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Thomson scattering experiments on a 100 MHz inductively coupled plasma calibrated by Raman scattering


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A new calibration method to obtain the electron density from Thomson scattering on an inductively coupled plasma is discussed. Raman scattering of nitrogen is used for recovering the Rayleigh scattering signal. This has the advantage that no corrections are necessary for stray light, like with other calibration methods, using the direct measured Rayleigh scattering signal on a well-known gas. It is shown that electron densities and electron temperatures can be measured with an accuracy of about 15% in density and of about 150 K in temperature. © 1995 American Institute of Physics.

I. INTRODUCTION

Fundamental studies of inductively coupled plasmas (ICP) are necessary to improve their applications in spectrochemical analysis and in the use for light sources. Since the electron temperature and electron density are central plasma parameters, a Thomson scattering setup is designed for these parameters on a 100 MHz argon ICP. The construction of the diagnostic is similar to the one realized by Van de Sanden et al.1 One of the features of this diagnostic is a detector, consisting of a photodiode array, in combination with a holographic grating with 2000 lines/mm. This results in a high spectral resolution in combination with a small apparatus profile.

Since the ICP is an atmospheric plasma of small dimensions, it is necessary to pay more attention to the method of measuring Thomson profiles and performing the calibration. The presence of atmospheric conditions causes an extremely intense Rayleigh scattering signal compared to the Thomson scattering signal. This large number of Rayleigh photons, together with stray light, will cause blooming on the photodiode array, which disturbs the spectral broader Thomson signal. To eliminate the large number of disturbing photons on our detector, the detector is physically darkened for the channels that would otherwise cause blooming. By blocking these channels, it is possible to measure an undisturbed Thomson scattering profile, from which only a limited central part is missing.

The small dimensions of the plasma and its surroundings are the origin of the laser stray light that can enter the detector and looks similar to the Rayleigh scattering profile, with the shape of the apparatus profile. Therefore, the Rayleigh scattering signal cannot be measured separately. In order to unravel Rayleigh scattering from the stray light component one has to measure Rayleigh scattering on two different gases with different cross sections. The difference between the ratio of the measured intensities and the ratio of the known scattering cross sections can be used to estimate the amount of stray light. This method of calibrating the setup takes a long time. Moreover, since we darkened the detector for this signal, it is not possible to use this calibration method as performed by Hieftje et al.,2 that is, measuring the Rayleigh signal. Their apparatus3 has less spectral resolution, but has the advantage that blooming is absent.

In the past it was suggested by Rühr4 that a Raman spectrum can be used for calibration. This calibration method5 for Thomson scattering experiments on a tokamak uses Raman scattering on hydrogen. In our case, a procedure was developed to recover the Rayleigh intensity from the Raman spectrum of nitrogen. The advantages of this calibration method are that stray light no longer plays any role and that the procedure can be applied easily and quickly.

This new method of calibrating the Thomson measurements on an atmospheric plasma is described in this paper.

Further, the results of experiments on a 100 MHz ICP are presented, where a large skin effect might be responsible for the difference with Thomson experiments on a 27 MHz ICP as previously published by Huang et al.6

II. RAYLEIGH AND RAMAN SCATTERING OF NITROGEN

To use the Raman spectrum for calibration, the intensity ratio of a specific Raman transition to the Rayleigh signal has to be presented as a conversion factor. It has to be noticed that the calibration will be done on nitrogen at room temperature, while the Thomson measurements to obtain electron density \(n_e\) and electron temperature \(T_e\) are carried out on free electrons in the plasma. Since the cross sections for the different Raman transitions depend on more parameters than the Rayleigh transitions, we chose for recovering the Rayleigh scattering signal of nitrogen instead of the direct approach which does not discern the Rayleigh signal and converts the Raman scattering intensity directly into the sensitivity of the setup. Ultimately, the discussed procedure leads to a conversion factor between the several Raman transitions in nitrogen and the Thomson scattering signal. In the derivation given below no explicit formulas are used to avoid large expressions which contain parameters of little interest for the final explicit expressions (see Sec. II C).

