Self-excited nonlinear plasma series resonance oscillations in geometrically symmetric capacitively coupled radio frequency discharges

Z. Donkó,1,a J. Schulze,2 U. Czarnetzki,2 and D. Luggenhölscher2
1Research Institute for Solid State Physics and Optics, Hungarian Academy of Sciences, P.O. Box 49, H-1525 Budapest, Hungary
2Institute for Plasma and Atomic Physics, Ruhr-University Bochum, Bochum 44780, Germany

Received 20 February 2009; accepted 10 March 2009; published online 1 April 2009

At low pressures, nonlinear self-excited plasma series resonance (PSR) oscillations are known to drastically enhance electron heating in geometrically asymmetric capacitively coupled radio frequency discharges by nonlinear electron resonance heating (NERH). Here we demonstrate via particle-in-cell simulations that high-frequency PSR oscillations can also be excited in geometrically symmetric discharges if the driving voltage waveform makes the discharge electrically asymmetric. This can be achieved by a dual-frequency \((f+2f)\) excitation, when PSR oscillations and NERH are turned on and off depending on the electrical discharge asymmetry, controlled by the phase difference of the driving frequencies. © 2009 American Institute of Physics. [DOI: 10.1063/1.3110056]

Capacitively coupled radio frequency (CCRF) discharges are of paramount importance for plasma processing applications ranging from chip and solar cell manufacturing to the creation of biocompatible surfaces. The kinetics and the heating of electrons largely determine the properties of the plasma. Therefore, a detailed understanding of the electron heating mechanisms is particularly important. Plasma processing CCRF discharges are often geometrically asymmetric (different surface areas of powered and grounded electrodes) and are operated at low pressures. Most models of electron heating\(^1\)–\(^5\) assume a sinusoidal rf current waveform and neglect high frequency self-excited nonlinear plasma series resonance (PSR) oscillations. This is justified in case of symmetric discharges, or generally at high pressures, but not in asymmetric discharges operated at low pressures, typically below about 10 Pa. Following some earlier investigations on the PSR,\(^6\)–\(^11\) it has recently been demonstrated that self-excited nonlinear PSR oscillations lead to a drastically enhanced electron heating at low pressures in geometrically symmetric CCRF discharges via nonlinear electron resonance heating (NERH).\(^12\)–\(^15\) PSR oscillations of the rf current and NERH have also been observed experimentally in such discharges.\(^16\)–\(^19\)

The PSR phenomenon can be understood in the frame of a simple voltage balance.\(^15\) The applied rf voltage \(V_\infty\) and the self bias \(\eta\) must be balanced by the sum of the two sheath voltages and the voltage across the bulk \(\Phi_b\). Using proper normalization, similar to Ref. 15, the PSR equation is

\[
\eta + V_\infty = -q^2 + \varepsilon (q_i - q)^2 + \Phi_b,
\]

where \(q\) is the unbalanced charge in the sheath at the powered electrode, \(q_i\) is the total unbalanced charge in the discharge, \(\varepsilon = (A_p/A_g) \left( \frac{n_{sp}}{n_{ig}} \right)\) is the symmetry parameter,\(^20\) and \(\Phi_b = -2(\partial^2 q/\partial \tau^2 + \kappa \partial q/\partial \tau)\).\(^15\) Regarding \(\Phi_b\), the first term represents electron inertia and the second term the resistive part. The experimentally verified\(^16\) nonlinear charge voltage relation of the sheath \(q^2\) and the inertia term \(\partial^2 q/\partial \tau^2\) in Eq. (1) form a nonlinear oscillator. This leads to high-frequency harmonics (more than an order of magnitude higher than the driving rf frequency) of the current with large amplitudes, (i) if collisional damping is sufficiently low, i.e., at low pressures and (ii) if the nonlinearity \(q^2\) in Eq. (1) does not cancel. In geometrically symmetric discharges, the areas of the powered and grounded electrodes \(A_p\) and \(A_g\) and the mean ion densities in the respective sheaths \(n_{sp}\) and \(n_{ig}\) are equal and \(\varepsilon = 1\). Although the individual sheaths are still nonlinear, the nonlinearity cancels for the sum of both sheath voltages. On the other hand, if \(A_p \gg A_g\), \(\varepsilon\) approaches zero and the sheath nonlinearities do not cancel. Clearly, this condition is optimum for the generation of PSR oscillations, which until now have been observed only in strongly geometrically asymmetric discharges.

The recently discovered electrical asymmetry effect \(\text{(EAE)}\)\(^20\)–\(^22\) now provides a means to achieve \(\varepsilon \neq 1\) even in geometrically symmetric discharges so that PSR oscillations become possible. It has been shown that the simultaneous application of a fundamental frequency \(f\) and its second harmonic (from here on we proceed with non-normalized expressions for \(V_\infty\) and \(\eta\) given in Volts)

\[
V_\infty(t) = V_0 [\cos(2 \pi f t + \Theta) + \cos(4 \pi f t)],
\]

leads to the establishment of a dc self-bias even in geometrically symmetric discharges. Here \(\Theta\) is a (variable) phase angle, and \(V_0\) is the amplitude of each harmonic given in Volts. This self-bias ensures a period averaged charge balance at each electrode and depends almost linearly on the relative phase \(\Theta\) between the applied voltage harmonics. At low pressures this variable dc self-bias leads to different mean ion densities in both sheaths and, therefore, to \(\varepsilon \neq 1\). In this way an asymmetry in the sheaths is created electrically instead of geometrically.

