

Ultraviolet filtered Rayleigh scattering temperature measurements with a mercury filter

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We report the development of ultraviolet filtered Rayleigh scattering as a diagnostic tool for measurements of gas properties. A frequency-tripled narrow-linewidth Ti:sapphire laser illuminates a sample, and Rayleigh scattered light is imaged through a mercury-vapor absorption filter. Working in the ultraviolet improves the signal-to-noise ratio compared with that previously obtained in the visible as the result of an enhanced scattering cross section as well as the nearly ideal properties of the mercury filter. Tuning the laser through the absorption notch of the filter is a means of probing the scattering line shape, which contains temperature information. Temperature measurements of air are shown to have uncertainties of less than 3%. © 1999

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Over the past several years, filtered Rayleigh scattering (FRS) has been employed as a diagnostic in a variety of fluid environments for nonintrusive flow visualization as well as for quantitative measurements of fluid properties. Solving the problem of performing nonintrusive temperature measurements is important for improved understanding of many areas of physics, for example, supersonic flows, weakly ionized plasmas, combustion processes, and even the atmosphere. Depending on the nature of the problem under investigation, a wide array of optical methods is available. FRS is attractive because, by focusing the laser to a sheet, it has the potential to provide a full two-dimensional, quantitative map of the flow field.¹ Typically, spectral measurements, such as absorption and fluorescence, are path integrated. Coherent anti-Stokes Raman spectroscopy and laser-induced thermal acoustics² can be used for point measurements, but they are inherently not well suited for two-dimensional imaging. Raman techniques are species specific and do permit imaging but suffer from low signal levels. Laser-induced methods are amenable to imaging; however, in many cases fluorescence quenching and saturation complicate quantitative analysis. FRS can be used to capture quantitative planar measurements of gas temperature, pressure, density, and velocity; it is nonresonant and thus is unaffected by quenching and saturation, and a high signal-to-noise ratio can be obtained because of the strong background suppression provided by the filter.

In the visible region the FRS technique has already shown its utility in a variety of environments, for example, for temperature measurement in combustion³ and for velocimetry and flow visualization in high-speed flows.^{1,4} Those earlier experiments used primarily frequency-doubled Nd:YAG sources paired with molecular-iodine filters (532 nm); see e.g., Refs. 1, 3, and 4. FRS has also been used in the ultraviolet with an excimer laser with an atomic iron filter (248 nm),⁵ but only modest success has been achieved because of limitations of the laser. Related research by workers in the lidar and other communities have paired narrow-linewidth lasers and filters in the

visible–infrared regions, for example, an alexandrite laser with a potassium filter (770 nm) (Ref. 6) and a dye laser with a cesium filter (389 nm).⁷

The focus of this study is the extension of the FRS technique from the visible to the ultraviolet portion of the spectrum, where the scattering is stronger and the more ideal mercury-vapor filter can be used. Preliminary research characterizing the mercury filter was performed in our laboratory by Finkelstein.⁸ To perform temperature measurements, one images Rayleigh scattering from a gas sample through a mercury filter, and a model is used to fit for gas properties. Because of the frequency dependence of the scattering cross section as well as favorable properties of the filter, higher signal-to-noise ratios are possible in the ultraviolet than in the visible or the infrared.

The FRS concept was described earlier,¹ so only a brief summary is given here. A narrow-linewidth laser is used to illuminate a sample volume, and the scattered light is imaged through a narrow-band atomic- or molecular-vapor absorption filter onto a detector. The amount of scattered light that arrives at the detector depends on the spectral overlap of the scattered light and the filter absorption profile. The scattered light consists of an elastic background component from window scattering and stray reflections as well as of Rayleigh light scattered from the sample volume. The light scattered by the molecules in the sample volume has a Rayleigh–Brillouin line shape that depends primarily on temperature, with a weaker dependence on pressure. One can determine temperature by measuring the scattering line shape with an absorption filter. Tuning the laser causes the Rayleigh–Brillouin line shape to sweep across the absorption filter profile, causing the transmitted light to vary in intensity. Because the absorption profile of the filter is known, the transmitted intensity profile can be deconvolved to yield the Rayleigh–Brillouin line shape, from which the temperature can be determined. This concept is shown schematically in Fig. 1. Note that when an appreciable background is present one may either perform a background subtraction or alternatively take data only in a smaller spectral

