Electron-temperature and electron-density profiles in an atmospheric-pressure argon plasma jet

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Line-shape analysis of the electron feature of Thomson-scattered laser light has been used to directly determine electron-temperature and electron-density profiles of an atmospheric-pressure argon plasma jet. Measured center line values of the electron temperature are in excess of 20 000 K at the torch exit, and remain essentially constant with increasing axial distance even though the plasma is recombining. These results are shown to be inconsistent with Saha equilibrium and classical electron-ion momentum transfer theory.

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INTRODUCTION

Thermal plasmas are well suited for a variety of material processing applications including plasma chemistry, plasma deposition, the manufacture of nanostructure materials, welding, and the destruction of hazardous wastes [1]. Modeling and diagnostics of thermal plasmas are understandably important to the development of this technology, and for gaining a fundamental knowledge of atmospheric-pressure nonequilibrium plasma physics.

Emission spectroscopy is the most straightforward and commonly applied diagnostic method used to determine plasma gas and electron temperature, and electron density. In order to interpret data from emission spectroscopy, it is often necessary to assume that the plasma is in local thermodynamic equilibrium (LTE). Recent experiments using line-shape analysis of laser light Thomson scattered by electrons in the plasma to directly determine the gas-temperature distribution in an atmospheric-pressure argon plasma jet [2], and gas temperature, electron temperature, and electron-density distributions in a free-burning argon arc discharge [3] indicate that the assumption of LTE in plasmas when electron densities are 10^{23} m^{-3} or lower is generally not valid.

Direct measurement of heavy-particle kinetic temperature, electron temperature, and electron density in plasmas is, to the authors' knowledge, only possible by line-shape analysis of Thomson-scattered laser light. If the degree of ionization of the plasma is low, Rayleigh scattering dominates and only heavy-particle kinetic temperature can be measured. For electron densities greater than 10^{-22} m^{-3}, the scattered-light line shape consists of a central narrow ion feature and a broad symmetric electron feature. Analysis of this line shape yields heavy-particle temperature, electron temperature and electron density. The application of line-shape analysis of the ion and electron feature of Thomson-scattered laser light to atmospheric pressure thermal plasmas has not been extensive, although some results have been reported [2–6].

Even if LTE is not valid in thermal plasmas, it seems reasonable to expect the existence of partial local thermodynamic equilibrium (PLTE) [7]. If the plasma is in PLTE, the upper atomic excited states are in thermal equilibrium with the electrons, and the temperature determined from emission spectroscopy is the electron temperature. Line-shape experiments performed on a free-burning argon arc discharge [3,5] suggest that PLTE occurs in the arc column. Until now, however, the existence of PLTE in atmospheric-pressure plasma jets has not been investigated directly.

This paper presents axial and radial electron-temperature and electron-density profiles of an atmospheric-pressure subsonic argon plasma jet discharged into ambient air, determined from line-shape analysis of the electron feature of Thomson-scattered laser light. Electron-temperature profiles are compared with temperature profiles determined from LTE emission spectroscopy and gas-temperature profiles determined from line-shape analysis of the ion feature of Thomson-scattered light and enthalpy-probe measurements. The results are discussed in terms of an argon relaxation-kinetics model of the plasma that calculates the time evolution of the electron and heavy-particle kinetic temperatures and species densities as the plasma decays by way of collisions and spontaneous emission of radiation.

THEORY

The expression for the line shape of Thomson-scattered light, discussed in detail elsewhere [8], is

$$S(\mathbf{k},\omega)=\frac{2\pi}{k}\left|1-\frac{G_e}{\epsilon}\right|^2 f_{0e}\left[\frac{\omega}{k}\right]+\frac{2\pi Z}{k}\left|\frac{G_e}{\epsilon}\right|^2 f_{0i}\left[\frac{\omega}{k}\right], \quad (1)$$

where k is the magnitude of the difference between the scattered wave vector and the incident wave vector ($\mathbf{k}=\mathbf{k}_s-\mathbf{k}_0$), ω is the difference between the angular frequency of the scattered light and the incident laser ($\omega=\omega_s-\omega_0$), 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

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

and

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

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 1 for this experiment. It is assumed that the velocity distribution functions for electrons and ions are Maxwellian. The first term of Eq. (1) is the electron feature of the scattered light spectrum, and the second term is the ion feature. Because of the large mass difference between electrons and ions, the electron feature is about two orders of magnitude broader than the ion feature.

