f$ are complex constants. The complex amplitudes can be found as above using the laws for the continuity of pressure and acceleration across the interface.

The solution of the wave equation gives the photoacoustic pressures inside and outside of the cylinder as

$$p_c(\hat{q}) = \frac{i \lambda}{\hat{q}} \left\{ 1 + \frac{\hat{\rho} \hat{c} J_0(\hat{q} \hat{r}) H_1^{(1)}(\hat{c} \hat{q})}{[J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q}) - \hat{\rho} \hat{c} J_0(\hat{q}) H_1^{(1)}(\hat{c} \hat{q})]} \right\} e^{-i\hat{q}\hat{t}} \quad (53)$$

$$p_f(\hat{q}) = \frac{i \lambda}{\hat{q}} \left[\frac{J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q} \hat{r})}{[J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q}) - \hat{\rho} \hat{c} J_0(\hat{q}) H_1^{(1)}(\hat{c} \hat{q})]} \right] e^{-i\hat{q}\hat{t}}, \quad (54)$$

where an amplitude factor λ , a dimensionless frequency $\hat{q}$, and a dimensionless time $\hat{t}$ have been defined as

$$\lambda = \frac{\bar{\alpha} \beta I_0 c_c a}{C_P}, \quad \hat{q} = \frac{\omega a}{c_c}, \quad \text{and} \quad \hat{t} = \frac{c_c t}{a},$$

and where, as before, a dimensionless density parameter $\hat{\rho}$ and a dimensionless sound speed parameter $\hat{c}$ have been defined as

$$\hat{\rho} = \rho_c / \rho_f \quad \text{and} \quad \hat{c} = c_c / c_f.$$

It can be seen that outside the cylinder there is both a near and far field depending on the value of the product $\hat{c}\hat{q}\hat{r}$. To determine the acoustic pressure in the far field, the zero order

Hankel function in the expression for p_f can be expanded⁴⁷ for large values of the argument as $H_0^{(1)}(z) \cong (2/\pi z)^{1/2} \exp(iz - i\pi/4)$ giving

$$p_f(\hat{q}) = \lambda \sqrt{\frac{2i}{\pi \hat{c} q^3 \hat{r}}} \left[\frac{J_1(\hat{q})}{J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q}) - \hat{\rho} \hat{c} J_0(\hat{q}) H_1^{(1)}(\hat{c} \hat{q})} \right] e^{-i\hat{q}\hat{\tau}_0}, \quad (55)$$

which can be expressed either in terms of the retarded time from the z axis $\hat{\tau}_0$ or the retarded time from the edge of the cylinder $\hat{\tau}$ which are given by

$$\hat{\tau}_0 = \frac{c_c}{a} \left(t - \frac{r}{c_f} \right) \quad \text{and} \quad \hat{\tau} = \frac{c_c}{a} \left(t - \frac{r-a}{c_f} \right).$$

Thus, only in the far field is the expected $r^{-1/2}$ dependence of the pressure amplitude found.

Of further note is the photoacoustic pressure when the cylinder and the surrounding fluid have identical sound speeds and densities. Through use of the expression for the Wronskian of Bessel functions of the first and second kind⁴⁷, it is easy to show that $J_1(z)H_0^{(1)}(z) - J_0(z)H_1^{(1)}(z) = 2i/\pi z$. The expression for the acoustic pressure when $\hat{\rho} = \hat{c} = 1$ thus reduces to

$$p_f(\hat{q}) = \frac{\lambda \pi}{2} [J_1(\hat{q}) H_0^{(1)}(\hat{q} \hat{r})] e^{-i\hat{q}\hat{\tau}}, \quad (56)$$

which in the far field becomes

$$p_f(\hat{q}) = \lambda \sqrt{\frac{\pi}{2 i \hat{r}}} \left[\frac{J_1(\hat{q})}{\sqrt{\hat{q}}} \right] e^{-i\hat{q}\hat{\tau}_0}. \quad (57)$$

For $\hat{\rho}$ and $\hat{c}$ equal to unity, the photoacoustic pressure show rounded maxima, and minima caused by constructive and destructive interference as would be found in an integration of the contributions of the source amplitudes over the volume of the cylinder. The general appearance of the amplitude curve is qualitatively the same as that found for the case of the fluid sphere as discussed previously; it is however the shape of the peaks and the positions of the nulls along the $\hat{q}$ axis that distinguish the response of one body from the other.

Figure 9

Fig 9: Magnitude and phase of the photacoustic pressure from Eq. 7 with $\hat{\rho} = 10$ and $\hat{c} = 1$, $\lambda = 1$, and $\hat{r} = 10$.

