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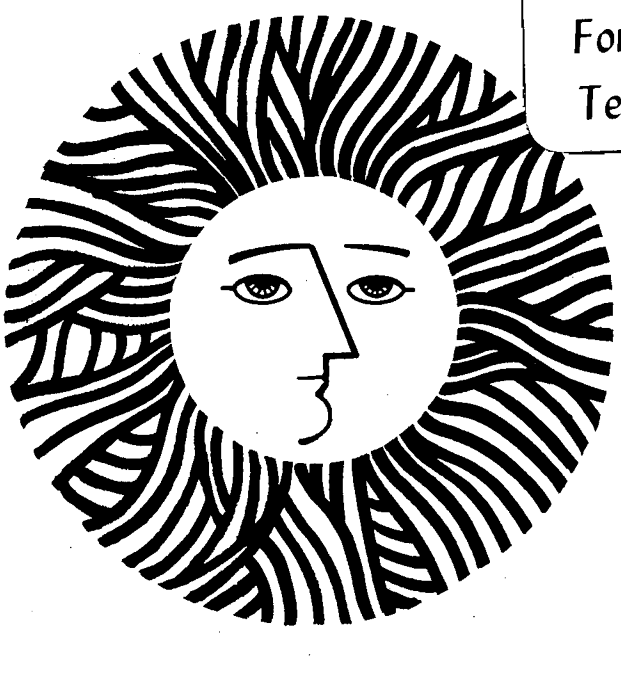

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The Scattering Contribution to Photoacoustic Signal

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Abstract

Using radiative transfer theory we show that under certain conditions the contribution of light scattering to the photoacoustic signal can be significant.

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Insensitivity to scattering has generally been accepted as an inherent characteristic of photoacoustic detection. The physical reasoning underlying this assumption has been that the photoacoustic signal arises from the heat generated following optical absorption, and thereby is a measure of the cross section of such absorption. However, the thermal energy generated by the absorption of light is a function of the intensity distribution within the sample, which depends on its scattering characteristics. Consequently, deviation from Beer's Law should, in principle, affect the thermal signal generated. We will show that for highly scattering media, the photoacoustic signal is strongly dependent on the scattering parameters. On the other hand, for optically thin solids and liquids the photoacoustic signal is insensitive to light scattering up to a scattering coefficient ($\lambda\alpha$) of $\sim 1/\ell$ where ℓ is the sample thickness, λ is the fraction of light scattered, and α is the total attenuation coefficient of a coherent beam.

Most photothermal detection techniques respond to the surface temperature averaged over the sample surface, and/or a weighted integral of sample temperature over its volume. We analyze the coupled heat diffusion and radiative transfer equations. We rigorously show that, for a wide range of experimental conditions, the coupled equations can be solved exactly yielding a photoacoustic signal S

$$S \propto 1 - (R+T) \qquad (1)$$

where R is the reflectance (diffuse and specular) and T is the transmittance (diffuse and coherent) of the sample of interest. Hence, Eq. (1) proves the intuitive conclusion that the photoacoustic signal is proportional to the absorptance of the sample.

In the case of gas-cell-microphone detection of solids, Eq. (1) is valid for i) $\ell_{th} >> \ell$, or ii) $\ell_{th} >> \ell_{opt}$, where ℓ_{th} is the thermal diffusion length and $_{opt}$ is the optical thickness of the sample. This conclusion also holds for transverse photothermal deflection, for piezoelectric photoacoustics of solids.

For the case where $\ell_{th} \leq \ell$ or $\ell_{th} \leq \ell_{opt}$, an approximate Schuster-Schwarzchild analysis of the radiative transfer equations is used to obtain an analytical expression for the photoacoustic signal. In the general case, the scattering characteristics of the medium enters the expression in a complicated form. An interesting limiting case is $\ell_{th} << \ell_{opt}$, ℓ for which

$$S \propto (1-\lambda)\alpha \left[1 + \frac{2(1+r_i)}{(1-r_i)} R\right] \tag{2}$$

where $(1-\lambda)\alpha$ is the absorption cross section and r_i is the internal reflection coefficient for diffuse light at the sample-gas boundary. It can be seen that the photoacoustic signal is proportional to the sample absorption cross section (within a thermal length), with the proportionality constant considerably increased from its value of 1 for no scattering. This is due to the increase in photon flux at the sample-gas boundary.

The solution for the coupled heat diffusion and radiative tranfer equations can be employed in one of two ways to evaluate the scattering contribution to the photoacoustic signal: i) one may write the solution in terms of R and T which can then be determined experimentally using integrating sphere techniques. Combining the measurement of R and T with the detected photoacoustic signal yields information on such sample properties as its internal reflectance which is otherwise experimentally inaccesible; or ii) one many solve the radiative transfer equation for R and T in terms of the scattering coefficient $\lambda\alpha$, the attenuation coefficient, and the internal reflectance. By inserting this solution in the general solution for the photoacoustic signal, one obtains the dependence of the photoacoustic signal on the scattering and the absorption of the sample.

