Plasma Sources Sci. Technol. **9** (2000) 541–544. Printed in the UK PII: S0963-0252(00)15494-5

Nonlinear radio-frequency potential in an inductive plasma

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Received 25 May 2000, in final form 2 July 2000

Abstract. By reducing the driving frequency of an inductive discharge, a nonlinear regime in the skin layer was reached where the nonlinear radio-frequency (rf) Lorentz force acting on electrons prevailed over the electric field force. Under these conditions a large second harmonic potential was found in the skin layer. A corresponding electric field at the second harmonic was found to be much larger than the discharge maintenance electric field at the fundamental frequency. This nonlinear electric field was successfully modelled as an rf Hall effect induced by the cross product of the rf magnetic field and the rf drift velocity.

Nonlinear effects in inductively coupled plasmas (ICPs) associated with radio-frequency (rf) components of the magnetic field are a subject of discussion in the current literature [1–12]. Theoretical analysis of a one-dimensional ICP [4, 5] has shown that the rf magnetic field changes the direction of the electron acceleration and may affect the electron collisionless heating rate in the skin layer. It has also been noted that electron oscillatory motion in the inhomogeneous rf field of the ICP skin layer may lead to the ponderomotive force [1–3] affecting the plasma density profile and to a second harmonic force resulting in rf polarization potential [4, 5]. Both the ponderomotive and second harmonic forces have a common origin and are due to nonlinear electron motion caused by the Lorentz force ($\boldsymbol{v} \times \boldsymbol{B}$) and by the $\boldsymbol{v} \cdot \mathrm{grad}\, \boldsymbol{v}$ part of the inertial force [8, 10].

To the best of our knowledge, both the ponderomotive force effect and the polarization potential at the second harmonic have not been observed in experiments with inductive plasmas. One reason for this is the negligibly small nonlinear effect that occurs at the typical ICP driving frequency $f = \omega/2\pi = 13.56$ MHz [2]. On the other hand, the ponderomotive force, as well as the second harmonic force, scale as $(\omega^2 + \nu_{eff}^2)^{-1}\,\mathrm{grad}\, E^2$ [12], therefore these forces are more pronounced at lower gas pressures (where the electric field E and the drift velocity are large) and at lower driving frequencies (where the rf magnetic field is large). Note that over a wide range of gas pressure p and driving frequency ω, the rf electric field E is a weak function of p and ω, and in an ICP where $E \propto \omega B$, then $B \propto \omega^{-1}$. As discussed in [8, 10], the nonlinear effects in an ICP are essential when the Lorentz force acting on electrons in the ICP skin layer is comparable to or larger than the electric force. This condition corresponds to the nonlinear skin effect and is equivalent to $\omega_B^2 > \omega^2 + \nu_{eff}^2$, where ω_B is the electron cyclotron frequency of the rf magnetic field and ν_{eff} is the effective electron collision frequency accounting for both the collisional and stochastic (collisionless) heating.

We report here on the observation of a second harmonic polarization potential in the skin layer of a low-pressure ICP operated at low frequency. Our experiments were carried out in a cylindrical ICP (20 cm in diameter and 10.5 cm in length) with a planar induction coil as described in detail in [8]. This system differs from that in [8] because the driving frequency was reduced to 0.9 and 0.45 MHz, with a corresponding change in the induction coil and matching circuit. The induction coil has the same geometry and dimensions (3.8 cm inside diameter (ID) and 12.7 cm outside diameter (OD)) as in [8], but it consists of 20 turns of litz wire. Since capacitive coupling is negligible at these low frequencies, no electrostatic screen was used.

The rf plasma potential was measured with a small Langmuir probe moved along the discharge axial direction at the discharge centre ($r = 0$) and at the fixed radial position ($r = 4$ cm) corresponding to the maximum in the radial distribution of the azimuthal rf electric $E_{\theta\omega}(r)$ and the radial rf magnetic $B_{r\omega}(r)$ fields. To prevent capacitive shunting by the probe shield and the input capacitance of the rf probe connected to the spectrum analyser, the Langmuir probe was positively biased with respect to the plasma potential via a resistor, $R = 50$ kΩ. As a result, the probe–plasma impedance was reduced by two orders of magnitude (compared with a floating probe) and was always much smaller than R and the shunting capacitive reactance. The same Langmuir probe was used to measure the electron energy distribution function (EEDF) and the dc plasma potential and to evaluate basic plasma parameters: plasma density n, electron temperature T_e and electron–atom collision frequency ν_{en}. Additionally, a two-dimensional differential magnetic probe [13] was used to evaluate the axial distribution of the azimuthal rf electric field $E_{\theta\omega}(z)$ and current density $J_{\theta\omega}(z)$ at the fixed radial position, $r = 4$ cm.

