Thomson scattering measurements in atmospheric plasma jets

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Electron temperature and electron density in a dc plasma jet at atmospheric pressure have been obtained using Thomson laser scattering. Measurements performed at various scattering angles have revealed effects that are not accounted for by the standard scattering theory. Differences between the predicted and experimental results suggest that higher order corrections to the theory may be required, and that corrections to the form of the spectral density function may play an important role. [S1063-651X(99)08202-1]

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I. INTRODUCTION

Temperature and density measurements using Thomson laser scattering have been extensively used since the theory of electron density fluctuations in plasmas was first established by Salpeter [1,2] in the 1960s. When this diagnostic technique was applied to the measurement of electron temperatures in atmospheric plasma jets, the results have indicated a strong discrepancy with other well established diagnostic methods such as emission spectroscopy or enthalpy probe measurements [3]. In particular, temperatures derived from Thomson scattering have been found to be considerably higher than those obtained with the more traditional techniques [3–6]. At the same time, electron density values derived from Thomson scattering measurements have been similar to those derived from other techniques, approximatively 10^{17} cm^{-3} for atmospheric pressure plasma jets close to nozzle exit. The validity of Thomson scattering experiments as well as other plasma diagnostics has been questioned by several authors [4–6], mainly because, if correct, Thomson results would suggest a strong departure of the plasma jet from the condition of local thermodynamic equilibrium (LTE). Plasma jets are not the only case where strong discrepancies between results from Thomson scattering measurements and from emission spectroscopy have been found. Microwave driven torches [7] and thermal arcs [6] have been reported to exhibit Thomson temperatures considerably higher than expected. On the other hand, LTE should be approached in plasmas with electron densities as high as the values reported by the experiments [8]. Snyder *et al.* [4] proposed a mechanism which in principle could account for non-LTE conditions. Three-body collisions, where one electron and one ion recombine during the encounter with another electron, leave the free electron with an excess energy which could be a significant part of the recombination energy. Therefore, electrons would be heated to a higher temperature than the ions, if the three-body electron-ion recombination rate would be sufficiently high. Detailed calculations, however, have shown that this heating process is negligible [4]. In addition, many effects in the experiment which could perturb the results were also considered by Snyder *et al.* [4]. These include linear inverse bremsstrahlung, influence of electron collisions, and fluctuations of the plasma jet. Taking all these corrections into account, significant differences from LTE would still remain.

In this paper we address, in addition to the aforementioned problems of discrepancies in electron temperature measurements, another inconsistency in the values for the temperature obtained by Thomson scattering. Our scattering measurements have shown an electron temperature dependence on the scattering angle. The standard theory [1,2,9–11] takes into account a change in the scattering angle by a proper modification of the spectral density function, which in turn describes the scattered power from the plasma electrons. Assuming the correctness of such an approach, we have to conclude that strong asymmetric conditions exist in the plasma jet. However, conditions of strong asymmetry were never reported in such plasmas, a fact that, in addition to the previous discussion, leads to the conclusion that some other effects, not considered in the standard theory, may indeed play an important role in determining the scattering profile in thermal plasmas.

II. EXPERIMENT

The experimental setup is shown in Fig. 1. A continuum *Q*-switched frequency doubled pulsed neodymium-yttrium aluminum garnet (Nd:YAG) laser is used to excite the plasma electrons. The pulse duration is 10 ns with a repetition frequency of 20 Hz, and the laser is operating at a wavelength of 532 nm. The plasma is generated with a Miller SG100 torch operating at atmospheric pressure with argon.

