CHARACTERIZATION OF THERMAL PLASMAS BY LASER LIGHT SCATTERING

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ABSTRACT

Characterization of an atmospheric pressure free-burning arc discharge and a plasma jet by lineshape analysis of scattered laser light is described. Unlike emission spectroscopy, this technique provides direct measurement of plasma gas temperature, electron temperature and electron density without the assumption of local thermodynamic equilibrium (LTE). Plasma gas velocity can also be determined from the Doppler shift of the scattered laser light. Radial gas temperature, electron temperature and electron density profiles are presented for an atmospheric pressure argon free-burning arc discharge. These results show a significant departure from LTE in the arc column, contradicting results obtained from emission spectroscopy. Radial gas temperature and gas velocity profiles in the exit plane of a subsonic atmospheric pressure argon plasma jet are also presented. In this case, the results show the plasma jet is close to LTE in the center, but not in the fringes. The velocity profile is parabolic.

INTRODUCTION

It is not possible to determine thermal plasma gas temperature from emission spectroscopy unless the plasma is in local thermodynamic equilibrium (LTE). Because electron densities are high ($> 10^{23}$ m^{-3}), kinetic processes of atmospheric pressure free-burning arc discharges and thermal plasma jets are dominated by electron collisions. It has therefore been generally assumed that LTE exists in these plasmas [1-3], and emission spectroscopy could be used to accurately measure the gas or kinetic temperature of the plasma. A more realistic description of the plasma is probably that of partial local thermodynamic equilibrium (PLTE), which assumes that the free electrons are in equilibrium with the populations of the upper excited states, but not necessarily with the ground state [4,5]. If the plasma is in PLTE, emission spectroscopy can determine the electron temperature, but not the gas temperature.

Analysis of the lineshape of laser light scattered by the plasma is probably the only direct and unintrusive method of determining gas temperature, electron temperature, electron density, and gas velocity without the assumption of LTE and with a high degree of spatial resolution. We describe in this paper the characterization of thermal plasmas by lineshape analysis of scattered laser light. Radial gas temperature, electron temperature and electron density profiles determined by this technique are presented and discussed for an atmospheric pressure argon free-burning arc discharge. Radial gas temperature and gas velocity profiles in the exit plane of a subsonic atmospheric pressure argon plasma jet are also presented and discussed.

THEORY

Scattering of electromagnetic radiation by a medium is due to density fluctuations within the medium [6]. In the case of ionized gases, density fluctuations of the atoms and ions give rise to Rayleigh scattering, while density fluctuations of the free electrons give rise to Thomson scattering. The total scattered light signal from a thermal plasma is therefore a combination of Rayleigh scattering from atoms and ions, and Thomson scattering from free electrons. The lineshape of Thomson scattered light has two components. One component, called the electron feature, is due to the density fluctuations of the free electrons themselves. The other component is known as the ion feature and is due to the electrostatic influence of the density fluctuations of the ions on the free electrons. The lineshape of Rayleigh scattered light is Gaussian for our experimental conditions [7]. The spatial radiation pattern of both Rayleigh and Thomson scattered light is that of the oscillating electric dipole.

The Thomson lineshape can be written as [6]

$$S(\vec{k},\omega) = \frac{2\pi}{k}\left|1 - \frac{G_e}{\epsilon}\right|^2 f_{oe}\left(\frac{\omega}{k}\right) + \frac{2\pi Z}{k}\left|\frac{G_e}{\epsilon}\right|^2 f_{oi}\left(\frac{\omega}{k}\right), \tag{1}$$

where k is the magnitude of the difference between the scattered wavevector and the incident wavevector $(\vec{k} = \vec{k}_s - \vec{k}_o)$, ω is the difference between the angular frequency of the scattered light and the incident laser $(\omega = \omega_s - \omega_o)$, and $\epsilon = 1 + G_e + G_i$ is the longitudinal dielectric constant of the plasma. The functions G_e and G_i are screening integrals defined by

$$G_e(\vec{k},\omega) = \lim_{\gamma\to 0}\int_{-\infty}^{\infty} \frac{4\pi e^2 n_e}{m_e k^2} \frac{\vec{k}\cdot\frac{\partial f_{oe}}{\partial \vec{v}}}{\omega - \vec{k}\cdot\vec{v} - i\gamma}\, d\vec{v}, \tag{2}$$

