Response of the electron density and temperature to the power interruption measured by Thomson scattering in an inductively coupled plasma

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Electron density and temperature measurements during a power interruption of the generator are performed on a 100-MHz flowing-argon inductively coupled plasma. The Thomson scattering setup for measuring the density and temperature is triggered in such a way that temperatures and densities are obtained as a function of time while the power is interrupted. The response of the electron temperature to the power interruption shows a jump downwards within 5 μs, presumably towards the heavy particle temperature. Therefore, the method can be used to determine the heavy particle temperature. Furthermore, insight is obtained as to the behavior of the electron density in the center of the plasma; it increases rather than decreases after the power interruption.

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I. INTRODUCTION

In this paper we present the results of Thomson scattering experiments while the power of an atmospheric 100-MHz inductively coupled plasma (ICP) is interrupted. This type of ICP is widely used as a spectrochemical analytical instrument. In the future, a closed version might be used for lighting applications. Research on this type of plasma can improve their application and performance. Since the plasma is created by an EM field in an argon flow, we will find ionizing and recombining parts in the plasma. Using the technique of power interruption, the plasma can be studied while every part is recombining. This technique was developed by Gurevich and Podmoshenskii [1] and improved by Fey *et al.* [2]. The essence of this technique is that instantaneously the EM field is dropped to zero by switching off the generator for a period of typically 100 μs, resulting in an absence of energy input for that period. The technique of power interruption can be used for studying plasma processes such as recombination rates, heat transfer, transport, and deviations from equilibrium. In past years, a lot of measurements were performed using passive emission spectroscopy [2,3]. This paper presents a new method: a combination of the power interruption technique and the Thomson scattering measurements. Thomson scattering has proved to be a valuable technique for measuring local electron densities and temperatures [4–6]. The opportunity to combine these two methods gives a lot of additional information such as the spatially resolved decay of the electron density and temperature as a function of time. Moreover, in contrast to line emission spectroscopy, it is a direct and spatially resolved observation of the electron gas during the power interruption. The results show that after switching off the generator the electron temperature decreases instantaneously (within 5 μs) to a lower level, presumably the temperature of the heavy particles. This is followed by a decrease of the electron density in the active plasma parts with a typical time scale of 100 μs. More remarkably, it is found that the electron density in the center of the plasma increases during the power interruption.

II. THE POWER INTERRUPTION TECHNIQUE

Since a plasma can exist by the grace of the production of electrons, information on the properties of this plasma component can help us to understand and model the ICP. Especially the process of ionization, recombination, and diffusion are very closely connected to the kinetics of the electrons. Therefore, a sudden change in the kinetics of the electrons can provide a lot of information about these processes. This is the main goal of the power interruption technique.

For the stationary plasma, where the electron temperature (T_e) is higher than the heavy particle temperature (T_h), there is a stepwise energy balance that schematically can be presented by

$$\text{EM field} \rightarrow \{e\} \rightarrow \{h\} \rightarrow \text{surroundings} . \qquad (1)$$

The field heats the electrons $\{e\}$ which are cooled by the heavy particles $\{h\}$. These, on their turn, give their energy to the environment. The transfer from electrons to heavy particles is only possible if $T_e > T_h$.

Since the presence of local thermodynamic equilibrium (LTE) implies that (a) the temperature of the species is equal, thus $T_e = T_h$, and (b) the internal state distribution of the particles obeys the Saha-Boltzmann relation, we should realize that due to the energy balance given above (1), the plasma can, strictly speaking, not be in LTE. Interrupting the power of the generator gives insight in the degree of equilibrium departure. To that account we study the following sequence of events:

(1) Cooling. Immediately after the sudden power interruption the first step of the balance disappears, the EM field is removed, so the electrons are not heated anymore. This induces the decay in T_e towards T_h with a typical time scale of a few microseconds [1,7,8]. After this time scale, the first aspect of LTE (a) is reached. The change

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in T_e can be measured indirectly by following the line radiation in the cooling mechanism [1,2]. The technique as presented in this paper follows T_e directly.