In general, both the Rayleigh and the Raman scattering intensity \(I\) are proportional to

\[I \propto \lambda^{2-\alpha} \exp(-\mu z),\]

where \(\alpha\) is the absorption index, \(\mu\) is the absorption coefficient, and \(z\) is the path length. The factor \(\lambda^{2-\alpha}\) accounts for the different wavelength dependence of the two scattering processes. The factor \(\exp(-\mu z)\) accounts for the attenuation due to absorption as a function of path length. In the case of Rayleigh scattering, the absorption coefficient is negligible, whereas in the case of Raman scattering, it is not.

Therefore, the intensity ratio of the Raman signal to the Rayleigh signal is given by

\[\frac{I_{\text{Raman}}}{I_{\text{Rayleigh}}} = \frac{\lambda^{2-\alpha}_{\text{Raman}}}{\lambda^{2-\alpha}_{\text{Rayleigh}}} \exp(-\mu_{\text{Raman}} z),\]

where \(\lambda^{2-\alpha}_{\text{Raman}}\) and \(\lambda^{2-\alpha}_{\text{Rayleigh}}\) are the wavelength dependence of the Raman and Rayleigh signals, respectively, and \(\mu_{\text{Raman}}\) is the absorption coefficient of the Raman signal.

By measuring the intensity ratio of the Raman signal to the Rayleigh signal, it is possible to determine the electron density and electron temperature of the plasma.
with $N_J$ the number density of particles in the states with rotational number $J$. This density is predicted by the Boltzmann relation, given by

$$N_J \propto N g_J (2J+1) e^{-E(J)/kT},$$

where $N$ is the total number density of particles, $g_J$ is the nuclear degeneracy [for nitrogen $g_J=2$ ($J$ even) and $g_J=1$ ($J$ odd)], $k$ is the Boltzmann constant, $T$ is the temperature, and $E(J)$ is the energy of the rotational level $J$ in the vibrational ground state. We neglect any vibrational dependent part, since more than 99.9% of the nitrogen molecules are in the vibrational ground state at $T=300$ K.

The scattered light has two polarization dependent contributions, one parallel and one perpendicular to the polarization of the incoming beam. Since the efficiency of the diagnostic is less for the perpendicular part than for the parallel part, an instrumental correction factor $C_0$ is introduced. The difference is caused by the difference in efficiency of the grating for the two polarizations (for the grating used $C_0=2.21$ at 532 nm).

### A. Rayleigh scattering

The Rayleigh signal arises from the rovibrational transitions for which $\Delta \nu=0$ and $\Delta J=0$.

Using the Rayleigh formulas for diatomic molecules, the differential Rayleigh cross section is calculated to be

$$\left( \frac{d\sigma}{d\Omega} \right)^J_{\text{Rayl}} \propto \frac{1}{45} \left( \frac{3}{\rho N_2} + 3C_0 \right) b_{J,J} \gamma^2,$$

where $\gamma$ is the anisotropic part of the polarizability tensor. The often used averaged dipole polarizability, $\alpha$, is here expressed by $b_{J,J}$, and $\rho N_2$, $\rho N_2$ is the depolarization ratio $(2.7\times10^{-3}$ for nitrogen). The factor $b_{J,J}$ is given by

$$b_{J,J} = \frac{J(J+1)}{(2J-1)(2J+3)}.$$

The total Rayleigh signal is given by a summation over all states $J$, and is found by combining Eqs. (1) and (3),

$$I_{\text{Rayl}}^{\text{tot}} \propto \left( \frac{3}{45 \rho N_2} + \frac{3C_0}{45} \right) \gamma^2 N \sum_{J=0}^{\infty} b_{J,J}(2J+1) g_J \exp -E(J)/kT.$$

Introducing the parameters $A_{\text{Rayl}}$ and $S$, Eq. (4) can be rewritten as

$$I_{\text{Rayl}}^{\text{tot}} \propto A_{\text{Rayl}} S \sum_{J=0}^{\infty} b_{J,J}(2J+1) g_J \exp -E(J)/kT.$$

Note that in the case of measuring free electrons in the plasma, the difference in the differential scattering cross section for nitrogen and for free electrons has to be taken into account ($d\sigma_{\text{Thomson}}/d\Omega = 7.95 \times 10^{-30}$ m$^2$ and $d\sigma_{\text{electron}}/d\Omega = 6.90 \times 10^{-32}$ m$^2$ at a wavelength of 532 nm).

