Temporal analysis of short laser pulses using degenerate four-wave mixing

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The temporal characteristics of picosecond and subpicosecond laser pulses have been investigated with a number of different techniques.\textsuperscript{1,2} Streak cameras provide the temporal profile of an optical pulse directly but are limited to a time resolution of \(\sim 1\) psec. The two-photon fluorescence method\textsuperscript{3} using two counterpropagating replica pulses suffers from a one-pulse-induced background fluorescence that limits the precision of the temporal measurement. By using second-harmonic generation (SHG), a background-free measurement of the pulse duration can be made. In the original version,\textsuperscript{4} a variable time delay is introduced between the two replicas of the original pulse and for a strictly reproducible pulse train yields the time autocorrelation function of the original pulse. This method is limited to measuring the average characteristics of the pulse train, and, because of phase-matching considerations, the geometry must be changed for different optical wavelengths. A more recent version of the SHG method that is suitable for single-pulse measurements uses a time-to-space mapping of the interaction region of two noncollinear replica pulses.\textsuperscript{5,6} Again this technique is limited because of phase-matching problems.

In this Letter we introduce a new method for the measurement of the temporal profile of a single optical pulse that is based on degenerate four-wave mixing (DFWM).\textsuperscript{7} Among the numerous applications of DFWM,\textsuperscript{8} that most closely related to our present proposal is the time-domain correlator described by O'Meara and Yariv.\textsuperscript{9} Our technique produces a background-free measurement and has none of the phase-matching difficulties associated with the SHG methods. Simple changes in the length of the nonlinear medium will allow measurements of pulse widths to be made in the subnanosecond to subpicosecond time interval.

We assume that two replica pulses of the original optical pulse are constructed with a simple beam splitter and mirror arrangement. The two replicas, with complex envelope functions \(A_1(r,t)\) and \(A_2(r,t)\), counterpropagate in a nonlinear optical medium as shown in Fig. 1. A quasi-cw probe field \(A_3(r,t)\) illuminates the overlap volume of \(A_1\) and \(A_2\). The probe field is of frequency \(\omega\), equal to the carrier frequency of the pulse fields. The phase conjugate wave \(A_3(r,t)\) originating in the overlap region will be shown to have a transverse spatial amplitude distribution which is characteristic of the temporal profile of the pulses \(A_1\) and \(A_2\).

The four coupled wave equations for \(A_1, A_2, A_3\), and \(A_4\) decouple and simplify considerably by assuming nondepletion of \(A_1, A_2,\) and \(A_3\) in the slowly varying envelope approximation. The single equation for the phase conjugate field \(A_3\) is

\[
\left( \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) A_3(r,t) = \frac{i 2\pi \omega x}{cn} A_1 A_2 A_4^*,
\]

where \(x\) is the instantaneous third-order nonlinear susceptibility. We assume arbitrary input amplitude functions of the form

\[
A_1(r,t) = f(\omega t - k x) g(y,z),
\]

\[
A_2(r,t) = f(\omega t + k x) g(y,z),
\]

\[
A_3(r,t) = h(x,y).
\]

Using these functions in Eq. (1) and assuming that the overlap region is completely contained in the nonlinear medium, we obtain a formal solution to Eq. (1) of

\[
A_3(r,t) = \frac{i 2\pi \omega x}{cn} \int h(x,y) \left[ f(\omega t - k (z + x - z')) \right]

\times \left[ f(\omega t - k (z - x + z')) \right] g^2(y,z') dz'.
\]

We will use the asymptotic limit of this solution, valid far from the interaction region where the upper limit of the integral goes to plus infinity. The energy distribution on the detector is then

\[
W_3(x,y) = \frac{1}{8\pi} \int_{-\infty}^{+\infty} A_3(r,t) A_3^*(r,t) dr.
\]