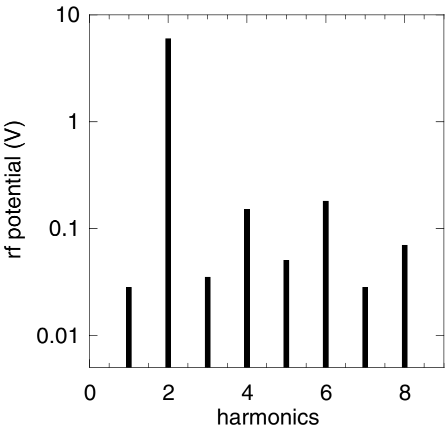

Figure 1. Plasma potential spectra measured in the skin layer. $P = 200$ W, $p = 1$ mTorr, $f = 0.45$ MHz, $z = 1$ cm and $r = 4$ cm.

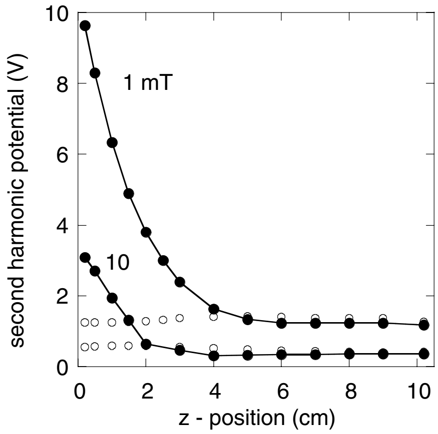

Figure 2. Axial distribution of the second harmonic potential for $f = 0.45$ MHz and $P = 200$ W. The full circles correspond to $r = 4$ cm and the open circles correspond to $r = 0$.

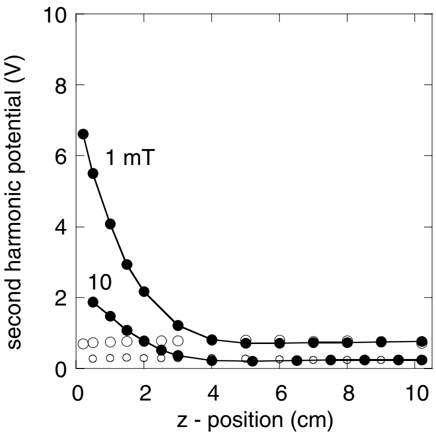

Figure 3. The same as in figure 2 but for $f = 0.9$ MHz.

These measurements were made in argon gas at pressures of 1 and 10 mTorr and at a power dissipated in the discharge between 50 and 400 W.

Prior to discussing our results, let us briefly describe the specific features of our ICP. At 1 mTorr the discharge operates near the ion free path regime ($\lambda_i \approx L, R$) with an electron mean free path λ_e much larger than the chamber length L and radius R. Under this condition the ICP operates in a strongly non-local regime (anomalous skin effect) where the thermal electrons pass through the skin layer in a small fraction of the rf field period, $v_t/\delta_\omega \gg \nu_{en}, \omega$, where v_t is the electron thermal velocity, $v_t = (2T_e/m)^{1/2}$ and δ_ω is the skin depth. Due to the low driving frequency, plasma screening has little effect on the electromagnetic field distribution, which is mainly defined by the chamber geometry. The electromagnetic field distribution within the discharge is similar to that under vacuum, with $\delta_\omega \approx \delta_0 = 2.42$ cm, which corresponds to the rf electric field distribution in the vacuum. At 200 W and 1 mTorr, the plasma density and electron temperature in the plasma centre ($r = 0$, $z = 5$ cm) were 1.3×10^{11} cm^{-3} and 6 eV respectively, independent of the driving frequency. These same parameters in the plasma heating zone ($r = 4$ cm, $z = 1$ cm) were about 5×10^{10} cm^{-3} and 7 eV, respectively. From magnetic probe measurements we found $\omega_B^2 \gg \omega^2 + \nu_{en}^2$ in the skin layer, thus the Lorentz force greatly exceeded the electric force there. Thus, in this case the skin effect in our ICP is not only anomalous but also nonlinear.

The frequency spectra of the plasma potential in the middle of the skin layer, 1 cm from the quartz window ($z = 1$ cm, $r = 4$ cm), in an ICP driven at 0.45 MHz is shown in figure 1. In this figure and hereafter, root-mean-square (rms) values are used for all of the oscillating quantities and the rf potential is referenced to the grounded discharge chamber. Some ICP parameters: the azimuthal electric field $E_{\theta\omega}$, the radial magnetic field $B_{r\omega}$, the characteristic decay length of the fundamental δ_ω and the second harmonic $\delta_{2\omega}$ fields and the rf drift velocity $v_{\theta\omega}$ are given in the table 1. As shown in figure 1, the first (fundamental) harmonic, probably induced by finite capacitive coupling from the induction coil, is the smallest one while the second harmonic exceeds the electron temperature and dominates. Figure 1 also shows that all even harmonics are significantly larger than the odd harmonics. The magnitudes of all harmonics (except the second) are much smaller than the electron temperature, and thus, they do not affect the discharge operation or interfere with the Langmuir and magnetic probes.