In this letter we demonstrate that the EAE allows the generation of PSR oscillations even in geometrically symmetric CCRF discharges since it leads to a symmetry parameter different from unity. It is the remaining nonlinearity in Eq. (1) that leads to the high frequency oscillations, although \(A_p = A_g\). As \(\varepsilon\) is a function of \(\Theta\), we also show that PSR
oscillations and NERH can be switched on and off by changing the phase.

Our investigations are carried out for a one-dimensional Ar discharge, simulated by a self-consistent bounded (1d3v) particle-in-cell code, complemented with a Monte Carlo treatment of collision processes. The voltage waveform applied to the powered electrode is given by Eq. (2), the other electrode is grounded. We use \( f = 13.56 \text{ MHz} \) and an electrode gap of \( L = 2.5 \text{ cm} \). The self-bias voltage \( \eta \) is determined in the simulations in an iterative way to ensure that the charged particle fluxes to the two electrodes, averaged over one lower frequency period \( (T = 1/f) \), become equal. Details of the simulation can be found elsewhere.\(^ {22} \)

The calculated self-bias voltage, the symmetry parameter, and the temporally averaged ion density profiles are displayed in Fig. 1 as a function of the phase angle \( \Theta \), for a pressure of 3 Pa and \( V_0 = 1000 \text{ V} \). By adjusting \( \Theta \), (i) the bias can be tuned over a wide range and the role of the two (powered and grounded) electrodes can be reversed and (ii) the position of the plasma bulk and the sheath lengths can be changed. \( \epsilon - 1 \) varies similarly to the bias, as can be seen in Fig. 1(a). The main advantages of this effect are that at fixed electrode geometry (i) the control of the position of the density profile provides a unique opportunity for controlling the relative fluxes of radicals (created in the plasma bulk) to the electrodes and that (ii) the variable dc self-bias provides a way to control the energy of the ions reaching the electrode surfaces.

Figure 2 shows the development of PSR oscillations with decreasing pressure, at fixed voltage amplitude \( V_0 = 1000 \text{ V} \) and phase angle \( \Theta = 7.5^\circ \). At a high pressure of \( \rho = 10 \text{ Pa} \), the PSR oscillations are strongly damped by collisions and the well-known pattern of electron heating/cooling is observed. At 5 Pa slight modulations appear at both the heating and cooling regions at both electrodes during the first quarter of the low frequency rf period. These modulations of the heating rate (and similarly of the electron current, not shown) become very pronounced at 3 Pa. Finer details are revealed in Fig. 2(d). The values of the symmetry parameter \( \epsilon \) are displayed, too. Figure 2 also shows (i) the heating of electrons due to an electric field reversal near the powered electrode during sheath collapse \( [t/T \approx 0.85 \text{ in Figs. 2(a)-(c)]} \) and (ii) the formation of electron beams during sheath expansion \( [\text{tilted excitation maxima in Fig. 2(e)}] \).\(^ {15-18} \) Both phenomena are related to the rapid expansion and constriction of the sheath. Figure 2 agrees qualitatively well with experimental investigations of NERH.\(^ {16-18} \)

Figure 3 illustrates how the PSR modulations of the electron heating are turned on and off as a function of the electrical discharge asymmetry induced and controlled by the EAE. The control parameter is the phase angle \( \Theta \) between the driving voltages (see Fig. 1 for \( \eta \)). Compared to the case of \( \Theta = 0^\circ \) (strong electrical asymmetry) PSR oscillations become less pronounced at \( \Theta = 30^\circ \) and practically vanish at \( \Theta = 51.8^\circ \), where the bias voltage \( \eta \) vanishes as well, and the electrical asymmetry is minimum. At phase angles above \( \Theta = 51.8^\circ \), the bias becomes positive and PSR oscillations move to the second quarter of the rf period, with NERH heating occurring near the grounded electrode.

The PSR oscillations lead to a rapid expansion of the sheath, which is much faster than by just the fundamental rf frequencies. Thereby, fast beams of electrons traveling from the sheath region toward the plasma bulk are created. The
density of these fast electrons equals the sheath density and is much lower than the bulk density. Therefore, their density can exceed by far the density of the bulk electrons at comparable energy. Thereby ionization can be strongly enhanced by the PSR effect. This enhancement is not necessarily adequately represented in the temporally averaged heating rate. It is a purely kinetic effect; in fact, few hot electrons are more efficient in ionizing than many warm electrons. Here, the energy gain of beam electrons by the PSR oscillations is well visualized in the heating rate plots.

Obviously, the electrons can gain energy only within the sheath region, where large fields are necessary to drive the current with a low density of electrons. In the bulk, the electron density is much higher, the field is much lower, and no substantial gain in energy occurs. However, the fast electrons from the sheath continue ballistically into the bulk and eventually become isotropic after about one mean free path. The trajectories in the bulk are visible in the excitation rate that probes energetic electrons.

The support by the Hungarian Fund for Scientific Research (Grant Nos. OTKA T048389, IN69892, and K77653) and the DFG (Grant No. GRK 1051) is gratefully acknowledged.