PHYSICAL REVIEW E VOLUME 61, NUMBER 2 FEBRUARY 2000

Dependence on the scattering angle of the electron temperature and electron density in Thomson-scattering measurements on an atmospheric-pressure plasma jet

S. C. Snyder, D. M. Crawford, and J. R. Fincke
Idaho National Engineering and Environmental Laboratory, P.O. Box 1625, Idaho Falls, Idaho 83415

(Received 13 September 1999)

Electron temperature and electron density measurements were made in an atmospheric-pressure argon plasma jet by line-shape analysis of Thomson-scattered laser light. The dependence of electron temperature and electron density on the scattering angle was investigated. Measurements were made using incident laser wavelengths of 532 and 355 nm. At 532 nm, the electron-ion collision frequency exceeds the Landau damping rate for shallow scattering angles, and the electron plasma wave resonance structure in the Thomson line shape is broadened. This resulted in dramatic increase in the apparent electron temperature as a function of decreasing scattering angle. At 355 nm, collisions do not affect the Thomson line shape. In this case, an angular dependence of the measured electron temperature is not expected and was not observed. Data taken at 532 nm at larger scattering angles are consistent with the 355-nm results, and show that the electrons are not in thermodynamic equilibrium with the heavy particles.

PACS number(s): 52.25.Rv, 52.70.Kz, 52.75.Hn, 42.62.Fi

I. INTRODUCTION

The determination of electron temperature and electron density of atmospheric-pressure plasmas by line-shape analysis of Thomson-scattered light has great appeal mainly because assumptions about local thermodynamic equilibrium (LTE) are not required to interpret the data. The standard method of emission spectroscopy requires an assumption of LTE, or at least partial local thermodynamic equilibrium (PLTE), where electrons are in thermodynamic equilibrium with the atomic excited states, in order to determine temperature. One can argue that the electron density in atmospheric-pressure plasmas is high enough to achieve at least PLTE [1], and consequently emission spectroscopy gives realistic values for the electron temperature.

Electron temperature measurements in an atmospheric-pressure argon plasma jet and transferred arc made by line-shape analysis of Thomson-scattered laser light have been reported [2–4]. Peak electron temperatures were found to be around 20 000 K. Heavy particle temperatures were around 12 000 K in the plasma jet to around 14 000 K in the transferred arc. Temperatures determined from emission spectroscopy are around 14 000 K in the plasma jet [2] to around 16 000 K in the transferred arc [4]. These results show a great departure from LTE and PLTE in atmospheric-pressure plasmas and indicate that emission spectroscopy does not give reliable values of the electron temperature. Similar results have been reported in an atmospheric-pressure microwave torch [5]. It has been suggested that electron temperatures must be greater than heavy particle temperatures in order to sustain plasmas that have steep temperature and density gradients [5]. Nevertheless, the discrepancy between electron temperature values determined from Thomson scattering and from emission spectroscopy remains.

Work has recently been published that measured the electron temperature and electron density in an atmospheric-pressure argon plasma jet by line-shape analysis of the electron feature of Thomson-scattered laser light as a function of scattering angle at a laser wavelength of 532 nm [6]. A clear dependence of the electron temperature and electron density on the scattering angle was found. Such a result was not expected since the expression for the line shape is a function of the scattering angle. Electron temperature and electron density should therefore be independent of scattering angle. It was concluded that previously unreported asymmetric conditions exist in the plasma jet, and that this asymmetry has a great influence on the line shape of the scattered light. A source of this asymmetry was proposed to be the steep density gradients within the scattering volume of the plasma. These gradients affect the description in Fourier space of the density fluctuations of the free electrons. Taking these steep gradients into account, an expression relating the measured electron temperature to the scattering angle and the actual electron temperature was derived. Fitting this expression to their experimental results, these authors found an actual electron temperature of around 10 500 K. This is in much better agreement with emission spectroscopic measurements, and is evidence of the existence of at least PLTE in the plasma.

We present in this paper measurements of electron temperature and electron density in an atmospheric-pressure argon plasma jet by line-shape analysis of Thomson-scattered light as a function of scattering angle at laser wavelengths of 532 and 355 nm. We also observed a dependence of the electron temperature on the scattering angle in our raw data at 532 nm. No clear dependence of electron temperature or electron density on the scattering angle was observed at 355 nm. Our results are compared with those of Gregori *et al.* [6]. After considering the effects of collisions on the line shapes, there is little evidence suggesting that the straightforward interpretation of the electron feature previously reported [2,3] is in error.