When the value of $\hat{\rho}$ becomes large, the rounded maxima appear as a series of sharp peaks as shown in Fig. 9. Further calculations show that the peaks become progressively sharper as $\hat{\rho}$ gets larger. For large values of $\hat{\rho}$ the cylinder can be considered as executing radial vibrations in contact with a low density gas. As the gas density decreases, its ability to react against the motion of the cylinder is diminished so that in the limit of large $\hat{\rho}$, the response curve of the cylinder shows a series of maxima at the radial eigenfrequencies. A plot of amplitude versus modulation frequency for small values of $\hat{\rho}$ possesses as its salient feature a single peak at low frequencies. As $\hat{\rho}$ is reduced, the position of the peak is found to

shift to smaller values of $\hat{q}$, and to decrease in width. A single peak in the frequency domain plot at a small value of $\hat{q}$ is, of course, characteristic in the time domain of a low frequency oscillation of the cylinder.

For low modulation frequencies Eq. 54 can be expanded using the first order approximations to the Bessel and Hankel functions $J_0(z) \cong 1$, $J_1(z) \cong z/2$, $H_0^{(1)}(z) \cong (2i/\pi) \ln z$, and $H_1^{(1)}(z) \cong -(2i/\pi z)$ to give

$$p_f(\hat{q}) = \frac{\lambda \pi}{2} \left[\frac{\hat{q} H_0^{(1)}(\hat{c} \hat{q} \hat{r})}{\hat{q}^2 \ln \hat{c} \hat{q} + 2\hat{\rho}} \right] e^{-i \hat{q} \hat{t}} . \quad (58)$$

Provided the condition $\hat{q}^2 \ln \hat{c} \hat{q} \ll 2\hat{\rho}$ is fulfilled, the photoacoustic pressure in the far field can be approximated as

$$p_f = \frac{\lambda}{\hat{\rho}} \sqrt{\frac{\pi}{8 \hat{c} \hat{r}}} \hat{q}^{\frac{1}{2}} e^{-i \hat{q} \hat{t}_0 - i \pi/4} , \quad (59)$$

which shows the pressure amplitude to increase as $\hat{q}^{\frac{1}{2}}$ and to be out of phase with the optical modulation by a factor of $\pi/4$. The low frequency response given by Eq. 59 differs from that for an infinite fluid layer where the pressure is in phase with the modulation and is independent of the modulation frequency, and that for a fluid sphere where the pressure is out of phase with the optical modulation by a factor of $\pi/2$ and increases in amplitude linearly with the modulation frequency. *The differences in the low frequency response of the layer, the cylinder, and the sphere are geometric characteristics of these bodies and account for their different responses to long optical pulses.*

C. Time Domain Response

1. Arbitrary Densities and Sound Speeds

The response of the cylinder to a delta function heating pulse in time can be found by Fourier transformation of Eq. 54 which gives the photoacoustic pressure in the fluid as

$$p_f(\hat{t}) = \frac{i\kappa}{2\pi} \int_{-\infty}^{\infty} \frac{J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q} \hat{r}) e^{-i \hat{q} \hat{t}}}{\hat{q} [J_1(\hat{q}) H_0^{(1)}(\hat{c} \hat{q}) - \hat{\rho} \hat{c} J_0(\hat{q}) H_1^{(1)}(\hat{c} \hat{q})]} d\hat{q} , \quad (60)$$

where the amplitude factor κ is defined as above and where E_0 is the energy per area in the laser pulse. In the far field, the photoacoustic pressure can be expressed in terms of the

retarded time from the perimeter of the cylinder as

$$p_f(\hat{r}) = \frac{\kappa}{2\pi} \sqrt{\frac{2i}{\pi \hat{c} \hat{r}}} \int_{-\infty}^{\infty} \frac{J_1(\hat{q}) e^{-i\hat{q}(\hat{r}-\hat{c})}}{\hat{q}^{\frac{3}{2}} [J_1(\hat{q}) H_0^{(1)}(\hat{c}\hat{q}) - \hat{\rho} \hat{c} J_0(\hat{q}) H_1^{(1)}(\hat{c}\hat{q})]} d\hat{q} . \quad (61)$$

The theoretical traces used for comparison with the experimental waveforms described below are calculated from Eq. 60 using the fast Fourier transform. It is noteworthy that the periodic form of the integrand in Eq. 60, and the analogous expressions found for the sphere or layer as well, permits a rapid and accurate numerical evaluation of the pressure using the fast Fourier transform algorithm.