![Fig. 1. Schematic diagram for pulse duration measurements by degenerate four-wave mixing. The spatial distribution of the conjugate wave \(A_3\) contains information about the temporal structure of replicas \(A_1\) and \(A_2\) of the pulse to be measured. The energy contained in \(A_3\) is deposited on the detector (photoplate, diode array, etc.) after reflection from the semitransparent beam splitter. To observe the near-field distribution of the conjugate beam, a one-to-one imaging objective lens should be positioned between the beam splitter and detector plane.](image-url)
We consider two cases of practical interest for which Eq. (4) may be evaluated. In the first case, the transverse dimensions of the replica pulses are assumed to be small compared with their longitudinal dimension. In addition, we assume that the probe field is spatially constant throughout the overlap region. Then Eq. (4) becomes

$$W_g(x) \approx \int_{-\infty}^{\infty} I(\omega t - kx)I(\omega t + kx)dt,$$

where $I$ is the longitudinal intensity profile of the pulse. We note that $W_g$ is a symmetrized spatial image of this pulse intensity profile about $x = 0$, from which the pulse duration can be determined. In this limit of small transverse dimensions, a modified replica of the original pulse can be used as the probe field, which is especially convenient for measurements with tunable picosecond lasers.

For the second case, we consider Gaussian profiles for both the temporal and spatial envelopes of the replica pulses,

$$f = a \exp\left(-\alpha(\omega t \pm kx)^2\right),$$
$$g = \exp\left[-\beta k^2(y^2 + z^2)\right],$$

where $\alpha$ and $\beta$ are related to the pulse width (FWHM) $T$ and the beam waist $w$ by $T = (2 \ln 2/\omega a)^{1/2}$ and $w = (\beta k^2)^{-1/2}$.

With these expressions in Eq. (3) and keeping the probe field spatially constant, the energy distribution of the conjugate wave on the detector is

$$W_g(x,y) \approx \exp[-4k^2(ax^2 + by^2)].$$

The spatial dimension (FWHM) $H$ of this elliptical Gaussian measured along the $x$ direction gives the pulse duration $T$ simply as

$$T = \sqrt{2} H/\alpha,$$

where for generality $\alpha$ is the group velocity of the pulses.

The time resolution of this DFWM method is limited by considerations similar to those found for the SHG time-to-space mapping method.10-12 The temporal spreading of the pulse wave packet due to the dispersive nature of the medium clearly limits the precision of the measurement. This effect can be minimized by keeping the length of the medium as small as possible. The divergence and alignment of beams 1 and 2 do not have a first-order effect on the mapping, but the alignment should be optimized to ensure adequate strength of the conjugate field. Diffraction effects due to the finite size of the interaction region will broaden the image on the detector although beam walk-off problems are not present. For ease of measurement, the spot size on the detector can be increased by using a converging probe beam. The lens is best placed before the beam splitter of Fig. 1. The spectral content of the probe field should be sufficiently large to cover the components of the pulse to enhance the strength of the conjugate field. Given the broad frequency distribution of the fields and especially in the case of frequency chirped or nontransform-limited pump pulses, the observation of the phase conjugate beam should be limited to near field to avoid substructures in the image related to the finite coherence time of the pump pulse.12 To achieve a background-free measurement, backreflection of the probe beam onto the detector should be avoided by slightly tilting the surfaces of the medium. With these limitations in mind, we anticipate an experimentally achievable time resolution of better than 0.1 ps.

In summary, we have presented a method for investigating the temporal structure of short optical pulses using a time-to-space mapping technique employing DFWM. The method is suitable for single-pulse or accumulated multiple-pulse measurements and is background free. Since the technique has no phase-matching difficulties, the geometry is invariant to changes in the pulse wavelength. The temporal resolution is estimated to be <0.1 psec.

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References

Anisotropic 2-D scattering. 3: The effects of incident laser wavelength

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This Letter presents the continuation of the experimental works reported in Refs. 1–3. In Ref. 1 the effects of a finite medium on multiple 2-D scattering from latex paint particles were presented. Multiple scattering from a semi-infinite medium of spherical polystyrene particles of fixed diameter was reported in Ref. 2. An important finding of Ref. 2 was that the intensity distribution curves for different particle sizes collapse to an effective isotropic solution by using (1 - g) as a correlation parameter with the radial optical thickness. $g$ is the asymmetry factor.4 In Refs. 1–3, the incident light was from a He–Ne laser (0.6328 μm), while in Refs. 1 and 2 the carrier of the scattering centers was double distilled water. The effect of the carrier (i.e., its index of refraction) on the scattering of red light was reported in Ref. 3. That is, in addition to suspending the scattering centers in water, high purity ethylene glycol was used. The results of that investigation were consistent with both Refs. 1 and 2, and the two different particle carriers produced essentially the same results. This would indicate that the carrier index of refraction appears to be a relatively insensitive parameter of the backscattered intensity.