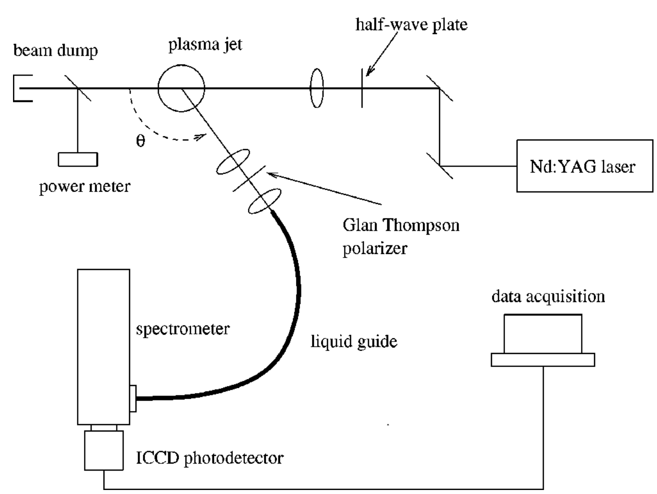

FIG. 1. Experimental layout.

In the experiment, a gas flow rate of 35.4 l/min and an arc current of 700 A at 30–35 V have been used. Data are taken in the plasma jet emanating from the 8-mm-diameter anode nozzle 4 mm from the nozzle exit. To be able to compare with previous results, the experimental setup has been chosen to be similar to the arrangement of Snyder *et al.* [4]. Therefore, details of the torch schematic and laser characteristics can be found there. Data collection is performed at various scattering angles by adjusting the position of a 2m-long visible-light liquid-guide, which ensures higher transmission than standard optical fiber bundles. The transmitted light is then imaged onto the 200-μm entrance slit of a 1m Acton Research AM510 monochromator. To disperse the light spectrally, we used a $140\times 120\ \mathrm{mm}^2$, 1800 groove/mm holographic grating. The line profile is measured with a Princeton Instrument two-dimensional intensified charge-coupled device gated array detector. The plasma jet is aligned perpendicularly to the scattering plane, and, to maximize the signal, the direction of polarization of the incident laser beam has been rotated along the direction of the jet axis with a half-wave plate. Since Thomson scattered light preserves the same polarization direction as the incident beam, a Glan-Thompson polarizer has been used to reduce unpolarized background light from the plasma.

It is known [12,13] that linear inverse bremsstrahlung can significantly heat up the plasma electrons if the laser energy flux is high enough. To eliminate this effect, Snyder *et al.* [4] performed measurements at various laser energies to be able to extrapolate the correct temperature for a weak nonperturbing electromagnetic field in the plasma. In our experiment, a Coherent *Labmaster Ultima* powermeter has been used to measure the laser energy for every pulse. In addition to this technique, we have carried out our measurements with a defocused beam at the jet axis, where data were gathered. A spot size of 2-mm diameter was used.

III. THEORY AND RESULTS

To derive values for the temperature and the density from the scattered light intensities, the experimental line profile has to be fitted with a theoretical line shape. According to the theory, the scattered power is proportional to the spectral density function $S(\mathbf{k},\omega)$, which, approximating the plasma as an electron gas with a neutralizing background of positive charges, takes the form [14]

$$S(\mathbf{k},\omega)=\frac{n_e}{|\epsilon(\mathbf{k},\omega)|^2}\int d\mathbf{v}\, f_e(\mathbf{v})\,\delta(\omega-\mathbf{k}\cdot\mathbf{v}), \tag{1}$$

where n_e is the electron density, ϵ is the plasma permittivity, f_e is the electron energy distribution function, $\mathbf{k}=\mathbf{k}_s-\mathbf{k}_i$ is the difference between the scattered and incident wave vector, and $\omega=\omega_s-\omega_i$ is the difference between the incident and scattered frequencies. In the nonrelativistic regime, $k=(4\pi/\lambda)\sin(\theta/2)$, where λ is the laser wavelength (532 nm in our case) and θ is the scattering angle, defined as the angle between the $\mathbf{k}_i$ and $\mathbf{k}_s$ vectors. In general, f_e is assumed to be a Maxwellian distribution at a temperature T_e, since electron-electron encounters are highly efficient in thermalizing the electron ensemble [8]. Within the Vlasov regime (collisionless plasma), Eq. (1) can be written as [9–11,14]