(2) Decay. While the plasma as a whole cools down very slowly due to energy transfer to the surroundings, the electrons recombine and diffuse away. By studying these processes, insight is obtained in the Saha-equilibrium disturbing mechanisms that violate aspect (b) of LTE. This violation is expected to be generated by diffusion and recombination. It is also expected that diffusion is dominant over recombination and that the electron density (n_e) decreases with a typical time scale of 100 μs [7]. This time scale τ can easily be estimated with the ratio of the square of the gradient length (Λ^2) and the diffusion rate (D),

$$\tau=\frac{\Lambda^2}{D} . \quad (2)$$

For the ICP one can use 10^{-2} m^2/s for D and 10^{-3} m for Λ, finding indeed the typical time scale of 100 μs. This is confirmed in previous spectroscopic studies. However, with the present technique we follow the electron density locally, directly, and more precisely. In this way it is found that the decay time is lower in the presence of large gradients. In the center of the plasma an increase rather than decay is found in n_e.

(3) Heating. Immediately after the power is switched on again, the opposite of cooling takes place, restoring $T_e > T_h$.

(4) Ionization. The interruption cycle is finished by a rather slow increase of the n_e back to the steady state value.

In this paper we present the measurements on the first two stages, the cooling and decay stages. By applying a trigger circuit, we are able to use Thomson scattering to measure n_e and T_e as a function of time while the power is interrupted. In this way, we are able to get direct and spatially resolved insight in the properties of the electron gas.

Figure 1 shows, as an example, how the electron temperature is expected to change during the interruption of the power from $t=0$ to 100 μs. Since after interruption of the generator, the decay of the electron temperature presumably towards the heavy particle temperature (T_h) takes place within a few microseconds, whereas there are no other mechanisms which drive T_e and T_h apart from each other, we expect that during the power interruption T_e remains equal to T_h. This implies that measuring the electron temperature just after switching off the generator can be used as a method to obtain the heavy particle temperature for the steady state condition.

III. EXPERIMENTAL SETUP

A. The description of the plasma

The power interruption is carried out on a pure argon plasma. This plasma is created in a RF coil, fed by a 100-MHz generator developed by Philips with the additional feature of the capability to switch the generator on and off by a transistor-transistor logic signal. It takes about 2 μs before the EM field is decreased down to 5% of the stationary field. The power supply is kept on to make a fast restore of the EM field possible. The RF coil has two windings with a diameter of 35 mm and a total height of 15 mm. The plasma torch is placed in the center of the coil and consists of three concentric quartz tubes (see Fig. 2). This enables the separate control of the three argon gas flows. The measurements are performed using an outer flow of 12 l/min, an intermediate flow of 0.3 l/min, and a central flow of 0.6 l/min. The standard operating input power is 1.2 kW. In principle, it is possible to introduce a nebulized aqueous solution into the plasma, but this is not done in the present work. The standard position where the measurements are carried out lies 7 mm above the upper winding of the load coil (ALC), just above the hottest area in the plasma.

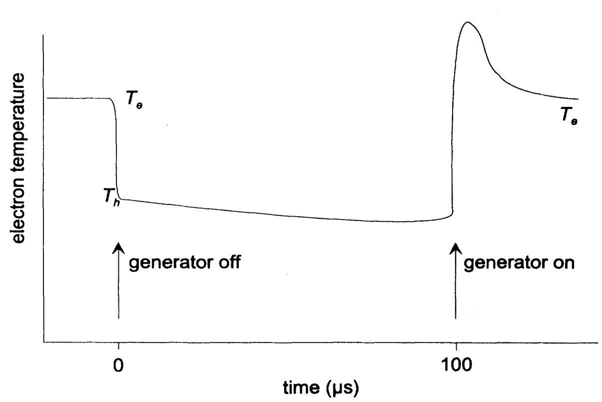

FIG. 1. The expected effect of the power interruption on the electron temperature. At the time of switching on the generator, a peak above the stationary T_e value is expected (this is subject for future research).