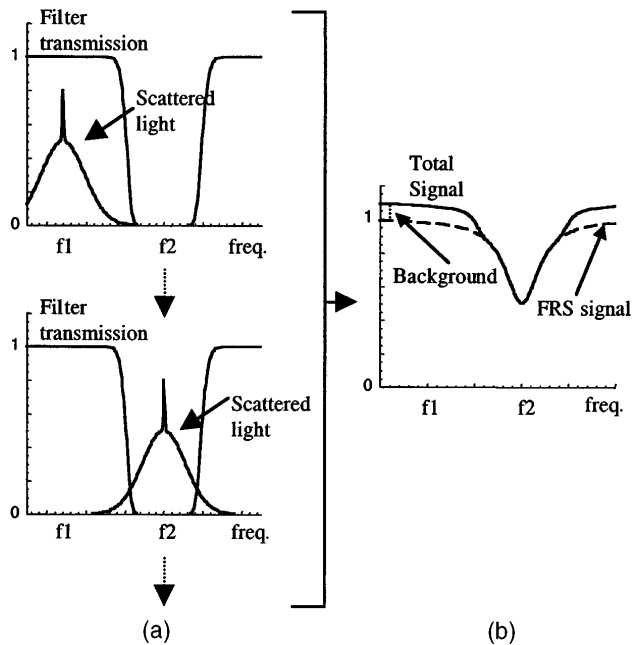


Fig. 1. (a) Spectral overlap of the scattered light with the filter transmission profile at two laser frequencies, f_1 and f_2 . The scattered light has two components, a broadband Rayleigh-Brillouin component and a narrow-band background component. (b) Tuning the laser through the absorption notch yields the FRS signal as a function of frequency. A model is used to fit the FRS data for gas properties.

region about the absorption line center where all background is strongly absorbed. The total Rayleigh scattering signal is proportional to density, so when the gas is in pressure equilibrium and the pressure is known, the temperature can also be determined from the density by use of the ideal gas law.

In FRS, the absorption filter serves two critical purposes: background suppression and probing of spectral information. Desirable filter properties are high out-of-band transmission, high in-band absorption, steeply sloping walls, and flexibility in selection of the absorption width. Mercury has an optically accessible strong ground-state transition at 253.7 nm and is well suited as a filter material. A model for mercury absorption has been developed earlier.⁸ Unlike iron and lead, mercury has a high vapor pressure (e.g., ~ 0.001 Torr at $\sim 20^\circ\text{C}$), so useful vapor number densities can be attained at manageable temperatures. In a 5-cm cell at low vapor pressure (of the order of 0.01 Torr), lines from each of mercury's six naturally occurring isotopes form separate notches with widths up to several gigahertz and filter walls that rise from 10% to 90% transmission over hundreds of megahertz. At a higher vapor pressure (of the order of 1 Torr), the isotopic lines blend together and form a notch with a width of tens of gigahertz. The out-of-band transmission is close to 100% (when filter window losses are neglected), and the in-band optical suppression is predicted to be ten orders of magnitude or more. We control the vapor pressure by setting the temperature of a side arm that contains a small amount (several grams) of liquid mer-

cury. The sidearm is immersed in a temperature-controlled liquid bath (water or mineral oil). The main body of the filter is a quartz tube of 5-cm diameter and 5-cm depth. To prevent any condensation in the main tube we heat the body of the tube to a temperature slightly higher than that of the sidearm.

A custom-built Ti:sapphire laser system⁸ is used as the illumination source for the ultraviolet FRS measurements. The laser is injection seeded and operates with a novel cavity-locking scheme, which ensures almost transform-limited narrow-linewidth single-mode output. The laser runs at 10 Hz and yields as much as 40 mJ of energy per pulse in the ultraviolet (third harmonic). The third-harmonic output from the laser system is delivered along the axes of a cylindrical cell with antireflection-coated windows that houses the sample gas. A 50-cm focal-length lens is used to focus the beam to a waist of $\sim 100\ \mu\text{m}$. To minimize any background light we pass the beam through several irises. A half-wave plate is used to ensure the correct orientation of the linearly polarized beam, and a quarter-wave plate compensates for any elliptical polarization introduced at the window. All the beam-shaping optics are antireflection coated. An antireflection-coated 5-cm focal-length lens is placed 8 cm from the beam and is used to image the Rayleigh scattered light at an observation angle perpendicular to the beam and cell axes. An iris of ~ 1 -mm diameter is placed between the lens and the cell and serves to define the sample region as well as further to reduce background light. The scattered light is passed through the mercury-vapor filter and then passed through a ISA H20 monochromator set to 254 nm, which acts as a broad passband filter. Finally, the scattered light is detected with an R-960 Hamamatsu photomultiplier tube (PMT). A quartz flat is used to pick off a fraction of the beam, after it has passed through the cell, to serve as a power and frequency reference. The picked-off beam is incident upon a diffuser, and elastically scattered light from the diffuser is measured with a photodiode for power normalization as well as passed through a second mercury-vapor absorption filter and measured with a second PMT to serve as a frequency reference. Thin-film interference filters (Corion G25-254-F) are placed in front of the reference photodiode and the PMT such that only the 254-nm light will be detected. The signals from both PMT's and the photodiode are collected with a Stanford Research Systems boxcar unit and a personal computer. A Stanford Research Systems Model 330 preamplifier unit is used at $5\times$ magnification to amplify the signals from the FRS PMT as well as the photodiode. Great care is taken to ensure linearity of the entire detection scheme.

