Novel Analysis of Thomson Scattering Data from Thermal Plasma Jets

G. Gregori, U. Korshagen, J. Heberlein, E. Pfender
Department of Mechanical Engineering, University of Minnesota
111 Church St. S.E., Minneapolis, MN 55455, USA

Abstract

In this paper we present a novel analysis technique of the Thomson scattered light from a thermal plasma jet that goes beyond the standard random phase approximation (RPA) and provides more consistent measurement values for the electron temperature and density in the weakly coupled and highly collisional plasma regime. Our approach is based on a memory function formalism (MFF) for the spectral density function with the use of the three lowest order frequency-moment sum rules. The proposed scattering technique is compared with the RPA result and with the standard BGK collisional model for the dynamic structure. It is shown that the obtained electron temperature values are close to local thermodynamic equilibrium (LTE), in contrast with previous findings.

1. Introduction

Electron density and electron temperature measurements in thermal arcs and plasma jets using Thomson laser scattering have been often questioned in the past,\textsuperscript{1,2,3} since they seem to indicate a large departure from the condition of local thermodynamic equilibrium (LTE), a result not confirmed from other well established diagnostics, such as emission spectroscopy or enthalpy probes. In addition, some authors\textsuperscript{2} have recently reported a dependence of the measured electron temperatures on the scattering angle, suggesting that a straightforward interpretation of light scattering data based on the commonly used random phase approximation (RPA) may not always be accurate. Inclusion of electron-ion collisions\textsuperscript{4,5} or correction for inhomogeneities in the scattering volume\textsuperscript{2} have been proposed as possible mechanisms responsible for the observed angular dependence in the analysis of measured data using the RPA. However, such approaches are still heuristic without rigorous theoretical justifications.

The degree of inter-particle coupling in typical plasma jets of electron temperature $T$ and electron density $n$ is $\Gamma=\gamma d_{D}^{3}$, where $d=(3/4\pi n)^{\frac{1}{3}}$ is the ion-sphere radius. This corresponds to a few electrons in the Debye sphere. It is then clear that the classical picture of ideal plasmas ($\Gamma<<1$) may start to break down, even if we are far from the region of strong coupling ($\Gamma>>1$). In addition, typical electron-ion collision frequencies are comparable to the plasma frequency, thus the main assumptions used in the RPA (namely, an ideal, collisionless plasma) become invalid.

In this paper we will show that the use of a theory for the electron density fluctuations that is valid in the region of interest, can indeed resolve the discrepancies previously reported in the measurements.

2. Experiment

The experimental setup is shown in Fig. 1. A dc torch operating at atmospheric pressure with argon has been used to generate the jet, which is probed 4 mm downstream from the
nozzle exit with a Q-switched frequency-doubled (532 nm) Nd:Yag laser. The pulse duration is 10 ns with a repetition rate of 20 Hz. The torch has been operated with a gas flow rate of 35.4 l/min and an arc current of 600-700 A at 30-35 V. The jet diameter at the nozzle exit is approximately 8 mm. Data collection is performed at various scattering angles with a visible-light liquid-guide and then imaged onto the entrance slit of a monochromator equipped with a 140 x 120 mm², 1800 groove/mm holographic grating. The line profile is then measured with a 2-D intensified charge-coupled device (ICCD) 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 light has been rotated along the direction of the jet axis with a half-wave plate. A Glan-Thompson polarizer has been used to reduce unpolarized background light from the plasma. Additional details of the experimental setup can be found in Gregori et al.²

The characteristic time-scale for electron heating in a laser pump field is given by $\tau_{ei} = M/2m\nu_{ei}$, where $\nu_{ei}$ is the electron-ion collision frequency. For typical plasma conditions in our experiment $\tau_{ei}=10$ ns, of the same order as the laser pulse duration. Collisional (bremsstrahlung) electron heating can then be significant¹, and inhomogeneous distribution of the pump field intensity may broaden the observed line-shapes differently at different scattering angles. Since this effect is linear with the field intensity, extrapolation of the measured temperature values to zero pump energy has been carried out, as described by Snyder et al.¹

![Diagram](image)