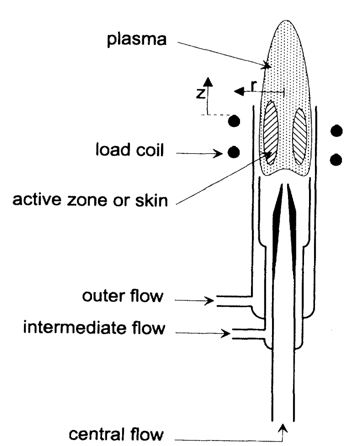

FIG. 2. A schematic view of the inductively coupled plasma. Note that an asymmetry as induced by the windings of the coil is reflected in the Thomson scattering values (cf. [6]).

B. The Thomson scattering diagnostics

Thomson scattering in the ICP originates from the scattering of incident light by the free electrons present in the plasma. Since this process has a very low efficiency, an intense light source and a very sensitive detector have to be used. The experimental setup is depicted in Fig. 3. The light source is a frequency doubled pulsed Nd:YAG (where YAG denotes yttrium aluminum garnet) laser at 532 nm (GCR 3 Quanta Ray, $E_{\text{pulse}}=0.45$ J, $t_{\text{pulse}}=7$ ns, $f_{\text{rep}}=10$ Hz). Two prisms and a lens L_1 focuses the laser light into the detection volume. With the lenses L_2 and L_3 the plasma and detection volume are imaged on the entrance slit of the monochromator. The detector is an intensified photodiode array with 1024 pixels and is cooled down to $-20\,^\circ$C. The full spectral range of the system is 13 nm with a resolution of 0.14 nm. The intensifier is gated, which allows us to measure during the short laser pulse only. In this way the plasma light is reduced by a factor of about 10^6 and can be neglected compared to the Thomson scattering signal. The Thomson scattering signal is broadened by the Doppler effect of the thermal electron motion. Therefore, the temperature of the electrons can be taken from the width of the Thomson scattering profile. After an absolute calibration of the setup using Raman scattering on nitrogen, the total scattering signal can be used for obtaining the n_e. For more details about the used Thomson scattering diagnostic and calibration technique we refer to [6].

Since the laser pulse is relatively short, it determines the point in time of the actual measurement of the density and temperature of the electrons. Therefore, a variable trigger circuit is necessary to choose the delay between switching off the generator and pulsing the laser. In Fig. 4 the signals dealing with the triggering are depicted. The trigger output of the flash lamps of the laser is used to trigger the generator. This signal precedes the actual laser light pulse by about 200 μs, taking into account that the generator itself has an internal delay of 40 μs. A variable delay device (VDD) is designed, with 0.1-μs resolution, to choose the time of interruption. In this way, the trigger circuit can be used to perform measurements conveniently at several points in time after the switching off of the generator. This happens with a repetition frequency of 10 Hz, the repetition frequency of the Nd:YAG laser. The final results are obtained by the integration of 1000 laser shots, which means 100 s to measure one position in the plasma for a certain moment after the switching off of the generator.

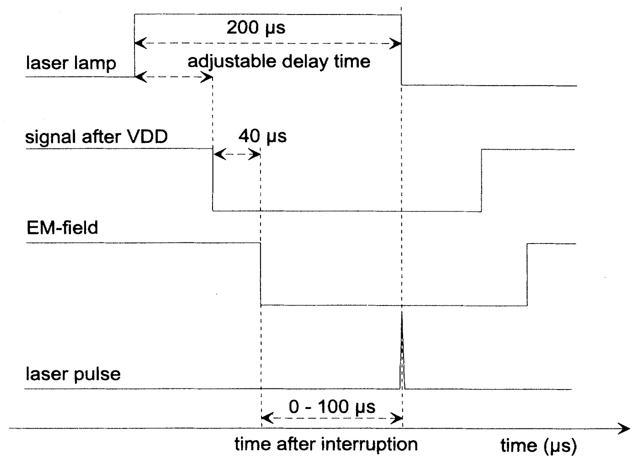

FIG. 4. The signals dealing with the triggering of the measurements.

