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Thomson scattering on non-thermal atmospheric pressure plasma jets

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Abstract
To characterize non-thermal atmospheric pressure plasmas experimentally, a large variety of methods and techniques is available, each having its own specific possibilities and limitations. A rewarding method to investigate these plasma sources is laser Thomson scattering. However, that is challenging. Non-thermal atmospheric pressure plasmas (gas temperatures close to room temperature and electron temperatures of a few eV) have usually small dimensions (below 1 mm) and a low degree of ionization (below $10^{-4}$). Here an overview is presented of how Thomson scattering can be applied to such plasmas and used to measure directly spatially and temporally resolved the electron density and energy distribution. A general description of the scattering of photons and the guidelines for an experimental setup of this active diagnostic are provided. Special attention is given to the design concepts required to achieve the maximum signal photon flux with a minimum of unwanted signals. Recent results from the literature are also presented and discussed.

Keywords: Thomson scattering, laser diagnostic, atmospheric pressure plasma, low temperature plasma

(Some figures may appear in colour only in the online journal)

1. Introduction
The scattering of photons on material particles is a vast topic. On unbound electrons it is called Thomson scattering, a mechanism that can be seen as the non-relativistic limit of Compton scattering. The elastic scattering on bound electrons is known as Rayleigh scattering and the inelastic scattering of photons on molecules as Raman scattering. Mie scattering occurs on larger particles of the dimensions of the photon’s wavelength. To study ionized gases, Thomson scattering (TS) is by definition ideal for experimental plasma characterization, as it delivers direct information about the density and energy distribution of free electrons. Due to the low scattering probability, TS was not observed experimentally until the late 1950s, when an anomalous reflection of radio waves in the earth’s ionosphere was explained by this mechanism (Forsyth et al 1953). The major impact for laboratory applications came with the development of the first high power ruby lasers that allowed the use of TS on Tokomak plasmas (Peacock et al 1969). A ruby laser can deliver single pulses with energies of tens of joules. Such a laser, operated in burst mode, is well suited for the study of fusion-related plasmas, even in real time (van der Meiden et al 2006). However, for smaller and more reproducible laboratory plasmas, accumulation of many scattering events by a laser with high average output is preferable. The
advent of high peak power Nd:YAG type lasers with faster pulse repetition rates led to the breakthrough in the use of TS on non-thermal laboratory plasmas, where non-thermal refers to a much lower gas than electron temperature (Huang et al. 1992, de Regt et al. 1995, Kempkens and Uhlenbusch 2000, Muraoka et al. 1998).

During the laser-plasma interaction, the electrons oscillate in the electric field of the laser beam. Due to these oscillations, the accelerated electrons will emit dipole electromagnetic radiation, which will be around the wavelength of the incident laser. If the electron density is sufficiently low so that an electron’s oscillation is unaffected by neighbouring electrons this is described as incoherent Thomson scattering (Salpeter 1960). This will be the case for the non-thermal atmospheric plasmas discussed here since, for the typical electron temperature of 1 eV, coherent Thomson scattering only becomes significant for values of electron densities of about \( n_e > 3 \times 10^{21} \). The signal processing from incoherent scattering is substantially simpler as the observed scattered spectrum is then a sum of individual scattering events (Evans and Katzenstein 1969).

High spatial resolution is achieved since only photons generated in the small interaction volume of the optical axis and the laser beam are detected. This volume can have diameters of less than 100 \( \mu m \) (Kono and Iwamoto 2004). The temporal resolution is determined by the laser pulse length or the gating of the detection system, whatever is shorter; TS with temporal resolution of down to 0.3 ns is reported in literature (Kieft et al. 2004). These capabilities of TS are unmatched by all other diagnostic techniques for free electron parameters in atmospheric pressure plasmas.

However, significant issues for the use of TS originate from the very small value of the Thomson scattering cross section and the dipole-like scattered wavefront. While this will be explained more in detail later, the problem is illustrated in figure 1 which shows an estimate of the fraction of photons per second generated by a typical Nd:YAG laser and a TS collection system (see (Carbone et al. 2015)). It is clear that for low electron densities, even with very high grade optics and a high quantum efficiency camera, only a very small signal can be expected.

With reproducible plasmas, the integration time can be increased substantially. The detection limit is considerably lowered and it is then given by the signal to noise ratio of the Thomson scattering signal with respect to the noise of the stray light, Rayleigh and Raman scattering. So, for example, TS in a few Pascal gas pressure plasma-reactors of about 0.5 m diameter has been used successfully to probe low temperature argon plasmas with electron densities as low as a few \( 10^{16} \) m\(^{-3} \) and electron energies up to about 14 eV (Hori et al. 1998, Crintea et al. 2009).

2. Thomson scattering theory and realization

2.a. Laser plasma interaction

The spectral power \( P_s(\Delta \Omega) \) of the scattered radiation collected in the solid angle element \( \Delta \Omega \) is given by

\[
P_s(\Delta \Omega) = P_{pl}L \frac{d\sigma}{d\Omega} \Delta \phi_\lambda(\lambda) \tag{1}
\]

where \( P_s \) is the incident laser power, \( L \) the length over which the observed laser-medium interaction takes place, \( n \) the density of the scattering particles, \( d\sigma/d\Omega \) the differential cross section, while \( \phi_\lambda(\lambda) \) contains the spectral information, assuming that \( \int \phi_\lambda(\lambda) d\lambda = 1 \). In the case of incoherent TS, the spectral information arises from the Doppler shift induced by the free electron motion. The wavelength shift is related to the 2D velocity plane given by the laser beam and the collection optics. The projection of this velocity distribution onto the 1D line-of-sight gives \( \phi_\lambda(\lambda) \). It can be derived from basic principles (Evans and Katzenstein 1969) that for a Maxwellian distribution of the electrons the observed 1D velocity distribution is equivalent to the actual 3D distribution.

