Temperature measurements by coherent Rayleigh scattering

Xingguo Pan

Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544

Peter F. Barker

Department of Physics, Heriot–Watt University, Edinburgh EH14 4AS, UK

A. Meschanov

Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544

J. H. Grinstead

NASA Ames Research Center, Moffat Field, California 94035

Mikhail N. Shneider and Richard B. Miles

Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544

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We demonstrate, for the first time to our knowledge, the utility of coherent Rayleigh scattering (CRS) for temperature measurements in low-density gases and weakly ionized plasmas by measuring the translational temperature of neutral argon in a glow discharge. By analysis of the near-Gaussian spectral profile of the CRS signal, we determine temperatures with an uncertainty of ≤3%. We also investigate the intensity range over which this simple Gaussian analysis can be used for temperature measurements and discuss its potential for gas diagnostics. © 2002 Optical Society of America

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Fast and accurate optical measurements of temperature are often required in combusting or high-speed flows for which high temperatures or pressures or highly reactive environments preclude the use of physical probes.[1,2] Spontaneous Rayleigh scattering is one technique that has found wide application for measurements of translational temperature.[3,4] It is nonresonant, and the temperature can be determined by a relatively simple spectral analysis based on its Gaussian spectral profile in the Knudsen regime. For Rayleigh scattering, the Knudsen regime corresponds to the case in which the mean free path is much greater than the fringe spacing formed between the incident and the scattered fields and depends on the angle of observation. In this Letter, we report on the application of a new technique called coherent Rayleigh scattering[5] (CRS) for temperature measurements in the Knudsen regime. According to Ref. 5 and our numerical calculations, the CRS spectral profile is well approximated by a Gaussian function whose width is 9.75% wider than the spontaneous Rayleigh scattering profile. The width of the profile is proportional to $\sqrt{T/M}$, where T is the temperature and M is the atomic or molecular mass of the species.

The CRS signal results from phase-matched scattering from a laser-induced density grating that is created in a gas or a plasma by the electrostrictive forces from within the interference pattern of two counterpropagating pump beams with electric fields $E_1(x, t)$ and $E_2(x, t)$ and frequencies ω_1 and ω_2. This periodic force perturbs the spatial velocity distribution of particles that are moving close to velocity $v = \Omega/q$, forming a volume density grating that travels at the same speed. The beat frequency of the electric field is given by $\Omega = \omega_1 - \omega_2$, and $q = |\mathbf{k}_1 - \mathbf{k}_2|$ is the wave vector of the beat wave, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of pump beams 1 and 2, respectively. The magnitude of the perturbation at each beat frequency (velocity) is proportional to the square of the density at that frequency (velocity) in the distribution function.

The CRS signal is formed by Bragg scattering of a probe beam from the induced gratings, and the intensity of the scattered CRS signal, I_4, is given by[5]

$$I_4 \sim \alpha^4 L^2 \delta\rho^2 I_1 I_2 I_3 , \tag{1}$$

where α is the polarizability of the atom or molecule, L is the interaction length, I_1 and I_2 are the intensities of the two pump beams, and I_3 is the intensity of the probe beam. Density perturbation $\delta\rho = \rho_0 \int_{-\infty}^{\infty} \delta f \mathrm{d}v$ is derived from the perturbation δf to velocity distribution function f_0, and its variation with velocity determines the spectral profile of the scattered light. Perturbation δf is calculated from the one-dimensional, collisionless Boltzmann equation

$$\frac{\partial f}{\partial t} + \nu \frac{\partial f}{\partial x} + \frac{F}{M}\frac{\partial f}{\partial \nu} = 0 , \tag{2}$$

where the distribution function is given by $f = \delta f + f_0$ and F is the electrostrictive force generated by the counterpropagating pump beams, which has the form

$$F = \frac{\alpha}{2}\frac{\partial}{\partial x}(E_1 + E_2)^2 . \tag{3}$$

To demonstrate the utility of CRS as a temperature diagnostic we measured the temperature of an argon glow discharge. The experimental setup is shown in Fig. 1. A frequency-doubled Q-switched Nd:YAG laser was used to produce two broadband pump beams

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that simultaneously induce a superposition of stationary and traveling-wave density perturbations. The multimode bandwidth of this laser was measured to be approximately 60 GHz full width at half-maximum at 532 nm. The laser was split into two beams by a 50/50 beam splitter, and each beam was focused into the tube by two 500-mm focal-length spherical lenses. The beams passed off axis through each lens and intersected at the center of the plasma tube at an angle of approximately 178°. The energy of each pump beam was approximately 20 mJ/pulse, and the polarization of each beam was parallel to the plane of the page. The frequency-doubled output of a narrow-frequency injection-seeded Nd:YAG laser was used to produce the probe beam, which propagated counter to pump beam 1. It was orthogonally polarized to the pump beams and was offset in frequency (~30 GHz) to prevent interference from other four-wave mixing processes.[6] This probe laser was tunable over a frequency range of ~9 GHz. The generated CRS signal propagated counter to pump beam 2 and was orthogonally polarized to the pump beams. A thin-film polarizer placed in the path of pump beam 2 extracted this beam for analysis.