Using a two flux model for R and T, the results of evaluating the solutions for various absorption constants and internal reflectance coefficients are shown in Fig. (1). Physically for $\lambda\alpha\ell < .1$, the effective light path length within the sample is equal to the sample thickness and the photoacoustic signal is independent of scattering. When $\lambda\alpha\ell \sim 1$, the mean path length of the light increases as the scattering increases, so does the absorption. At still higher scattering, the signal saturates since all the light is scattered without further increase in the effective path lengths. For very high scattering samples, the reflectance becomes larger as the scattering increased leading to a decrease in the light intensity within the sample. Hence, the signal decreases.

Our results provide a complete picture of the role of light scattering in photoacoustic detections. We show that whereas under certain conditions photoacoustic spectroscopy is insensitive to scattering, under others it can significantly affect photoacoustic spectra.

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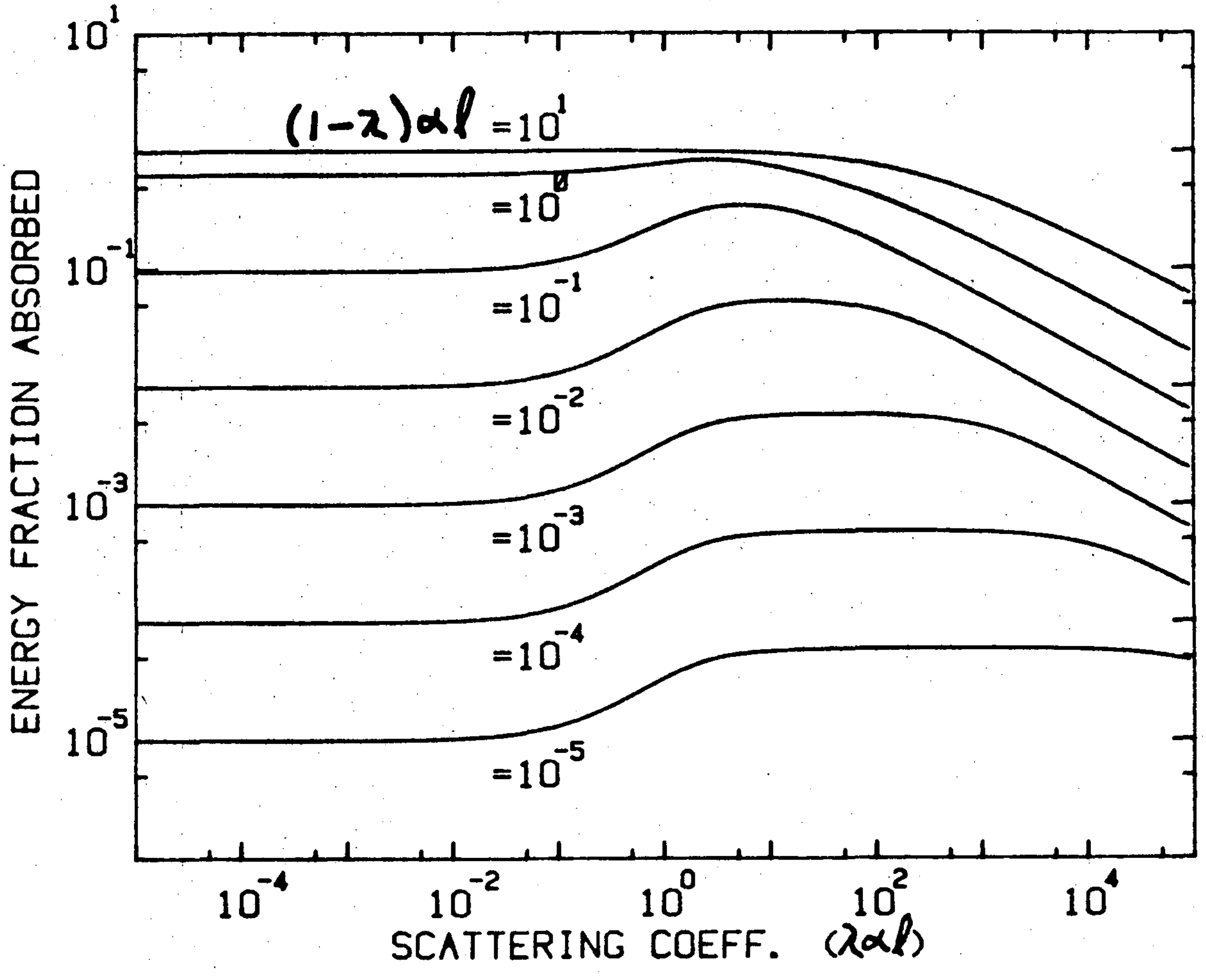

Fig. (1)

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