2. Identical Densities and Sound Speeds

When the density and sound speed parameters are equal to one, it is possible to evaluate the far field photoacoustic pressure in closed form. First, Eq. 60 is written as

$$p_f(\hat{r}) = \frac{\kappa}{2\pi} \sqrt{\frac{\pi}{\hat{r}}} \int_0^{\infty} \frac{J_1(\hat{q})}{\sqrt{\hat{q}}} [\cos \hat{q}(\hat{r}-1) - \sin \hat{q}(\hat{r}-1)] d\hat{q} , \quad (62)$$

which can be integrated to give the photoacoustic pressure in terms of hypergeometric functions as

$$\begin{aligned} p_f(\hat{r}) &= \frac{\kappa}{2\pi} \sqrt{\frac{\pi}{\hat{r}}} \left[\frac{\Gamma(\frac{3}{4})}{2^{\frac{1}{2}} \Gamma(\frac{5}{4})} {}_2F_1\left(\frac{3}{4}, -\frac{1}{4}, \frac{1}{2}; (\hat{r}-1)^2\right) \right. \\ &\quad \left. - \frac{2^{\frac{1}{2}} \Gamma(\frac{5}{4})}{\Gamma(\frac{3}{4})} (\hat{r}-1) {}_2F_1\left(\frac{5}{4}, \frac{1}{4}, \frac{3}{2}; (\hat{r}-1)^2\right) \right] \\ &\hspace{15em} 0 < \hat{r} < 2 \\ p_f(\hat{r}) &= -\frac{\kappa}{2\pi} \sqrt{\frac{\pi}{\hat{r}}} \frac{\Gamma(\frac{3}{2})}{2^{\frac{3}{2}} \Gamma(2)} (\hat{r}-1)^{-\frac{3}{2}} \left[{}_2F_1\left(\frac{3}{4}, \frac{5}{4}, 2; (\hat{r}-1)^{-2}\right) \right. \\ &\quad \left. + {}_2F_1\left(\frac{5}{4}, \frac{3}{4}, 2; (\hat{r}-1)^{-2}\right) \right] \\ &\hspace{15em} \hat{r} > 2 . \end{aligned} \quad (63)$$

These equations show that the unusual shape of the two dimensional waveforms seen in the experiments are actually hypergeometric. Evaluation of Eq. 63 gives the curve previously obtained from Eq. 60 using the numerical Fourier transform, with $\hat{\rho} = \hat{c} = 1$ or using the two-dimensional mapping given above. The solution given by Eq. 63 shows that the

photoacoustic pressure has a discontinuity at $\hat{\tau} = 2$, as the numerical Fourier transform solution (see Fig. 10 below) would seem to indicate, and that for large values of $\hat{\tau}$, the photoacoustic pressure has a $\hat{\tau}^{-\frac{3}{2}}$ dependence.

Figure 10

Fig. 10: Calculated and experimental photoacoustic waveforms for cylinders with $\hat{\rho} \neq 1$. Left column: pressure calculated from Eq. 13 with $\kappa = 1$ and $\hat{\tau} = 10$ versus dimensionless retarded time $\hat{\tau}$. Right column: experimental pressure versus time. (a) n-Pentane cylinder in water ($\hat{\rho} = 0.6266$, $\hat{c} = 0.6979$, $a = 220 \mu\text{m}$, $A = 1.24 \text{ cm}^{-1}$ and $E_0 = 82 \text{ J/m}^2$). The voltage and time scales in the oscilloscope trace are 50 mV/div and 200 ns/div , respectively. (b) Dibromomethane cylinder in water ($\hat{\rho} = 2.477$, $\hat{c} = 0.6440$, $a = 140 \mu\text{m}$, $A = 2.36 \text{ cm}^{-1}$ and $E_0 = 91 \text{ J/m}^2$). The voltage and time scales in the oscilloscope trace are 100 mV/div and 200 ns/div , respectively. (c) Water cylinder in n-Hexane ($\hat{\rho} = 1.508$, $\hat{c} = 1.415$, $a = 250 \mu\text{m}$, $A = 5.5 \text{ cm}^{-1}$ and $E_0 = 267 \text{ J/m}^2$). The voltage and time scales in the oscilloscope trace are 50 mV/div and 200 ns/div , respectively.