Collisions are influence the line shape of the electron feature in several ways [4,8,9]. Interruption of the scattering process by electron-ion or electron-electron collisions can broaden the line shape if the collision frequency is comparable to the Doppler width ω_D of the one-dimensional Maxwellian velocity distribution, which is given by

$$\omega_D=k\left[\frac{2k_B T_e}{m_e}\right]^{1/2}, \quad (4)$$

where T_e is the electron temperature and k_B is Boltzmann's constant. The electron-ion collision frequency $\nu_{\rm ei}$ is given by [8]

$$\nu_{\rm ei}=2.92\times 10^{-6} n_e[\mathrm{cm}^{-3}](T_e[\mathrm{eV}])^{-3/2}\ln\Lambda\ , \quad (5)$$

where

$$\Lambda=1.53\times 10^{10}\frac{(T_e[\mathrm{eV}])^{3/2}}{(n_e[\mathrm{cm}^{-3}])^{1/2}}\ . \quad (6)$$

The electron-electron collision frequency $\nu_{\rm ee}$ is given by

$$\nu_{\rm ee}=\frac{\nu_{\rm ei}}{2^{1/2}}\ . \quad (7)$$

Collisional effects are not important if $\nu_{\alpha\beta}/\omega_D \ll 1$, where $\alpha\beta$ denotes electron-electron or electron-ion collisions.

Line-shape structure that appears when scattering from electron plasma waves (collective or coherent scattering) is important can be broadened by collisions if collisional damping of the electron plasma waves is comparable to collisionless Landau damping of the waves [8]. The Landau damping rate is given by [10]

$$\gamma=\omega_p \pi^{1/2}\frac{k_D^3}{k^3}\exp[-(k_D/k)^2]\ , \quad (8)$$

where $\omega_p=(n_e e^2/m_e\epsilon_0)^{1/2}$ is the electron plasma frequency, ϵ_0 is the permittivity of free space, and $k_D=(m_e\omega_p^2/k_B T_e)^{1/2}$ is the Debye wave number, which is the reciprocal of the Debye length λ_D. Collective effects become important when the parameter $\alpha=k_D/k \gg 1$.

EXPERIMENT

The argon plasma jet studied was generated by a Miller Model SG-100 plasma torch. The torch operating current ranged from 300 to 900 A. Torch voltages were about 23 V for all operating currents. The argon flow rate was 35.4 l min^{-1}. A schematic of the torch is given in Fig. 1. The laser source was a Q-switched frequency-doubled pulsed neodymium-doped yttrium aluminum garnet (Nd:YAG) laser operating at a wavelength of 532 nm. The pulse duration was 10 ns and the pulse rate was 10 Hz. The scattered laser light was spectrally resolved using a 1.3-m monochromator equipped with a 600-groove mm^{-1} grating. The entrance slit width of the monochromator was 150 μ. Line shapes were detected using a thermoelectrically cooled gated 576×384-pixel two-dimensional intensified charge-coupled device (ICCD) diode array detector. The gated intensifier was triggered by the firing of the laser. The incident laser beam was focused into the plasma using a 1.5-m focal length lens to reduce perturbation of the plasma by the laser beam. The diameter of the waist of the focused laser beam was about 200 μ. Data were taken at laser energies from 50 to 225-mJ pulse^{-1}. The laser energy was adjusted using a half-wave plate and Brewster's-angle polarizer in order to maintain a constant beam divergence and hence constant spot size of the focused laser beam. To maximize the signal, a second half-wave plate was used to rotate the polarization of the incident laser light to be parallel with the entrance slit of the monochromator. The scattered light was collected and transferred to the monochromator using two 40-mm-diameter, 300-mm focal length achromatic lenses. A Glan-Thompson polarizer was used to reject the randomly polarized plasma background radiation while transmitting the vertically polarized scattered light. The torch was operated verti-

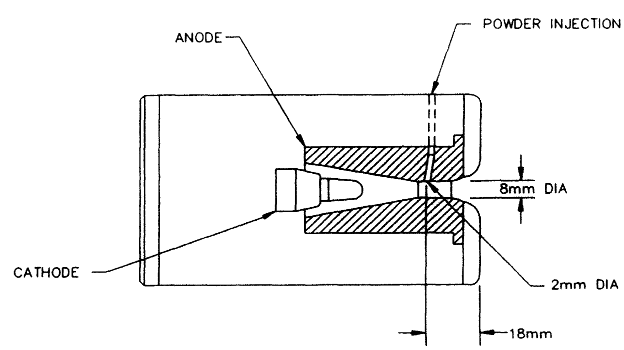

FIG. 1. Torch schematic.