**Figure 3**: Schematic of the experimental setup.

3. Theory

In a Thomson scattering experiment, the intensity of the scattered radiation is proportional to $S(k,\omega)$, the electron density-density correlation function, or spectral density function, which represents the spectrum of the longitudinal density fluctuations. In the low energy (non-relativistic) limit $k=|k_i-k_s|=\frac{4\pi}{\lambda_i}\sin(\theta/2)$, where $k_i$ and $k_s$ are the incident and scattered wavenumber, respectively, $\lambda_i$ is the incident laser wavelength, and $\theta$ the scattering angle. The difference between the scattered and the incident photon frequency is $\omega = \omega_s - \omega_i$. In the high frequency limit, $\omega \gg v_i$, where $v_i=(k_T/m)\frac{1}{2}$ is the electron thermal speed, $S(k,\omega)$ is well approximated by $S_{zz}(k,\omega)$, the charge-charge correlation function. This is the relevant structure factor that determines the high frequency line-shape of the light scattered signal.
from free electrons in the plasma. Since a weakly non-ideal plasma does not have short-range order, characteristic of highly correlated systems, it is reasonable to assume a simple Debye-Hückel form for the pair correlation function. The charge-charge correlation function then satisfies the following frequency-moment sum rules for the typical plasma conditions we encounter:

\[
\Omega_0 = \int S \omega Z(k, \omega) d\omega = S(k) = \frac{k^2}{k^2 + k_D^2};
\]

\[
\Omega_2 = \int \omega^2 S \omega Z(k, \omega) d\omega = \frac{1}{2} (kv_c)^2;
\]

\[
\Omega_4 = \int \omega^4 S \omega Z(k, \omega) d\omega = \frac{1}{2} \left[ 3(kv_c)^4 + (kv_c)^2 \omega_p^2 \right]
\]  \hspace{1cm} (1)

where \( k_D = (8\pi e^2 / k_B T)^{1/2} \) is the inverse of the Debye length and \( \omega_p \) is the plasma frequency. We can notice that the zeroth order momentum represent the total power at a given scattering wavenumber, \( \Omega_2 \) is a restatement of the conservation of the number of particles, and \( \Omega_4 \) includes the effect of the particle interactions. Using a memory function formalism, the charge-charge correlation function can be expressed in a very general form:\n
\[
S \omega Z(k, \omega) = \frac{(kv_c)^2 N(k, \omega)}{\sqrt{\omega^2 - (kv_c)^2}/2S(k) - \omega^2 N\omega(k, \omega) + [\omega N(k, \omega)]};
\]

where \( N(k, \omega) \) and \( N\omega(k, \omega) \) are the “memory functions”. Here, \( N(k, \omega) \) represents the damping of the electron plasma wave and \( N\omega(k, \omega) \) its dispersion. In a phenomenological approach, the memory functions are chosen such as the correct three lowest order frequency-moment sum rules are exactly reproduced by the charge-charge correlation function. The advantage of such a representation is that we do not need anymore an exact microscopic theory to derive the spectrum of the longitudinal density fluctuations. Conversely, the spectrum is obtained in a form that is phenomenologically self-consistent. Assuming that the memory functions are much simpler objects than the density correlation itself, we adopt the following gaussian form for the damping function:\n
\[
N(k, \omega) = \sqrt{\pi} \tau (\omega_0^2 - \omega_1^2) \exp(-\tau^2 \omega^2);
\]

with \( \tau \) the relaxation time for the collective modes and

\[
\omega_0^2 = \frac{\Omega_2}{\Omega_1};
\]

\[
\omega_1^2 = \frac{\Omega_4}{\Omega_2};
\]

The dispersion memory function is then obtained from \( N(k, \omega) \) with the help of the Kramers-Kronig relation:\n
\[
N\omega(k, \omega) = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{N(k, \omega')}{\omega - \omega'} d\omega' = 2\pi (\omega_0^2 - \omega_1^2) \exp(-\tau^2 \omega^2) \int_{-\infty}^{\infty} \exp(y) dy.
\]

This completes the set of equations required to describe the longitudinal density fluctuations. The charge-charge correlation function contains then three unknowns: the electron temperature \( T \), the electron density \( n \), and the relaxation time \( \tau \). Those are determined by fitting Eq. (2) with the experimental line-shapes.

4. Results & Discussion

We compare the temperature and density values obtained with three different models for the spectral density function: the standard RPA in the form given by Salpeter\(^1\), the BGK approximation\(^1\), and the memory function approach described above. The main difference between the RPA and the BGK approximation is that the former does not consider collisions,
thus the density fluctuations are simply obtained by the solution of the Vlasov equation. This is not true anymore in the BGK model. Here, an approximate form is introduced for the collision integral in the Boltzmann equation, which is solved for the density fluctuations by treating the collision term as a perturbation on the Vlasov’s solution. In addition, both the RPA and the BGK models calculate the spectral density function using the principle of superposition of dressed particles, which requires that each charged particle always carries a well established screening cloud. In other words, the condition \( \Gamma << 1 \) (ideal plasma) must be satisfied. We shall observe that at \( k << k_D \) (small scattering angles, or large laser wavelength) we probe the density fluctuations far in the collective regime. As noticed by Snyder et al., here collisional damping is probably dominant, and the use of either RPA or BGK models is questionable. Conversely, in the region \( k > k_D \) (large scattering angles, or small laser wavelength) there occurs the transition from a kinetic to a fluid behavior of the plasma particles. This is the regime where non-ideal effects are most important, and again both RPA and BGK models may become invalid. Instead, the phenomenological memory function approach that we have described in this paper does not rely on a particular microscopic theory for the derivation of the spectral density function, and it is valid at all scattering angles.