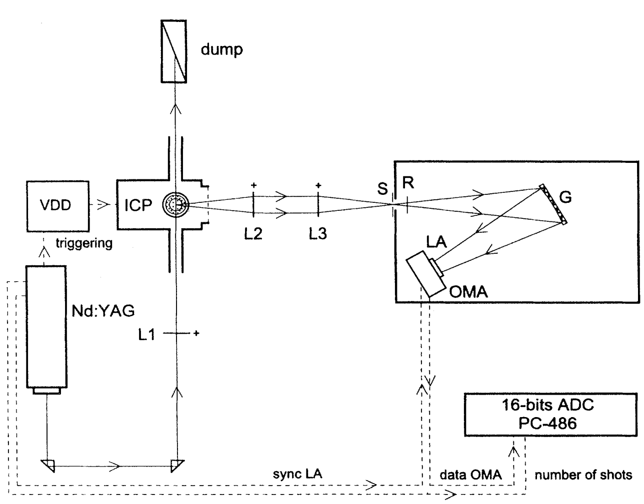

FIG. 3. The Thomson scattering setup, viewed from the top. L_1, L_2, and L_3 are focusing and detection lenses, S is the entrance slit, R is the mica retarder, G is the 2000 lines/mm grating, LA is the image intensifier, OMA is the photodiode array, and VDD is the variable delay device.

IV. RESULTS AND DISCUSSION

The measurements were performed at 7 mm and 13 mm ALC with an input power of 1.2 kW, for ten different radial positions. A second series was performed at 7 mm ALC and 4.5 mm from the center of the plasma with different power inputs, namely, 0.6, 0.9, 1.2, 1.5, and 1.8 kW. All the series of measurements start with a measurement without interrupting the generator. Immediately after this, Thomson measurements are carried out 5, 10, 20, 30, 40, 60, and 80 μs after switching off the generator. In this way, a set of measurements is obtained for several radial positions as a function of the time.

In Fig. 5 the electron temperatures and in Fig. 6 the electron densities are depicted for five of the ten radial positions as a function of time for a power of 1.2 kW. Note that at 7 mm ALC and for 1.2 kW input power the center of the active area lies about 4.5 mm from the center. Here we will find the highest n_e and also the highest heavy particle temperatures, as we will in Sec. IV A. The edge of the plasma lies at about 8 mm from the center, as can be seen in Figs. 5 and 6, where it is

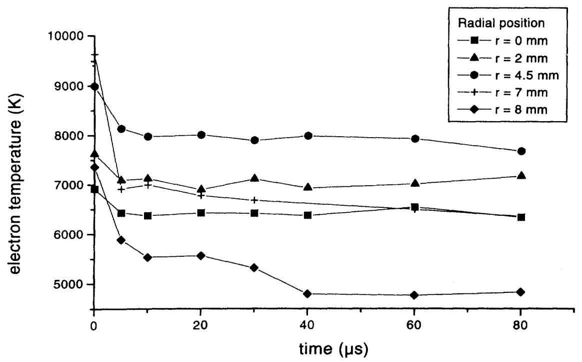

FIG. 5. The electron temperature as a function of time after switching off the generator for several radial positions at 7 mm ALC and with an input power of 1.2 kW.

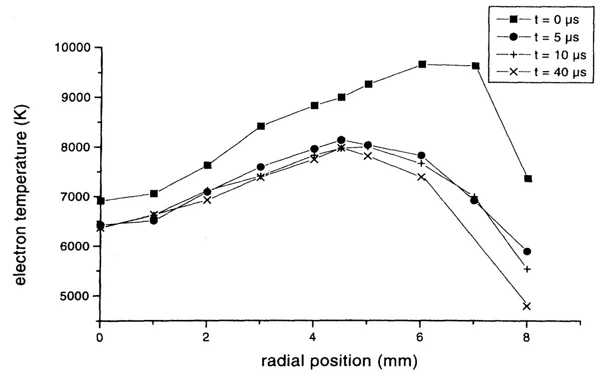

FIG. 7. Radial profiles of the electron temperature at 0, 5, 10, and 40 μs after switching off the generator, measured at 7 mm ALC, the input power equals 1.2 kW.

shown that the lowest values for n_e and T_e are found for $r=8$ mm. Efforts to measure even further at the edge of the plasma failed due to the presence of nitrogen at that position. Here, the measured signal shows a weak Thomson profile, because of the low densities, with a superposition of a strong Raman scattering spectrum, because the presence of some nitrogen.