One might speculate that the observed second harmonic potential originates in the sheath at the plasma boundary as a result of the sheath nonlinearity and the capacitive coupling from the induction coil. However, the fact that the second harmonic is two orders of magnitude larger than the fundamental harmonic rules this out. In what follows we will focus only on the characteristics of the second harmonic potential (SHP) since it is the dominant effect observed.

The axial distributions of the SHP at $r = 4$ cm and on the discharge axis ($r = 0$) are shown in figures 2 and 3 for the argon pressures of 1 and 10 mTorr and the driving frequencies of 0.45 and 0.9 MHz. At 1 mTorr and $r = 4$ cm (near the maximal electromagnetic field) these figures show that the SHP drops sharply in the skin layer, away from the window ($z = 0$), and reaches a background level behind the skin layer. The rf potential distribution implies that the source of the second harmonic voltage is located in the skin layer. The background plasma potential, accounting for about 10% of

Table 1. ICP parameters for $P = 200$ W, $p = 1$ mTorr, $r = 4$ cm and $z = 1$ cm.

f (MHz)	$E_{\theta\omega}$ (V cm^{-1})	δ_ω (cm)	$V_{2\omega exp}$ (V)	$V_{2\omega theor}$ (V)	$E_{2\omega}$ (V cm^{-1})	$\delta_{2\omega}$ (cm)	$B_{r\omega}$ (G)	$v_{\theta\omega}$ ($\times 10^{-7}$ cm s^{-1})
0.45	0.83	2.24	6.5	8.1	4.6	1.42	14	5.8
0.9	0.97	1.94	4.1	6.9	3.3	1.25	9.8	8.0

the total second harmonic voltage, is across the chamber wall sheath.

The axial electric field $E_{z2\omega}$ of the measured SHP (see table 1) appears to be significantly larger than the azimuthal electric field at the fundamental frequency $E_{\theta\omega}$ that maintains the discharge. Thus, at 0.45 MHz and 1 mTorr in the middle of the skin layer ($z = 1$ cm), $E_{z2\omega}/E_{\theta\omega} = 5$. Note that $E_{z2\omega}$ is the rf component of the ambipolar field and therefore does not produce a conductive current nor electron heating. Indeed, we were able to measure a small axial second harmonic current density $J_{z2\omega}$ in this discharge that was similar to those at 3.39 and 6.78 MHz [10], but the magnitude of $J_{z2\omega}$ appeared to be two to three orders of magnitude smaller than the conductive current calculated using the measured values of $E_{z2\omega}$. The polarization field $E_{z2\omega}$ and the second harmonic current $J_{z2\omega}$, both having nonlinear origins, are produced by different parts of the nonlinear force [11] ($E_{z2\omega}$ is due to the potential part and $J_{z2\omega}$ is due to the solenoidal (inductive) part) and they have different directions and space distributions [10]. Thus, the large second harmonic potential observed here is a result of plasma polarization due to the rf Lorentz force as it was predicted in [4, 5]. Note that under the conditions of our experiment, the centrifugal inertial force $mv_{\theta\omega}^2/r$ was always much less than the Lorentz force.

We found that in the skin layer $E_{\theta\omega}(z)$ and $E_{z2\omega}(z)$ both decay exponentially, but have different decay lengths, $\delta_\omega > \delta_{2\omega}$, see table 1. This suggests a nonlinear origin for the second harmonic field; note that a local quadratic effect proportional to $E_{\theta\omega}^2$ should have a space decay length of a half of $E_{\theta\omega}$. In our experiment the ratio of $\delta_\omega/\delta_{2\omega}$ was found to be 1.6 for both frequencies, which implies some 'non-quadratic' nonlinearity. Such a behaviour is typical for the anomalous skin effect when the electron thermal motion is important. The electron thermal motion mixes the electron drift velocities along the z-direction (space dispersion) so that in the skin layer $v_{\theta\omega}(z)$ is less than its local value $v_{loc} = eE_{\theta\omega}(z)/m(\nu_{en} + j\omega)$ and $v_{\theta\omega}(z) > v_{loc}(z)$ behind the skin layer, and the characteristic length of the azimuthal drift velocity decay δ_v is greater than δ_ω [14]. Therefore, for an rf polarization field induced by the Lorentz force in the nonlocal regime, one has to expect $\delta_{2\omega}^{-1} = \delta_\omega^{-1} + \delta_v^{-1}$ so that $\delta_\omega/\delta_{2\omega} = 1 + (\delta_\omega/\delta_v) < 2$. The electron thermal motion is also expected to bring non-locality into the plasma polarization and further increase $\delta_{2\omega}$.