When the plasma tube was filled with argon without a discharge, the CRS signal could easily be detected with the naked eye on a white card at atmospheric pressure. We used a temperature controlled plane-parallel Fabry–Perot etalon to spectrally resolve the CRS signal. This etalon had theoretical finesse of 75 and a free spectral range of 14 GHz. The convolution of the instrument function and the laser linewidth was found to have a full width at half-maximum of 150 MHz. Approximately 4% of the CRS signal was directed to another photomultiplier tube for signal normalization. We determined the spectral profile of the CRS scattered light (~3 GHz) by scanning the narrow-band probe laser across the transmission profile of the etalon, and the signal was detected by a photomultiplier tube. The CRS signal for each laser pulse was integrated on a boxcar averager and recorded on a computer at each frequency step of the laser.

The plasma tube had an inside diameter of 2 cm and was operated at a pressure of 5000 Pa and a current of 10 mA. Because of the shallow crossing angle in the discharge tube, the interaction region had a transverse spatial resolution of 100 μm and a longitudinal resolution of 1 cm. The ionization ratio of the glow discharge was estimated to be 10^{-7}, and therefore the coherent light scattered from electrons or ions could be neglected.

Figure 2 is a plot of the coherent Rayleigh scattering signal from the argon discharge, the recorded instrument function, and a best fit to the data. A nonlinear Levenberg–Marquardt fitting routine was used to fit a convolution of a Gaussian function and the measured instrument function to the data. The raw spectra for each graph (not shown) had a 250-MHz modulation owing to the longitudinal mode structure of the pump laser. This modulation was observed as a well-defined frequency in the power spectrum of the recorded profile, which was filtered out in the frequency domain of a Fourier transform of the recorded data. The sharp peak in the CRS spectrum near line center is due to elastic scatter of the probe laser from the plasma tube walls. It does not coincide with the center of the CRS spectrum because it is produced off axis, entering the etalon at a slightly different angle to the CRS signal. A neutral plasma temperature of 479 ± 13 K was derived from the fitted width of the Gaussian function. As a check on our analysis we also measured the temperature of argon in the plasma tube without a discharge. A plot of this spectrum, the instrument function, and the fit to the data are shown in Fig. 3. Using the same analysis as described above, we derived a temperature of 294 ± 7 K. The gas temperature was measured directly by a thermocouple to be 293 ± 1 K, in good agreement with the CRS value.

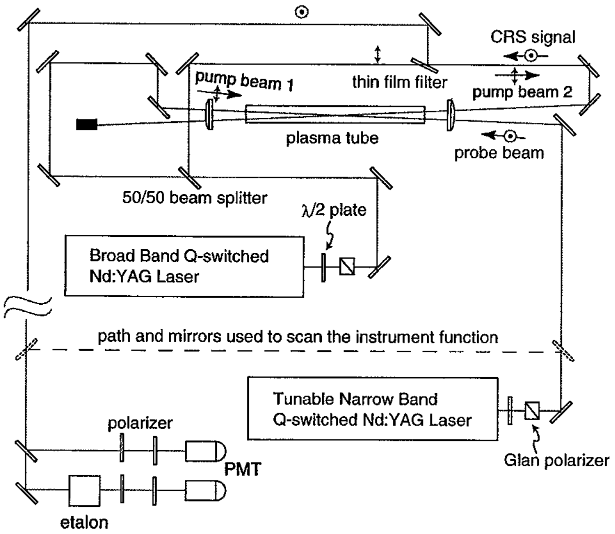

Fig. 1. Schematic of the coherent Rayleigh scattering experiment used to measure the temperature of an argon glow discharge: PMT, photomultiplier tube.

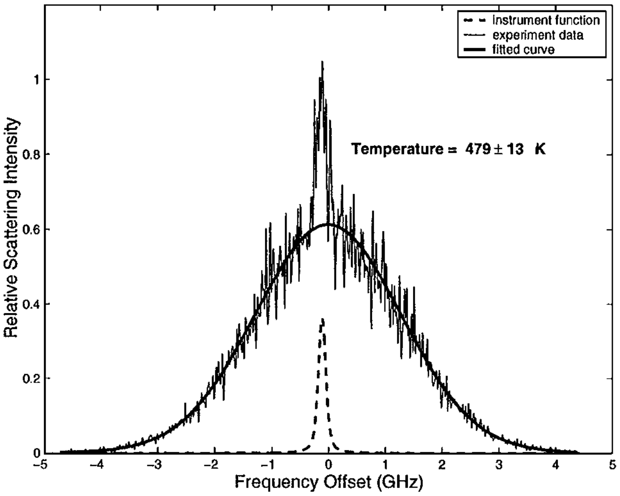

Fig. 2. Spectrally resolved CRS signal from an argon glow discharge at 5000 Pa and a current of 10 mA. The instrument function and a curve fitted to the data are also shown, as is the temperature calculated from the curve fit.