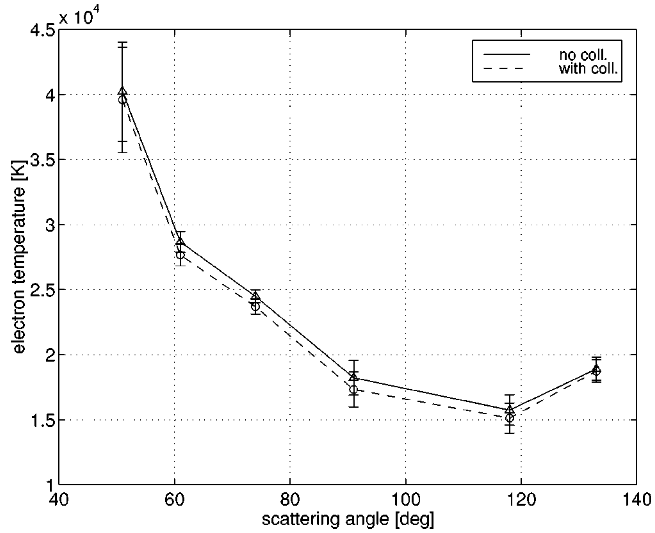

FIG. 2. Electron temperature T_e vs scattering angle. Collisionless model (solid line) and collisional model (dashed line).

$$S(\mathbf{k},\omega)=\frac{n_e}{\sqrt{2}v_t}\frac{e^{-x_e^2}}{|1+\alpha^2 W(x_e)|^2}. \tag{2}$$

Here $v_t=(k_B T_e/m)^{1/2}$ is the electron thermal speed, where k_B is the Boltzmann constant and m the electron mass. The plasma permittivity is given by [15]

$$\epsilon=1+\alpha^2 W(x_e), \tag{3}$$

with

$$\alpha=\frac{1}{k\lambda_D}, \tag{4}$$

where λ_D is the Debye length, and

$$x_e=\frac{\omega}{\sqrt{2}kv_t}, \tag{5}$$

$$W(x_e)=1-2x_e e^{-x_e^2}\int_0^{x_e} e^{y^2}dy+i\sqrt{\pi}x_e e^{-x_e^2}, \tag{6}$$

where k is the wave number and y a dummy variable.

As mentioned before, to obtain electron temperature and electron density measurement, we need to match the experimental line profile with the one given in Eq. (2), by properly adjusting the values for T_e and n_e. This nonlinear fitting is performed using the Levenberg-Marquardt algorithm, as described by Press *et al.* [16]. In comparison to previous studies, here we have performed measurements at different scattering angles. The results are given in Figs. 2 and 3. Typically, one should expect that the electron density and electron temperature are independent of the scattering angle. Clearly, the electron temperature shows a strong dependence on the scattering angle. Since the dependence on the scattering angle is accounted for in Eq. (2), our measurements reveal an effect that, to our knowledge, has not been reported in previous publications of measurements in atmospheric plasma jets. We also notice that a scattering angle of 90°

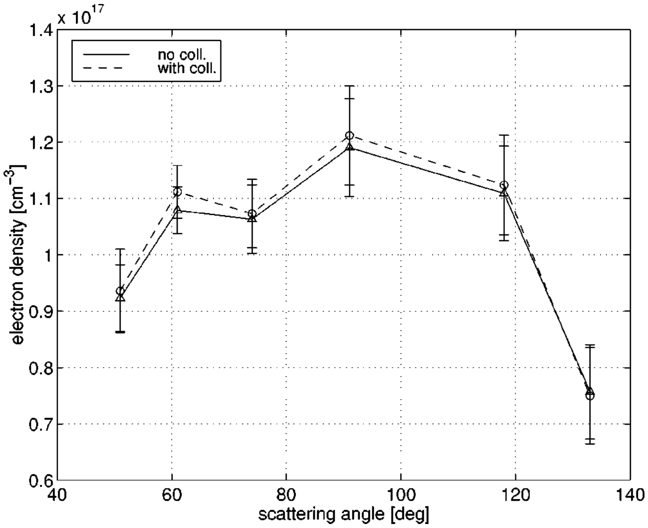

FIG. 3. Electron density n_e vs scattering angle. Collisionless model (solid line) and collisional model (dashed line).