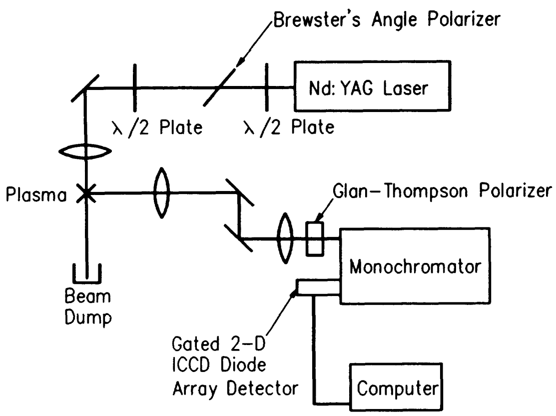

FIG. 2. Experimental schematic.

cally, and the scattering angle was 90° to both the plasma axis and incident laser beam. Emission spectroscopy data consisting of neutral and singly ionized argon lines were taken using the same optical and detector setup except that a 90° image rotator was placed in the optical path to orient the plasma jet image horizontally on the monochromator entrance slit. A schematic of the scattering experiment is presented in Fig. 2.

RESULTS AND DISCUSSION

Typical electron features taken at the radial position $r=0$ mm and axial positions of $z=2$ and 20 mm from the torch face are presented in Figs. 3(a) and 3(b), respectively. The torch current in both cases was 900 A. These line shapes were accumulated over 50 laser pulses to improve the signal-to-noise ratio. The intense peak at 0 GHz is the unresolved ion feature. The smooth line is the nonlinear least squares fit of Eq. (1) to the data, excluding the ion feature. Electron temperature and electron density were determined from this fit. The symmetry of the line shapes implies that the electron velocity distribution functions are Maxwellian as assumed [11]. The effect of convolution of the instrument response function with the line shapes was minor, changing the fitted values of the electron temperature and density by $<2\%$.

Significant laser heating of the electrons by linear inverse bremsstrahlung was observed. This made it necessary to measure electron temperature as a function of incident laser pulse energy. The unperturbed or actual electron temperature was then found by extrapolating the linear fit of the data to the 0-mJ pulse^{-1} laser energy [5,12]. A representative laser heating curve, taken at $r=0$ mm and $z=2$ mm with a torch current of 900 A is given in Fig. 4. Electron-density values were not observed to be significantly affected by the laser energy.

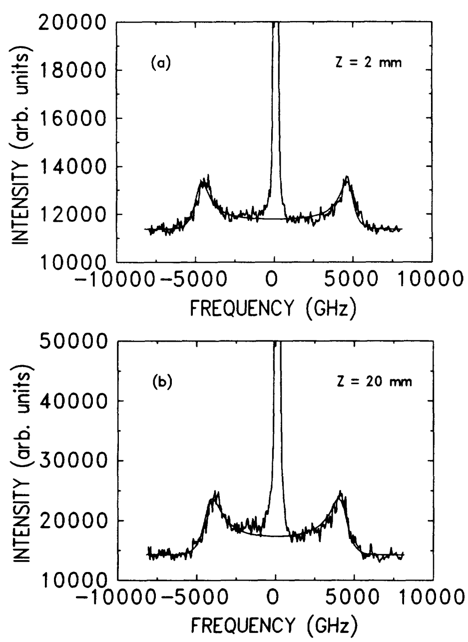

FIG. 3. Experimental electron features at the $r=0$-mm and $z=2$- (a) and 20-mm (b) positions. The electron temperatures and electron densities determined from these line shapes are 26 750 K$\pm3\%$ and 1.40×10^{23} $m^{-3}\pm3\%$, respectively, at $z=2$ mm and 24 600 K$\pm3\%$ and 9.70×10^{22} $m^{-3}\pm3\%$, respectively, at $z=20$ mm. The electron temperature values are strongly affected by laser heating.