An exact solution to the problem of a uniformly irradiated infinite cylinder surrounded by

a fluid with identical acoustic properties can be obtained by using a Greens function solution of the wave equation for the velocity potential. Details of the calculation are omitted, but the result can be expressed as

$$\begin{aligned}\phi_f(\hat{r}) &= \frac{\kappa a}{\pi c \rho} \int_0^{[\hat{r}(\hat{r}+2\hat{r}-2)]^{\frac{1}{2}}} I(\hat{r}, \hat{r}, \hat{z}') d\hat{z}' & 0 \leq \hat{r} < 2 \\ \phi_f(\hat{r}) &= \frac{\kappa a}{\pi c \rho} \int_{[(\hat{r}+2\hat{r})(\hat{r}-2)]^{\frac{1}{2}}}^{[\hat{r}(\hat{r}+2\hat{r}-2)]^{\frac{1}{2}}} I(\hat{r}, \hat{r}, \hat{z}') d\hat{z}' & 2 < \hat{r}\end{aligned}\quad (64)$$

where

$$\begin{aligned}I(\hat{r}, \hat{r}, \hat{z}') &= \sin^{-1} \left\{ \frac{2 \hat{r}(\hat{r}-1+\hat{r}) - (\hat{z}'^2 + 1) + (\hat{r}-1)^2}{2 \hat{r}[(\hat{r}+\hat{r}-1)^2 - \hat{z}'^2]^{\frac{1}{2}}} \right\} \\ &- \sin^{-1} \left\{ \frac{2 \hat{r}(\hat{r}-1+\hat{r}) - \hat{z}'^2 + (\hat{r}-1)^2 - \{\hat{r} - [(\hat{r}+\hat{r}-1)^2 - \hat{z}'^2]^{\frac{1}{2}}\}}{2 \hat{r}[(\hat{r}+\hat{r}-1)^2 - \hat{z}'^2]^{\frac{1}{2}}} \right\}.\end{aligned}$$

The velocity potential from Eq. 64 can be calculated through straightforward numerical integration; the far field pressure, determined by numerical differentiation of ϕ_f , is found to be identical with that calculated from the numerical Fourier transformation, the hypergeometric functions or through use of the mapping described above. The solution given by Eq. 64 is somewhat easier to evaluate than the solution given in terms of hypergeometric functions or the integral that arises in the case of the mapping.

3. Response to Long Pulses

In the limit where the duration of the optical pulse is long compared with the transit time of sound across the cylinder, only the low frequency response of the cylinder is needed since the optical pulse contains only low frequency components. Thus, the Fourier transform of Eq. 54, gives the low frequency response of the cylinder p_l as

$$p_l(\hat{t}) = \frac{i \kappa}{8 \hat{\rho}} \frac{d}{d \hat{t}} \int_{-\infty}^{\infty} H_0^{(1)}(\hat{c} \hat{q} \hat{r}) e^{-i \hat{q} \hat{t}} d\hat{q}, \quad (65)$$

which can be integrated to give

$$p_l(\hat{t}) = \frac{\kappa}{2 \hat{\rho}} \frac{d}{d \hat{t}} \left\{ \frac{u(\hat{t} - \hat{c} \hat{r})}{\sqrt{\hat{t}^2 - (\hat{c} \hat{r})^2}} \right\} \quad (66)$$

where $u(\hat{t})$ is the Heaviside function. For an exciting light intensity profile of the form $I(t) = (E_0/\Theta)S(t/\Theta)$, where Θ is a pulsewidth parameter, convolution of Eq. 66 with $I(t)$ gives the photoacoustic pressure in the far field as

$$p(\hat{\tau}) = \frac{\kappa}{\tilde{\Theta}^{\frac{3}{2}} \hat{\rho} \sqrt{2^3 \hat{c} \hat{\tau}}} \int_{-\infty}^{\hat{\tau}} \frac{dS(T)/dT}{\sqrt{\hat{\tau} - T}} dT \quad (67)$$

where a dimensionless pulsewidth parameter $\tilde{\Theta}$ and a dimensionless retarded time $\hat{\tau}$ have been defined as $\tilde{\Theta} = c\Theta/a$ and $\hat{\tau} = (\hat{t} - \hat{c}\hat{r})/\tilde{\Theta}$. The photoacoustic waveform for long pulse excitation, according to Eq. 67, is thus proportional to the $\frac{1}{2}$ 'th derivative^{13,14} of $I(t)$. *It can be seen that the expression for the photoacoustic pressure given by Eq. 67 in the limit where $\hat{\rho} = \hat{c} = 1$ reduces to the expression given in Section I as a theorem.*