results in values that are similar to the ones given by Snyder *et al.* [4]. Since the theory used to calculate the spectral density function is based on the assumption of collisionless plasmas, we have been interested in seeing if the inclusion of electron-ion collisions could account for this dependence on the scattering angle. However, the inclusion of such a collision term in the Boltzmann equation leads, in general, to solutions which cannot be expressed in a closed form, except for some simplified cases. Modification of the spectral density function for a Balescu-Lenard collision integral has been studied by Jasperse and Basu [17,18]. Their work has been shown to agree with the quantum mechanical calculation of DuBois, Gilinsky, and Kivelson [19]. The plasma permittivity in the regime of dominant electron-ion collisions takes the form [17]

$$\epsilon = 1 + \alpha^2 [W(x_e) + i\,\eta_{\rm ei} I_{\rm ei}(\sqrt{2}x_e)], \tag{7}$$

where $\eta_{\rm ei} = \nu_{\rm ei}/kv_t$, with $\nu_{\rm ei}$ the electron-ion collision frequency, and $I_{\rm ei}$ is a complex integral which is given by Jasperse and Basu [17], and for an argon plasma can be written as

$$I_{\rm ei}(z) = -\frac{z}{2}\int_0^1 W_2\left(\frac{z}{\sqrt{1-\mu^2}}\right)\frac{\mu^2}{(1-\mu^2)^2}\,d\mu, \tag{8}$$

with

$$W_n(\xi) = i^n \int_0^\infty \exp(i\xi t - t^2/2)\,t^{n+1}\,dt, \tag{9}$$

with μ and t dummy variables. Results for n_e and T_e have been obtained with this collision term, and are also plotted in Figs. 2 and 3. As we can see, the introduction of the collision term in the spectral density function does not significantly change the angular behavior of the electron temperature. This indeed agrees with the preliminary calculations of Snyder *et al.* [4], who showed that electron collisions seem to have little influence on the line shape in thermal plasmas. In conclusion, our results show that a straightforward interpretation of Thomson scattering results in our plasma is highly questionable due to the surprising angle dependence obtained by application of the standard scattering theory.

IV. DISCUSSION

A possible explanation of this unexpected behavior of the electron temperature is based on the consideration that such an effect has to be correlated with strong density gradients that change the plasma properties within the region from which scattered light is collected. The spectral density function (1) is obtained by a proper ensemble average of the Fourier transform of the point-particle density distribution, which for the electrons in a box of unit volume with periodic boundary conditions takes the form [14]

$$\rho(\mathbf{r}) = \sum_{j=1}^{n_e} \delta(\mathbf{r}-\mathbf{r}_j), \tag{10}$$

where r_j is the (time dependent) position of the jth electron, and n_e is the mean number of electrons in the box. Expanding $\rho(\mathbf{r})$ into Fourier series, we have

$$\rho(\mathbf{r}) = \sum_{\mathbf{k}} \rho_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}, \tag{11}$$

where $\mathbf{k}$ is a vector whose components are integers, with the sum extended over the entire range of wave numbers. The Fourier components of $\rho(\mathbf{r})$ are given by

$$\rho_{\mathbf{k}} = \int d\mathbf{r}\,\rho(\mathbf{r})e^{-i\mathbf{k}\cdot\mathbf{r}} = \sum_j e^{-i\mathbf{k}\cdot\mathbf{r}_j}. \tag{12}$$

In the limit of the random phase approximation (RPA), Pines and Bohm [20] derived an equation of motion for $\rho_{\mathbf{k}}$ in an electron plasma with a uniform background of positive charges

$$\ddot{\rho}_{\mathbf{k}} + \omega_{\rm pe}^2 \rho_{\mathbf{k}} = -(kv_t)^2 \rho_{\mathbf{k}}, \tag{13}$$

where $k = |\mathbf{k}|$ and $\omega_{\rm pe} = (4\pi e^2 n_e/m)^{1/2}$ is the electron plasma frequency. The collective regime will dominate if $\omega_{\rm pe} \gg (kv_t)$, and, in the opposite limit, electrons will behave as free individual particles. The RPA holds when nonlinear interactions among density fluctuations are negligible, which is usually the case if we have a large number of particles randomly distributed in the volume [20]. The theory derived by Pines and Bohm is strictly valid only in a region where the average electron density is constant. We may notice that the typical length scale for the kth fluctuation mode is of the order of $1/k$. If Λ is the characteristic length of density inhomogeneities, then we may avoid the treatment of the plasma boundary provided that $\Lambda \gtrsim 1/k$. Assuming Λ to be of the order of the radius of the jet nozzle, this condition is satisfied for the scattering angle we have considered.