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As a next step, the effects of the irradiation wavelength on the multiple scattering in a medium of finite thickness were considered. Consequently, a He–Cd laser beam (0.4414 μm) was required to be incident normal to the scattering medium. In all other respects, particularly in terms of particle size (0.261 μm), particle carriers (distilled water and high purity ethylene glycol), and magnitude of particle concentration (optical depth requirements), this experiment was identical to that presented in Ref. 3.

Therefore, the purposes of this investigation were for incident blue light (1) to demonstrate again that the anisotropic intensity distribution curves will collapse to an effective isotropic solution for a given value of \( \tau_0 \) by using \((1 - g)\) as a correlation parameter and (2) to detect the effect of the refractive index of the liquids on the data received. Thus by comparing these results with those of Ref. 3 it will be possible to determine if the change in the irradiation wavelength adversely affects the data received as compared to theory.

In this experiment, it was assumed that, except for the very slight absorption character of the water or ethylene glycol (liq), the particle-water or the particle-ethylene glycol solutions possess only scattering properties (s). Thus the optical thickness for this investigation was defined as

\[
\tau = \tau_s + \tau_{\text{liq}},
\]

where

\[
\tau_s = \eta C_s L.
\]

Note that \( \eta \) (the volume of scattering particles per total volume of liquid plus scattering particles) and \( L \) (the radius or depth of the medium) are measurable. \( C_s \) (the effective scattering cross section per particle volume) must be calculated.

The laboratory apparatus used to acquire the experimental data is the same as that discussed in Refs. 1–3. Thus the experimental setup and procedure followed exactly parallels that described in Ref. 3 and involved acquisition of data sets, where the sets were distinguished by the volume of scattering particles, the liquid carrier used, and its depth.

The ethylene glycol index of refraction (1.472) was measured; the indices of refraction of water (1.3313) and the polystyrene spheres (1.6201) were calculated. The asymmetry factor \( g \) and \( C_s \) were determined from Mie theory (for water \( g = 0.73 \) and \( C_s = 27992 \text{ cm}^{-1} \); for ethylene glycol \( g =

Fig. 1. Experimental (symbols) and theoretical backscattered nondimensional intensities vs the radial optical thickness for double distilled water medium with a black bottom and 0.4414-μm incident light (○, 5 ml; ▲, 10 ml of spherical particles solution).

Fig. 2. Experimental (symbols) and theoretical backscattered nondimensional intensities vs radial optical thickness for an ethylene glycol medium with a black bottom and 0.4414-μm incident light (○, 20 ml; ▲, 40 ml of spherical particle solution).

Fig. 3. Experimental (symbols) and theoretical backscattered nondimensional intensities vs radial optical thickness for an optical depth parameter of 2, a black bottom, and 0.4414-μm incident light (○, 5 ml in water; ▲, 10 ml in water; ▼, 20 ml in ethylene glycol; ■, 40 ml in ethylene glycol).

Fig. 4. Experimental (symbols) and theoretical backscattered nondimensional intensities vs radial optical thickness in an ethylene glycol medium for an optical depth parameter of 2 and a black bottom (○, 5 ml with 0.4414 μm; ▲, 10 ml with 0.4414 μm; ▼, 5 ml with 0.6328 μm; ■, 10 ml with 0.6328 μm).
0.75 and \(C_x = 7403 \text{ cm}^{-1}\). Thus the relative indices of refraction of latex in water and ethylene glycol are 1.217 and 1.100, respectively. The corresponding size parameters \((n \pi d/\lambda)\) are 2.473 and 2.734. The liquid depth \(z\) was calculated from an imposed restriction that the asymmetric optical depth \(\tau_g\) be limited to values of \(\tau_{90}\), 1, 2, 4, 5, and 10 where

\[
\tau_g = (1 - g)\tau_{90} = (1 - g)nC_xz. \tag{3}
\]

Equation (3) was used under the assumption that \(\tau_g > \tau_{90}\). (The resulting error was <5%.)

A detector probe was moved radially, and detector signal levels were recorded. These output voltages are proportional to the amount of backscattered power transmitted to the PMT via the detector probe. The incident radiant power level was measured periodically and was found to be stable for the time required to take the data.