II. THEORY

The line shape of laser light scattered by density fluctuations of the free electrons in a plasma is described analytically by the Salpeter equation [7]. The Salpeter equation has two components. One component, referred to as the ion feature, arises from the influence of ions on the electron density fluctuations. The other component is called the electron feature and is due to the density fluctuations of the free electrons themselves. The electron feature is a function of the electron temperature, electron density, and the wave number $k=|\vec{k}_s-\vec{k}_0|=(4\pi/\lambda_0)\sin(\theta/2)$, where $\vec{k}_s$ is the wave vector of the scattered light, $\vec{k}_0$ is the wave vector of the incident laser light, λ_0 is the wavelength of the incident laser light, and θ is the scattering angle.

Collisions can greatly influence the electron feature. Interruption of the scattering process by electron-ion or electron-electron collisions can broaden the distribution function describing the electron density fluctuations [8]. The Doppler width of the distribution function is $\omega_D=kv_t$, where $v_t=(2k_BT_e/m_e)^{1/2}$ is the electron thermal velocity, k_B is Boltzmann's constant, T_e is the electron temperature, and m_e is the electron mass. The electron-ion collision frequency is given by [7]

$$\nu_{ei}=2.92\times 10^{-6} n_e (T_e)^{-3/2}\ln\Lambda, \tag{1}$$

with n_e the electron density measured in cm^{-3} and

$$\Lambda=1.53\times 10^{10}\frac{(T_e)^{3/2}}{(n_e)^{1/2}}. \tag{2}$$

In Eqs. (1) and (2), T_e is measured in eV. The electron-electron collision frequency is $\nu_{ee}=\nu_{ei}/2^{1/2}$. If $\nu_{\alpha\beta}/\omega_D\ll 1$, where $\alpha\beta$ denote electron-ion or electron-electron collisions, then collisional broadening of the distribution function is not significant. It is seen that $\nu_{\alpha\beta}$ is independent of the laser wavelength and scattering angle.

If the wavelength of the density fluctuation for a specific k value is greater than the Debye length, collective effects are important [2]. In this case, the electron feature has a resonance structure due to scattering from electron plasma waves. Collisions can affect this structure. The height, full width at half maximum (FWHM), and position of this resonance are dependent on the scattering angle. It has been shown that the shape of the resonance structure is approximately Lorentzian with a FWHM given by the sum of the Landau damping rate and a term dependent on the collision damping rate [9]. The Landau damping rate is given by [10]

$$\gamma_L=\omega_p\pi^{1/2}\left(\frac{k_D}{k}\right)^3\exp[-(k_D/k)^2], \tag{3}$$

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, e is the charge of an electron, and $k_D=(m_e\omega_p^2/k_BT_e)^{1/2}$ is the Debye wave number. If $\nu_{\alpha\beta}/\gamma_L\ll 1$, collisional broadening of the resonance structure is not important [7,9].

III. EXPERIMENT

Our experimental conditions were chosen to be similar to those of Gregori *et al.* [6]. The atmospheric-pressure argon plasma jet was generated with a Miller Model SG-100 direct current plasma torch. A Q-switched, injection-seeded Quanta-Ray GCR-4 neodymium-doped yttrium aluminum garnet (Nd:YAG) laser was the laser source. The laser pulse rate was 10 Hz and the pulse width was 10 ns. Experiments were conducted using 532 and 355 nm wavelengths from the Nd:YAG laser. In both cases, the laser pulse energy was 100 mJ pulse^{-1}. In order to minimize laser heating of the electrons by inverse bremsstrahlung [2], the laser beam was defocused to a spot size of about 1 mm in the plasma. The Thomson-scattered light was collected with a 50.8-mm-diameter 200-mm focal length lens, collimated and focused with another 50.8-mm-diameter 200-mm focal length lens on to the entrance slit of a 1-m focal length McPherson Model 2061A monochromator with a 600 groove mm^{-1} grating blazed at 500 nm. The entrance slit was opened to 150 μm. The electron feature was detected using a Princeton Instruments Model ICCD-576G/RB gated two-dimensional intensified charged-coupled device (ICCD) array. The gate of the detector was triggered by the firing of the Q-switch of the Nd:YAG laser. The gate width of the detector was 25 ns. A Glan-Thompson polarizer was used to reduce the unpolarized background light from the plasma. A half-wave plate rotated the polarization of the incident laser beam to be parallel with the entrance slit to maximize the Thomson signal.

