Photoacoustic Monopole Radiation in One, Two, and Three Dimensions

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We show that the photoacoustic pressure in one, two, and three dimensions can be found as mappings of the optical deposition of heat in space for short optical radiation pulses. In addition, we find the photoacoustic pressure to be proportional to the zeroth derivative in one dimension, the one-half derivative in two dimensions, and the first derivative in three dimensions of the optical radiation intensity for long pulses. Experiments with fluid layers, cylinders, and droplets give ultrasonic wave forms that are in general agreement with the theorems.

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Investigation of the photoacoustic effect [1] in fluids has been motivated to a large extent by its numerous applications in spectroscopy, chemical kinetics and analysis, and trace detection where a knowledge of the amplitude dependence of the effect is essential for the interpretation of experimental data. Recently, experiments with fluid bodies [2] have shown that information regarding the dimensions, geometry, sound speed, and density of an irradiated body can be found from the shape of its photoacoustic wave form. Such experiments point up the question of precisely how ultrasonic wave forms are governed by both the physical characteristics of the irradiated body and the properties of the optical exciting pulse.

Here, we derive three theorems for short-pulse excitation of fluid bodies that give the temporal profiles of photoacoustic waves as mappings of the spatial distribution of heat created by the absorption of optical radiation, thus obviating solution of the wave equation for pressure [3–7]. Furthermore, we prove theorems in one, two, and three dimensions that describe the temporal profiles of photoacoustic waves excited by long optical pulses. We also report experiments with fluid layers, cylinders, and droplets.

The photoacoustic effect is governed by a wave equation for pressure [8,9], which can be written in terms of a velocity potential ϕ as

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

where p is determined from $p=-\rho_f\,\partial\phi/\partial t$, 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. Here, we use a Green's-function solution [10] to the wave equation for the velocity potential,

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

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

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

Consider a heating function of the form $H(\hat{\xi},t)=\alpha E_0\hat{h}(\hat{\xi})\delta(t)$, where α 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

$$p(\hat{\tau}_0)=\tfrac{1}{2}\kappa\hat{h}(\hat{\tau}_0)\,, \tag{4}$$

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 $\kappa=\alpha\beta E_0 c^2/C_P$.

Consider the analogous problem 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 traveling 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

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

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

$$p(\hat{\tau}_0)=(\kappa/2\hat{r})\hat{\tau}_0[\hat{h}(-\hat{\tau}_0)+\hat{h}(\hat{\tau}_0)]\,. \tag{6}$$

For a cylindrically symmetric source, we find the photoacoustic wave generated by a δ-function heating pulse by expressing Eq. (6) in terms of cylindrical coordinates z and ρ [i.e., $r=(\rho^2+z^2)^{1/2}$] and integrating the result over the z axis. This procedure gives the photoacoustic pressure as

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

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 δ-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

$$\int_{\hat{\rho}}^{\infty}d\hat{\zeta}f'(\pm\hat{\zeta})(\hat{\zeta}^2-\hat{\rho}^2)^{-1/2}=\tfrac{1}{2}\hat{h}(\hat{\rho})\,,$$

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 inversion [7,11,12] which gives

$$f(\hat{\eta})=-\frac{1}{\pi}\int_{\hat{\eta}}^{\infty}d\hat{\rho}\,\frac{\hat{\rho}\hat{h}(\hat{\rho})}{(\hat{\rho}^2-\hat{\eta}^2)^{1/2}}\,. \tag{8}$$

As examples of the use of the three theorems for short optical pulses, we consider an optically thin, infinite fluid layer, a fluid cylinder, and a droplet. 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 (see Fig. 1) for $\hat{\tau}>0$ in one, two (with $\hat{\rho}\gg 1$), and three dimensions as

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

$$p(\hat{\tau})=\frac{\kappa}{\pi(2\hat{\rho})^{1/2}}\int_{-\hat{\delta}}^{0}d\hat{\eta}\,\frac{\hat{\eta}+1}{(\hat{\eta}+\hat{\tau})^{1/2}[1-(\hat{\eta}+1)^2]^{1/2}}\,, \tag{10}$$

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

respectively, where $\hat{\tau}$ is the dimensionless retarded time from the edge of the body defined as $\hat{\tau}=(c/\xi_0)[t-(\xi-\xi_0)/c]$, and $\hat{\delta}$ is the lesser of 2 or $\hat{\tau}$.