and

$$G_i(\vec{k},\omega) = \lim_{\gamma\to 0}\int_{-\infty}^{\infty} \frac{4\pi Z e^2 n_i}{m_i k^2} \frac{\vec{k}\cdot\frac{\partial f_{oi}}{\partial \vec{v}}}{\omega - \vec{k}\cdot\vec{v} - i\gamma}\, d\vec{v}, \tag{3}$$

where the f's are the generalized one dimensional velocity distribution functions for electrons and ions, denoted by the subscripts e and i respectively, the m's are the electron and ion masses, the n's are the electron and ion number densities, e is the electron charge, and Z is the ion charge, equal to one for this experiment. It is assumed that the electrons and ions have Maxwellian velocity distributions characterized by electron and ion temperatures T_e and T_i. We have in one dimension for electrons

$$f_{oe}(v) = \left(\frac{m_e}{2\pi k_B T_e}\right)^{1/2} \exp\left(-\frac{m_e v^2}{2 k_B T_e}\right), \tag{4}$$

and for ions

$$f_{oi}(v) = \left(\frac{m_i}{2\pi k_B T_i}\right)^{1/2} \exp\left(-\frac{m_i v^2}{2 k_B T_i}\right), \tag{5}$$

where k_B is Boltzmann's constant. It is also assumed that the ion temperature equals the gas temperature. The first term in Eq. (1) is the electron feature and second term is the ion feature. The electron feature is considerably broader than the ion feature, scaling roughly as $(m_i/m_e)^{1/2}$. Plots of the electron feature and ion feature where the scattering angle is 90° and the incident laser wavelength is 532 nm for a hypothetical argon plasma with T_e = 15000 K, T_i = 13000 K, and $n_e = 6.40 \times 10^{22}$ m^{-3} are presented in Figs. 1 and 2, respectively. The two peaks in the electron feature are due to scattering from electron waves in the plasma. The effect of scattering from ion-acoustic waves is seen in the two humps of the ion feature.

The plasma gas velocity $\vec{v}$ can be determined directly from the Doppler shift of the ion feature relative to the frequency of the incident laser by

$$\Delta\omega_D = \vec{k}\cdot\vec{v} \ . \quad (6)$$

EXPERIMENTAL

Lineshape measurements were made in a vertical free-burning arc and in the jet of a commercial subsonic plasma spay torch, both operated with argon at atmospheric pressure (640 mm Hg). The arc was generated by a standard gas-tungsten arc (GTA) welding torch using a 2.4 mm diameter thoriated-tungsten cathode ground to a 60° included tip angle, and was operated over a water-cooled copper anode. The cathode-to-anode gap was 9 mm and the arc current was 100 A. The plasma spay torch was operated 900 A with a gas flow rate of 35.4 l min^{-1}. The torch nozzle diameter was 8 mm.

Because of the intense plasma background radiation and relatively weak signal strengths, the use of a high-powered pulse laser and gated detection is necessary to measure the scattered light signal.

Resolving the electron feature does not require high resolution spectroscopy. For these measurements, a frequency-doubled pulsed Nd:YAG laser generating 10 ns pulses at a wavelength of 532 nm and a scanning 1.3 m monochromator with a 1200 groove mm^{-1} grating were used. The laser pulse rate was 10 Hz. The scattered laser light pulses were detected with a photomultiplier tube (PMT) and boxcar averager synchronized to the firing of the laser. The output of the boxcar averager, averaged over 10 laser shots, was digitized by an analog-to-digital (A-D) converter and stored on a computer. A spectral range of ~ 16 nm centered about 532 nm was scanned for a typical electron feature. The laser beam was normally incident to the flow axis of the arc discharge and the scattering angle was 90° to the flow axis and incident laser beam. The GTA torch was mounted on a translation stage, and radial positions were determined by translating the torch relative to the incident laser beam. The spatial resolution of the measurements was $\sim 3 \times 10^{-3}$ mm^3. A half-wave plate was used to rotate the polarization of the incident laser beam to maximize the scattering signal and to verify that the dependence of the signal intensity on the polarization angle was characteristic of oscillating electric dipoles and not stray light. A schematic of the setup to measure the electron feature is given in Fig. 3.

