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Thermally launched photoacoustic waves

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Abstract: Photoacoustic waves can be generated by submicron sized particles that absorb radiation and transmit heat to a surrounding fluid. When the thermal expansion coefficient of the absorbing body is small, a photoacoustic effect is not produced until heat diffuses into the surrounding fluid. The time evolution of the acoustic wave produced by a suspension of particles can be monitored using the transient grating technique. Experiments are reported for a reverse micelle solution.

1. INTRODUCTION

Consider the photoacoustic effect generated by irradiating a slurry of particles with a short light pulse. Immediately after the radiation is absorbed, *N*-shaped photoacoustic waves are emitted (1,2) by the individual particles. Since the particles in a slurry are located randomly in space, the individual acoustic waves do not add coherently; and, since in each wave a compression must be accompanied by a rarefaction (3), the pressure waves add to zero at a field point outside the excitation region. However, the thermal expansion of both the particles and the surrounding fluid add coherently throughout the volume of the slurry to give an acoustic wave. The production of the acoustic wave through thermal expansion is complicated since the particles undergo a sharp temperature increase from the laser heating followed by a gradual cooling through heat conduction to the surrounding fluid. The fluid, on the other hand, is heated through heat diffusion from the particles. In a description of the photoacoustic effect, the time dependent expansions and contractions of both the particle and fluid must be taken into account.

Consider a somewhat simpler problem where the absorbing body has a negligible expansion coefficient and the acoustic waves are generated solely by thermal expansion of the fluid. In the experiment the photoacoustic effect from the fluid expansion is monitored using the transient grating technique. In order to generate acoustic waves, a laser beam is split into two equal intensity beams which are recombined to produce an optical standing wave. The deposition of heat at the antinodes of the optical beam causes a standing acoustic wave to be generated whose frequency is determined by the optical fringe spacing. For visible or ultraviolet light beams, the acoustic frequencies are generally in the *GHz* range. Because of the short acoustic wavelength, the preferred method of detection of the ultrasonic wave is through diffraction of a probe laser beam directed at the Bragg angle with respect to the grating. The time evolution of the acoustic wave is monitored by recording the intensity of the diffracted light signal which is proportional to the square of the acoustic density.

A description of the photoacoustic effect requires determination of the heat per unit volume and time entering the fluid from the laser heated particle, and a solution to the hydrodynamic equations for the time dependent acoustic density when heat is deposited sinusoidally in space and instantaneously in time. The frequency domain temperature of the fluid τ_f is found by solution to the heat equation and can be written,

$$\tau_f = \tau_M \hat{T}_f h_0(\kappa_f^t r) e^{-i\omega t}, \quad (1)$$

where h_0 is a spherical Hankel function, r is the radial coordinate, ω is the modulation frequency, t is the time, and where τ_M is the frequency domain temperature amplitude given by $\tau_M = i\bar{\alpha}I_0/\rho C_P\omega$, where $\bar{\alpha}$ is the particle absorption coefficient, I_0 is the laser beam intensity, ρ is the particle density, C_P is the particle specific heat capacity. The parameter $\hat{T}_f$ in Eq. 1 is a dimensionless temperature amplitude defined by

$$\hat{T}_f = \frac{(\kappa_f^t a) \left(\frac{\sin \kappa_s^t a}{\kappa_s^t a} - \cos \kappa_s^t a \right) e^{-i\kappa_s^t a}}{[(\hat{K} - 1) \frac{\sin \kappa_s^t a}{\kappa_s^t a} - \hat{K} \cos \kappa_s^t a + i \hat{\chi}^{\frac{1}{2}} \sin \kappa_s^t a]},$$

where $\hat{K}$ and $\hat{\chi}$ are ratios of the thermal conductivities and diffusivities of the material inside the sphere to that outside the sphere respectively, κ_f and κ_s are the thermal wavevectors in the surrounding fluid and the particle respectively, and a is the particle radius. The thermal wavevectors κ^t , with s and f referring to quantities inside the sphere and in the fluid, respectively, are given by

$$\kappa^t = \frac{i\omega}{l_h c} = \frac{i\omega}{\chi},$$

where the χ is the thermal diffusivity and l_h is the heat conduction length ($l_h = \chi/c$, where c is the sound speed). The result given by Eq. 1 has been derived previously by Isakovich (4), and has been discussed recently by Zozula and Puchenkov (5) with regard to generation of the photoacoustic effect by a suspension of particles excited by a Gaussian laser beam.