The incident radiation was assumed to be scattered anisotropically in the particles–liquid suspension dependent upon the effective optical thickness. Raw data pairs (voltage \(v\) and radial distance \(R\)) were recorded for various depths, liquid type, and dispersed particle volumes. A data reduction program from Ref. 8 transformed this raw data into a nondimensional scattered intensity as a function of radial optical thickness \(\tau_R\) (with optical depth \(\tau_g\) as parameter):

\[
\frac{(R/R_0)^2}{I/I_0} = 538.86vR^2 \times 10^{-6}/PL. \tag{4}
\]

Figures 1 and 2 illustrate the program output. The heavy lines are theoretical results.9 These figures point out that the experimentally determined quantities do not exactly match the theoretical results (i.e., radial optical thickness are larger than the corresponding theoretical values and the experimental nondimensional intensities are low). Even so, the appropriate trends are exhibited by the experimental data in terms of slope, shape as a function of \(\tau_R\), and separation due to \(\tau_g\). Figure 3 is a typical comparison of \((R/R_0)^2(I/I_0)\) vs \(\tau_R\) for both liquids for given optical depths \(\tau_g\). Notice that there is fair agreement for all the data presented.

Figure 4 is included to demonstrate that the incident radiation wavelength does not adversely affect the backscattered radiation. This particular figure is typical for ethylene glycol—for water there is a little more spread in the data. The 0.6328-\(\mu\)m data are from Ref. 3.

The data follow the same trends as predicted by the theory, although not exactly. The errors which cause these discrepancies appear to be independent of the medium and the irradiation wavelength. They are assumed to be due to the absorption of the particles and the scattering of the liquid.

In comparing the results found using water as the medium with those of ethylene glycol (Fig. 3), the results are very similar. Also Fig. 4 indicates that the data presentation is not a function of this incident radiation wavelength. Thus, for this wavelength (0.4414 \(\mu\)m), as it was for the 0.6328-\(\mu\)m incident wavelength of Ref. 3,

1. the anisotropic nondimensional intensity distribution depends upon the asymmetry factor \(g\), and, even though the range tested is small, \((1 - g)\) is a correlation parameter in an anisotropic backscattering from a medium of finite depth as it was with the anisotropic backscattering a semi-infinite medium; and

2. the particle carriers produce very similar results.

Therefore, it may be concluded that changing of the incident radiation wavelength does not adversely effect the received data or the agreement between experimental and theoretical intensities.

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References


**High-speed retardation modulation ellipsometer: erratum**

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Equation (12) of this paper1 was misprinted. It should be as follows:

\[
\delta\Psi = \sum_{i=1}^{m} \frac{1}{l} \left( \Psi_{ij} - \Psi_{ij}^0 \right)^2 
\]

Reference


**Significance of the extinction/backscatter ratio and the boundary value term in the solution for the two-component lidar equation**

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Recently, Klett2 provided a stable inversion solution for the lidar equation which yields the quantitative extinction profile as a function of range. Fernald2 also proposed a so-
olution for the two-component lidar equation (including aerosol and molecular terms) in a more general form and qualitatively stated that this solution is most sensitive to errors in the aerosol extinction/backscatter ratio and boundary value at the calibration level when applied to moderately turbid atmospheres. This Letter describes the characteristics of this solution and dependence of its accuracy upon extinction/backscatter ratio and boundary value by means of numerical simulation.

Fernald's solution is written as follows:

$$\alpha_1(z) = -S_1 \alpha_2(z) + \frac{X(z) \exp \left[ \frac{S_1}{S_2} \int_{z}^{z_e} \alpha_2(z') \, dz' \right]}{X(z_e) \exp \left[ \frac{S_1}{S_2} \int_{z}^{z_e} \alpha_2(z') \, dz' \right] + 2 \int_{z}^{z_e} X(z) \exp \left[ \frac{S_1}{S_2} \int_{z}^{z'} \alpha_2(z'') \, dz'' \right] \, dz}$$

where $\alpha$ and $S$ are the volume extinction coefficient and extinction/backscatter ratio, respectively, and the suffixes 1 and 2 are quantities related to aerosol and molecule, respectively. $X(z)$ is the range-squared lidar signal, and $z_e$ is the range from the lidar at which a boundary condition is assigned. Molecular extinction coefficients are, in general, considered to be known from available meteorological data or appropriate standard model atmospheres.