4. Low Density Response

The dimensionless oscillation frequency $\hat{\omega}_0 = \omega a/c_c$, where ω is the oscillation frequency, occurs where the denominator in Eq. 54, expanded for small $\hat{\rho}$, is a minimum. The expansion gives a dimensionless damping parameter $\hat{\gamma}$,

$$\hat{\gamma} = \frac{\pi \hat{\omega}_0}{4 (-\ln \hat{c} \hat{\omega}_0)} \quad (68)$$

with dimensionless oscillation frequency $\hat{\omega}_0$ given as a solution to the transcendental equation,

$$\hat{\omega}_0^2 \ln \hat{c} \hat{\omega}_0 = -2 \hat{\rho} .$$

Both the frequency and the damping thus depend on the density and sound speed parameters.

Consider the time evolution of the motion of the cylinder. First, the pressure inside the cylinder, which takes on the properties of a gas bubble, rises rapidly following the absorption of radiation. Owing to the small inertia of the cylinder compared with an equal volume of the surrounding fluid, the outward expansion of the cylinder is slow. Under the assumption of no heat conduction made in the derivation of the wave equation for pressure, the expansion is adiabatic as well. Now, the oscillation frequency is determined by the adiabatic compressibility of the cylinder, and its effective mass. The latter can be found by calculating the radiation force for radial oscillations of an arbitrary cylinder. For

any harmonic cylindrically symmetric radiator, the pressure outside the cylinder must be a solution to the wave equation, and hence must be of the form

$$p_f = P_0 H_0^{(1)}(\hat{c}\hat{r}\hat{q}) e^{-i\hat{q}t}, \quad (69)$$

where P_0 is an amplitude factor. For harmonic oscillation of the cylinder, the velocity and acceleration at $r = a$ can be written as $\dot{r}_0 \exp(-i\hat{q}t)$ and $\ddot{r}_0 \exp(-i\hat{q}t)$, respectively, where $\dot{r}_0$ and $\ddot{r}_0$ are constants. The two constants are related through the equation $\ddot{r}_0 = \dot{r}_0(-i\hat{q}c_c/a)$. Through use of the relation $\ddot{r} = -(\frac{1}{\rho_f a}) \frac{\partial p}{\partial r}$, where a dot indicates a time derivative, it is possible to show that P_0 is given in terms of $\dot{r}_0$ as $P_0 = -(ic_f \rho_f / H_1(\hat{c}\hat{q})) \dot{r}_0$. Since the force per length on the cylinder is $2\pi a$ times the pressure at $r = a$, the force per length f becomes

$$f = -2\pi i a c_f \rho_f \dot{r}_0 \left[\frac{H_0^{(1)}(\hat{c}\hat{q})}{H_1^{(1)}(\hat{c}\hat{q})} \right] e^{-i\hat{q}t}. \quad (70)$$

The Hankel functions can be written out in terms of Bessel functions of the first kind J and of the second kind Y to give

$$f = \left[\left(\frac{2\pi a^2 \rho_f}{\hat{c}} \right) \hat{F}_1(\hat{c}\hat{q}) \ddot{r}_0 + (2\pi a \rho_f c_f) \hat{F}_2(\hat{c}\hat{q}) \dot{r}_0 \right] e^{-i\hat{q}t}, \quad (71)$$

where the frequency dependent factors $\hat{F}_1$ and $\hat{F}_2$ are given by

$$\begin{aligned} \hat{F}_1 &= \frac{J_0(\hat{c}\hat{q}) J_1(\hat{c}\hat{q}) + Y_0(\hat{c}\hat{q}) Y_1(\hat{c}\hat{q})}{\hat{q}[J_1^2(\hat{c}\hat{q}) + Y_1^2(\hat{c}\hat{q})]} \\ \hat{F}_2 &= \frac{Y_0(\hat{c}\hat{q}) J_1(\hat{c}\hat{q}) - J_0(\hat{c}\hat{q}) Y_1(\hat{c}\hat{q})}{J_1^2(\hat{c}\hat{q}) + Y_1^2(\hat{c}\hat{q})}. \end{aligned}$$