### B. Raman scattering

Analog to Rayleigh scattering, the Raman scattering intensity $I_{\text{Raman}}$ can be calculated with Eq. (1), where the Raman scattering contains the parallel and perpendicular polarized parts with respect to the incoming beam. The perpendicular part can easily be found in the equations because it is accompanied by the instrumental factor $C_0$.

During the measurements the $S$-branch as well as the $O$-branch pure rotational Raman spectrum is detected. The Raman cross section for the $S$-branch ($\Delta J=+2$), is given by

$$\left( \frac{d\sigma}{d\Omega} \right)^{J+2}_{\text{Raman}} \propto \frac{1}{45} \left( 4+3C_0 \right) b_{J+2,J} \gamma^2 = A_{\text{Raman}} b_{J+2,J} \gamma^2,$$

with $A_{\text{Raman}}=(4+3C_0)/45$ and

$$b_{J+2,J} = \frac{3(J+1)(J+2)}{2(2J+1)(2J+3)}.$$

For the $O$-branch ($\Delta J=-2$)

$$\left( \frac{d\sigma}{d\Omega} \right)^{J-2}_{\text{Raman}} \propto \frac{1}{45} \left( 4+3C_0 \right) b_{J-2,J} \gamma^2 = A_{\text{Raman}} b_{J-2,J} \gamma^2,$$

with

$$b_{J-2,J} = \frac{3(J-1)}{2(2J+1)(2J-1)}.$$

These equations describe the Raman intensity for the different transitions that can be measured with the experimental setup. Finally, a combination of the above-mentioned formulas will give a conversion factor that transfers the measured Raman intensities to the Rayleigh intensity.

### C. The conversion factors from Raman to Rayleigh scattering intensity

Dividing the total Rayleigh intensity by the single peak Raman intensity, the two separate conversion factors for $\Delta J = +2$ and $\Delta J = -2$, which depend only on the temperature and on $J$, are given by

$$\Delta J = +2: \frac{I_{\text{Rayl}}}{I_{\text{Raman}}} = \frac{A_{\text{Rayl}}}{A_{\text{Raman}}} S \frac{2(2J+3)}{3(J+1)(J+2)} g_J^{-1} \exp E(J)/kT,$$

$$\Delta J = -2: \frac{I_{\text{Rayl}}}{I_{\text{Raman}}} = \frac{A_{\text{Rayl}}}{A_{\text{Raman}}} S \frac{2(2J-1)}{3(J-1)} g_J^{-1} \exp E(J)/kT.$$
FIG. 1. A schematic view of the used quartz torch. Note the asymmetry of the rf coil which is found back in the results.

each of these transitions the formulas can be applied (see Sec. III C). In this way for several of the peaks (six in total), a corresponding Rayleigh scattering intensity can be calculated. Averaging gives the final value of the Rayleigh scattering intensity.

III. EXPERIMENT

A. Description of the setup

The Thomson diagnostic is developed for measuring the electron temperature and density in an argon ICP. This plasma is created by a high frequency field of 100 MHz in a coil within a quartz tube with an inner diameter of 18 mm, see Fig. 1. Note the geometry of the coil that makes the plasma asymmetric. Through this torch three separately controlled flows of argon are streaming. We apply an outer flow of 12 s/min, an intermediate flow of 0.3 s/min, and a central flow of 0.6 s/min. There is no water introduced into the central channel. The standard input power settings are 1.2 kW. The standard location of measuring lies 7 mm above the upper winding of the load coil and in the center of the skin, the hottest area in the plasma.

B. Thomson scattering diagnostic

The experimental setup is depicted in Fig. 2. A frequency doubled Nd:YAG laser (GCR 3 Quanta Ray, $E_{\text{pulse}}=0.45 \text{ J}, \tau_{\text{pulse}}=7 \text{ ns}, f_{\text{rep}}=10 \text{ Hz}$) at 532 nm is used. Two prisms lead the beam into the detection volume. The beam is focused by a lens $L_1$ ($f=1000 \text{ mm}$). Finally, the laser beam is absorbed in a laser dump. The detection branch starts with two lenses $L_2$ and $L_3$ ($f=600 \text{ mm}$) that image the detection volume onto the entrance slit (300 $\mu$m) of the

FIG. 2. The Thomson scattering setup, viewed from the top, with $L_1$, $L_2$, and $L_3$ the focusing and detection lenses, $S$ the entrance slit, $M$ the mica retarder, $G$ the 2000 lines/mm grating, $L_A$ the image intensifier, and OMA the optical multichannel analyzer.