Let us assume that electrons are distributed over a volume V, which is subdivided into two regions V_1 and V_2 of uniform electron density. In both regions we can write an equation for the density fluctuations of the type of Eq.(13). Thus, multiplying each equation with the corresponding volume and adding them together, we obtain

$$V\ddot{\rho}_{\mathbf{k}}+\frac{4\pi e^2}{m}(V_1 n_{e,1}\rho_{\mathbf{k},1}+V_2 n_{e,2}\rho_{\mathbf{k},2})$$
$$=-V_1(kv_{t,1})^2\rho_{\mathbf{k},1}-V_2(kv_{t,2})^2\rho_{\mathbf{k},2}, \tag{14}$$

where the subscripts 1 and 2 refer to corresponding volumes. Here, $\rho_{\mathbf{k}}=(V_1\rho_{\mathbf{k},1}+V_2\rho_{\mathbf{k},2})/V$. Similarly we can define an average electron density in the volume V as $n_e=(V_1 n_{e,1}+V_2 n_{e,2})/V$. If we now write

$$n_{e,1}=n_{e,2}+\nabla n_e\cdot\mathbf{s}, \tag{15}$$

$$\rho_{\mathbf{k},1}=\rho_{\mathbf{k},2}+\nabla\rho_{\mathbf{k}}\cdot\mathbf{s}, \tag{16}$$

where $\mathbf{s}$ is the displacement vector from the volume V_1 to V_2, and if we assume that the electron temperature is constant in V, we obtain

$$\ddot{\rho}_{\mathbf{k}}+\omega_{\mathrm{pe}}^2\rho_{\mathbf{k}}(1+\delta)=-(kv_t)^2\rho_{\mathbf{k}}, \tag{17}$$

with

$$\delta=\frac{V_1V_2}{V^2}(\nabla\ln n_e\cdot\mathbf{s})(\nabla\ln\rho_{\mathbf{k}}\cdot\mathbf{s}). \tag{18}$$

In Eq. (17) the plasma frequency is evaluated with respect to the average electron density in the total volume V. The constant temperature approximation inside the volume V needs some justification. In general, such an approach holds if the electron density profile is steeper than the temperature profile along the jet radius. From spectroscopic measurements reported by Snyder *et al.* [4], we see that this condition applies in atmospheric jets.

The expression for δ takes a simpler form in the case of a rotationally symmetric plasma jet, and a rotationally symmetric scattering volume. Even if the plasma jet tends to oscillate [21], due to fluid turbulence and change in the position of the anode attachment, on a time scale of several seconds (which is the usual time required to collect enough light from single-shot laser pulses), the jet is very stable and the approximation of cylindrical symmetry holds. On the other hand, the scattering volume is often very far from being cylindrical, and such an approximation may not be satisfied. In addition to that, we must also consider the fact that averaging over a long period of time may indeed introduce an additional source of error, since it could alter the value of density variations inside the scattering volume. The consequence of this time averaging is a smoothing of the density profile, thus apparently reducing the effect of density gradients.