One approach to collecting the electron feature is to accumulate the line shape over a number of successive laser shots. This is done to increase the signal-to-noise ratio (SNR) of the line shape. Strictly speaking, this is not the best method for data collection. The electron feature is a complicated function. Two or more electron features do not superpose to form a third electron feature. At best, an accumulation of successive single-shot line shapes gives an approximation, actually a rather good approximation [2], of an electron feature. Nevertheless, our approach to this work was to acquire single-shot line shapes when possible, and calculate essentially instantaneous electron temperatures and electron densities from these line shapes. Using conventional optics instead of fiber optics, which have a lower collection efficiency, it was possible to record single-shot line shapes with an adequate SNR at the laser wavelength of 532 nm. However, at 355 nm, several argon-ion emission lines interfere with the Thomson line shape. This spectral interference was subtracted from the line shape, but doing so decreased the SNR of the single-shot line shapes. Consequently, it was

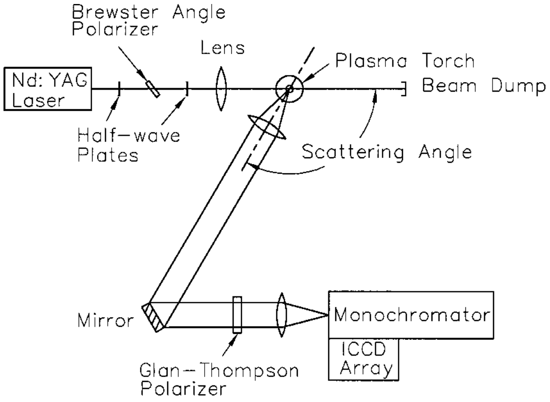

FIG. 1. Schematic of the experimental setup. The half-wave plate and Brewster angle polarizer at the output of the laser are used to adjust the laser power while maintaining a constant beam divergence.

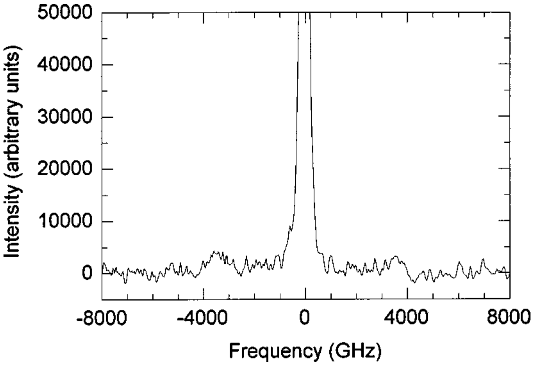

FIG. 2. An example of a single-shot electron feature. The incident laser wavelength was 532 nm and the scattering angle was 50°. The central peak is the unresolved ion feature. The electron temperature and electron density determined from this line shape are $37\,900 \pm 1000$ K and $9.0\times10^{22} \pm 0.2\times10^{22}\ \mathrm{m}^{-3}$, respectively. Instrumental broadening has not been deconvolved from this line shape.

necessary to accumulate a line shape over 10 laser shots. A schematic of the experiment is shown in Fig. 1.

IV. RESULTS AND DISCUSSION

All data were taken at an axial location 5 mm downstream from the exit plane of the plasma jet at the radial position of $r=0$ mm. The argon flow rate was 35.4 $\mathrm{l\,min}^{-1}$ and the torch current was 900 A. At a given scattering angle, 30 single-shot line shapes were recorded when 532 nm was the laser wavelength. The electron temperature and electron density from each line shape were determined by fitting the Salpeter equation to the data. A weighted average of the electron temperature and electron density was then calculated. When 355 nm was used for the laser wavelength, 10 line shapes, each the accumulation of 10 laser shots, were recorded. A weighted average of the electron temperature and electron density was calculated from this data. An example of a single-shot electron feature taken at a scattering angle of $\theta=50°$ using a laser wavelength of 532 nm is presented in Fig. 2. A line shape taken at a scattering angle of $\theta=50°$ using a laser wavelength of 355 nm after background subtraction is given in Fig. 3. The central peak in the line shapes is the unresolved ion feature, and it gives a measurement of the instrument response function (IRF) of our experimental apparatus of about 350 GHz for 532 nm and 650 GHz for 355 nm. The smaller peaks around ±3500 GHz are the electron plasma wave resonance structure. The electron plasma wave resonance structure in the line shapes taken at 532 nm is broadened by convolution of the IRF with the line shape at scattering angles less than 80°. Convolution affects the line shapes taken at 355 nm to a much lesser extent, and only at scattering angles less than 60°. Instrument broadening is misinterpreted as an increase in the electron temperature and decrease in the electron density. For example, at 532 nm and $\theta=50°$, instrument broadening gives an apparent increase of the electron temperature of about 8000 K and a decrease of the electron density of about $2\times10^{22}\ \mathrm{m}^{-3}$, and 4000 K and $0.6\times10^{22}\ \mathrm{m}^{-3}$ for 532 and 355 nm, respectively. Plots of electron temperature and electron density as a function of scattering angle for laser wavelengths of 532 and 355 nm are given in Figs. 4 and 5, respectively. Instrument broadening effects have been accounted for in the plots. Still, the 532-nm data show an obvious increase in the electron temperature at scattering angles less than 80°. The electron density data seem to be independent of scattering angle. There is no significant angular dependence of electron temperature or