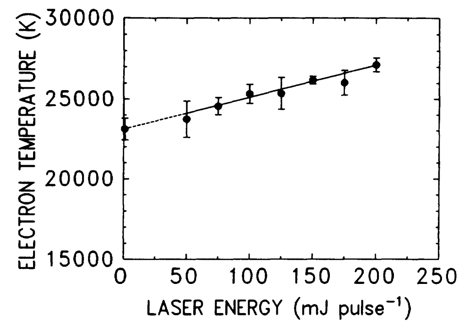

FIG. 4. A typical plot of electron temperature as a function of laser energy. The data were taken at $r=0$ mm and $z=2$ mm. Each individual data point is the average electron temperature calculated from six line shapes, each composed of the accumulation of 50 laser shots. The y intercept of the linear fit of the data is the unperturbed electron temperature.

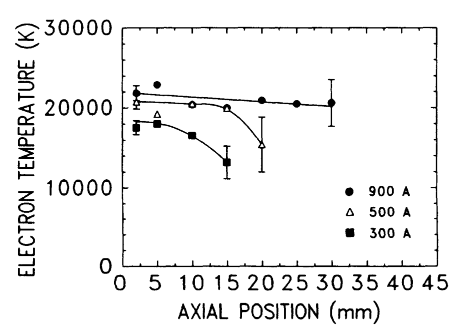

FIG. 5. Electron temperature as a function of axial position at $r=0$ mm for torch currents of 300, 500, and 900 A.

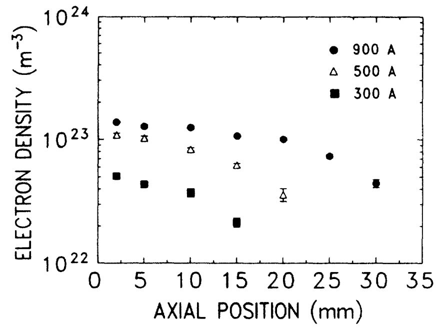

FIG. 6. Electron density as a function of axial position at $r=0$ mm for torch currents of 300, 500, and 900 A.

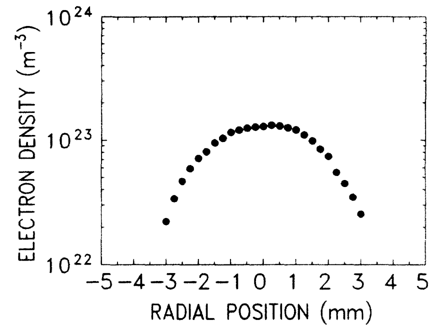

FIG. 8. Radial profile of electron density at $z=2$ mm with a torch current of 900 A.

Unless otherwise noted, all electron-temperature data reported have been corrected for laser-heating effects.

Centerline axial electron-temperature and electron-density profiles, taken at torch currents of 300, 500, and 900 A, are presented in Figs. 5 and 6, respectively. Radial electron-temperature and electron-density profiles, taken 2 mm downstream from the torch exit at a torch current of 900 A, are shown in Figs. 7 and 8, respectively. Plotted in Figs. 9 and 10 are the 900-A centerline axial electron-temperature and electron-density profiles, respectively. Also plotted in Figs. 7 and 9 are temperatures determined from emission spectroscopy, and gas temperatures determined from ion-feature line-shape analysis [2] and enthalpy-probe measurements [13]. Emission temperatures were calculated from absolute intensity measurements of the 696.5 and 738.4-nm neutral argon lines and the 480.6-nm singly ionized argon line assuming LTE.[14] The neutral argon lines were chosen because they are isolated from ionic transitions.

The electron temperatures measured from Thomson scattering are surprisingly high. The electron temperature is almost twice the gas temperature at the torch exit. This degree of non-LTE was not expected. These data also suggest that even PLTE does not exist. Equally surprising is the apparently weak connection between the electron temperature and the electron density. The axial dependence of the electron temperature remains almost constant even though the electron density decreases significantly with increasing axial distance as the plasma recombines, as shown in Figs. 9 and 10. Furthermore, the radial electron-temperature profile is noticeably asymmetric, while the radial electron-density profile is quite symmetric as shown in Figs. 7 and 8. Because of these results, the possibility that a systematic experimental error had occurred was examined in depth. This discussion follows.