From Eq. (1) it is clear that Klett's solution is a special case of Fernald's where the aerosol extinction/backscatter ratio $S_1$ is formally set equal to molecular extinction/backscatter ratio $S_2$, provided that the power $k$ is equal to unity in Eq. (6) of Klett's original paper. The aerosol extinction/backscatter ratio $S_1$, however, depends on the size distribution and complex refractive index of the aerosols and varies from $-10$ to $100$. Generally, the value $S_1$ is unknown, and an appropriate estimation is necessary. Furthermore, a boundary value for $\alpha_1$ at $z = z_e$ must be known by, for example, the slope method from lidar measurements.

In the following, some numerical calculations are carried out confining the situation to lidar measurements in the horizontal direction. Molecular extinction and backscattering coefficients are derived from the U.S. Standard Atmosphere with the laser wavelength of 0.532 μm. The distribution of aerosol properties are modeled as follows:

$$\alpha_1(z) = T \cdot \alpha_2 \left[ 1 + A \sin \left( \frac{2\pi z}{L} + \varphi \right) \right],$$

$$\beta_1(z) = \frac{\alpha_1(z)}{S_1},$$

where $\alpha$ indicates the model (true) quantity, $\beta_1$ is the aerosol backscattering coefficient, $T$ is an index for turbidity relative to the molecular extinction. $L (= 5 \text{ km})$, $A (= 1.0)$, and $\varphi (= 0.0)$ are the horizontal scale, the amplitude, and the phase of the aerosol distribution. The lidar signal is modeled with the following expression:

$$\epsilon = \frac{1}{N} \frac{\sum_{i=1}^{N} [\alpha_1(z_i) - \alpha_1(z_i)]^2}{\frac{1}{N} \sum_{i=1}^{N} \alpha_1(z_i)}$$

The value of $\epsilon$ is a function of $N$ because the degree of convergence depends mainly on the assumed boundary value and the distance from the boundary. In the present calculation, the summation is done over the entire range ($N = 1000$).

Reconstruction errors were calculated for various combinations of four parameters: (1) turbidity $T$; (2) true extinction/backscatter ratio $S_1$; (3) assumed extinction/backscatter ratio for Eq. (1) $S_1$; and (4) ratio of the assumed boundary value to the true one $B_c$. Some examples are shown in Fig. 2 for the cases of $T = 5$ (solid curve) and $T = 20$ (dashed curve). It can be seen from Fig. 2 that the solution is less sensitive to the boundary value ratio $B_c$ in turbid air ($T = 20$) than in clean air ($T = 5$). Similarly, the solution has a weak dependence on the difference of $S_1$ from $S_{10}$ in turbid air. This is why Klett's solution is better applied to turbid air.

The general tendency that the principal axes of the contours incline down on the right in this case implies that overesti-

![Fig. 1. Aerosol extinction coefficient profiles. Model profile with $S_1 = 50$ and $T = 10$; solution profiles with $S_1 = 90$ and 8.5 ($= S_2$). Boundary values are correctly given.](image)
The aerosol extinction coefficient for the boundary condition is usually estimated by the slope method, which is necessarily an averaged quantity over some range. Consequently, it is important to introduce an averaged value for $X(z_c)$ over the same range interval to reduce the boundary term error due to local inhomogeneities and noise. To this end, the following smoothing procedure would be effective:

$$X(z_c) = \frac{1}{M} \sum_{i=1}^{M} X(z_i) \exp(2 \cdot \tilde{\alpha} \cdot Z_i) \exp(-2 \cdot \tilde{\alpha} \cdot z_{ic}),$$

where $M = (i_2 - i_1 + 1)$ and $i_c = (i_1 + i_2)/2$, and $\tilde{\alpha} = (\alpha_1 + \alpha_2)$ means the averaged extinction coefficient estimated by the slope method applied to the range from $z_{i_1}$ to $z_{i_2}$.