The total reactive force on the cylinder can thus be broken up into an inertial term proportional to the acceleration and a resistive term proportional to the velocity. In the limit of small $\hat{q}$, $\hat{F}_1$ reduces to $-\hat{c} \ln \hat{c}\hat{q}$, so that the effective mass becomes $2\pi a^2 \rho_f (-\ln \hat{c}\hat{q})$. From the definition of the adiabatic compressibility κ_s it is easy to show for radial expansion of an infinite cylinder that $df = (-4\pi/\kappa_s) dr$, so that $4\pi/\kappa_s$ represents the effective force constant for the adiabatic oscillations. If the oscillation frequency is taken as the square root of the force constant divided by the effective mass then the above values for these two quantities

give the dimensionless oscillation frequency $\hat{\omega}_0$ as a solution to $\hat{q} = [2\hat{\rho}/(-\ln \hat{c}\hat{q})]^{1/2}$. In addition, the factor $\hat{F}_2(\hat{c}\hat{q})$ in the velocity dependent term at low frequency reduces to $\pi\hat{c}\hat{q}/2$; thus, the dimensionless damping, given by the product of a/c_c and one half of coefficient of $\ddot{r}_0$ divided by the coefficient of $\ddot{r}_0$, becomes $\pi\hat{q}/4(-\ln\hat{c}\hat{q})$. These expressions for the oscillation frequency and damping, are identical to those found from the low frequency limit for the photoacoustic pressure as given by Eq. 68 above.

D. Experiments and Results

As described above, fluid cylinders were made by forcing a stream through a fine glass capillary into a second immiscible liquid. In the case of n-pentane, which has a density significantly lower than that of water, a U-shaped capillary was employed so that the n-pentane flowed upward through the water. The liquid forming the cylinder was dyed with Dupont Oil Red for all the liquids except water and formamide, which were dyed with Methyl Blue. The dyes chosen were soluble in only one liquid so that the optical radiation was absorbed solely by the cylinder.

The cylinders were irradiated with a few *mJ* of the 532 nm output of a frequency doubled Nd : YAG laser. The laser beam was directed into a cubic photoacoustic cell 20 cm on a side where it irradiated an approximately 1 cm long portion of the cylinder. Pressure waves were detected at perpendicular incidence to the direction of the laser beam with a 52 μm thick polyvinilidene fluoride film transducer as described previously using essentially the same detection and recording electronics.

Experiments were conducted to record photoacoustic pressure versus time for liquids that have a density close to that of water, or liquid mixtures whose composition was adjusted to have a density approximately that of water. From the recorded waveforms (not reproduced here) it became clear that for the cylinders with densities equal to those for the surrounding fluid, the photoacoustic wave is not simply a succession of identical waves repeated in time and multiplied by a numerical factor, as is the case for those generated by a spherical droplet with $\hat{\rho} = 1$, or the infinite fluid layer.

Fig. 10 shows experimental photoacoustic waveforms along with calculated waveforms from Eq. 60 where $\hat{\rho}$ differs from unity. Measurements of the diameters of the cylinders were made with an estimated error of $\pm 10\%$ using the shadow cast by the cylinder on a calibrated scale. In all cases, the diameters of the cylinders determined by the positions of the cusp in time (together with a knowledge of the sound speeds in the cylinders) agreed with those determined from the optical measurements to within the error limits of the latter.

An accurate description of the rather complicated time profiles of the waves requires evaluation of Eq. 60. However, a few qualitative features of the waves can be found without recourse to the integral formulas. First, each waveform at long time has a slow rise to the baseline; this response is characteristic of two-dimensional waves²⁴. Each of the waves possesses both compressions and rarefactions in accord with the principle that the time integral of the pressure in such two-dimensional waves must be zero²⁴. Also, it can be said that in addition to the primary wave, successive waves are produced whenever $\hat{\rho}$ or $\hat{c}$ differs from unity owing to reflections of the initial outgoing wave at the interface between the two fluids. A quantitative discussion of this last point is given in the Discussion Section.

A close examination of the experimental photoacoustic oscilloscope traces shows that the high frequency features of the calculated waveforms are not precisely reproduced by the experiment. There are several factors in the experiments reported here that act to degrade the high frequency features of the waveforms including integration of the cylindrical wave over the plane surface of the detector, misalignment of the detector with respect to the cylinder, non-uniform heat deposition in the cylindrical stream owing to a mismatch in the indices of refraction of the cylinder and the surrounding fluid, excitation with a finite pulsewidth laser, and nonuniformity in the diameter of the cylinder along its irradiated length. Although the oscilloscope traces in Fig. 10 show general agreement with the calculated waveforms; clearly, more sophisticated experiments where these factors are controlled are possible.