Let us consider a cylindrical plasma column of length l and radius r. Its volume is thus $V_1=\pi r^2 l$. Suppose we add an infinitesimal layer of thickness dr and volume $V_2=2\pi rl\,dr$, then consider the total volume $V=V_1+V_2\approx V_1$; we have $(V_1V_2)/V^2=(2/r)dr$. Similarly, if Λ is the characteristic length of spatial gradients, we may approximate

$$\nabla n_e\cdot\mathbf{s}\approx-\frac{n_e}{\Lambda}r, \tag{19}$$

$$\nabla\rho_{\mathbf{k}}\cdot\mathbf{s}\approx-\frac{\rho_{\mathbf{k}}}{\Lambda}r. \tag{20}$$

With these results, the density correction term (18) can be written as $d\delta=(2r/\Lambda^2)dr$, where the differential points out the fact that only an infinitesimal layer has been added. Assuming that the region of the plasma under investigation has a radius R_s, then by integration we obtain the total density correction term

$$\delta=\int_0^{R_s}\frac{2r}{\Lambda^2}dr=\left(\frac{R_s}{\Lambda}\right)^2. \tag{21}$$

Let us return to Eq. (17), which has a natural physical interpretation if we write the dispersion relation for the wave associated to the kth fluctuation mode. Assuming $\rho_{\mathbf{k}}\sim e^{i\omega_k t}$, we obtain

$$\omega_k^2=\omega_{\mathrm{pe}}^2(1+\delta)+(kv_t)^2. \tag{22}$$

Setting $\delta=0$ yields the standard Langmuir relation. Therefore, the effect of density inhomogeneities requires introducing a correction term in the dispersion relation. For a particular wave frequency, the wave number k is higher than it would have been in the absence of the density perturbation. We can then introduce an effective wave number

$$\tilde{k}=k\left[1+\frac{\omega_{\mathrm{pe}}^2}{(kv_t)^2}\delta\right]^{1/2}=k(1+\alpha^2\delta)^{1/2}, \tag{23}$$

which accounts for the presence of large scale gradients in the plasma volume under consideration. It is interesting to notice that, due to the $1/k^2$ dependence, the correction term is considerably stronger at smaller wave numbers, i.e., large α, while for $\alpha\ll 1$ the correction becomes unimportant. It is crucial to remark that the quantity k in an actual experiment is considered constant since the scattering angle is fixed by the chosen experimental arrangement. Under these conditions, a change in the effective wave number will appear as a change in the quantity kv_t, and the experiment will *measure* an electron temperature that formally accounts for the physical effect of the change in the wave number. We can easily determine this *effective* temperature measured by Thomson scattering as

$$\tilde{T}_e=T_e\left[1+\frac{\omega_{\mathrm{pe}}^2}{(kv_t)^2}\delta\right]=T_e(1+\alpha^2\delta) \tag{24}$$

from the requirement $\tilde{k}v_t=k\tilde{v}_t$.

V. CONCLUSIONS

The corrected temperature (24) has an angular dependence that is similar to the one shown in our experiment. In a very simple approximation we may try to regard the term δ given by Eq. (18) as a constant. Such an approach would be completely justified only if the scattering volumes are the same at different scattering angles. However, it is clear that by changing the scattering angle, the sampling volume is modified too, and the assumption of constant δ is not valid. Figure 4 shows how the collection volume changes with the

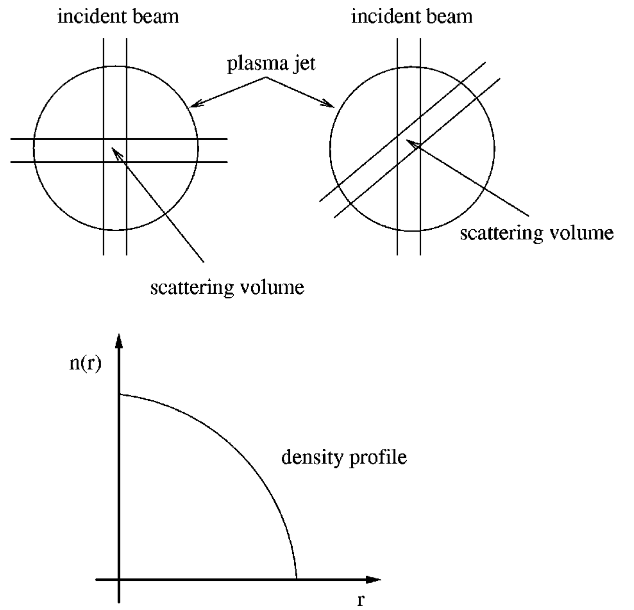

FIG. 4. Change in the collection volume at different scattering angles.