Expression (1) can be used to obtain the electron density and temperature by inserting the corresponding differential cross sections for TS

\[
\frac{d\sigma_{TS}}{d\Omega} = r_e^2(1 - \sin^2 \theta \cos^2 \psi) \tag{2}
\]

Here, the scattering angle \( \theta \) is the angle between the incident and the scattered wave vector, \( \psi \) the angle between the plane of scattering and the polarization of the incident beam and \( r_e = e^2/(4\pi\varepsilon_0m_e) \) the classical electron radius. It is obvious that the setup should be chosen such that the polarization is perpendicular to the detection at \( \psi = 90^\circ \). Then the scattering angle can be chosen freely. However, with a \( \theta = 90^\circ \) arrangement, the stray light is minimized and the spatial resolution is at its optimum, as the detection beam is not skewed. The spatial resolution may be further improved by magnifying the projection of the detection volume so that it fully occupies the entrance slit of the spectrometer. Then, (2) equals \( d\sigma_{TS}/d\Omega \) = \( r_e^2 = 7.94 \times 10^{-30} \) m\(^2\)sr\(^{-1}\).

Equation (1) for the scattered laser light is also valid for Raman (RmS) and Rayleigh (RyS) scattering provided the appropriate cross sections are inserted. The strength of the different scattering mechanisms is thus primarily determined by the differential cross section. For illustration different values of \( d\sigma/d\Omega \) are given in table 1.

The elastic scattering on atoms and molecules (RyS) depends strongly on the gas species, more precisely on their polarizability and on the wavelength (Sneep and Ubachs 2005). For atmospheric plasma jets, the ionization degree is low with about \( n_e/N = 10^{-4} \) to \( 10^{-7} \), where \( N \) is the neutral gas density. That means that the Rayleigh signal exceeds the TS signal by 3–5 orders of magnitude (see table 1). The RyS can be removed taking advantage of the fact that its spectral width is much smaller than that of TS. This is due to fact that both RyS and (incoherent) TS are based on the Doppler effect. The atoms being much heavier than electrons have a much smaller thermal velocity range. The removal of RyS signal can be done by for example using multiple spectrometers as will be explained more in detail later. Other techniques employ notch filters such as volume Bragg gratings (Glebov et al. 2012, Klarenhaar et al. 2015), interference filters (Wesseling and
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Kronast 1996) or gas vapours (Bakker and Kroesen 2001, Lee and Lempert 2002). Another way to reduce the influence of Rayleigh scattering is to use an infrared laser, since RyS has a $\lambda^{-4}$ dependence. This approach is however obstructed by the price of low sensitivity IR detectors (Bowden et al 1999) and the fact that working at invisible wavelengths requires care.

For the interference of TS with Raman scattering (RmS) the situation is more complicated. Again the RmS signal can be typically 1–3 orders of magnitude stronger than that of TS but the spectral shape of the RmS signal is much larger than that of RyS. RmS can have a similar spectral width as TS (see figure 2). Thus the RmS and TS signals can easily overlap in air containing plasmas and a careful identification of both signals is thus necessary (van Gessel et al 2012). The interference between TS and RmS strongly depends on the plasma conditions. High power densities will lead to high values of the electron density which in turn lead to high gas temperatures. This results in an enhanced dissociation of molecules which, combined with the high $T_e$ and the Boltzmann distribution of the rotational states, leads to a very low RmS signal. The overlap of RmS with TS will thus be limited in plasmas of high power (density).

Laser induced fluorescence (LIF) can occur and might overlap largely within the spectral range of the TS signal. Either LIF is produced by surfaces in the vicinity of the laser or in optical components (Wesseling and Kronast 1996). That can be subtracted but its noise will reduce the detection limit. Another issue is LIF on plasma species. In He plasmas with a 532 nm laser LIF was observed (Wesseling and Kronast 1996, Hübner et al 2014), however the originating species is unknown; it could be N$_2$(B$^2\Sigma \rightarrow X^2\Sigma, \nu >10$) or a He excimer. In very strong LIF environments the choice of the laser wavelength might have to be reconsidered. In most cases the LIF decay is much slower than the immediate scattering events such as TS so that the signals can be temporally resolved recorded and disentangled (Soltwisch and Kaczor 1999).

2.b. Experimental realization
2.b.1. Laser delivery. In this section, the key issues of the implication of Thomson scattering measurements performed on non-thermal atmospheric pressure plasma jets are addressed through a description of a generic experimental setup. A basic assumption in our approach is that the plasma is reproducible so that the signal can be accumulated over many thousand laser shots. This requires that the plasma has no or little drift over a period of hours and implies that stochastic plasmas, such as moving filaments in a Dielectric Barrier Discharge are excluded. The ultimate goal of single shot TS on atmospheric pressure plasmas has so far only been realized in more dense systems such as an arc plasma at atmospheric pressure (Uchino et al 1982) or a microwave torch with $n_e$-values of about $10^{21}$ m$^{-3}$ (van der Mullen et al 2007).

It is also necessary to be ensured that the laser itself does not perturb the plasma; for example too high laser power can heat the plasma by inverse Bremsstrahlung. It was found by
Carbone et al (2012) that taking into account electron-neutral and electron–ion inverse Bremsstrahlung leads to low theoretical values of the critical laser fluency (laser energy per area).

Additionally, in a cold non-thermal plasma with an electron temperature of about 1.6 eV, with a laser beam spot of about 150 μm diameter and at a laser pulse energy of 100 mJ, they found by experiment that the electron temperature is increased by 10%. However, due to the high thermal conductivity, electrons can transfer heat to adjacent regions. Consequently in such a case of non-isolated electrons the increase of $T_e$ is probably reduced significantly not only by heat conduction but also by ionization processes during the finite laser pulse length (de Vries et al 2005). Apart from laser-plasma heating one should realize that a laser can change the degree of ionization by photo detachment of negative ions or multi-photon dissociation, excitation or ionisation (Noguchi et al 2002, Muraoka and Kono 2011). All this can be readily checked by ensuring that there is a constant width and linear intensity response of the TS signal to the laser power.