FIG. 3. A typical Thomson profile with a fitted Gauss corresponding to $n_e=(2.7 \pm 0.4) \times 10^{21} \text{ m}^{-3}$ and $T_e=8925 \pm 150 \text{ K}$. Notice the absence of a signal in the central channel, which is caused by blocking these pixels, in order to avoid blooming.

FIG. 4. A measured Raman spectrum of nitrogen at room temperature. As an example the fitting of a Raman transition is depicted. This fitting is based on the known apparatus profile and shows that the Raman peaks are superimposed on a Rayleigh and stray light background. Present at room temperature calibration conditions, this background will disappear under plasma conditions.
monochromator. Behind the slit a half-lambda mica retarder $R$ is placed, since the grating efficiency is higher for a normal polarized beam. The dispersive element is the concave holographic grating $G$ [Jobin Yvon, 2000 lines/mm, $(100 \times 100)$ mm and $f=1$ m]. To reject the stray light, the prisms are positioned at about 3 m from the detection volume and the focusing lens $L_1$ has an antireflection coating for 532 nm. Anyhow, in the case of measuring close to the torch, the most interesting area, the largest source of scattering, is the quartz ICP torch itself, which, of course, cannot be eliminated.

The detector consists of three parts: a gated image intensifier $L_A$ ($10^4$ times amplification, $\tau_{\text{gate}}=100$ ns, imposed by laser synchronization problems), a cooled ($-20^\circ\text{C}$) optical multichannel analyzer (OMA) (photodiode array of 1024 pixels, $25 \, \mu\text{m} \times 2.5 \, \text{mm}$ each, total length 25.6 mm), and an IBM 486 PC with a 16 bit AD converter. The full spectral range of the detector is 13 nm, the apparatus profile has a $1/e$ width of 0.14 nm. To prevent the array from blooming during the measurements due to the intense Rayleigh and stray light, the OMA is physically darkened for about 50 pixels in the center, which eliminates the core of the Rayleigh scattering profile. However, it should be noticed that apart from the core, there is also a contribution of Rayleigh and stray light photons in the wings of the profile. This originates from the finite contrast of the monochromator. For the plasma-on Rayleigh scattering this wing contribution turns out to be negligible. This is due to the high gas temperature and consequently low density.

In Fig. 3 a measured Thomson scattering profile (squares) and the fitted Gaussian profile (dashed line) are depicted. The perfect fit of the Gaussian profile shows that the Doppler broadened profile is indeed not disturbed by Rayleigh scattering or stray light. From the width of this Doppler broadened profile the electron temperature can be calculated. The surface of the profile is used for calculating the electron density. Therefore, the sensitivity of the setup has to be calibrated.

C. Calibration procedure

To get absolute results on the electron density by the Thomson scattering signal, the sensitivity of the setup has to be calibrated absolutely. Van de Sanden et al. used the Rayleigh scattering signal for this purpose. Therefore, measurements on an argon gas sample of well-known pressure were performed under equal conditions as during the plasma measurements. Knowing the ratio between the Rayleigh (at 532 nm) and the Thomson cross section (equal to $1/143$) and measuring the pressure and gas temperature of the sample,
using the ideal gas law, a relation was derived between the absolute density and the number of ADC counts.

In the case of measuring on an atmospheric plasma that shows a strong Rayleigh signal, and moreover measuring on a geometrically small plasma, causing stray light, the Rayleigh signal cannot be measured properly together with the Thomson signal using the detector mentioned above. Therefore, as already mentioned, the detector is blocked for Rayleigh and stray light photons.