E. DISCUSSION

For the fluid droplet and infinite layer it is possible to write the Fourier transform ex-

pression for the photoacoustic pressure in terms of a power series in the transmission and reflection coefficients of a spherical wave at a spherical interface for the droplet, and a plane wave at a plane interface for the layer. A similar manipulation of Eq. 60 for a fluid cylinder can be carried out given expressions for the reflection coefficient R and transmission coefficient T of a cylindrical wave at a cylindrical interface. Consider a unit amplitude, outwardly propagating, cylindrical wave P_I incident on a cylindrical interface with a radius of curvature a between two fluids, and an inwardly propagating, cylindrical wave P_R reflected from the interface. These two waves travel in a medium with acoustic properties denoted with a subscript c . The outwardly propagating, transmitted cylindrical wave P_T is considered to propagate in an infinite medium with properties denoted with subscripts f . Since all three waves must be a solution to the inhomogeneous wave equation for pressure, they must be of the form

$$\begin{aligned} P_I &= H_0^{(1)}(k_c r) e^{-i\omega t} \\ P_R &= R H_0^{(2)}(k_c r) e^{-i\omega t} \\ P_T &= T H_0^{(1)}(k_f r) e^{-i\omega t} . \end{aligned} \quad (72)$$

The requirements of the continuity of pressure and acceleration at a cylindrical interface at $r = a$ yield two equations for the complex transmission and reflection coefficients.

We do not include the details of the derivation, but the photoacoustic pressure given by Eq. 60 can be written as the expansion

$$p_f(\hat{r}) = \frac{\kappa}{4} \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} J_1(\hat{q}) H_0^{(1)}(\hat{c}\hat{q}\hat{r}) T(-R)^n e^{-i\hat{q}\hat{t}} d\hat{q} . \quad (73)$$

where

$$\begin{aligned} R &= - \frac{H_1^{(1)}(\hat{q}) H_0^{(1)}(\hat{c}\hat{q}) - \hat{\rho}\hat{c} H_1^{(1)}(\hat{c}\hat{q}) H_0^{(1)}(\hat{q})}{H_1^{(2)}(\hat{q}) H_0^{(1)}(\hat{c}\hat{q}) - \hat{\rho}\hat{c} H_1^{(1)}(\hat{c}\hat{q}) H_0^{(2)}(\hat{q})} \\ T &= \frac{H_0^{(1)}(\hat{q}) H_1^{(2)}(\hat{q}) - H_1^{(1)}(\hat{q}) H_0^{(2)}(\hat{q})}{H_1^{(2)}(\hat{q}) H_0^{(1)}(\hat{c}\hat{q}) - \hat{\rho}\hat{c} H_1^{(1)}(\hat{c}\hat{q}) H_0^{(2)}(\hat{q})} . \end{aligned} \quad (74)$$

The interpretation of Eq. 73 is identical to that for the analogous expression for the photoacoustic effect generated by a spherical droplet. Consider the first term in Eq. 73 where $n = 0$

and the factor $(-R)^n$ is unity. Here, the outgoing cylindrical wave is partially transmitted depending on T and appears at the field point at $\hat{r} = 0$. At the same time, a wave is reflected at the interface between the two fluids that propagates to the z axis where it is reflected and reappears at the perimeter of the cylinder delayed in time and with an inverted amplitude. This second wave appears at the field point at a later time with an amplitude and phase determined by the Bessel and Hankel functions in the integrand of Eq. 73 and the product of $T(-R)$. Higher powers of $(-R)$ in the integrand of Eq. 73 represent successive reflections of the wave at the interface between the two fluids and inversion at the z axis.

The derivations given for R and T for a cylindrical wave at cylindrical fluid-fluid interface, surprisingly, appears to be the first ever given! Further discussion of their interesting, but rather complicated, characteristics has been published by the authors in the reference given in the Introduction to this section.

Despite the complexity of the photoacoustic waveform characteristic of the uniformly irradiated cylinder, it is clear that the acoustic waveform generated by short pulse irradiation of the cylinder is highly indicative of its two-dimensional geometry, and contains information about the diameter of the cylinder, and its density and sound speed relative to the surrounding medium. If the density and sound speed of the surrounding fluid as well as the sound speed in the cylinder are known, the results given here indicate that, in principle, an analytical technique is possible where it is possible to determine the diameter of the cylinder and its density from an accurate recording of the photoacoustic wave.