Smaller pulse energies offered with a high repetition rate, can achieve high photon fluxes without such laser-plasma heating. One could think e.g. of a 100 kHz pulse rate Nd:YAG type laser systems producing pulses of 1 mJ.

However, working with high repetition rate laser systems implies that more plasma emission is collected. At the same time we should realize that plasma emission is normally not a big issue at non-thermal atmospheric pressure plasmas. That is in contrast to molecular low pressure plasmas or high power (close to thermal) plasmas, where in the first case molecular bands or in the second case continuum radiation readily dominates the TS signal (Muraoka and Kono 2011). Molecular radiation might be avoided by a tunable laser system.

The focusing of the laser beam into the plasma must ensure the best possible beam quality with a low beam divergence (low $M^2$ value). For that the use of apertures to block small angle scattered laser light is recommended. On the other hand the laser focusing is crucial to resolve the small atmospheric pressure plasmas. The focusing should be realized such that the whole radial extent of the laser beam is within the detection volume of the collection optics. This makes the signal independent of the beam diameter and small plasma displacements. The focusing has to be balanced against possible laser-plasma heating. The exiting laser beam should be effectively apertured and dumped to minimize scattered or reflected laser light. This is a major issue in micro-discharges (Kono and Iwamoto 2004) where there is inevitably a surface, for example an electrode, in the vicinity of the laser focus. The unwanted stray laser light can totally swamp the TS signal, as the latter has such a small scattering cross section. The stray light has more or less the same polarization as the laser, so it cannot be filtered effectively with polarizers. Here the choice of the scattering angle $\theta$ in relationship to surrounding solid surfaces can help. Where plasma electrodes form a very thin and narrow channel, a back-scattering confocal arrangement can be used with $\theta = 180^\circ$ as was reported by Belostotskiy et al (2008). In that way, only one dimensional access is needed. This is at the price of higher stray light intensities. If the plasma is in a closed chamber for a controlled atmosphere, Brewster angled windows combined with apertures placed as far as possible from the detection volume help to reduce scattered light.
As previously mentioned (see figure 1), the collected solid angle is a critical issue for the signal strength. High grade imaging optics with small F-numbers is preferable. However, the limitation related to on-axis achromatic lenses is usually given by spherical aberration while off-axis mirrors are rather limited by astigmatism. Moreover, it should be kept in mind that the opening angle of the first optical element determines it for all further elements, since the etendue (light emitting surface times solid angle) is conserved. For a triple grating spectrometer this means that at least eight lenses and quite a large space are required.

2.b.2. Detection system. Here, the description of the detection system will focus on a triple grating spectrograph (TGS) (see figure 3). However, depending on the demands on filtering of the laser photons, other systems could be more suitable.

The collected photons are focused onto the entrance slit of the spectrograph. The slit is parallel to the axis of the laser beam, which allows spatially resolved images in the dimension along the laser beam. Inside the TGS, the beam has to be rotated, creating a vertical image of the slit. That is because it is highly preferable to remain at a constant beam height above the optical table while the vertical slit image is dispersed horizontally by the gratings. As previously mentioned, the polarization of the laser beam has to be perpendicular to the scattering plane. However, as the gratings in the spectrometer are usually polarization dependent, the beam polarization should be rotated after the scattering by a λ/2 plate.

A single spectrometer typically might have a stray light rejection of $10^{-3}$ (ratio of incident photons compared to the ones received at e.g. $\delta \lambda = 0.5$ nm). However, in the previous section it was shown that a RyS suppression of a factor of at least $10^{-5}$ is required. The use of a double spectrometer can provide that, as illustrated in figure 4. The first two identical gratings (G1 and G2) and the blocking mask form a notch filter for the central laser wavelength. Suppression of more than 6 orders of magnitude for signals that are $\delta \lambda = 2.5 \text{ nm}$ apart is readily achievable (van de Sande Thesis 2002). After the second grating, imperfections of the beam are removed by an intermediate slit that is similar to the entrance slit. The third grating resolves the actual spectral information. The gratings should have the appropriate spectral wavelength range and focal length. For the typical case of $T_e = 1 \text{ eV}$ that corresponds to a TS signal FWHM of $\lambda_{\text{FWHM}} = 2.5 \text{ nm}$, a spectral range of about 10 nm is sufficient. That demands, with a typical 10–20 mm CCD detector, a dispersion of about $d = 2 \text{ mm nm}^{-1}$. With that the width of the blocking mask $w_{\text{mask}}$ can be calculated. The latter should be chosen to achieve the desired detection limit of the minimum electron energy. To record signals at $\delta \lambda = 0.5$ nm (0.1 eV) with a dispersion $d$ and an entrance slit width of $w_{\text{slit}} = 250 \mu \text{m}$, the mask must not exceed $w_{\text{mask}} = 2 \times \delta \lambda d + w_{\text{slit}} = 2.25 \text{ mm}$. However, the width of the mask might be chosen broader according to the stray light conditions as that will reduce unwanted light at the cost of low electron energy detection.

It is interesting to note that using an entrance slit of e.g. $w_{\text{slit}} = 250 \mu \text{m}$, to cover the whole laser beam, combined...
with the requirement for the spectral bandwidth to be below \( \lambda_{bw} = 0.3 \) nm, to be able to filter the laser wavelength and detect low energy electrons, requires a focal length of

\[
f = \frac{w_{dm} \cos(\alpha)}{m g \lambda_{bw}} = 45 \text{ cm}
\]

Here \( m \) is the order of diffraction, \( g \) the grating constant and \( \alpha \) the angle of incidence on the grating. These values are taken from van de Sande (Thesis 2002).