To demonstrate the technique we performed temperature measurements of air. The basic procedure was to scan the laser across the filter absorption and collect the FRS signal as a function of frequency. We normalized the FRS signal, using the reference photodiode, and the frequency axis was established with the reference PMT. A thermocouple inside the sample cell was used as a reference measure of temperature. Data obtained from a typical scan, in this case of air at 50 Torr and $295 \pm 2\ \text{K}$, are shown in

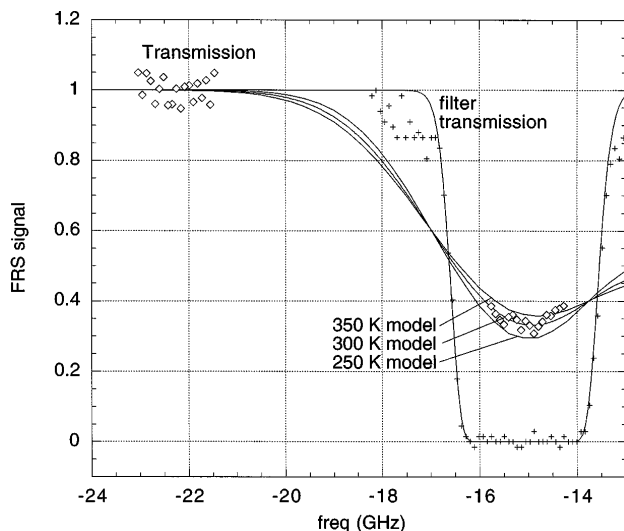


Fig. 2. Ultraviolet FRS data from air at $T = 295 \pm 2$ K and $P = 50$ Torr (open diamonds). The reference frequency axis data (crosses) are also shown, as well as model fits for air at $T = 250, 300, 350$ K and $P = 50$ Torr. The fitting program returned $T = 304 \pm 10$ K.

Fig. 2. The data shown are 100-shot averages. Using the mercury absorption model as well as a fitting routine originally developed for visible FRS,¹ we extracted temperatures from the data. For example, the fit to the data in Fig. 2 returned a temperature of 304 ± 10 K. The data shown in Fig. 2 are after background subtraction. We determined the background by bringing the test cell to vacuum. Preliminary measurements were made at three conditions: $T = 295 \pm 2$ K, $P = 1$ atm; $T = 295 \pm 2$ K, $P = 50$ Torr; $T = 330 \pm 3$ K, $P = 1$ atm. In all cases the fitting routine returned temperatures within 3% of the actual temperature. The model-and-data fitting procedure contains no free parameters or calibration points and does account for the laser line width as well as geometric effects. Uncertainties are determined by the fitting routine from noise in the data. An integral part of the fitting routine is the S6 model created by Tenti *et al.*⁹ for the Rayleigh-Brillouin line shape. The S6 model is designed for a single species of polyatomic atoms and neglects the presence of all branches of rotational Raman scattering. This model has been successfully used by several other researchers.^{1,4,7}

Comparing the signals obtainable with a frequency-doubled Nd:YAG system with those from a frequency-tripled Ti:sapphire system shows the benefit of working in the ultraviolet. The total scattering cross section can be given as¹

$$\sigma = [32\pi^3(n-1)^2/3\lambda^4N^2](6 + 3\rho_0/6 - 7\rho_0),$$

where n is the index of refraction, N is the number density of scatterers, λ is the excitation wavelength, and ρ_0 is the depolarization. The cross section scales as inverse wavelength to the fourth power, though this scal-

ing is offset by an energy per photon scaling, resulting in a cubic dependence of the number of scattered photons for a given beam energy. Decreasing the wavelength from 532 to 254 nm (a factor of 2.09) yields a gain of 9.19 from the wavelength cubed and a gain of 1.16 owing to the increased index of refraction and depolarization.¹⁰ Thus, for a given beam energy, one has 10.7 times more Rayleigh scattered photons at 532 than at 254 nm. Background absorption for the mercury filter is much stronger (suppression of 10^{10} and greater for a typical 5-cm cell) than for the iodine filter (limited to 10^5 by background continuum absorption), so there is a significant reduction in background noise. This is particularly important in cases in which background scattering is strong compared with the Rayleigh signal, such as when windows or walls are in close proximity to the sample volume or when many particulates are present.

In conclusion, we have used ultraviolet filtered Rayleigh scattering to perform accurate temperature measurements in gases, using a mercury absorption filter paired with a Ti:sapphire laser source. Measurements with uncertainties of approximately 3% were obtained under several conditions. The superior spectral properties of the mercury filter as well as the frequency dependence of the Rayleigh cross section indicate that higher signal-to-noise ratios are attained by shifting from the visible to the ultraviolet.

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