V. VISCOUS EFFECTS IN TRANSIENT GRATING EVOLUTION

A. Introduction

We have carried out a detailed analysis of the time response of a transient photoacoustic grating starting from the coupled equations for temperature and pressure. The focus of this work has been on determining the effects of viscosity and heat conduction. Since some of the results of this work are reproduced in the proposal accompanying this progress report, only a very abbreviated account of the research will be given here. Full details of the research

can be found in *J. Chem. Phys.* 97, 9324 (1993)

B Summary of Results

Starting from the coupled equations for pressure and temperature, which follow from the linearized hydrodynamic equations, we have solved for the density, pressure, and temperature changes produced by a delta function deposition of heat in time for an optical grating. The heating function is thus described in the frequency domain as

$$H_s = \bar{\alpha} I_0 (1 + \cos \hat{x}). \quad (75)$$

which describes the deposition of heat by two coherent optical beams interfering in space. We take the viscosity of the fluid to be a frequency dependent quantity and use a two parameter description of the viscosity. The expression for the (dimensionless) time dependent density is found to be

$$\delta(\hat{t}) = i \cos \hat{x} \times \int_{-\infty}^{\infty} \frac{(q + i/\hat{\tau}_v) e^{-i q \hat{t}}}{\left\{ q^4 + i[1/\hat{\tau}_v + \gamma \hat{l}_h] q^3 - [\hat{c}^2 + \gamma \hat{l}_h / \hat{\tau}_v] q^2 - i[1/\hat{\tau}_v + \gamma \hat{l}_h (\hat{c}^2 - 1) + \hat{l}_h] q + \hat{l}_h / \hat{\tau}_v \right\}} dq. \quad (76)$$

where $\hat{x}$ is the grating wavevector times the distance along the optical grating, $\hat{\tau}_v$ is the viscous relaxation time, $\hat{l}_h$ is the dimensionless heat conduction length, γ is the heat capacity ratio, and $\hat{c}$ is the sound speed ratio (the infinite to zero frequency sound speed ratio). Eq. 76 gives a description of the evolution of the grating, especially the effects of heat conduction and viscosity on the evolution of the acoustic standing wave; this equation was also used to fit transient grating data using Marquardt's method.

The experiments here were conducted with $CuCl_2$ dissolved in ethylene glycol. $CuCl_2$ was used in the experiments since Cu^{++} has a strong absorption in the near ultraviolet spectral region, and possesses, as well, a ground state recovery time shorter than the pulsewidths of the lasers used. Ethylene glycol was selected for these experiments since its viscosity varies over a wide range with only small changes in temperature. The experimental method used is standard and will only be described briefly; details of the apparatus can be found in Ref.

2. The transient grating was generated with either the 280 nm or 317 nm doubled output of a synchronously pumped dye laser. The dye laser was pumped with the frequency doubled output of a modelocked Nd:YAG laser Q-switched at 326 Hz. The output of the first dye laser at 633 nm was split into two beams; one beam was frequency doubled to produce approximately 2 μJ pulses at 317 nm. The remainder of the 633 nm beam was used as the grating probe beam. The widths of the 633 nm pulses (i.e. the full width at half maximum), as determined by autocorrelation, were typically 55 ps.

Samples of $CuCl_2$ with an optical density of 1.0 mm^{-1} at 317 nm were prepared by dissolution of $CuCl_2 \cdot 2H_2O$ in reagent grade ethylene glycol. No attempt was made to remove water from the solution. The solution was placed in a quartz cell that could be heated or cooled by a stream of dry N_2 . Temperatures in the cell were measured with a thermocouple placed in contact with the fluid and were controlled to within $\pm 1 K$. The 317 nm beams were focussed into the cell at a crossing angle of 50° to give spots of 550 μm in diameter and a grating wavevector k of $1.676 \times 10^7 m^{-1}$. The 633 nm probe beam was time delayed relative to the ultraviolet pulse by translating a corner cube reflector. One excitation beam was chopped at 163 Hz so that the diffracted probe beam could be detected with a lock-in amplifier.

Figure 11

Fig. 11: Diffracted probe beam intensity (in arbitrary units) versus time for Cu^{++} in ethylene glycol at several values of the temperature. Data in column (a) were taken with a wavevector k of $1.676 \times 10^7 \text{ m}^{-1}$; those in column (b) were taken with a wavevector of $9.245 \times 10^6 \text{ m}^{-1}$.

A second set of experiments was conducted with the same apparatus, but using the 280 nm frequency doubled output of a second dye laser as the excitation source, and the 633 nm laser as the probe source. The angle of intersection of the two 280 nm beams was 23.8° giving a grating wavevector of $9.245 \times 10^6 \text{ m}^{-1}$.

Figure 11 shows typical waveforms of diffraction intensity versus probe beam delay time taken at several temperatures for two different grating wavevectors. From these data relaxation time, zero frequency viscosity and sound speed versus temperature for ethylene glycol were determined. The experimental data could be fit with the two parameter model for relaxation.

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