Figures 2 and 3 show that there is no significant second harmonic potential along the axis of the discharge symmetry, where both $E_{\theta\omega}$ and $B_{r\omega} = 0$. Thus, it is concluded that the second harmonic rf field observed in this experiment is only observed where the rf magnetic and the induced electric field are strong. Figures 2–4 show that the second harmonic potential decreases with increases in gas pressure and driving frequency. This trend is consistent with the nonlinear nature

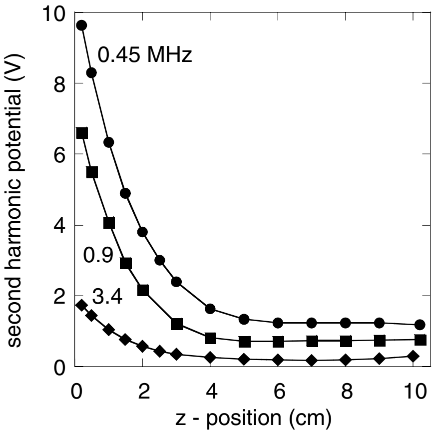

Figure 4. Axial distribution of the SHP for different driving frequencies. $P = 200$ W and $p = 1$ mTorr.

of the SHP, suggesting that $V_{2\omega}$ is proportional to $v_{\theta\omega}B_{r\omega}$. Indeed, with increasing gas pressure, the discharge ionization and the energy balance require a smaller rf electric field, $E_{\theta\omega} \sim \omega B_{r\omega}$, and a smaller drift velocity, $v_{\theta\omega}$ to induce the same electric field (while with increasing frequency a smaller magnetic rf field is required).

Measurements performed at different frequencies and over a wide range of the discharge power have shown that the second harmonic voltage generated in the skin layer and defined as $V_{2\omega} = V_{2\omega}(z = 0) - V_{2\omega}(z > \delta_{2\omega})$ depends very little on the discharge power (plasma density). Thus, at 1 mTorr and a driving frequency 0.9 MHz, the change in the discharge power from 50 to 400 W results in a change of $V_{2\omega}$ from 5 to 8 V. The insensitivity of the nonlinear effects to the rf power is due to a fundamental property of a weakly-ionized gas discharge plasma in a steady state. In such a medium, the discharge maintenance field and the drift velocity (in our case $E_{\theta\omega}$, $B_{r\omega}$ and $v_{\theta\omega}$) are held almost constant by the ionization and electron energy balance. The shrinkage of the skin depth with discharge power (which is very small in our experiment due to the very low frequency) just slightly increases the electromagnetic fields and drift velocity in the skin layer.

The rf driving frequency in our experiment is much lower than the ion plasma frequency ω_{pi}, but it is still much higher than the inverse ion transition time across the skin layer $v_i/\delta_{2\omega}$, ($v_i/\delta_{2\omega} \ll \omega \ll \omega_{pi}$). Therefore, both, ions and electrons are practically immobile in the second harmonic field. Thus, the second harmonic field is the rf component of the ambipolar field that preserves the plasma quasi-neutrality by limiting the electron displacement under the action of the time-varying Lorentz force [5].

One can interpret the second harmonic voltage found here as the rf Hall effect (caused by the $E \times B$ rf fields in the skin layer) and evaluate it by integration of the rf component of the Lorentz force along the z-direction. Note that the rf component of the Lorentz force does not depend on the phase shift between $B_{r\omega}$ and $v_{\theta\omega}$:

$$V_{2\omega} = \int_{z}^{L} 2^{-1/2} B_{r\omega} v_{\theta\omega}\, \mathrm{d}z. \quad (1)$$

Having measured the plasma density $n(z)$ and the azimuthal current density $J_{\theta\omega}$ [14] in this experiment we could find $v_{\theta\omega}$ and calculate $V_{2\omega}$. The values of $V_{2\omega\, theor}$ found are in reasonable agreement with the experimental values of $V_{2\omega\, ex}$ (see table 1).

There is also a dc component of the Lorentz force (referred to as the ponderomotive force) with a magnitude that is comparable to that of the second harmonic. In contrast to the second harmonic component, the ponderomotive force may modify the plasma density and the dc ambipolar potential in the skin layer [1–5]. To the best of our knowledge, such ponderomotive effects have not yet been demonstrated by experiment in weakly-ionized gas discharge plasmas. In this work we described the first observation of a nonlinear polarization field induced by the rf Lorenz force in a low-frequency ICP. We believe that the search for ponderomotive effects (which are closely related to the SHP) should be focused on low-frequency ICPs where the rf magnetic fields are large.

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