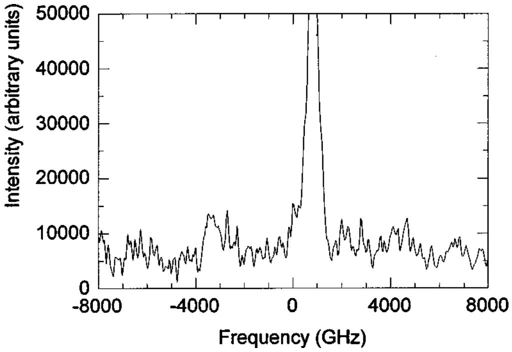

FIG. 3. An example of an electron feature accumulated over 10 laser shots. The incident laser wavelength was 355 nm and the scattering angle was 50°. The electron temperature and electron density determined from this line shape are $23\,400 \pm 900$ K and $1.02\times10^{23} \pm 0.03\times10^{23}\ \mathrm{m}^{-3}$, respectively. Instrumental broadening has not been deconvolved from this line shape.

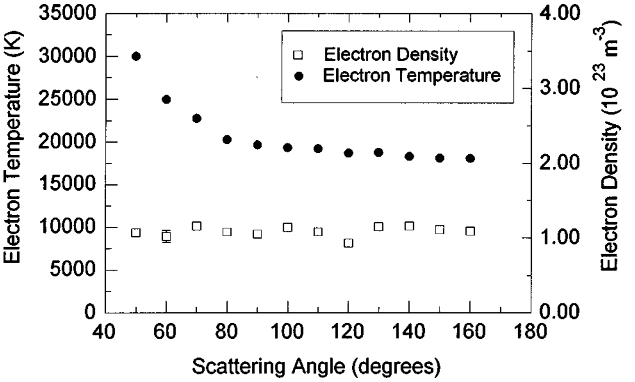

FIG. 4. Electron temperature and electron density as a function of scattering angle. The data were taken with a laser wavelength of 532 nm. Typical uncertainties of the data points are ~5%. Instrumental broadening has been deconvolved from the data.

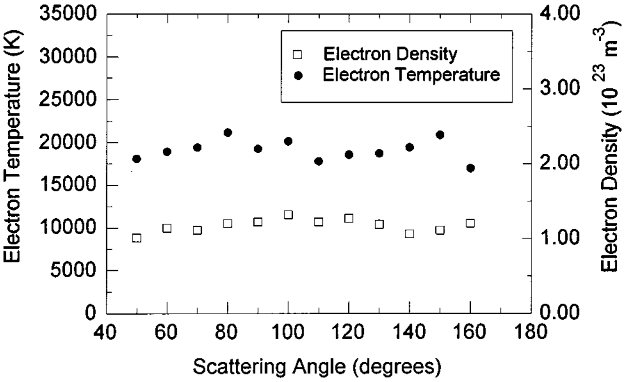

FIG. 5. Electron temperature and electron density as a function of scattering angle. The data were taken with a laser wavelength of 355 nm. Typical uncertainties of the data points are ~5%. Instrumental broadening has been deconvolved from the data.

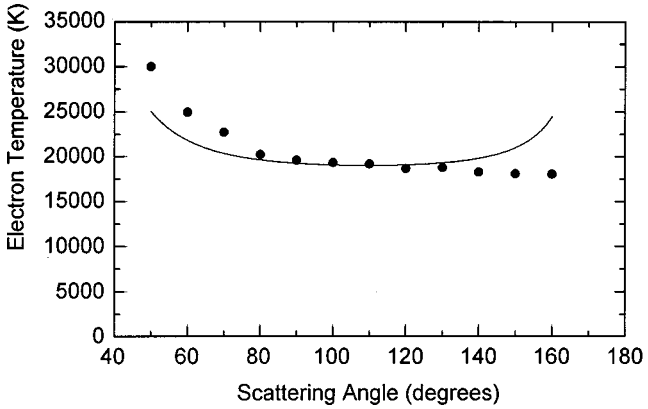

FIG. 6. A fit of Eq. (4) to the electron temperature as a function of scattering angle data taken with a laser wavelength of 532 nm.

electron density evident in the 355-nm data. These results are consistent with the prediction of the Salpeter equation that there should be no angular dependence of the data.