scattering angle. There is also another effect to take into account: as the sampling volume changes, the average electron density in the volume is modified as well. On the other hand, the effect of different scattering volumes at different angles can also be seen from Fig. 3, where the electron density tends to decrease for both small and large angles. In these situations the scattering volume is indeed larger than in a region close to 90°, resulting in lower values of the average electron density, which is integrated over the scattering region. We can account for this change in the electron density by writing

$$n_e = n_{e0} \sin\theta, \tag{25}$$

where n_{e0} is the average electron density at 90°. Reported in Table I are the values of n_{e0} obtained by fitting Eq. (25) with the experimental data for the density. In Fig. 5 the predicted angular dependence of the density measurements is compared with the experimental values obtained in the collisionless regime. Similarly, we can approximate the change in the effective spot radius as $R_s = R_{s0}/\sin\theta$, where R_{s0} is the spot size at 90°. In the limit of the approximation used to derive Eq. (21), we can then account for the change in volume by letting

$$\delta = \delta'/\sin^2\theta, \tag{26}$$

where $\delta' = (R_{s0}/\Lambda)^2$ is a true constant to be determined. We can thus estimate the corrected electron temperature by fitting our experimental data with the expression given in Eq. (24) with $\delta = \delta'/\sin^2\theta$, and, using for the electron density Eq. (25), we obtain the results given in Table I. In Fig. 6 this model is compared with the experimental electron temperature obtained in the collisionless regime. As we can see, the theoretical model we have presented is able to predict with reasonable accuracy the experimental dependence of the electron temperature with the scattering angle. Therefore, in the limit of this approach, we can conclude that the density gradient could account for a significant increase in the electron temperature measured by Thomson scattering. If correct, the theory we have presented could indeed resolve the discrepancy with the spectroscopic and probe measurements. In particular, the values reported in Table I are not too far from the electron temperature measured by emission spectroscopy [22,23], and are in reasonable agreement with numerical

TABLE I. Corrected electron temperatures and electron densities for the collisionless and collisional regimes.

	n_{e0} (10^{17} cm^{-3})	δ'	T_e (K)
no coll.	1.173 ± 0.032	0.096 ± 0.007	10656 ± 1117
with coll.	1.190 ± 0.036	0.094 ± 0.007	10088 ± 1259

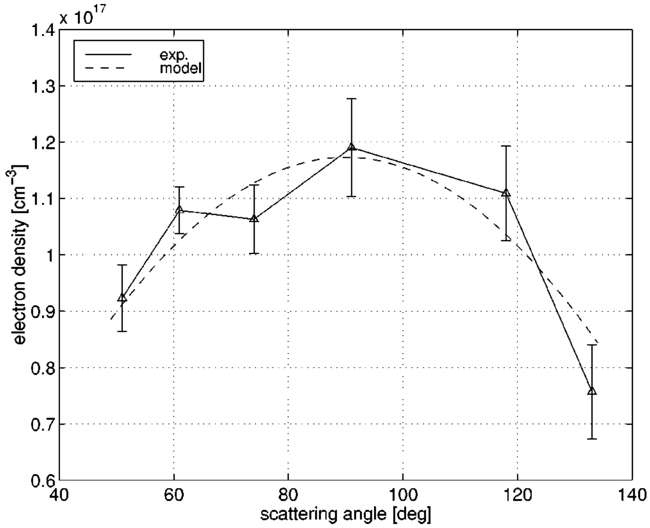

FIG. 5. Predicted electron density from Eq. (25) with $n_{e0} = 1.173 \times 10^{17}$ cm^{-3}. The experimental values have been obtained in the collisionless model.