Consider now the case 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 $H(\hat{\xi},t)=\alpha I_0\hat{h}(\hat{\xi})e^{-i\omega t}$, which, in one dimension, according to Eq. (3), gives the acoustic pressure as an integral over space 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

$$p_l=(\alpha\beta I_0\xi_l c/2C_P)e^{-i\hat{q}_l\hat{\tau}_l},\quad p_l(\hat{\tau}_l)=\tfrac{1}{2}\kappa\delta(\hat{\tau}_l)\,, \tag{12}$$

respectively, where $\hat{q}_l=\omega\xi_l/c$, $\hat{\tau}_l=(c/\xi_l)(t-\xi/c)$, 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

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

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 equation [10] 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 zeroth-order Hankel function of the first kind, J_0 is a Bessel function, and k is the wave vector. In the limit of low frequencies, $J_0(k\rho')\cong 1$, which gives the low-frequency photoacoustic pressure and its Fourier transform as

$$p_l=(\pi\alpha\beta I_0\xi_l c/2C_P)\hat{q}_l H_0^{[1]}(\hat{q}_l\hat{\rho}_l)e^{-i\hat{q}_l\hat{t}_l}\,,$$
$$p_l(\hat{t}_l)=\kappa\frac{d}{d\hat{t}_l}\frac{u(\hat{t}_l-\hat{\rho}_l)}{(\hat{t}_l^2-\hat{\rho}_l^2)^{1/2}}\,, \tag{14}$$

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

We find the time profile of the photoacoustic wave generated by a long light pulse by convoluting a normalized light intensity function of the form $I(t)=E_0S(t/\Theta)/\Theta$ 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

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

$$P(\tilde{\tau}_l)=\frac{\kappa}{\hat{\Theta}_l^{3/2}(2\hat{\rho})^{1/2}}\int_{-\infty}^{\tilde{\tau}_l}dT\,\frac{dS(T)/dT}{(\tilde{\tau}_l-T)^{1/2}}\,, \tag{16}$$

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

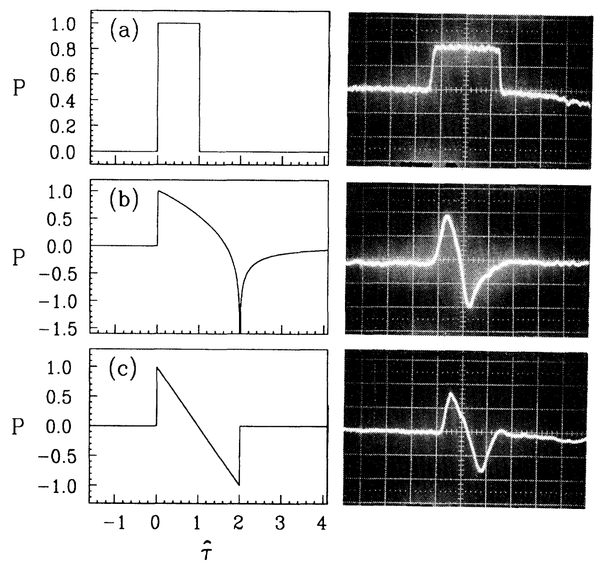

FIG. 1. Photoacoustic wave forms from short laser pulses. Left column: photoacoustic pressure P in arbitrary units vs dimensionless time $\hat\tau$ for (a) a fluid layer, (b) a cylinder, and (c) a sphere. The equations in the text were evaluated with $\kappa/2=1$, $\kappa/2\hat\rho^{1/2}=1$, and $\kappa/2\hat r=1$ in (a), (b), and (c), respectively. Right column: experimental wave forms 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 μsec/div and 20 mV/div, (b) 200 nsec/div and 20 mV/div, (c) 500 nsec/div and 50 mV/div. The laser fluences were 15, 60, and 20 J/m^2, and the absorbances of the bodies were 0.4, 0.5, and 1.4 cm^{-1}.

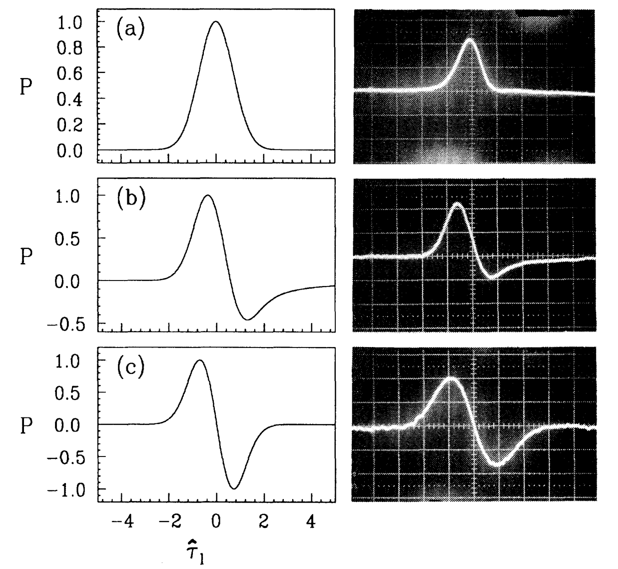

FIG. 2. Photoacoustic wave forms from long laser pulses. Left column: pressure P in arbitrary units vs dimensionless retarded time from the origin for bodies in (a) one dimension, (b) two dimensions, and (c) three dimensions excited by a Gaussian light pulse in time. Equations (15)–(17) were evaluated with $\kappa/2\hat\Theta_l\sqrt{\pi}=1$, $\kappa/(2\hat\rho)^{1/2}\hat\Theta^{3/2}=1.25$, and $\sqrt{2}\kappa/\sqrt{\pi e}\,\hat\Theta^2\hat r_l=1$ (a), (b), and (c), respectively. Right column: experimental wave forms 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 nsec/div and 100 mV/div, (b) 200 nsec/div and 50 mV/div, and (c) 200 nsec/div and 20 mV/div. The laser fluences (estimated from the fluence for a train of pulses) and optical absorbances were 10 J/m^2 and 80 cm^{-1} for the half space, 200 J/m^2 and 5 cm^{-1} for the cylinder, and 100 J/m^2 and 2 cm^{-1} for the sphere.