**References**


**Millimeter wave gas/aerosol spectrophone and application to diesel smoke**

Charles W. Bruce and N. M. Richardson

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Recent developments in radiative sources at millimeter wavelengths, coupled with attendant advantages for propagation through aerosols, have resulted in a requirement for absorption coefficients at these wavelengths. The need is made more acute by the absence of complex indices and applicable calculational techniques. In response, prototype in situ millimeter wavelength spectrophone (or acoustical) systems have been developed by one of the authors (Bruce). The spectrophone design of this Letter is conceptually related to earlier gas–aerosol systems developed for use at the major IR transmission window wavelengths. This system was designed primarily with aerosols in mind but can be used to measure absorption by gases, aerosols, mixtures of these, or to separate the gaseous from the aerosol contribution.

Increasing the wavelength of the probe source toward millimeter wavelengths results in a rapidly decreasing scattered proportion for many aerosols of interest (to the fourth power of the particle dimension $r$) since the particulate size spectrum becomes Rayleigh ($2\pi r/\lambda << 1$). Under the same condition, the magnitude of the absorption is often small rendering photoacoustical techniques, which have relatively high sensitivities, appropriate.

The millimeter wave system of this Letter is both acoustically and electrically tuned to obtain higher sensitivity per unit absorption and per unit microwave source power. An impact diode with Cerrire isolator was used operating at 34-GHz wavelength.

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The millimeter wave system of this Letter is both acoustically and electrically tuned to obtain higher sensitivity per unit absorption and per unit microwave source power density. An impact diode with Cerrire isolator was used operating at 34-GHz wavelength.
An output power of ~0.1 W was coupled first into a rectangular waveguide (WR-28) system where the wave was modulated and the forward power optimized. A flow-through cylindrical (circular) absorption cell was coupled to the rectangular portion of the waveguide through a transition. An electret microphone was coupled into the guide to detect the dominant acoustical mode. A ferrite modulator provided the time varying power and a reference signal for the phase sensitive detection system used. Forward and reverse waveguide power were controlled using an E/H tuner and crystal detectors mounted on a bidirectional (cross-guide) coupler.

Cylindrical absorption cells with various diameters have been used largely for the purpose of encouraging particular modes of propagation for the microwave energy. A desirable property for this energy is that its density (Poynting vector) be concentrated laterally in the center of the cell (waveguide cavity) rather than near the walls. This minimizes effects on the sensitivity which occur when particles collect on the cell wall. The cell used for the measurements of this paper had a 1.1-cm i.d. and 20-cm length. The aerosol was introduced at the end of the cell opposite the source of microwave power. This aerosol intake was designed to serve as a partial reflector for the formation of the electromagnetic and acoustical standing waves. Extraction of the aerosol at the opposite end of the absorption cell was accomplished through small holes in the circumference of the cylinder. A plenum over these holes and attached to the outer wall of the cylinder was connected to an aerosol filter and then to the metered flow system. Airflow velocities of ~50–70 cm/sec were employed for the aerosol absorption measurements. The intake portion of the cell was adjustable in position to permit tuning for resonance of the standing electromagnetic wave.

Longitudinal acoustical resonance was obtained by adjustment of the modulation frequency for the maximum acoustical signal with an absorbing gas in the cell. To prevent aerosols from circulating into the source region of the waveguide, a polymer window cut from commercial food wrap was inserted between waveguide flanges near the output end of the rectangular waveguide. A small counterflow of clean air was introduced between the window and absorption cell to prevent accumulation of aerosol particles in this region and particularly on the window.

The spectrophone system was calibrated using gaseous absorption. The extinction coefficients (which equal the absorption coefficients in the absence of appreciable scattering) of Freon 22 were measured in a one-pass transmission chamber which was carefully developed to be free of multipath effects which caused large errors in earlier results. The ratio of the resulting extinction coefficient to the acoustical cell detector signal voltage divided by a representative sample of the E-M wave power density in the guide determines the calibration factor. Calibrations performed at various levels of absorption confirmed linearity of the absorption with concentration in the range employed.

Signal to noise for a signal processing bandwidth of ~10^-1 Hz was found to be <0.05 km^-1 for a measurement (absorption signal minus base line signal), although, in the aerosol mode of operation, the airflow noise raises this to as much as 0.2–0.3 km^-1. While this is adequate for the purposes of this paper, a source of microwave power just received having 2.5 times the power used here is expected to increase the signal to noise by that proportion. The authors have observed that, without the flow-through requirement, the sensitivity can be considerably increased from the above values (i.e., by increasing cavity acoustical isolation, careful ratio-removal of source noise, and use of a higher cavity reflectivity or voltage standing wave ratio).