The heating function can be found from Eq. 1 by computing the heat flux from the particle, which is integrated over the area of the sphere, and multiplied by the number of spheres per unit volume in the slurry. The heating function for the diffusion of heat into the fluid, when expressed in dimensionless form, is given by,

$$h = iN \left[\frac{4\pi a^3}{(\kappa_f^t a)^3} \right] \hat{T}_f (1 - i\kappa_f^t a) e^{i\kappa_f^t a - i\omega t}, \quad (2)$$

where N is the number of particles per unit volume.

The response of the density δ to a sinusoidal deposition of heat in space that is a delta function in time is given by the Fourier transform integral (6),

$$\delta(\hat{t}) = \frac{i\rho_f\beta_f\theta_t}{2\pi} \int_{-\infty}^{\infty} dq \left\{ \frac{\cos kx}{[q^3 + iq^2(\hat{l}_v' + \gamma\hat{l}_h) - q(1 + \gamma\hat{l}_v'\hat{l}_h) - i\hat{l}_h]} \right\} h(q)e^{-iq\hat{t}}, \quad (3)$$

where $\hat{t}$ is a dimensionless time ($\hat{t} = ckt$, where k is the wavevector determined by the optical standing wave), θ_t is a temperature defined as $\theta_t = \bar{\alpha}E_0/\rho_f C_{Pf}$ and where β_f , ρ_f , and C_{Pf} are the thermal expansion coefficient, density, and specific heat capacity of the fluid, respectively, E_0 is the laser fluence, $\hat{l}_v'$ is the viscous length parameter, and x is the spatial coordinate. Equation 3 gives the density as a function of time following laser irradiation of the solution, which, when squared, is proportional to the diffracted light intensity. Note in Eq. 3 that when $h(q)$ is a constant,

corresponding to a delta function deposition of heat in the fluid, the factor in braces gives the effect of viscous damping and heat conduction on the thermal and acoustic modes. The heating function thus modifies the time response of the density to take into account the slow diffusion of heat from the particle.

2. EXPERIMENTS

Experiments were done by irradiating a solution of "water-in-oil" micelles made from water, isooctane, and the surfactant AOT. The exciting laser irradiated a 1 mm cuvette with 40 ps, 317 nm laser pulses. The grating was probed with a 633 nm pulse from the same laser that was delayed in time using an optical delay line. The interior of the micelles contained malachite green, a dye which is soluble in water but not in isooctane. Measurements of the diffracted light signal versus time from the acoustic grating are shown in Fig. 1.

Figure 1. Diffracted light intensity for (left trace) 2.5 nm radius micelles, and (right trace) 17 nm micelles. The grating wavevector is $1.676 \times 10^7 \text{ m}^{-1}$

The data for the 2.5 nm radius micelles indicate that heat is released rapidly; the signal appears as if the dye were uniformly dispersed in the isooctane. The waveform for the 17 nm micelles, on the other hand, shows a diminished acoustic signal with a gradual increase in the mean signal amplitude to its maximum value. Several simulations of data for large micelles show that the reduction in the acoustic signal along with a gradual rise in the mean value of the diffracted light signal is characteristic of a slow heat release from the interior of the micelle.

Both the preliminary experiments and several data simulations show that by fitting the intensity of the diffracted light signal to Eq. 3, it is possible to determine the diameter of the absorbing body, and its thermal conductivity and diffusivity relative to the surrounding fluid. The photoacoustic technique should thus form a diagnostic technique for determining particle radii and two thermal parameters for dilute slurries of particulate matter that can be excited by optical radiation.

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