respectively, where $\hat\Theta_l=c\Theta/\xi_l$ and $\tilde\tau_l=\hat\tau_l/\hat\Theta_l$. 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 zeroth, the one-half, and the first time derivative, respectively, of the intensity of the exciting optical pulse evaluated at the retarded time from the origin. Further analysis [7] shows that these properties are more general than has been found for the simple cases of bodies with spherical and cylindrical symmetry. Figure 2 gives plots of Eqs. (15)–(17) where S is a Gaussian function of time.

Experiments with short optical pulses were done using the frequency-doubled output of a Q-switched Nd-doped yttrium aluminum garnet (Nd:YAlG) laser. The acoustic waves were detected [2] with a polyvinylidene fluoride film transducer whose output was displayed on an oscilloscope after amplification by a factor of 40. A fluid layer was 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. 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 wave forms generated by a layer, a cylinder, and a sphere.

For the long-pulse experiments we used 1.06-μm radiation pulses from the Nd:YAlG laser with the quarter-wave plate associated with the Q switch removed. The laser generated a train of approximately 1-μs-long pulses, which were typically asymmetric, with slightly faster rise times 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 wave forms for the long-pulse experiments are shown in Fig. 2.

The one-dimensional mapping given here for δ-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. The two-dimensional mapping reproduces [7] the photoacoustic wave form 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 [21]. The three-dimensional mapping explains the origin of the N-shaped wave found in Refs. [2], [22], and [23] 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 [24,25]. We also believe that the δ-function response given here would lead to considerable simplification of some previous calculations in three dimensions [26,27] since the acoustic wave excited by arbitrary light pulses can be found in terms of simple convolution integrals.

The theorems for long light pulses given here generalize the results of Refs. [15] and [17] where waves proportional to the exciting radiation pulse were found in one-dimensional calculations. The authors of Ref. [18] hinted at the two-dimensional theorem given here when they 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 wave forms. We find the wave form in Fig. 2(b) 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. [21] using a completely different method. We also find agreement between the wave form predicted from the long-pulse theorem for three dimensions and the wave forms calculated in Refs. [23] and [28].

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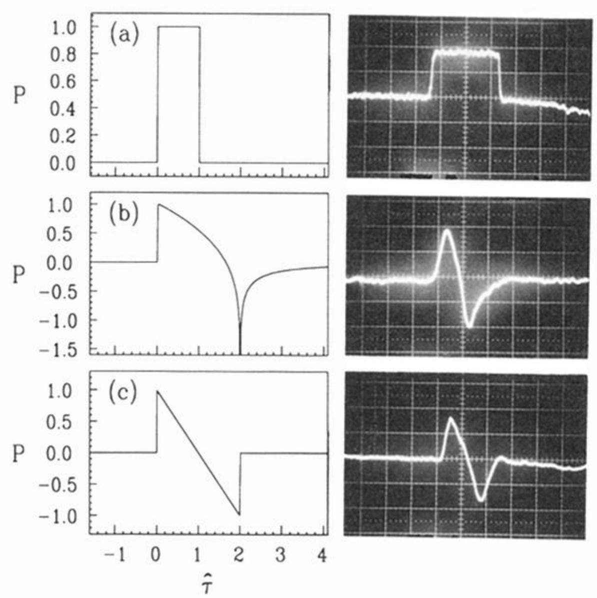

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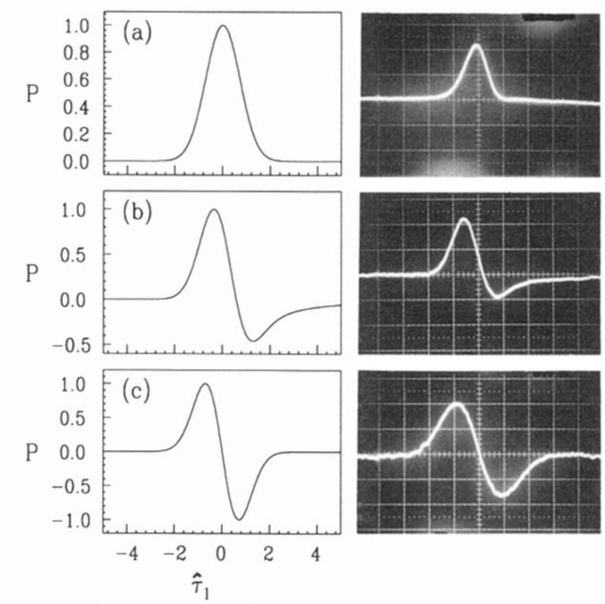

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