Obviously the whole spectrometer must be placed in a completely black box and to avoid cross signals by e.g. higher/lower order diffraction, screens can be placed appropriately to absorb the light.

In order to identify unambiguously the RmS or plasma emission it is highly desirable to obtain a continuous spectrum by each laser shot. Therefore, the signal should be recorded by a multichannel detector (Kono and Nakatani 2000). The detector could be an array of photomultiplier tubes but today is now more likely to be a charge coupled detector with an intensifier (ICCD). The camera should be chosen for highest quantum efficiency, high gain and low dark noise and drift, while the frame rate is of less importance. The 2D camera image then resolves the TS spectrum both spatially, in the direction along the plasma-laser intersection, and in wavelength.

In practice, the focusing of the scattered laser light in the TGS is a crucial element in aligning the detection system. This can be realized by placing a low power diode laser with the same wavelength as the TS laser at the exact position of the camera (Carbone et al 2015). Then, the gratings and slits can be aligned without lenses. Next, the lenses are inserted one by one and focused correctly. Eventually, the laser beam should form a spot at the detection volume in the plasma.

Instead of a TGS, a single spectrometer could be used (Nedanovska et al 2011, Adress Thesis 2014). The setup of a single spectrometer with no dedicated laser wavelength filter is tempting as it is more straightforward. The laser scattering arrangement is very similar to that described in the sections above; the difference is that the measured laser scattering spectrum also contains the contributions from Rayleigh scattering. Consequently the drawback is the difficulty or sometimes inability to identify the possible small TS contribution in the environment of the huge Rayleigh and Raman scattering components.

As mentioned above, options other than spectrometers are possible to filter the laser wavelength (see figure 4). Interference filters were employed by Wesseling and Kronast (1996). A possible reduction of stray light by a factor of \( 10^{-6} \) is reported. However, a potential difficulty is the laser induced fluorescence in the filters. The use of gas vapor absorption

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**Figure 4.** Simplified schemes for different laser light reduction techniques: (A) a triple grating arrangement with blocking mask; (B) a glass and interference filter set and (C) a vapor cell with a gas with a strongly absorbing transition which has a wavelength matched by a tunable laser. L denotes lenses and G gratings. The colors exaggerate the real wavelength spread.
lines was also successfully used (Bakker and Kroesen 2001, Lee et al 2002). However, in order to shift the laser wavelength onto the absorbing transition, a high power dye or Ti:sapphire laser is needed from which the amplified spontaneous emission needs to be blocked. The option of using volume Bragg filters has so far only been realized for Raman spectrometers (Klarenbaar et al 2015), but should be explored for Thomson scattering as well.

A general recommendation for the stray and Rayleigh scattered light suppression is difficult to give. In terms of low redistribution of light the TGS, or even more spectrometers, is unmatched. However, if compactness and easy handling is concerned the use of interference or Bragg filters might be more efficient. It is interesting to note that Raman scattering setups exhibit very similar issues concerning light suppression and top-notch systems employ also TGS or multiple filters.

2.c. Calibration of Thomson scattering

The rotational $J$–$J'$ transitions of molecules lead to peaks in the Raman scattering spectrum, as can be seen in figure 2. This makes such a molecular spectrum ideal for the absolute calibration of both the intensity and the wavelength by using a multi-component fit (de Regt et al 1995).

The RmS calibration can be performed with any molecule that has a rotational constant that matches the detection wavelength range. The allowed transitions of $J' = J ± 2$ lead to the red-shifted Stokes (+) and blue-shifted anti-Stokes (−) band with their characteristic intensity variation due to the Placzek–Teller coefficients. The total scattered power is

$$P_{\text{RmS}}(\Delta \Omega) = P_L \Delta \Omega \sum_{J' = J ± 2} n_0 \frac{d \phi_{\text{RmS}}(J,J',\lambda)}{d \Omega} \phi_{\text{RmS}}(J,J',\lambda)$$

(3)

where $\phi_{\text{RmS}}(J,J',\lambda)$ is also determined by the instrumental profile. The detection system does not need to resolve the individual rotational lines; an envelope fit is sufficient. However, if the signal intensity is expected to be high, better spectral resolution should be targeted. Nevertheless, because of the very little counts that TS provides, spectral signal binning is advised as the signal in each spectral wavelength interval must exceed the noise level.

In practice, the calibration is performed by using the ambient air or a vacuum vessel, with known gas pressure and temperature and exactly the same detection system. Consequently, the scattering signal is measured at a known gas density $n_0$ which can be related, together with the Boltzmann relation to obtain $n_T$. Using the respective cross sections, the wavelength integrated measured TS and RmS signals give the absolute electron density

$$n_e = n_0 \frac{P_{\text{TS}} d \sigma_{\text{RmS}} / d \Omega}{P_{\text{RmS}} d \sigma_{\text{TS}} / d \Omega}$$

(4)

With this calibration method, there is no need to determine the incident laser power, the plasma-laser interaction length or the solid angle $\Delta \Omega$ of the detection system. However, the absolute accuracy depends on the value of the respective Raman cross section. For example for $N_2$ an uncertainty of about 6% is expected based on the values for the polarizability given in the literature (Olney et al 1997). Using the Rayleigh signal (basically the $\Delta J = 0$ component) as a calibration is also possible, but then a careful measurement of the stray light contribution is needed. If so, this can be measured under vacuum conditions or in different gases with largely different RyS cross sections, with an extrapolation to a ‘zero’ cross section (Evans and Katzenstein 1969, Huang and Hieftje 1985).