The center of the plasma also has lower densities and temperatures compared to the skin, a well-known ICP feature originating from the limited skin-depth of the plasma for the EM field, the source of the ionization energy.

The further discussion of the results will consider first the behavior of the electron temperature and is continued by the response of the electron density to the power interruption. However, note that n_e and T_e are obtained from the same Thomson scattering profile.

A. The electron temperature during the power interruption

Looking at the behavior of the electron temperature as a function of time (cf. Fig. 5), we find large differences for the different radial positions. In the skin center, at $r=4.5$ mm, the response to the power interruption is as expected, namely, a sudden decrease of T_e towards a lower level, assumed to be the heavy particle temperature, and afterwards a slow decrease due to the cooling of the plasma by heat transfer to the surroundings. We will refer to this behavior with "typical response." The time scale of the jump of T_e to T_h is indeed within a few microseconds, as predicted by theory. Remarkable is the behavior as a function of time at the edges of the ionizing area, at $r=2$ and $r=7$ mm. Here we find from 0–5 μs a decrease followed by an increase of T_e in the next five microseconds. Since these jumps up in temperature are in the order of 100 K whereas the accuracy in T_e is about 150 K [6], the behavior can be due to inaccuracies of the diagnostics.

The temperature responses to the power interruption at the center ($r=0$ mm) and outer side ($r=8$ mm) of the plasma are comparable to that of the skin center ($r=4.5$

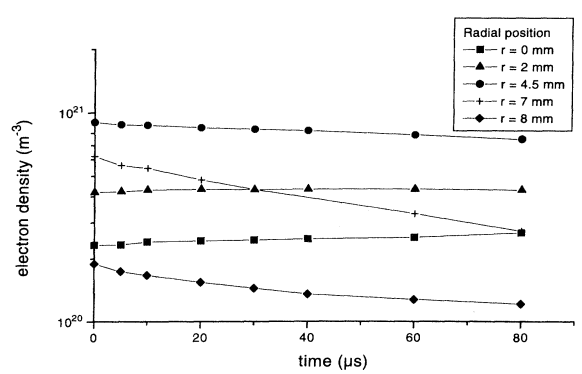

FIG. 6. The electron density as a function of time after switching off the generator for several radial positions at 7 mm ALC and with an input power of 1.2 kW.

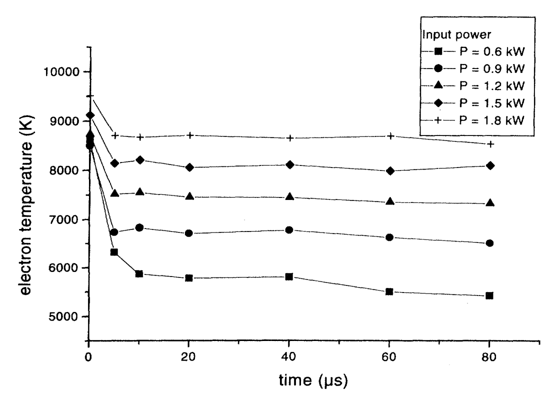

FIG. 8. The electron temperature as a function of time during the power interruption for five different power values. The measurements are performed at $r=4.5$ mm and at 7 mm ALC.