The apparent angular dependence in the 532-nm data is not as pronounced as that reported by Gregori *et al.* [6]. Furthermore, our electron density data do not exhibit the sin θ dependence observed by Gregori *et al.* [6]. The angular dependence of the electron temperature data observed by Gregori *et al.* [6] was attributed to an electron density gradient in the measurement volume in the plasma. They calculated that because of this density gradient, k increases to a value greater than it would be if the electron density were uniform in the measurement volume. This increase in k manifests itself as an apparent increase in the measured electron temperature. An expression relating the measured electron temperature to the actual electron temperature was given by

$$\tilde{T}_e = T_e\left[1+\frac{\omega_p^2}{(k\nu_t)^2}\,\delta\right]^{1/2}, \tag{4}$$

where $\delta = \text{const}/\sin^2\theta$. Fitting Eq. (4) to the electron temperature as a function of scattering angle data with T_e and δ adjustable parameters yielded $T_e \sim 10\,500\,\text{K}$. This temperature is in better agreement with values obtained from emission spectroscopy [2], and consistent with PLTE. A fit of Eq. (4) to our electron temperature data gives an effective electron temperature of $17\,725 \pm 1550\,\text{K}$ and $19\,885 \pm 600\,\text{K}$ for the 532-nm and 355-nm data, respectively. The fit of Eq. (4) to the 532-nm data, shown in Fig. 6, is not very good. The fit of Eq. (4) to the 355-nm data, shown in Fig. 7, is much better. However, since there is no clear angular dependence of electron temperature, there is no justification for fitting Eq. (4) to this data; a simple linear fit would be as credible. Nevertheless, the effective electron temperature determined from our data is much higher than the result of Gregori *et al.* [6], and consistent with previous results [2].

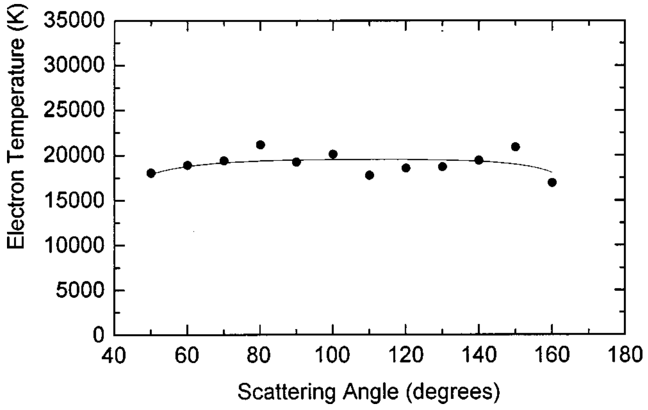

FIG. 7. A fit of Eq. (4) to the electron temperature as a function of scattering angle data taken with a laser wavelength of 532 nm.

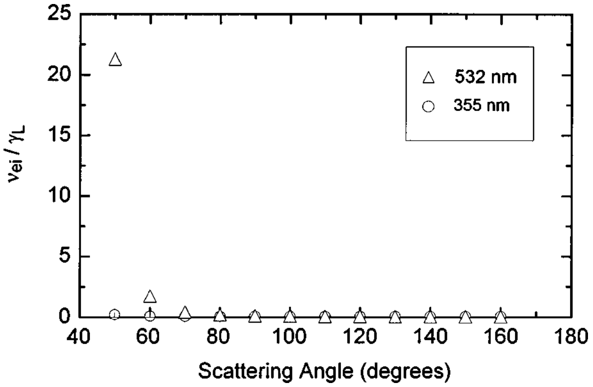

FIG. 8. A plot of the ratio of the collision frequency to the Landau-damping rate as a function of scattering angle.