Unrealistically high temperatures could result if accounting for the effects of laser heating of the electrons was done incorrectly. The electron temperature as a function of laser energy curve of Fig. 4 is quite linear. This strongly suggests that nonlinear laser-plasma interactions [11] are not occurring, and supports the assumption that

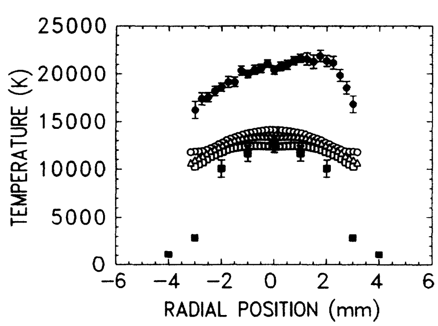

FIG. 7. Radial temperature profile at $z=2$ mm for a torch current of 900 A. The solid circles are electron temperatures determined from Thomson scattering. The solid squares are gas temperatures determined from ion feature measurements of Thomson-scattered light [2]. The open circles, triangles, and squares are temperatures determined from emission spectroscopy of the 480.6-nm singly ionized Ar transition, and the 738.4- and 696-5-nm neutral Ar transitions, respectively.

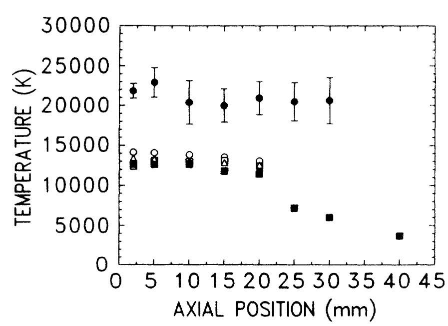

FIG. 9. Axial temperature profile at $r=0$ mm with a torch current of 900 A. The solid circles are electron temperatures determined from Thomson scattering. The solid squares are gas temperatures determined from enthalpy-probe measurements [13]. The open circles, triangles, and squares are temperatures determined from emission spectroscopy of the 480.6-nm singly ionized Ar transition, and the 738.4- and 696.5-nm neutral Ar transitions, respectively.

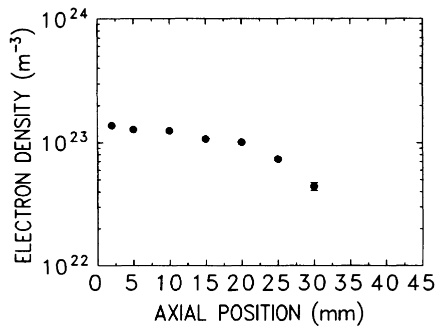

FIG. 10. Axial profile of electron density at $r=0$ mm with a torch current of 900 A.

the unperturbed electron temperature can be found by extrapolating this function to 0-mJ pulse^{-1}. As a further check, the incident laser beam was defocused, increasing the beam waist diameter by about a factor of 6. This decreased the beam intensity by a factor of 36 and should virtually eliminate laser heating of the electrons. This was indeed observed. Centerline electron temperatures measured at $z=2$ and 20 mm with a torch current of 900 A were independent of the laser energy, and were very consistent with the values presented in Fig. 5. We therefore conclude that laser-heating effects have been accounted for properly.

Line-shape theory predicts a narrowing of the spectrum of scattered light and an enhancement of the structure due to collective effects as the scattering angle is decreased. The experiment was reconfigured to collect light at a scattering angle of 70°. The resulting line shapes were significantly narrower, and the electron plasma wave resonance structure much more pronounced. Measurements of the centerline values of the electron temperature and electron density at $z=2$ and 20 mm at a current of 900 A were in good agreement with the 90° scattering-angle results.