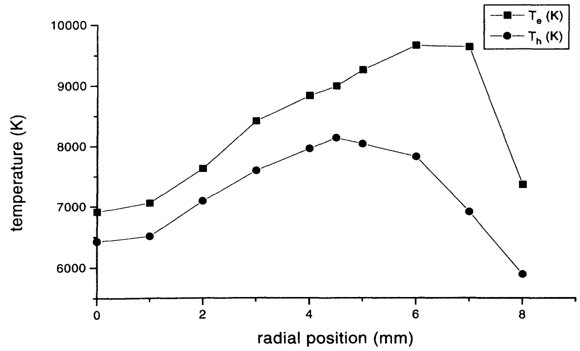

FIG. 9. The electron and heavy particle temperature as a function of radius obtained by combining the Thomson scattering diagnostic with the power interruption technique; the axial position was 7 mm ALC, the input power equals 1.2 kW.

mm). Figure 7 shows how the radial T_e profile evaluates as a function of time. After the jump downwards, we see globally a slow decrease of the temperature. Note that at the center of the plasma ($r=0$ mm) T_e remains constant after the small jump downwards immediately after the power switchoff.

The measurements at $r=4.5$ mm and $h=7$ mm ALC for different powers (cf. Fig. 8) show a similar response to the power interruption as the one given in Fig. 5. For five powers the response is measured. The measurement with 1.2 kW input power corresponds just to the measurement at $r=4.5$ mm in Fig. 5 if the mentioned accuracies are taken into account, but is measured one day later. Nevertheless, the reproducibility after having restarted the plasma is dominated by the accuracy of the power settings, which results in an estimated accuracy of about 500 K [6]. As a function of increasing input power, Fig. 8 shows that the size of the jump downwards decreases. This implies that the higher the input power

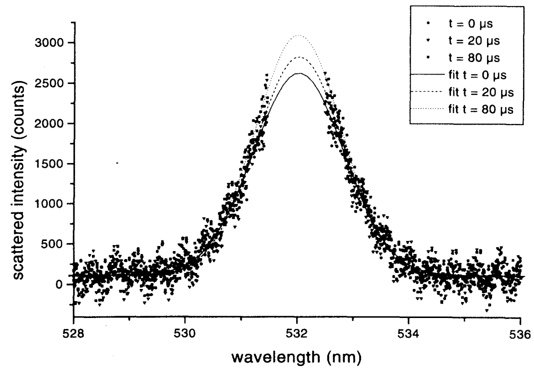

FIG. 11. Three Thomson scattering profiles obtained at 7 mm ALC and in the center of the plasma ($r=0$ mm). The lines in the figure are the corresponding fits, representing an accuracy better than 2% in width and surface. Note the increasing surface (density) as a function of time after switching off the generator. Note that the central part of the Thomson profile is absent, because this part is blocked on the detector to prevent it for blooming by the intense Rayleight scattering signal.

the smaller the temperature difference is between the electrons and heavy particles, or in other words, the plasma will be more in thermal equilibrium for higher input powers.

Assuming that the temperature of the electrons drops down to the heavy particle temperature within 5 μs after switching off the generator, the heavy particle temperature T_h for the steady state condition can be determined. In Fig. 9 the steady state values of T_e and T_h are given as a function of the radial position. In Fig. 10 T_e and T_h are shown as a function of input power at $h=7$ mm ALC and at $r=4.5$ mm.

We can conclude that this technique offers a good method to obtain heavy particle temperatures in an ICP. The more often used method uses Rayleigh scattering [9,10]. Rayleigh scattering can be used to measure the

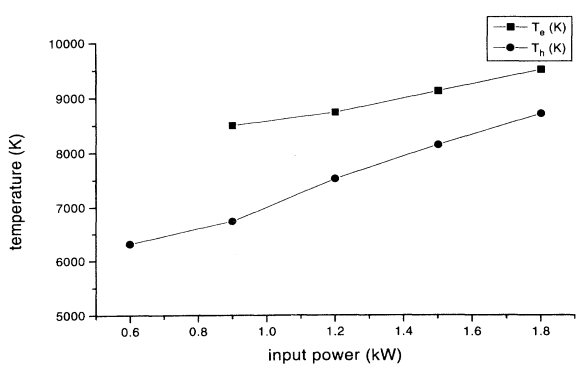

FIG. 10. The electron and heavy particle temperature for five different input powers obtained by combining the Thomson scattering diagnostic with the power interruption technique. The axial and radial positions are 7 mm ALC and 4.5 mm, respectively.