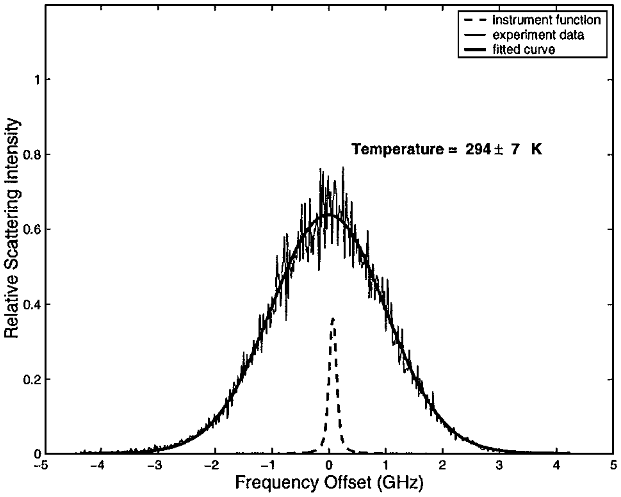

Fig. 3. Spectral profile of CRS signal for argon (without discharge) at a pressure of 50 mbars. The instrument function and a curve of best fit to the data are also shown. The temperature shown was derived from the width of the fitted function as described in text; it was independently measured by a thermocouple to be 293.0 ± 1 K.

For the analysis described above we have assumed that the perturbation to the velocity distribution function δf caused by the periodic electrostrictive force is small compared with Maxwell–Boltzmann distribution function f_0 for a particular traveling-wave velocity. Formally, we assumed that $\delta f \ll f_0$ and that $\partial(\delta f)/\partial v \ll \partial f_0/\partial v$. With these approximations the Boltzmann equation is linear and the perturbations at different frequency components are linearly independent. We verified by numerical integration of the Boltzmann equation that, when the electrostrictive potential well depth is a small fraction of the average kinetic energy of the gas ($1/2\alpha E_1 E_2 \ll kT$), our approximations are valid. For argon at $T = 300$ K, with a polarizability of $\alpha = 1.82 \times 10^{-40}$ cm^2/V, line shapes that differ significantly from the Gaussian shape predicted by our linear analysis[7] would require pump intensities greater than the above-threshold ionization intensity (10^{13} W/cm^2). Inasmuch as the polarizability of most atoms and molecules varies by only an order of magnitude, this analysis can be applied to most gases.[8]

We have demonstrated that one can use coherent Rayleigh scattering to make accurate measurements of temperature in weakly ionized plasmas, and we concluded that it is particularly suited to measurements in low-density plasmas, for which the Knudsen regime can be met with available laser sources and geometries. Like spontaneous Rayleigh scattering, CRS may also be used at high densities outside the Knudsen regime, but in this regime acoustic motion and thermal diffusion become important, and the simple Gaussian line shape analysis used here cannot be applied. We have found that elastically scattered light from the probe beam is one of the limiting interferences of this technique at low gas densities, primarily because of a difficulty in separating the narrow-bandwidth scattered probe light from the CRS signal. Spatial filtering of the signal beam, which was not required in this experiment, should reduce this interference.[9] The short period density grating created by the nearly counterpropagating pump beam geometry maximizes the Knudsen number and therefore the density range over which the simple Gaussian analysis described here is valid. This CRS geometry also maximizes the spectral width of the coherently scattered light. The multimode nature of the pump lasers and the temporal jitter (4 ns) of the probe laser necessitated long integration times to reduce the measurement uncertainty to that which has been achieved here. We believe that this uncertainty can also be achieved in a single or a few shot measurements when the spectral profile of the CRS signal is recorded on a two-dimensional detector and a modeless dye laser is used to form the pump beams.[10]

The application of CRS as an alternative to Rayleigh scattering appears promising in low-density environments in which a strong signal can be generated over a long interaction length. It also appears to be an attractive alternative to laser-induced grating techniques in the Knudsen regime, where the utility of these coherent techniques is poor.[11,12] Finally, the Rayleigh scattering cross section is orders of magnitude stronger than the Raman scattering cross section, and therefore it may find application for temperature measurement in environments in which coherent anti-Stokes Raman spectroscopy is currently used.

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