The temperature values obtained from these three different models at various scattering angles are plotted in Fig. 2. We clearly see that both the RPA and BGK models show a strong dependence of the temperature values on the scattering angle. This behavior is much less pronounced in the memory function formalism, where, at low scattering angles, we can still see variations of the temperature values, but errors are also considerably larger. Low scattering angles correspond to the region \( k_D/k \sim 6-7 \), where collisional broadening is important.

![Graph showing electron temperature variations](image)

**Figure 4:** Electron temperature values on the axis of an argon plasma jet, 4 mm downstream, obtained at various scattering angles with three different models for the dynamic structure: RPA (full circles), BGK (full triangles) and memory function formalism (open squares).

At larger scattering angles (\( k_D/k \sim 3 \)) the differences among the models are less pronounced, but, on average, the temperature values obtained from the memory function approach are 2000 K lower. As discussed, in this region we start seeing non-ideal coupling effects. Fitting the memory function results at all scattering angles to a straight line gives \( T = 18000 \pm 4300 \) K with reasonably high goodness-of-fit probability value (>0.2). Excitation temperatures close to 15000 K have been obtained from emission spectroscopy measurements in similar plasmas at the same axial position. Within the measurement errors, emission spectroscopy and Thomson scattering results then suggest that LTE should be approached in these plasmas. An opposite conclusion was reached by Snyder et al. using the RPA in the data analysis at large
scattering angles, an indication that a correct model for the spectrum of the density fluctuation is crucial for interpreting the physical properties of weakly coupled argon plasma jets. From previous studies on the effect of couplings $\Gamma$-0.05-0.1 in the structure of the spectral density function it was shown that the experimental electron features are broader than expected from the RPA. There it was concluded that the broadening was due to density inhomogeneities and, to some extent, collisions. However, no direct justification was offered and non-ideal effects may indeed have some influence. Even if large density inhomogeneities in the scattering volume may contribute to the spectral broadening, and be responsible for the observed angular dependence of the temperature values, as suggested by some authors, Snyder et al. have presented data that favor other broadening mechanisms for typical density variations in the jet. On the other hand, we should again stress the fact that, in the memory function approach, the details of physical mechanisms responsible for the line-shape broadening are irrelevant, thus the derived temperature values do not depend on the actual damping mechanism.

In Fig. 3 the density values obtained from the same three different approaches are compared. We see that the memory function formalism provides slightly higher density values than the other methods. Previous Stark broadening measurements conducted on similar jets have shown that electron densities at the same axial position are near to $1.4 \times 10^{23}$ m$^{-3}$, in closer agreement with the memory function results.

**Figure 5:** Electron density values on the axis of an argon plasma jet, 4 mm downstream, obtained at various scattering angles with three different models for the dynamic structure: RPA (full circles), BGK (full triangles) and memory function formalism (open squares). Errors in the reported values are of the order of 10% or less, in all cases.

5. Conclusions

In this paper we have discussed a novel technique to analyze Thomson laser light scattered data from thermal plasma jets. It is argued that the phenomenological description of the longitudinal density fluctuations based on the memory function formalism is more accurate than the standard random phase approximation when collisions or non-ideal plasma effects are significant in determining the dynamic structure of correlated systems. Indeed the preliminary results shown here seem to confirm that some of the problem reported in the past with Thomson scattering measurements of electron temperature and density can be avoided if the proposed approach is followed. In particular, electron temperatures obtained with this method are much less dependent on variations of the scattering angle, with average values
close to the ones determined from spectroscopy measurements of line intensity. This would then favor conditions of LTE or near-LTE in the plasma jet, contrary to the results that would have been obtained instead from the use of the RPA in modeling the spectrum of the density fluctuations. A memory function approach also shows better agreement with independent Stark broadening measurements of electron densities, confirming its overall validity in describing the dynamic structure of these plasmas.

Acknowledgements

This work was supported by the Engineering Research Program of the Office of Basic Energy Sciences of the US Department of Energy under grant FG02-85ER-13433.

References