In case of the incoherent scattering, the spectral line width of Thomson scattered photons $\phi_{\text{TS}}(\lambda)$ is directly related to the Doppler shift induced by the electron velocity distribution function. A Maxwellian electron energy distribution function (EEDF) leads to a Gaussian distribution of the scattered photons. Inserting the corresponding function of $\phi_{\text{TS}}(\lambda)$ into (1) gives (van de Sande Thesis 2002)

$$P_{\text{TS}} d \lambda = P_L d \sigma_{\text{TS}} \frac{1}{\Delta \Omega} \frac{\exp \left(-\frac{\delta \lambda}{\Delta \lambda_{1/e}} \right)}{\delta \lambda} d \lambda$$

(5)

with $\Delta \lambda_{1/e}$ being the width of the spectral function at a fraction of 1/e of the maximum and $\delta \lambda = \lambda_i - \lambda$. The electron temperature $T_e$ can be derived via the Doppler formula to give

$$T_e = \frac{m_e c^2}{8 k_B \sin^2 (\theta/2)} \left( \frac{\Delta \lambda_{1/e}}{\lambda_i} \right)^2$$

(6)

with $\lambda_i$ the input laser wavelength. In practice, it is helpful to plot the logarithm of $P_L$ as a function of $(\delta \lambda)^2$, which gives a graph of the linearized electron energy distribution function (EEDF/√E, known as the electron energy probability function, EEPF) (Huang et al 2000). With equation (6) the slope of the linear trend of this plot is equal to $-1/k_B T_e$ (see figure 5).

For the electron density, the spectral function $\phi_{\text{TS}}(\lambda)$ does not need to be known, only the total scattered signal as found from (4). That is because the number of scattered photons is directly proportional to $n_e$. As the signal can be very noisy, from equation (5) a fit of a Gaussian function of the data is
recommended with at least two fitting parameters, height and width. This yields the electron density, via the spectral area, and the temperature via equation (6).

As discussed in the work of Bowden et al (1999), the energy range over which the EEDF can be measured depends on the total electron density and the electron energy distribution in the context of the signal to noise ratio in the measured signals. In the case of their work for low pressure systems, the noise in the plasma emission or that of the TS signal itself determines the detection limit. However, as discussed above for plasmas in ambient air, also the noise of the Raman scattering plays a crucial role.

Effectively the method used in obtaining the data in figure 5 makes it possible to probe the mean energy and deviations from it, but does not provide information on the tail of the distribution function, simply because electrons with energies higher than a few $k_B T_e$ have much lower densities. This means that with TS in such plasmas it will be difficult to get insight into the effect of ionization processes on the EEDF and vice versa, at least not for plasmas with relatively low $n_e$ and $T_e$ values.

2.d. Comparison, advantages and disadvantages

There is a wide variety of experimental methods available to determine electron parameters in non-thermal atmospheric pressure plasmas. (Gigosos et al 2003, Ivkovic et al 2004, Chichina et al 2005, Balcon et al 2007, Ito et al 2010, Overzet et al 2010, Palomares et al 2010, Qian et al 2010, Hofmann et al 2011, Palomares et al 2012, Shashurin et al 2012, Sretenovic et al 2012, Bruggeman and Brandenburg 2013). However, the laser scattering techniques benefit from the straightforward interpretation of the raw data without much theoretical framework. A related example is RyS, where RyS gives direct values of the gas density $N$ from which $T_e$ can be obtained via the pressure. To validate this method, the rotational distribution of a molecular gas can be measured by optical emission or absorption spectroscopy or laser induced fluorescence. That only provides $T_e$ if specific assumptions are satisfied, such as equilibrium distribution for the rotational states. This problem is addressed more extensively by Bruggeman et al (2014).

A technique that delivers information about the free electrons and that is easier to implement than TS is Stark broadening (Gigosos et al 2003, Ivkovic et al 2004, Palomares et al 2012). This spectroscopic technique relates the line broadening of an optical transition to the electric micro-field surrounding the emitter. A priori knowledge of the line shape as function of the electron density, temperature and perturber mass is necessary in order to use this method (Gigosos et al 2003).

As argued in Hübner et al (2014), the conditions for Stark broadening measurements in cold atmospheric pressure plasmas are not easy to satisfy. Especially in the case of a low gas temperature the Van der Waals broadening usually dominates over Stark broadening. For ambient conditions the hydrogen $\beta$-line has a pressure broadening of $\Delta \lambda_{\text{WAV}} \approx 100$ pm, while the Stark line width is about $\Delta \lambda_S \approx 2$ pm for $n_e = 10^{18} \text{m}^{-3}$, and so it is even smaller than the corresponding fine structure. Moreover, this is a line-of-sight method and to measure local line broadenings out of the measured cumulative line shape, inverse Abel transformation has to be accomplished of many line shapes that are integrated over a non-uniform and unbounded plasma.

Because of the steep plasma gradients the demands for TS in terms of spatial resolution are as strict (resolution $< 100 \mu\text{m}$) as for Stark broadening. However, this is possible to tackle in TS as the laser focus defines the spatial resolution. Kono and Iwamoto (2004) achieved TS with a laser beam waist of about 25 $\mu\text{m}$ to resolve small gradients while keeping the laser-plasma interaction low with a laser pulse energy of 4 mJ.

In Palomares et al (2010) it was found that the electron density measured in a microwave driven atmospheric plasma jet by passive methods such as $H_3$ Stark broadening and continuum emission agreed to within the experimental uncertainties to those determined under the same conditions with TS. They also found that the use of absolute emission line intensities with a to determine $T_e$ deviated from those determined by TS. This is of interest since line emission probes the tail of the electron energy distribution function, above the excitation potential of the emitting atoms, whereas TS probes the mean electron energy region (van de Sande et al 2002).