An important concern that must be addressed when performing line-shape experiments on a fluctuating system such as a plasma jet is the effect accumulation of many individual line shapes has on the measurement. Two different Thomson line shapes do not superpose to produce a third Thomson line shape. If the plasma fluctuations are not great, the accumulated line shape is a very good approximation of the individual line shapes. However, if shot-to-shot fluctuations are significant, structure due to collective effects becomes smeared out, effectively reducing the value of α. The electron temperature and electron density determined from this accumulated line shape will be in error. In this experiment, centerline values of the electron density were sufficiently high that a line shape with an adequate signal-to-noise ratio was obtainable from a single laser pulse. From 30 to 50 single pulses were recorded on the centerline at $z=2$ and 20 mm with a torch current of 900 A. After analysis of the single-pulse line shapes, average values of the electron temperature and electron density were calculated. The individual line shapes were then added together, and the resulting accumulated line shape was analyzed. In all cases, single-pulse data agreed very well with the results of the accumulated line shape. However, this was not the case when the experiment was repeated at larger radii. For example, results of measurements made at $z=2$ mm and $r=2.5$ mm with a torch current of 900 A were that the average value of the single-pulse electron temperature data was 17 200 K$\pm$6% while the result obtained from accumulating the individual line shapes was 20 500 K$\pm$6%. The average value of the single-pulse electron-density data was 7.80×10^{22} m$^{-3}\pm$5%, and the value found from the accumulated line shape was 6.00×10^{22} m$^{-3}\pm$4%. The observed variation was due to large scale fluctuations in the mixing layer surrounding the jet. This effect was difficult to evaluate at even larger radii because scattering signals were weak. An example of a single-pulse line shape is given in Fig. 11.

The influence of collisions on the line shapes was examined using the peak values of the electron temperature and electron density of 23 000 K and 1.40×10^{23} m^{-3}, respectively. It can be seen from Eqs. (4)–(8) that $\nu_{ei}/\omega_D=0.05$, and $\nu_{ei}/\gamma=0.18$. We conclude from this that collisions did not affect the line shapes.

It must be pointed out that the general line-shape theory expressed by Eq. (1) was derived assuming that there are many electrons in a Debye sphere. This is to say that $n_e\lambda_D^3\gg1$. Excellent agreement between the line-shape theory and experimental results has been demonstrated for plasmas with $n_e\lambda_D^3=2-10$ [5,15,16]. Theoretical calculations indicate that the line-shape theory is valid when $n_e\lambda_D^3>0.1$ [17]. For our experimental conditions, $n_e\lambda_D^3>3$, and the line-shape theory is considered to be correct. This is further supported by the

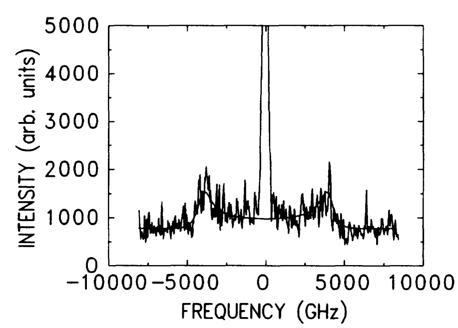

FIG. 11. A typical electron feature from a single laser pulse. The line shape was taken at the $r=0$ mm and $z=20$ mm positions with a torch current of 900 A. The smooth curve is the fit of the theory to the data, and gives an electron temperature of 20 950 K$\pm$7% and electron density of 9.50×10^{22} m$^{-3}\pm$6%. In this case, the incident laser beam was defocused to a spot size of $\sim$1.2 mm to eliminate the effects of laser heating. The average electron temperature and electron density determined from 30 consecutive single-pulse line shapes was 22 980 K$\pm$4% and 8.90×10^{22} m$^{-3}\pm$3%, respectively. The electron temperature and electron density determined by superposing the 30 single-pulse line shapes was 23 300 K$\pm$4% and 8.94×10^{22} m$^{-3}\pm$4%, respectively.

very good fit of the theory to our experimental line shapes, as well as the good agreement of the gas temperatures in the exit plane of this torch previously determined from ion-feature analysis and enthalpy-probe measurements [13].