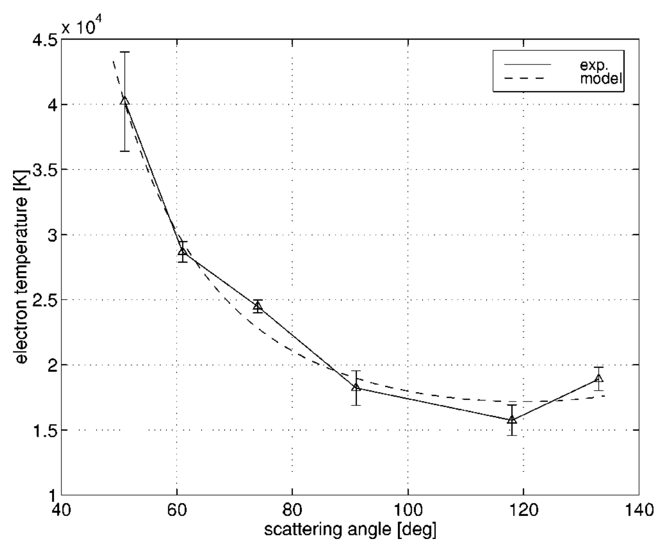

FIG. 6. Predicted electron temperature from Eq. (24), with Eq. (25) for the electron density and Eq. (26) for the density correction. We used $n_{e0} = 1.173 \times 10^{17}$ cm^{-3}, $T_e = 10656$ K, and $\delta' = 0.096$. The experimental values have been obtained in the collisionless model.

simulations of the plasma jet [22,24]. We can observe that the measurements taken at larger scattering angles ($\gtrsim 90°$) seem to be more accurate. This is evident from the fact that in this regime k is large, thus reducing the effects of both electron ion collisions and density gradients, as seen from Eqs. (7) and (24). However, the determination of T_e still requires measurements at different scattering angles and a fit to determine δ'.

It should be pointed out that we cannot test the accuracy of our theory with the present experimental apparatus. This can be seen if we, for example, write down the apparent temperature incremenent due to density inhomogeneities for a rotationally symmetric plasma using Eqs. (21) and (24),

$$\Delta T_e = \frac{4\pi e^2 n_e}{k_B}\left(\frac{R_s}{k\Lambda}\right)^2, \tag{27}$$

where the same limitations apply for the assumption of a constant δ, as previously discussed. For a simple estimate, we may take $n_e \approx 10^{17}\ \mathrm{cm}^{-3}$, and a typical length scale for the density variations of the order of the jet nozzle radius $\Lambda \sim R = 0.4$ cm. In Fig. 7, ΔT_e is plotted for various scattering angles versus the spot diameter R_s. As stressed before, Eq. (27) is only a crude approximation, and only some qualitative conclusions can be inferred. First, the temperature increment seems to be a strong function of the scattering volume. However, in a real experiment it is difficult to have a direct control on the collection volume. This is also because of the jet shot-to-shot fluctuations, which may change the value of the density gradients within the volume itself.

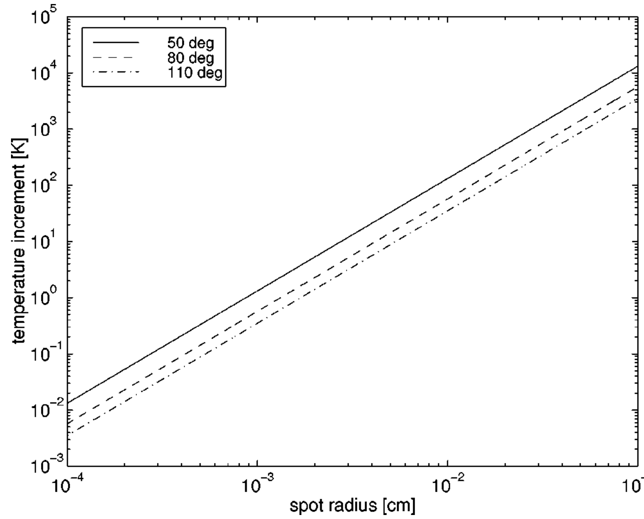

FIG. 7. Temperature increment ΔT_e vs spot radius R_s for various scattering angles.

Taking all these effects into account, we may conclude that to have a negligible effect on the temperature, a spot size of a few μm would be required. Such a dimension is well beyond our present capabilities.

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