The electron density can also be obtained by current or impedance measurements or by using a simplified global model (Balcon et al 2007, Overzet et al 2010). However, these approaches usually rely on many uncertain values, such as the electron velocity, the plasma jet diameter, and the spatial distribution of the electron density and so provides only an estimate of the electron density.

Finally, electron densities can be also obtained from the change of the refractive index induced by the plasma. This can be measured by sending an electromagnetic wave (or laser) into the plasma. Scattering, absorption or phase shifting of microwaves is a well-known technique (Hutchinson 1990). However, as the dimensions of many atmospheric pressure plasmas are smaller than the wavelength of the electromagnetic waves typically used, either the scattering volume must be known precisely (Shneider and Miles 2005) or a shorter wavelength must be chosen. The former approach was successfully implemented by Sashurin et al (2015), while the latter was put into practice by Ito et al (2010) by using mm-wave heterodyne interferometry. Volume-integrated (in the first case) and line-integrated (in the second case) electron densities were obtained with good temporal resolution (down to ns). However, one should keep in mind that a variation in gas temperature or mixture might also change the refractive index and mask the effect of the electrons (Leibold et al 2000). Furthermore, the use of electromagnetic waves can only provide line- or volume-integrated electron densities. Despite the obvious advantages of Thomson laser scattering, it should be kept in mind that the TS systems are not simple to be implemented and therefore unlikely to be applicable to systems in industrial environments (e.g. lack of access
to the plasma or the considerable investment in equipment).
However in this context it can find value calibrating other less complicated techniques or plasma models.

3. Results of Thomson scattering measurements

For reviews on TS measurements of the electron properties in dense hot atmospheric pressure plasmas such as arcs (Uchino et al. 1982, Snyder et al. 1994), plasmas created by laser induced breakdown (Nedanovska et al. 2011) or inductively coupled plasmas (de Regt et al. 1995) the reader is directed to the reviews of Warner and Hieftje (2002) about spectro-chemical plasmas, or Muraoka and Kono (2011) on low temperature plasmas in general. This brief review will focus on published work in non-thermal atmospheric pressure plasmas. The material has been selected to illustrate facets of the use of TS in obtaining electron densities, temperatures and electron energy distribution functions. A summary is given in table 2.

Non-thermal plasma jets are generally created in an inert gas, often with up to a few percent added molecular species to generate reactive species for specific applications. The gas is generally flowed through a dielectric tube, often quartz or alumina with diameters from below 1 mm up to several mm. An electric field is applied within the vicinity of the tube using different driving circuits or electrode configurations. The frequency, pulse shape and magnitude of the applied voltages tend to define the features of the plasma. Irrespective of the driving pulse shapes and frequencies, gases or flow rates, the defining property of all the sources discussed here is, that there is a luminous plume, extending into the ambient air or controlled atmosphere from the exit of the primary plasma source. Most, but not all, reported TS measurements are made in this plume region. To date TS measurements have been reported in plasma jets driven by microwave, radio or kHz frequencies, sometimes in combination.

3.a. Microwave driven plasma jets

An example of a microwave driven plasma jet is the Surfatron as shown in figure 2, which is similar to one used in the measurements of van Gessel et al. (2012). A 2.45 GHz MW cavity launches radiation along the jet axis, creating and sustaining the plasma by generating a cylindrically symmetric surface wave (Moisan and Zakrzewski 1991) in a 0.8 mm diameter, air cooled quartz tube through which argon is flowing at 1 slm into the ambient air. The generator delivered a power of 50 W but the absorbed power in the plasma is unknown. Spatially resolved TS measurements were obtained for distances up to 10 mm from the end of the tube and with up to 10% air added to the argon gas flow.

Figure 6(A) shows some of the TS-derived results. It clearly shows the presence of a plasma with electron densities up to a

<table>
<thead>
<tr>
<th>Plasma</th>
<th>Gas</th>
<th>( n_e ) ( \times 10^{20} \text{m}^{-3} )</th>
<th>( T_e ) [eV]</th>
<th>Method</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 kHz DBD-jet</td>
<td>He</td>
<td>0.005–0.15</td>
<td>0.1–1.6</td>
<td>532 nm + TG</td>
<td>Hübner et al. 2014</td>
</tr>
<tr>
<td>5 kHz Needle-jet</td>
<td>Ar</td>
<td>0.01–0.7</td>
<td>0.1–3</td>
<td>532 nm + TG</td>
<td>Hübner et al. 2013</td>
</tr>
<tr>
<td>RF 14.5 MHz, pulsed jet</td>
<td>Ar, Ar/air</td>
<td>0.05–0.2</td>
<td>0.2–2</td>
<td>532 nm + TG</td>
<td>Van Gessel et al. 2013</td>
</tr>
<tr>
<td>DC microgap</td>
<td>Ar</td>
<td>0.3–0.9</td>
<td>0.7–1</td>
<td>526.5 nm + TG Backscattering</td>
<td>Belostotskiy et al. 2008</td>
</tr>
<tr>
<td>MW 2.45 GHz jet</td>
<td>Ar</td>
<td>0.2–8</td>
<td>1.2–5</td>
<td>532 nm + TG</td>
<td>Van Gessel et al. 2012</td>
</tr>
<tr>
<td>MW 2.45 GHz microgap</td>
<td>He/N2, Air</td>
<td>3–18</td>
<td>0.6–1.5</td>
<td>532 nm + TG</td>
<td>Kono et al. 2004</td>
</tr>
<tr>
<td>RF 27 MHz ICP</td>
<td>Ar, Ar/H2O</td>
<td>1–20</td>
<td>0.3–0.7</td>
<td>532 nm + SG</td>
<td>Huang et al. 1992</td>
</tr>
<tr>
<td>MW 2.45 GHz ICP</td>
<td>Ar</td>
<td>1–22</td>
<td>0.8–1.9</td>
<td>532 nm + TG</td>
<td>van der Mullen et al. 2007</td>
</tr>
<tr>
<td>LIBS</td>
<td>He</td>
<td>100–800</td>
<td>1.4–1.6</td>
<td>532 nm + DG</td>
<td>Nedanovska et al. 2011</td>
</tr>
</tbody>
</table>