If the calibration on gas (plasma off) would be performed on a nonblocked detector, Rayleigh scattering would be measured together with stray light and separating them needs additional effort. Moreover, removing the strip that is used to cover the detector for measuring the Rayleigh signal would increase the inaccuracy of the calibration and take a lot of time. This is not done in our study, so the only signal that can be measured with this setup at atmospheric conditions is Raman scattering (see Fig. 4), with peaks at wavelengths which are different from that of the laser and stray light peak. Fitting the surface under a Raman peak for a value of the rotational quantum number $J$ with an apparatus profile, a corresponding Rayleigh intensity can be recovered using the conversion factors of Eqs. (8) and (9). The fits are performed with a fixed width, the width of the apparatus profile, and using only the upper-half of the profile. This has the advantage that a small overlap of the Raman peaks and a background by, for example, Rayleigh scattering does not influence the results. As an example, the fit of one Raman peak is depicted in Fig. 4. The origin of the “background” lies in a small overlap of the Raman peaks and in a flank of the Rayleigh and stray light profile. Under plasma conditions, this flank of Rayleigh scattering and stray light can be neglected because the intensity of Rayleigh scattering decreases by at least a factor of 10 as a result of the higher temperature and therefore lower density of heavy particles. During the calibration, when the plasma is not lit, the amount of stray light is small compared to the Rayleigh scattering signal. This can be concluded from the Thomson measurements. The perfect Gaussian fit through the Thomson measurements shows that stray light has a negligible influence, so that it appears to be small compared to the Thomson profile as well as the Raman scattering measurement. To increase the accuracy of calibration, six Raman peaks are treated and the recovered Rayleigh signals are after this conversion is averaged. In this way it is possible to recover the eliminated Rayleigh signal from the Raman measurements with an accuracy of better than 9% (see Sec. II C) and to calibrate the setup without inaccuracies caused by stray light.

![FIG. 9. The measured $n_r$ as a function of height at a radial position of 4.5 mm.](image1)

![FIG. 10. The measured $T_r$ as a function of height at a radial position of 4.5 mm.](image2)

![FIG. 11. The measured $n_r$ as a function of power at a radial position of 4.5 and 7 mm ALC.](image3)

![FIG. 12. The measured $T_r$ as a function of power at a radial position of 4.5 and 7 mm ALC.](image4)
IV. RESULTS

In Figs. 5 and 6 sets of measurements for different radial positions at 7 mm above the coil (ALC) are presented. The plasma power settings are 1.2 kW. The asymmetry of the radial dependent electron density and electron temperature profiles are due to the asymmetric coil. Further, the lower density and lower temperature in the center of the plasma are well-known features of the ICP. Here, the advantage of Thomson scattering is directly shown by the spatially resolved information. There is no need for mathematical routines to reconstruct local information out of line-of-sight measurements (Abel-Inversion). Comparing these results with measurements performed by Huang et al.\textsuperscript{6} we see a much larger difference in density and temperature between the center and the hottest area of the plasma. In general this difference is due to the skin effect. So we conclude that in our plasma a larger skin effect is present. This might be explained by our high frequency generator of 100 MHz instead of the normally used frequency of 27 MHz.

In Figs. 7 and 8 the electron temperatures and electron densities are shown at a height of 13 mm ALC. Since this region is farther away from the ionizing part of the plasma, we see lower values. Moreover, the skin effect is decreased as well. The radial profiles are much smoother. The measurements of Figs. 9 and 10 show also the change in electron density and electron temperature by increasing the height above the load coil at a radial position of 4.5 mm. Finally, the dependence of the power input on the density and temperature is presented in Figs. 11 and 12.

The highly dispersive grating in combination with the gated image intensifier and the 1024 channel photodiode array is responsible for a very accurately measured Thomson profile (within 1%), see also Fig. 3. But still the accuracy of these measurements is about 15% in the electron density and about 150 K in the electron temperature. The inaccuracies in electron temperature do not include reproducibility, which is an important source of inaccuracies in the electron temperature since the power settings can only be reproduced by about 0.1 kW. The inaccuracy imposed by this problem is about 500 K. For the influence of the power on the electron temperature see Fig. 12.

The main inaccuracy in the electron density is caused by the calibration of each Thomson measurement. Beside minor inaccuracies in the conversion of the Raman to the Rayleigh intensity, the most important sources are the instabilities in the setup and the measured Raman spectrum itself. The Nd:YAG laser has a power stability within 3%. Other origins of variations in the intensity of the detected signal can be found in a slight drifting of the optical components, for example, by heating. These variations are directly responsible for the inaccuracies since the electron density depends on the surface under the Thomson profile. To avoid these instability effects, a rather long period of about 1 h waiting time is necessary to get the whole setup stable. Further, the more often a calibration on nitrogen is carried out, the more accurate the electron density will be. In practice calibrating each hour will do. Inaccuracies in the Raman measurements will in the future be lowered by a better conditioned nitrogen gas sample above the torch instead of flowing nitrogen through the torch.

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