The smoke aerosol was generated externally, injected into a small aerosol chamber (volume ~70 liters), and stirred lightly throughout the measurements. The diesel fuel was burned on a short cylindrical wick (diam = 1.2 cm) immersed in a small cup having the same diameter. Above the wick, an aluminum cone funneled the smoke through a copper tube, cooling the smoke, into the chamber.

The aerosol chamber (volume ~70 liters) was sampled by the flow-through spectrophone and Gelman filter particle collection systems. Measurements of the smoke were continuously obtained using the spectrophone and periodically using the flow-through filter sampling technique. Separate aerosol filters were used for two purposes though the filter material, and pore sizes were the same, i.e., Nuclepore, polycarbonate with 0.2-μm pore size. One set of filters was used for determination of aerosol mass loading and the other for SEM photos of the particles forming the distribution. To determine the mass loading, each filter was placed in a small hermetically sealed container and weighed. Then the filter was inserted into the sampling system and a timed sample obtained. Posttest weighing, divided by the sampling time and the calibrated flow rate, yielded the mass density of the smoke. The absorption coefficient divided by this number gives the absorption per unit mass density, i.e., the mass absorption coefficient.

The second set of filters was loaded very lightly using low flow rates and short sampling periods to avoid deceptive appearances of in situ coagulation. A sample photo is shown in Fig. 1.

These particles appear predominantly to be stringy (a high length-to-radius ratio). Although those on the filters are generally of the order of 2 μm long, a few are quite large, including those with sufficient extent to be visible to the eye.

It was determined that the particle chemical content is largely carbonaceous (77.5% by weight). The remainder consists of high molecular weight hydrocarbons, mainly alkanes of the general formula C_nH_{2n+2} with 12 ≤ n ≤ 34.4

The mass absorption coefficient for diesel smoke obtained from the measurements described is 6.1 × 10^{-3} m^2 g^{-1}, ±4 × 10^{-4} m^2 g^{-1}, where the latter number is based on the repeatability of the net result. The estimated probable error is higher at 12% emphasizing systematic errors.

Possible absorption contributions due to gaseous products of the combustion process were then investigated by use of a...
removable filter between the smoke chamber and spectrophotometer. This filter was installed so that the insertion itself did not affect the system parameters. When the filter was in the flow circuit the absorption was negligible relative to the level previously obtained for the smoke. Measurement of the total absorption as a function of time after the smoke generation previously obtained for the smoke. Measurement of the total flow circuit the absorption was negligible relative to the level not affect the system parameters. When the filter was in the phone. This filter was installed so that the insertion itself did not contribute.

Although it is desirable to obtain a value for the mass absorption coefficient from theory for comparison with that of the spectrophotometer, the lack of values for the complex refractive index and the complexity of the aerosol with regard to shape and composition (including inhomogeneities) render this very difficult. A very rough approximation can be obtained by assuming that the complex index for the carbonaceous aerosol is constant from visible through millimeter wavelengths. Comparisons of theory and measurements for similar smokes is constant from visible through millimeter wavelengths. If the absorption coefficient is assumed to be inversely proportional to the wavelength, the extrapolated result is of the order of $10^{-5} \text{ m}^2 \text{ g}^{-1}$. No explanation for the difference is apparent.

References

Frequency domain measurements of lag on charge injection devices
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An ideal optical detector illuminated by an impulse of light would yield an electrical signal at the first reading and zero residual signal in subsequent readings. In reality, most detectors such as charge injection devices (CID) do not respond instantaneously but rather exhibit lag.1,2 When a CID pixel is exposed to a unit impulse of light and its output signal held from sample to sample in a sequence of reads, the waveform shown in Fig. 1 results. Notice that the first reading is not unity (assuming 100% detector performance efficiency), and the signal decays exponentially with base $\alpha$. This $\alpha$, which satisfies $0 < \alpha < 1$, is the quantity which we use to describe lag. The lag is of great concern for the CID as an optical detector, because it not only degrades the impulse response but also reduces the charge storage capacity of the detector.