Measurement of the ion feature does require high resolution spectroscopy. The use of a high-powered pulse laser and gated detection is still necessary, but in this case, the incident laser bandwidth must be less than the bandwidth of the ion feature. The fairly recent availability of injection-seeded Nd:YAG lasers fulfills these requirements. Accordingly, the

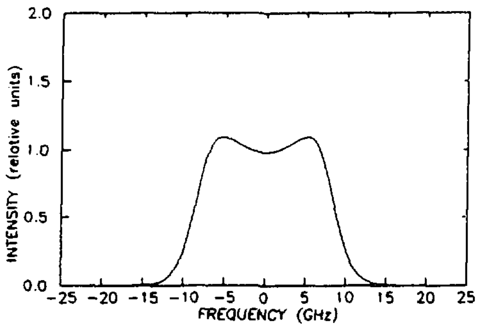

Figure 1. *Ion feature for a hypothetical plasma with* T_e = 15000 K, T_i = 13000 K, and n_e = 6.4 x 10^{22} m^{-3}.

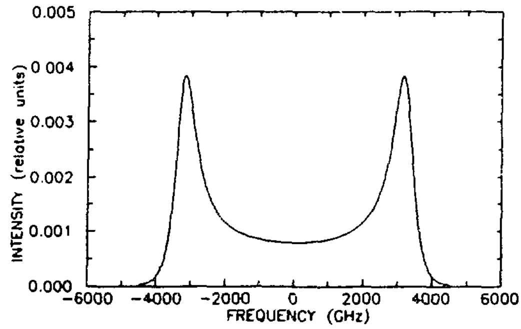

Figure 2. Electron feature for a hypothetical plasma with T_e = 15000 K, T_i = 13000 K, and n_e = 6.4 x 10^{22} m^{-3}.

experimental setup for resolving the ion feature was similar to the electron feature experiment, except that a pulsed frequency-doubled injection-seeded Nd:YAG laser operating at a wavelength of 532 nm with a pulse duration of 10 ns and pulse rate of 20 Hz was used as the laser source. The bandwidth of the laser is < 100 MHz. Furthermore, a scanning tandem Fabry-Pérot interferometer (F-PI) was used for spectral analysis of the scattered light. The scattering angle for ion feature measurements made in the free-burning arc was also 90° and normal to the flow axis and incident laser beam. The plasma jet was operated vertically with the scattered light collected at an angle of 10° from the flow axis in the plane formed by the flow axis and the incident laser wave vector which was normal to the flow axis. This gave a scattering angle of 80°. More details about the measurement of the ion feature can be found elsewhere [7]. The schematic for the ion feature experiment is presented in Fig. 4.

RESULTS AND DISCUSSION

Both electron and ion features were recorded in the free-burning arc, but only the ion feature was recorded in the plasma jet. An experimentally resolved electron feature taken in the free-burning arc is shown in Fig. 5. The electron temperature and electron density were determined from a nonlinear least squares fit of Eq. (1) to this data. This fit is represented by the solid curve. The value of T_e stated in Fig. 5 is strongly influenced by laser heating, and the data must be corrected for this, as described elsewhere [8]. Ion temperatures and electron densities are not effected by laser heating. A typical experimental ion feature from the plasma jet is shown in Fig. 6. In general, these lineshapes are treated as a superposition of Thomson and Rayleigh scattering. The solid curve now represents a nonlinear least squares fit of Eq. (1) superposed with a Rayleigh component described by a Gaussian function to the raw lineshape data. In this case, the contribution from Rayleigh scattering to the total lineshape is < 1%. The ion or gas temperature was determined from this fit. The peak in the center, which was fitted by a Gaussian superposed with the total lineshape function, was the response of the F-PI to the incident laser light. This provides a reference frequency from which Doppler shifts are measured and a measurement of the F-PI instrument response function for deconvolution purposes. The lineshape shows a definite Doppler shift due to the flow velocity of the jet relative to the reference frequency. The gas velocity was determined from this shift using Eq. (6).

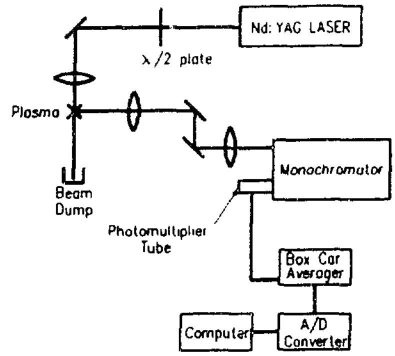

Figure 3. Experimental setup to measure the electron feature.

