Enhanced metastable population through evaporation cooling and recombination in the argon afterglow

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Measurements, modelling and numerical simulations performed in a pulsed inductively coupled argon plasma at low pressures (1—5 Pa) show that very low electron temperatures are achieved on a characteristic time scale of a few tens of µs through evaporation cooling. This allows for recombination resulting in the observed increase of the metastable density in the afterglow phase. The previously observed super-linear scaling with the electron density of the electron decay time is well reproduced analytically by assuming that microfield limited electron-stabilized three-body recombination into highly excited Rydberg-states takes place. This hypothesis is strongly supported by experimental results from various diagnostic techniques.

1. Introduction

Modulated low pressure RF plasmas exhibit a variety of effects being highly interesting for both, research and industrial applications. For example Johnson et al already in 1950 [1] observed an increase of the plasma emission after power termination (the so called “afterglow”). Strauss et al [2] observed a similar effect and demonstrated via absorption spectroscopy an enhanced population of metastable argon atoms. The reestablishment of metastables, however, is still contradictory discussed. Many authors attribute this to the classical electron-stabilized three body \( \text{Ar}^+ + e + e \rightarrow \text{Ar}^{**} + e \) recombination [3] with a decay rate \( \nu_{3b} = -(1/n_e)(\partial n_e/\partial t) \) scaling as \( n_e^2 \).

In a recent investigation [4] the electron density decay time – obtained via a newly developed absorption spectroscopic method – varied as a function of the electron density indicating already recombination. However, in that work the electron density decay rate exhibited a scaling with the electron density which is weaker than quadratic but still stronger than linear. This can not be explained by the conventional three-body recombination discussed above. Furthermore, an increase of the metastable density and of the emission upon pulse termination is also observed.

In general, recombination presumes very low electron temperatures due to the inefficient electron capture at high electron energies. A very efficient electron cooling mechanism is the so called “diffusive” or “evaporation” cooling, consisting of the loss through diffusion only of the high energy electrons which are able to overcome the potential barrier of the wall sheath. This has been investigated theoretically [5,6] and also verified experimentally [7].

In the present work, we identified evaporation cooling as an efficient electron cooling mechanism taking place within tens of microseconds and leading to electron temperatures as low as the gas temperature, thus creating suitable conditions for recombination. Further it is proposed, that the three-body recombination into highly excited Rydberg states [8] where the upper quantum state number is limited due to Stark splitting in the ionic micro-field could explain the experimental observations.

2. Analytical model

2.1. Evaporation cooling

The fluid dynamic volume averaged approach outlined here aims to estimate the energy decay time in the afterglow of the inductively coupled plasma (ICP) by accounting for the evaporation cooling. The losses due to elastic collisions are important only at later times for establishing the final value of the electron temperature \( T_e \). The inelastic collisions are not important due to the fast energy decrease.

Assuming a Maxwellian distribution function with spatially homogeneous temperature \( T_e \), the
solution to the volume averaged fluid electron-energy balance equation takes the form:

$$\kappa T_e(t) = \frac{\kappa T_e(0)}{1 + t/\tau_e}$$  \hspace{1cm} (1)$$

with $\kappa$ the Boltzmann constant, $T_e(0)$ the electron temperature at power-switch-off time ($t = 0$) and the time constant $\tau_e$ given by:

$$\frac{1}{\tau_e} = \frac{\pi^4 \kappa T_e(0)}{2m_iL^2\nu_{in}} \left[1 + \ln \left(\frac{L\nu_{in}}{\pi u_B} \sqrt{\frac{m_i}{2\pi m_e}}\right)\right].$$  \hspace{1cm} (2)$$

Here $\nu_{in}$ is the ion-neutral elastic collision frequency, $L$ is the chamber dimension and $u_B$ is the Bohm velocity. For our experimental conditions equation (2) gives $\tau_e = 10\mu$s.

It should be noted that the analytical solution for the temperature decay (equation (1)) remains valid also in the case of a non-maxwellian distribution. Only the value for the energy decay time (2) would then be different which could explain the observed discrepancies between calculated and measured decay times, presented further on.

2.2. Recombination

Due to the Stark splitting in the ionic microfield, the atomic discrete levels transform in the continuum already at a finite quantum number $n_m$. Accounting for this effect, it can be shown that the recombination rate is given by:

$$\nu_r = C \left[ \frac{E_{16/15}}{E_{14/15}} \frac{n_e^{16/15}}{n_e^{14/15}} \frac{n_e^{1/3}}{n_e^{2/3}} \psi \right] \hspace{1cm} (3)$$

Here $C = 4.0 \times 10^{-30} \text{ cm}^6 \text{s}^{-1} \text{eV}$, $n_{IT} = 10^{23.19} \text{ cm}^{-3}$ is the scaling density resulting from the Inglis-Teller formula, the ionization energy $E_i$ and the electron temperature are in eV, the density in cm$^{-3}$ and the parameter $\psi = (E_i/T_e)(n_e/n_{IT})^{4/15}$. Typically for our experimental conditions $\psi \ll 1$ resulting in the slightly superlinear $n_e$-scaling of the electron decay rate of 4/3 in excellent agreement with the experimental data [4].