The angular dependence of the 532-nm data must still be explained. The version of the Salpeter equation used to analyze our data does not include collisional effects. From Eqs. (1) and (2), we see that for a plasma with an electron temperature of 20 000 K, and electron density of $1\times10^{23}\,\text{m}^{-3}$, the electron-ion collision frequency is about 600 GHz. For a laser wavelength of 532 nm and a scattering angle of 50°, the Doppler width of the electron density fluctuation distribution function is $\omega_D = 7770\,\text{GHz}$. Since this is the minimum Doppler width encountered in this work, the requirement that $\nu_{\alpha\beta}/\omega_D \ll 1$ is met for all conditions of this experiment. From Eq. (3), a plot of ν_{ei}/γ_L as a function of scattering angle for laser wavelengths of 532 and 355 nm was calculated and is given in Fig. 8. It is seen that, for the 532-nm wavelength, the condition that the ratio $\nu_{ei}/\gamma_L \ll 1$ for collisional broadening of the electron plasma wave resonance structure to be negligible is not met at scattering angles less than around 70°. For the 355-nm wavelength, collective effects are much less prominent. The ratio $\nu_{ei}/\gamma_L \ll 1$ for all scattering angles, and the line shapes can be considered unaffected by collisions for all scattering angles. We therefore attribute the difference between the 532- and 355-nm data to collisional broadening of the electron plasma wave resonance structure. This broadening becomes significant in the 532-nm data at the shallower scattering angles, and is wrongly interpreted as an increase in the electron temperature. Instead, it is our opinion that the comparison of the 532-nm data with the 355-nm data gives an experimental measurement of the degree to which collisions affect the electron feature.

V. CONCLUSIONS

Electron temperature and electron density measurements were made as a function of scattering angle by analysis of the electron feature of Thomson-scattered laser light. Laser wavelengths used were 532 and 355 nm. Electron temperature data taken at 532 nm show a definite angular dependence; temperatures increase with decreasing scattering angles at angles less than 80°. At shallow scattering angles and an incident laser wavelength of 532 nm, the damping rate of electron plasma waves by collisions exceeds the Landau-damping rate. This causes a broadening of the electron plasma wave resonance structure of the electron feature. This broadening is misinterpreted as an increase in electron temperature as a function of scattering angle. However, at 355 nm, the Landau-damping rate exceeds the collision-damping rate for all scattering angles. One therefore expects collisional effects on the electron feature to be negligible at this wavelength, and electron temperature values should be independent of the scattering angle. This was observed. We conclude that a comparison of the 532-nm data at shallow scattering angles with the 355-nm data gives a quantitative assessment of collisional effects on the electron feature.

Electron temperature values determined from line shapes unaffected by collisions at 532 nm are in good agreement with those measured at 355 nm, and range from 18 000 to 19 000 K. The electron temperature is considerably greater than the heavy particle temperature and the temperature determined from emission spectroscopy. Evidently, neither LTE nor PLTE exists in this plasma. Electron density values determined from the line shapes are apparently independent of the scattering angle for both 532- and 355-nm incident laser wavelengths. The electron density measured at 532 nm agrees with that taken at 355 nm and is around $1 \times 10^{23}\ \mathrm{m}^{-3}$.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy under Contract No. DE-AC07-94ID 13223. The Department of Energy funding was provided by the Department of Energy Office of Science, Office of Basic Energy Sciences, Engineering Research Program.

[1] M. I. Boulos, P. Fauchais, and E. Pfender, *Thermal Plasmas, Fundamentals and Applications* (Plenum, New York, 1994), Vol. 1.
[2] S. C. Snyder, L. D. Reynolds, J. R. Fincke, G. D. Lassahn, J. D. Grandy, and T. E. Repetti, Phys. Rev. E **50**, 519 (1994).
[3] S. C. Snyder, G. D. Lassahn, and L. D. Reynolds, Phys. Rev. E **48**, 4124 (1993).
[4] R. E. Bentley, J. Phys. D **30**, 2880 (1997).
[5] J. Jonkers, L. J. M. Selen, J. A. M. van der Mullen, E. A. H. Timmermann, and D. C. Schram, Plasma Sources Sci. Technol. **6**, 533 (1997).
[6] G. Gregori, J. Schein, P. Schwendinger, U. Kortshagen, J. Heberlein, and E. Pfender, Phys. Rev. E **59**, 2286 (1999).
[7] J. Sheffield, *Plasma Scattering of Electromagnetic Radiation* (Academic, New York, 1998).
[8] O. Theimer and R. Theimer, Plasma Phys. **15**, 837 (1973).
[9] D. E. Evans and J. Katzenstein, Rep. Prog. Phys. **32**, 207 (1969).
[10] R. O. Dendy, *Plasma Dynamics* (Clarendon, Oxford, 1990).