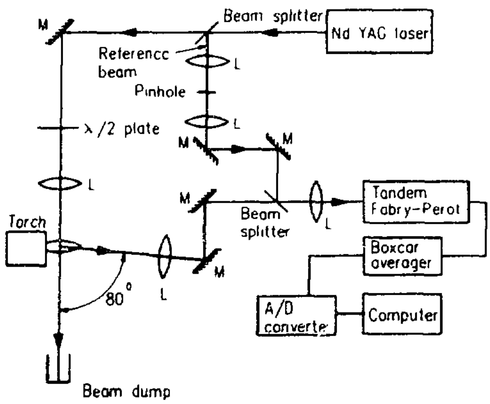

Figure 4. Experimental setup to measure the ion feature.

Radial profiles of gas temperature and electron temperature, corrected for laser heating, of the free-burning arc at 2 mm below the cathode determined from analysis of scattered laser light are presented in Fig. 6. A significant departure from LTE is evident throughout the arc, especially at the center. These results clearly contradict the generally accepted assumption that LTE exists in the column of atmospheric pressure arcs [1-3].

The radial profile of electron density at 2 mm below the cathode tip determined from analysis of the electron feature of the scattered laser light is presented in Fig. 7. These results agree reasonably well with electron density profiles of similar arcs determined from Stark broadening, which is independent of LTE [3].

The radial gas temperature profiles of the plasma jet with an operating current of 900 A determined from lineshape analysis is compared with the radial temperature profile determined from standard emission spectroscopy [7] in Fig. 9. It is evident from this data that the plasma jet is close to LTE in the center, but not in the outer regions. This is probably due to radiation trapping by ground state argon atoms and electron diffusion [4].

Until now, reliable exit plane velocity profile data of plasma jets has not been available. By analogy with incompressible laminar flow in a cylindrically symmetric channel, computational modelers [9] postulate that the velocity profiles are nearly parabolic. To examine this, the radial velocity profile at 2 mm downstream from the torch exit from $r = 0$ to $r = 3$ mm was fit to a parabola after being reflected about the flow axis, and is plotted in Fig. 10. As can be seen, the fit is quite good and justifies the assumption of parabolic velocity profiles.

CONCLUSIONS

Lineshape analysis of laser light scattered by a plasma allows the direct measurement of plasma gas temperature, electron temperature and electron density, and plasma gas velocity. This method is unintrusive, has a high degree of spatial resolution, does not require an Abel inversion, and, most importantly, does not rely on an assumption of LTE to interpret the data. Using this technique, we have for the first time, to our knowledge, directly investigated the existence of LTE in atmospheric pressure free-burning argon arcs and atmospheric pressure, subsonic argon plasma jets. We have found strong evidence that LTE does not exist in the arc column of the free-burning argon arc, contrary to expectations. We have also found that the core region of an plasma jet is close to LTE, but there is a severe departure from LTE in the fringes. The radial velocity profile in the exit plane of the plasma jet is very nearly parabolic, as postulated.

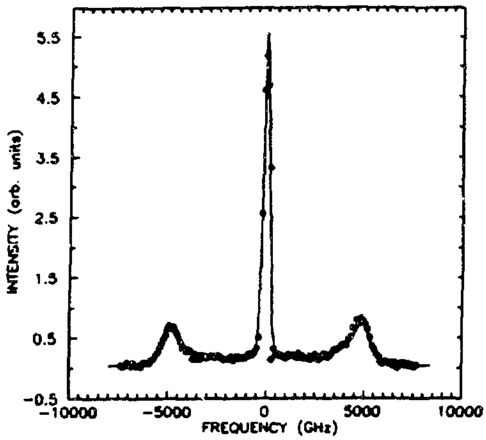

Figure 5. Electron feature in a free-burning arc at 2 mm below the cathode at the radial position $r \approx 0$ mm. $T_e = 28240$ K ± 3% and $n_e = 1.62 \times 10^{23}$ m^{-3} ± 3%.