3. Experimental

3.1. Experimental apparatus

The experimental setup is described in detail elsewhere [4,9] and here only a brief summary is given. The stainless steel vacuum vessel (Fig. 1) consists of an upstream plasma chamber (i.d. 350 mm) and a downstream remote chamber (i.d. 500 mm). A 2.5 turn four-fold inductive planar antenna (200 mm diameter) driven at 13.56 MHz creates the plasma. Power to the antenna is supplied by a Dressler Cesar 1312 RF generator. For impedance matching a standard L-type matching unit is used. The RF power is pulsed at a repetition frequency of 5, 10 or 20 Hz (depending on the discharge conditions) and a duty cycle of 85%. The fall time of the RF current in the generator amounts to less than 1 $\mu$s.

The metastable atom density is monitored by a tunable diode laser absorption spectroscopy system and consists of a compact grating-stabilized external cavity diode laser (ECDL) and the adherent optics. The laser is tuned at 696.73 nm corresponding to the transition from the Ar1s$_5$ state to the Ar2p$_2$ state (in Paschen notation). The time-dependent measurements of the ion velocity distribution function (IVDF) were performed by the use of a retarding field energy analyzer (Impedans Ltd., Semion RFEA Probe) placed at the discharge bottom. For the time-resolved measurements of the electron density a 26.5 GHz Ka-band microwave interferometer (Miwitron Ltd., MWI 2650) was used.

3.2. Experimental results

Figure 2 presents the measured IEDFs in the afterglow. A peak is clearly visible, corresponding to the ion energy gain from traversing the wall.
sheath. Since the potential drop in the sheath is proportional to the mean electron energy (or temperature in case of a Maxwellian distribution), an estimate could be obtained for the time evolution of the electron temperature in the postdischarge.

Figure 3 presents the inverse of the obtained in this way electron temperatures. According to (1) it should be a straight line with a slope depending on \( \tau_\varepsilon \). The obtained results confirm this nicely. The resulting time constant of 26 \( \mu s \) is also in a good agreement with the predictions of (2) and the results from a 2D numerical fluid simulation.

It should be noted that the exact value of the electron temperature (Fig. 3) might be disputable in case of non-maxwellian distribution functions but the experimentally obtained time constant will nevertheless remain the same.

Figure 4 shows the temporal decay of the electron density as a function of power at a gas pressure of 1 Pa measured by the microwave interferometer. Three different intervals with different decay rates are clearly visible. The first interval immediately after pulse termination is dominated by the rapid diffusional decay due to the loss of hot electrons (evaporation cooling). In the second interval the density decay is dominated by recombination. At higher powers the density is higher and, thus, the recombination is better pronounced, resulting in a steeper decay of the electron density. In the third interval at very late times, when the electron density is too low for the recombination to play a role and the electron temperature is already very low, a slow diffusional decay is observed. In contrast to the recombination interval, where the decay time exhibits a pronounced power dependency, the late diffusion interval varies only weakly with power.

Figure 5 shows the temporal evolution of the normalized metastable density as a function of the applied power at 1 Pa. At lower powers the metastable density monotonously decreases, while at higher powers an increase of the metastable density and an afterpeak are observed. Immediately after pulse termination metastable quenching dominates. At \( t \approx 500 \mu s \) the electrons are already cooled efficiently by evaporation cooling and recombination becomes efficient. According to the proposed recombination mechanism, the recombination results in highly excited Rydberg states, which then radiatively decay to the metastable levels, resulting in their population and an increase of the emission (afterglow). For
lower powers, however, the electron densities are lower and the recombination is not efficient, explaining the observed monotonous decay of the metastable density.

Figure 6 shows a comparison of results from a numerical modelling and measurements. More details for the simulation could be found in [9]. A correlation between the simulated Rydberg atoms density and the observed emission and a good agreement in the evolution of the metastables are seen, which confirms the above explanation for the presence of the afterglow peak in the metastable density.

4. Conclusions

It was shown that an efficient electron energy decrease through evaporation cooling in the afterglow of a low pressure ICP takes place on a time scale of tens of microseconds. This provides favourable conditions for recombination as indicated by measured afterpeaks in the metastable density and the time decay of the electron density obtained via a microwave interferometer. The close-to-linear scaling of the density decay time with the electron density obtained experimentally is reproduced assuming three-body recombination into highly excited Rydberg states influenced by the ionic micro-field.

References