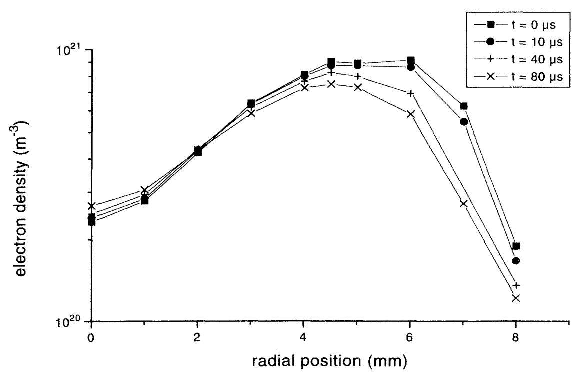

FIG. 12. Radial profiles of the electron density at 0, 10, 40, 80 μs after switching off the generator, measured at 7 mm ALC and with an input power of 1.2 kW.

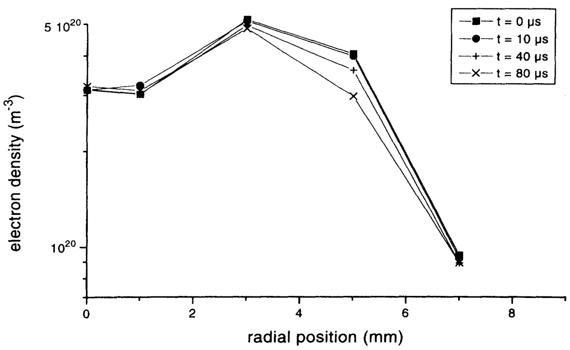

FIG. 13. Radial profiles of the electron density at 0, 10, 40, 80 μs after switching off the generator, measured at 13 mm ALC and with an input power of 1.2 kW.

heavy particle density and combining this with the known pressure (1 atm) the heavy particle temperature can easily be calculated using the ideal gas law. The temperatures measured by Marshall and Hieftje [9] show comparable results, but for a detailed comparison a further study is required.

B. The electron density during the power interruption

In Fig. 6 it can be seen that n_e decays slowly, which can be interpreted as a result of recombination and diffusion processes. The time scale in the hottest area ($r = 4.5$ mm) is about 100 μs. The decay of n_e is larger at the outer side of the plasma, at $r = 7$ and 8 mm, as can be deduced from the measurements. Surprisingly, the center of the plasma ($r = 0$ mm) is an exception. Here, we find an increasing electron density as can be concluded from the fitted raw data depicted in Fig. 11 as well. The Thomson scattering profiles are obtained at 7 mm ALC for 0, 20, and 80 μs after the generator is switched off. The rather low values of the electron densities cause profiles with noise. Nevertheless, the large number of measured points make the fits accurate within 2% for both width and total surface.

The increasing density indicates that there must be a large transport of electrons inwards. This could be driven and controlled by the large spatial n_e gradient present at stationary conditions. Before the plasma would be extinguished, there is a force that tries to smooth the electron density profile. This behavior can also be seen very clearly in Fig. 12, which shows that the density decreases everywhere but in the center, where it increases. Also at higher positions, at $h = 13$ mm ALC, a slowly increasing n_e is observed in the center of the plasma, see Fig. 13. Since the accuracy of the density measurements is within 15% [6], the response must be a physical effect.

V. CONCLUSIONS

The combination of the Thomson scattering diagnostics and power interruption enables us to study the response of the electron density and temperature to the power interruption of the EM field. The typical response is a sudden decrease of the electron temperature to the temperature of the heavy particles, followed by a slow decay due to cooling of the plasma as a whole. This typical response is predicted by theory. Furthermore, after the power of the generator is switched off, the electron density is expected to decay slowly, due to recombination and diffusion. This decrease is measured everywhere, except along the plasma axis, where an increase in electron density is observed, probably due to transport of electrons towards the center by the large spatial electron density gradient present at stationary conditions.

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