Figure 6. MW jet similar to that shown in figure 2. The jet was operated with a 1.0 slm flow of argon into ambient air. (A) The TS derived radial profile of the electron density and temperature measured at 5 mm downstream from the nozzle exit. (B) Measurement of the partial air pressure derived from the simultaneously measured Raman laser scattering. Reprinted with permission from van Gessel et al. (2012) © 2012, IOP Publishing.
few $10^{20}$ m$^{-3}$ and electron temperatures of a few eV. Figure 6 also illustrates the additional valuable information that can be obtained from TS measurements, since the Raman scattered signal from $N_2$ and $O_2$ is obtained simultaneously. This allows the determination of the spatial distribution of the air fraction as shown in figure 6(B) and the gas temperature (not shown). It clearly demonstrates that the plasma is confined to the argon gas stream. The Raman scattering also shows that the gas is heated up. Rayleigh scattering shows even that the plasma core gas temperatures reach up to 600 K. The lowest measurable electron density was of about $5 \times 10^{18}$ m$^{-3}$, determined mainly by the plasma emission and unwanted scattered light (RmS).

3.b. Radio frequency driven plasma jets

Atmospheric pressure, radio frequency driven plasmas are either operated with an EM-field perpendicular to the gas flow, such as in a plate-plate or coaxial configuration, or an EM-field that is more or less parallel to the flow. In the latter case, the powered electrode is a cylindrical electrode or an internal, rod-type electrode (needle) in a dielectric tube. These jets can operate without a secondary grounded electrode, a cylindrical metal electrode on the outer surface of the dielectric cylinder or an electrode in the jet’s effluent. Some of the first work on applying TS to a cold atmospheric plasma was reported by van de Sande et al. (2002) in such a coupled system where the powered of two cylindrical electrodes on the outside of a 4 mm diameter quartz tube was driven with a rectangular shaped voltage at a frequency of 112 kHz at an average power of about 35 W. The second electrode was grounded. Helium was flowed through the tube at 0.35 slm. In this case the laser was passed axially through the discharge and the scattered light was observed through the quartz tube. So here the measurement was in the plasma source rather than in the jet. The measured electron density was found to be modulated with the driving frequency between 0.75 and $2.5 \times 10^{19}$ m$^{-3}$. The electron temperature was modulated with values between 0.5 and 3.8 eV. However, the employed laser power of 400 mJ might have been sufficient to cause laser-plasma heating (see Carbone et al. 2012). Consequently, $T_e$ is probably systematically overestimated. A feature of this work was that the electron temperatures were also determined using absolute spectral line intensity measurements giving values that deviated from the TS values. As discussed above, TS can also be used to determine the electron energy distribution. Here the TS spectra, as shown in figure 7 was found to deviate at some point in the RF cycle from a single Gaussian distribution, indicating that the electron energy distribution was not Maxwellian as assumed in the spectral line intensity analysis.

A time modulated RF frequency driven plasma jet was studied by van Gessel et al. (2013). A pin electrode inside a 1.5 mm inner diameter glass tube was powered at 14.5 MHz which in turn was modulated with a 20 kHz square pulse with a 20% duty cycle. Two configurations of grounded electrode were used, one with a ring electrode around the tube and the other with a plate positioned 3 mm from the tube exit. Argon gas, with a few percent of admixed air, $N_2$ or $O_2$ was flowed through the tube at a rate of 1.0 slm. Figure 8 shows the TS results obtained at the end of the 20 kHz pulse. At least an order of magnitude higher electron density was found in the high electromagnetic field region between the electrodes, compared to what was found in the effluent region after the grounding plate.

3.c. High-voltage pulsed plasma jets

Plasma jets can also be generated in the geometries described above by applying high voltage sinusoids or pulses at 10’s kHz frequencies to one of the electrodes. Time resolved imaging shows that the resultant ionization can occur as a guided streamer. Traveling ionization waves form a plasma channel with the plasma extinguished before the next voltage pulse or half cycle of alternating voltage (Lu et al. 2012).

An example of TS measurements from a source operating with a 7 kV, 250 ns wide positive pulse at 20 kHz is shown in figure 9 based on Hübner et al. (2014). In this case, the powered electrode is a hollow capillary inside a quartz tube enclosing the 4.5 slm He flow. The quartz has an inner diameter of 2.1 mm and outer diameter of 7.1 mm; around the tube at the height of the powered electrode is a grounded electrode placed. The plasma jet emerges out of the quartz tube at about 7 mm downstream from the powered electrode. A hollow plasma channel is formed that collapses downstream. Interestingly, the maximum electron density was found further downstream and not at the nozzle exit. Note that the densities in figure 9 were measured at different times, corresponding to the temporal maximum at each axial position. Mean electron energies of up to 2 eV were found (not shown) which decayed rapidly after the ionization wave has passed.

Adress (Adress Thesis 2014) studied the plasma plume of another dielectric barrier discharge-based, atmospheric pressure plasma jet, operating into ambient air with helium feed gas at 3 slm. An assembly of two sequential, cylindrical electrodes is used; the upstream is grounded while the downstream is powered. The tube has an inner diameter of 4 mm and the
Figure 8. Images and spatially resolved electron density measurements from a modulated RF jet. Argon $+$ 1% air gas was flowed at 1.0 slm into ambient air. Top figure with grounded, concentric electrode close to the tube exit. Bottom figure with a grounded plate 3 mm from the tube exit, indicated by a dark bar on the electron density map. The end of the quartz tube is at axial position zero. Reprinted with permission from van Gessel et al (2013) © 2013, AIP Publishing LLC.