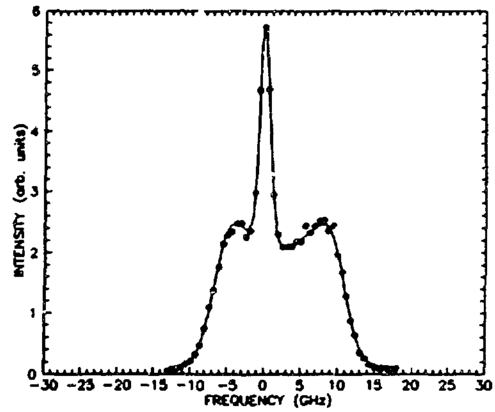

Figure 6. Experimental ion feature from the plasma jet at 2 mm downstream from the exit and at $r = 0$ mm. $T_i = 12630$ K ± 7% and the gas velocity $v = 1095$ m s^{-1} ± 3%.

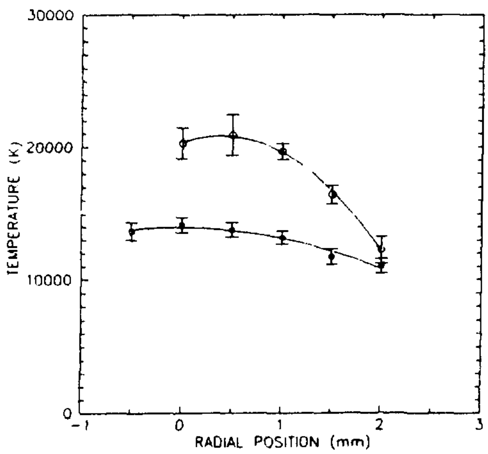

Figure 7. Radial temperature profiles of a free-burning arc at 2 mm below the cathode. The solid dots are the gas temperature. The open circles are the electron temperature.

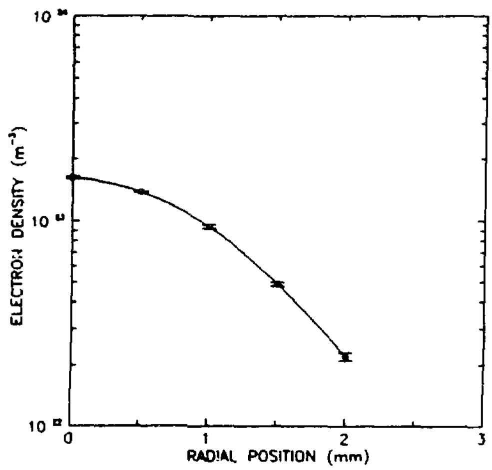

Figure 8. Radial profile of electron density in a free-burning arc at 2 mm below the cathode.

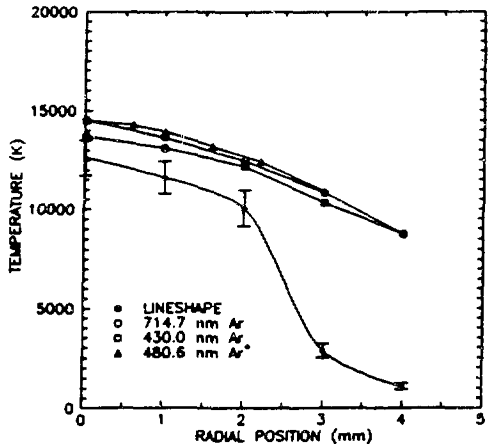

Figure 9. Radial temperature profiles of a plasma jet at 2 mm downstream from the torch exit determined from lineshape analysis and emission spectroscopy. Torch operating current was 900 A.

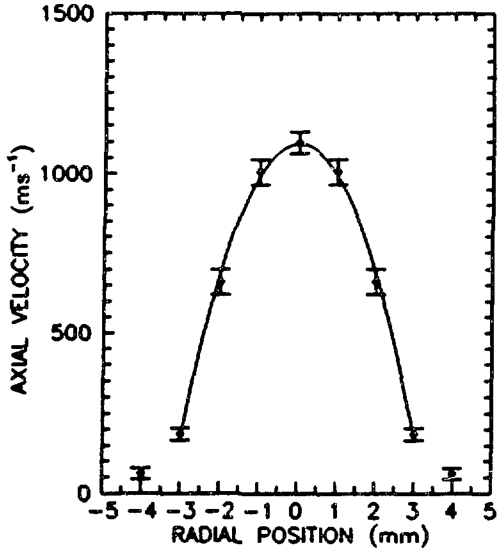

Figure 10. Radial velocity profile of a plasma jet at 2 mm downstream from the torch exit. The solid curve is a parabolic fit of the data. Torch operating current was 900 A.

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