The principle of the CID is that the photogenerated charge, integrated and stored in the metal–insulator–semiconductor (MIS) capacitors, is read out by collapsing the potential well which causes the charge to recombine in (or be injected into) the substrate, thereby inducing a current in an external circuit.1 Such drive line sensing readout is usually configured to operate with a high impedance voltage preamplifier. The physical mechanism of lag in the CID with voltage type readout is twofold: readout lag and device lag. The former is due to the injection pulse loading effect inherent in the voltage sensing scheme, whereas the latter arises from incomplete charge injection or recollection of the injected charge because of the traps or sticky charge in the potential well. Empirical results of an exponential response to a step-function excitation suggest that the trapped charge after each injection is linearly proportional to the charge population in the potential well before injection. Such lag mechanism is essentially a linear system and can be described by a constant $\alpha$.

An accurate measurement of lag in the time domain requires synchronization between the optical pulse and the CID pixel reading time. Furthermore, the residual signal due to lag must be large compared with the noise, which is especially demanding in the case of a low lag situation. Lag measurement in the frequency domain on the contrary is far superior. The idea is to analyze the way lag modifies the frequency response of the output of a detector such as CID. In this Letter, a derivation of the frequency domain lag model will be presented.

The CID is an integrating type of detector, and its output is normally displayed by a sample-and-hold circuit. Assuming a sinusoidal input signal (optical), the frequency response due to integration and sampling can be written as

$$A_s(f) = \lim_{N \to \infty} \frac{1}{N} \sum_{n=0}^{N-1} a(nT) \exp(-j2\pi f nT),$$

where $T$ is the sampling period, which corresponds to the integration time of the device, and $a(nT)$ is the detected signal at $t = nT$ given by

$$A_s(t) = \lim_{N \to \infty} \frac{1}{N} \sum_{n=0}^{N-1} a(nT) \exp(-j2\pi f nT).$$

![Fig. 1. Unit impulse response of a CID detector with laga = 20%.](image-url)
Eq. (2) is the signal integrated in the primary cycle, whereas the subsequent terms represent the delayed residual signal due to lag from previous cycles. The factor \((1 - \alpha)\) accounts for the lag for the corresponding cycle \(t = nT\).

We can now bring the effect of zero-order hold into the expression by writing \(A(f) = A_s(f)H_0(f)\), where \(H_0(f)\) is the transfer function of the zero-order hold with holding time \(T\) given by

\[
H_0(f) = \frac{\text{sinc}(fT)}{\exp(-j2\pi f T)}. \tag{3}
\]

The normalized amplitude frequency response for the whole system can then be expressed as

\[
\left| \frac{A_s(f)}{A(0)} \right| = \frac{(1 - \alpha) \sin^2(fT)}{\sqrt{1 - \alpha^2 + 4\alpha \sin^2(\pi f T)}}. \tag{3}
\]
With the implied normalization understood, the subscript \( n \) of \( A_n(f) \) can be dropped. A plot of Eq. (3) with \( \alpha \) as a parameter is shown in Fig. 2, where the horizontal axis is the normalized frequency \( f(t_f / T) \), \( f_c \) being the chopping frequency and \( t_f \) the integration time. By measuring the optical response at two chopping frequencies, the lags can be deduced from the ratio

\[
\frac{R(\alpha, f_1 / T)}{R(\alpha, f_2 / T)} = \frac{|A(f_1, \alpha)|}{|A(f_2, \alpha)|} \tag{4}
\]

using an iterative computer routine. The need for signal response normalization is thus eliminated. Table I lists the two-frequency response ratio of Eq. (4) as a function of lag for normalized frequency pairs of 0.1/0.3 and 0.2/0.4. Depending on the integration time, the chopping frequency can be properly chosen for good resolution in the lag measurement.

Since the optical signal is normally modulated by a chopper in the optical laboratory, a wave analyzer (e.g., HP302A, \( \Delta f = 6 \) Hz) can be used to select only the fundamental frequency component. The measurement thus involves a pure sinusoidal waveform making automated lag measurements not only possible but also easy to do. As a test of this frequency domain lag model, Fig. 3 plots the frequency response of two indium antimonide CID infrared detectors whose lag was measured to be 10 and 30%, respectively, using the impulse response method (time domain measurement). The fit of data points into the theoretical curves is remarkable, and the agreement is excellent between frequency and time domain approaches.

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References