Figure 9. Pulsed DBD-like jet with a capillary inner high voltage electrode and a grounded outer ring, operated with a flow of He into ambient air, adapted from (Hübner et al 2014). For each z-position TS was recorded at the time of the maximum electron density (waveform shown in figure 10). The spatial distribution of the electron density shows its collapsing evolution. The $z = 0$ position corresponds to the end of the quartz tube.
nozzle is also 4 mm downstream from the grounded electrode. The electrical waveform consists of a 6 kV positive pulse at 20 kHz with half width at half maximum of 2 \( \mu \)s.

Preliminary results indicate that electron densities at the centre of the plasma plume are about \( 5 \times 10^{19} \) m\(^{-3} \) and the electron temperature 0.16 eV. These values increase radially to values of about \( 1.3 \times 10^{20} \) m\(^{-3} \) and 0.25 eV at 2.2 mm from the centre. These results differ compared to (Hübner et al 2014), however, there are significant differences between the two experiments, for example, the electrode geometry and the voltage pulse shape.

Since TS provides temporal resolution in the nanosecond range, the ionization wave of the guided streamer can be probed directly. That gives unique insight into the excitation kinetics. In figure 10(A), the temporal evolution of a high voltage pulsed jet with an argon flow into ambient argon is shown (Hübner et al 2013). For different positions, the maximum value of the electron density and temperature of the guided streamer arrives at different times. In argon a steep decay of the energy and density of the electrons is found. However, different to that is the temporal decay of the He DBD-like jet in figure 10(B) (Hübner et al 2014). As in Ar the ionization front of the guided streamer arrives at different times, however a slower temporal decay and lower electron densities are found. The electrode configuration and applied voltage is not comparable, however, from the trends and other unpublished data it is concluded that the fundamental difference of the electron kinetics is due to the difference in feeding gas. Argon exhibits a more rapid decay than He, and indeed dissociative recombination rate coefficients are slightly faster for Ar, but as the densities are also much higher, electron-ion recombination reaction rates are faster. Another issue is the spatial distribution of the plasma, which is also different in both gases.

Shown is also the visible plasma emission which follows the electron temperature very closely. That can be understood if, in first approximation, the excitation balance consists only of electron impact excitation balanced by radiative decay, where the timescale of the latter is in the order of tens of ns.

3.d. Other micro discharges

Plasma sources which truly satisfy the word ‘micro’ are very challenging to access by TS. The small dimensions demand a well-defined small laser beam focus and high stray light suppression, as very strong signals from the surfaces in the laser vicinity will be produced.

Hassaballa et al (2012) performed TS on a modified plasma display panel cell. Two electrodes with a gap of 100 \( \mu \)m are embedded in glass coated with a protective MgO film. A dielectric-barrier discharge is then ignited with a 20 kHz, 240 V peak voltage on top of the dielectric at pressures of a few hundred mbar in Ne/Ar mixtures. Since the plasma is located very close to the surface, the laser beam path was prolonged and ‘cleaned’ with apertures. Electron densities were measured as
close as 60 μm from the surface. Another challenging setup is presented by Belostotskiy et al. (2008) as mentioned before with a backscattering arrangement using a periscope in the laser beam path. An electrode gap of 600 μm powered by dc (50 mA, 310–350 V) in 300–700 mbar argon was studied. Due to the high power density electron densities as high as 4 × 10^{19} m^{-3} are reported.

Even smaller plasma dimensions are reported in the work Kono et al. (2004). A micro gap was studied at atmospheric pressure consisting of two knife edges with 100 μm spacing in between. To insert the laser in that gap a very short focal length of 15 cm together with apertures along an extended laser beam path were used. That arrangement lead to a spatial resolution of 25 μm, however the issue of laser heating is likely to occur and therefore the laser power was reduced to 4 mJ per pulse. A microwave power of 100 W lead to peak pressures consisting of two knife edges with 100 mA, 310–350 V in air, i.e. already slightly coherent TS with a ≈ 0.3.

All three given examples employ a TGS to filter the stray and RyS light. That underlines the conclusion of section 2.b in respect to the choice of the TGS for a high performance filtering system.

4. Summary

This paper focuses on an overview of the implementation of Thomson scattering to micro-scale atmospheric pressure plasma jets. This is a complex diagnostic and so details and issues requiring consideration for its proper use are discussed. Some typical measurements, from a variety of plasma jet systems, are also shown and discussed to illustrate its power in providing fundamental data on the properties of the electrons in plasma jets.

In summary the crucial issues in implementing Thomson scattering are

(a) Scatter and collect as much of the incident photon flux as possible. The laser power should be as high as possible, without perturbing the plasma, but also the solid angle of the collection optics must be optimized.

(b) Have a stable and reproducible plasma source. That is required as multiple laser shots are averaged and any drift or a stochastic nature of the plasma will alter the measured values.

(c) Ensure proper reduction of the stray light and unwanted scattering signals. This requires consideration of issues, such as the laser beam quality and the capability of spectral filters to suppress photons at adjacent wavelengths. The wavelength dependence of a spectrometer can easily make the wings of the laser spectra appear as a false TS Gaussian distribution.

(d) If the TS signal can be fitted to a Gaussian fit, the resultant electron temperature gives a mean electron energy value.

In conclusion TS is currently a powerful technique for probing the spatial and temporal behaviour and properties of the major, lower energy component of the electrons in a variety of non-thermal atmospheric pressure plasma jets. These measurements can be invaluable in validating and as input to simulations and models for such plasma jets. There are still major challenges in being able to fully explore a broader range of the electron energy distribution and to work in high pressure plasma systems with short scale or non-reproducible spatial and temporal structure.

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