The axial electron-temperature and electron-density profiles seem to suggest the following. High-temperature electrons are generated in the discharge region of the torch. These electrons exit the torch with a bulk axial velocity of $\sim 1000\ \mathrm{m\,s^{-1}}$ [2]. In the absence of the discharge, the plasma recombines, and the electrons are heated by three-body electron-ion recombination [18]. However, the time scale is too short for the electrons to cool appreciably by electron-ion momentum exchange. The validity of this interpretation was investigated using an argon relaxation-kinetics model, described in more detail elsewhere [19]. Briefly, this model characterizes the behavior of the plasma as it relaxes from initial conditions by way of collisional excitation and ionization by electrons and heavy particles, Penning ionization, radiative and three-body recombination and spontaneous emission. Reverse reaction rates are calculated from the principle of detailed balance using the Boltzmann equation at the heavy-particle temperature T_g for heavy-particle excitations, and T_e for electron excitations or the Saha equation at T_e for ionization rates. The model includes neutral argon ground, 4*s*, and 4*p* states, singly ionized argon from the ground state to 4*p* state, inclusive, and electrons. The initial state of the plasma is specified by T_e, T_g, n_e, and the total pressure. The only energy-loss mechanism from the plasma is through photon emission. The model considers the plasma to be optically thick to the neutral argon ground state to 4*p* (resonance) transition. The dominant electron-cooling mechanisms are collisional excitation of the argon ground state, ionization of both ground-state and excited-state argon, and electron-ion momentum transfer. The dominant electron-heating mechanism is three-body electron-ion recombination. It is assumed that there is no current density in the plasma. The model predicts that the time required for a plasma with $T_e = 23\,000$ K, $T_g = 12\,500$ K, and $n_e = 1.40 \times 10^{23}$ $\mathrm{m^{-3}}$ to come to thermal equilibrium is on the order of 100 ns. During this time a volume element of the plasma has traveled 0.1 mm. The model also calculates that the maximum temperature difference between electrons and heavy particles that can be sustained by three-body electron-ion recombination is only ~ 1200 K. To sustain the temperature difference observed, the classic electron-ion momentum-transfer rate [20] must be decreased by a factor of at least 50. Furthermore, the excited-state Saha equation [7], using the emission results to determine excited-state densities and the measured electron temperatures, predicts that the plasma should be ionizing instead of recombining. This would rapidly cool the electrons and increase the electron density in contradiction to the observations.

At this point, we can only speculate about the large discrepancy between the results of the electron-feature measurements and the predictions of the kinetics model. The consistency of the experimental results tends to rule out any systematic errors. For these results to be incorrect would mean that the line-shape theory with the assumption of a Maxwellian electron velocity distribution function is, in spite of the previous discussions, not applicable to this type of plasma. The other possibility is that Saha equilibrium between the excited states and ions is not necessarily attained and/or that the classical description of electron-ion interactions overestimates the rate of momentum transfer. Attempts to describe long-range Coulomb interactions in terms of two-body collisions have been considered by some to be inadequate [21–25]. In the two-body approach, the scattering force on a test particle is calculated for one single particle, and the effects (energy transfer, for example), not the forces, are added for all of the field particles. In fact, the total scattering force on a test particle is usually much smaller than the force due to a typical neighboring field particle. The correct effect of this total scattering force is smaller than the sum of the effects of the separate contributions to the scattering force. Second, the application of the Saha equation to a system far from equilibrium is problematic. Its application to multitemperature systems can be shown to lead to a contradiction, as follows. The reaction considered is $\mathrm{Ar}^* \leftrightarrow \mathrm{Ar}^+ + e^-$, where Ar^* denotes excited-state argon, Ar^+ denotes singly ionized argon, and e^- denotes a free electron. The degree of excitation and the electron kinetic energy are assumed to be described by one (high) temperature, and the atom and ion kinetic energies are assumed to be described by a different (lower) heavy-particle temperature. The number of electrons is obtained from the Saha equation using the higher temperature. It is equally valid to obtain the number of ions using the same Saha equation with the lower temperature. For this system, the number of electrons should be the same as the number of ions, but these two applications of the Saha equation give different results and thus a contradiction.

CONCLUSIONS

Electron-temperature and electron-density profiles of an atmospheric-pressure argon plasma jet were made from analysis of the electron feature of laser light Thomson scattered by the plasma. These measurements suggest significant deviations from LTE persist throughout the plasma. Measured centerline electron temperatures are in excess of 20 000 K in the torch exit plane, while gas temperatures at the same location are about 12 500 K. Furthermore, the electron temperature remains almost constant with increasing axial distance even though the electron density decreases significantly. A comparison of the measured electron temperatures with temperatures determined from emission spectroscopy indicates that departures from PLTE also persist. There is a large discrepancy between the experimental results and the expected behavior predicted by an argon relaxation-kinetics model. The reason for this discrepancy is presently not understood.

ACKNOWLEDGMENTS

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