Instabilities in low-pressure inductive discharges with attaching gases

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Plasma instabilities at frequencies 1 Hz–900 kHz have been observed in low-pressure inductive processing discharges with attaching gases. Instability windows in pressure and driving power are found. A volume-averaged (global) model of the instability is developed, considering idealized inductive and capacitive energy deposition. As pressure or power are varied to cross a threshold, the instability is born at a Hopf bifurcation, with relaxation oscillations between inductive and capacitive modes causing modulations of charged particle densities, electron temperature, and plasma potential. The oscillations can be so strong that the potential collapses and negative ions flow to the walls. © 1999 American Institute of Physics. [S0003-6951(99)03849-8]

Although instabilities in nonmagnetized, attaching gas plasmas at high pressures (∼1 Torr) have been well studied in dc glows 1–3 and capacitive discharges, 4 there has been only one study reported 5 for low-pressure (∼20 mTorr) inductive discharges of the type increasingly used in plasma-assisted microfabrication technology. Planar or cylindrical coils in a low aspect ratio (length/diameter) discharge are generally used. In the planar configuration used in our experiments, a flat helical coil is wound from near the axis to the center of the discharge and as the optical emission decays, with peak relaxation current modulations of 50%.

In a previous experiment, 5 instabilities were found with a cylindrical coil using O₂ and Ar/SF₆ (85% Ar, 15% SF₆) gas feeds. The source was a 12-turn copper coil 30 cm in diam powered by a 0.46 MHz rf generator operated between 200 and 500 W. With pressures in the range 0.25–10 mTorr, instabilities were observed with frequencies in the range of 1–40 kHz. Typically, with increasing pressure the instabiliy grows and then decays, with peak relaxation current modulations of an unbiased Langmuir probe of 50%.

In preliminary experiments, using the planar coil configuration described above, we have also observed oscillations in O₂ and in Ar/SF₆ mixtures. The instabilities were observed on the current to an unbiased Langmuir probe in the center of the discharge and as the optical emission detected by a photomultiplier tube viewing a midplane diameter. The instabilities appeared within power and pressure windows (e.g., 320–600 W at 5.8 mTorr; 4–8 mTorr at 360 W in SF₆) and exhibited a wider range of frequencies (1 Hz–900 kHz) than in the experiments with the cylindrical coil. Relaxation oscillations were sometimes observed having essentially 100% modulation; i.e., the electrons were almost completely expelled during the oscillation. Figure 1 gives one example of an oscillation near 1 kHz at 540 W in a 5.8 mTorr SF₆ discharge. The optical emission versus time shown in Fig. 1(a) consists of a series of sharp pulses. The corresponding frequency spectrum of the probe current shown in Fig. 1(b) indicates the strongly anharmonic nature of the oscillations. Similar phenomena are seen in Ar/SF₆ mixtures and in O₂ discharges.

We have developed a model for the instability for a volume-averaged (global) cylindrical discharge (radius R, length l). We consider electrons (density ne, temperature Te), negative ions (density n−, temperature Te) and positive ions (density n+, temperature T i). We take Te = constant and T i ≥ T j; temperatures are given in equivalent voltage units. The densities are assumed to be uniform within the bulk plasma, dropping sharply at the plasma-sheath edge near the end and circumferential walls. The particle balance equations are:

\[ \frac{dn_e}{dt} = K_{ei} n_e n_g - K_{a_i} n_e n_g - \Gamma_e A_{eff}/V, \]

\[ \frac{dn_i}{dt} = K_{a_i} n_e n_g - K_{te} n_e n_g - \Gamma_{-i} A_{eff}/V, \]

![Figure 1](image-url)
where $K_{iz} = K_{iz0}e^{-\eta_{iz}/T_e}$ and $K_{att} = K_{att0}e^{-\eta_{att}/T_e}$ are Arrhenius forms for the ionization and attachment rate constants, $n_e$ is the neutral gas density, $\Gamma_e$ and $\Gamma_-$ are the electron and negative ion fluxes to the walls, and $V = \pi R^2 l$ is the plasma volume. The effective loss area is

$$A_{\text{eff}} = 2\pi R^2 h_I + 2\pi R h_R,$$

where the ratios $h_I$ and $h_R$ of edge-to-center densities are taken from low-pressure diffusion theory to be

$$h_I = 0.86/(3 + l/2\lambda_i)^{1/2}, \quad h_R = 0.8/(4 + R/l\lambda_i)^{1/2},$$

with $\lambda_i$ the ion-neutral mean-free path. The fluxes are functions of the plasma potential $\Phi$ (with respect to the walls) through the Boltzmann relations

$$\Gamma_e = \frac{1}{2}n_e\varphi e^{-\Phi/T_e}, \quad \Gamma_- = \frac{1}{2}n_-\varphi e^{-\Phi/T_0},$$

where $\varphi = (eT_e/\pi m)^{1/2}$ and $\varphi_- = (eT_0/\pi M_-)^{1/2}$ are the electron and negative ion mean speeds. A simplified model is used for the positive ion flux appropriate to a low-pressure diffusion equilibrium,

$$\Gamma_+ = n_u u_B + \frac{1}{2}\varphi_- \Gamma_-,$$

where $u_B = (eT_e/M_+)^{1/2}$ is the Bohm (sound) velocity. The conditions of quasi-neutrality in the bulk plasma, $n_+ = n_e + n_-$, and at the walls, $\Gamma_+ = \Gamma_e + \Gamma_-$, are assumed.

The energy balance equation is

$$\frac{d}{dt} \left( \frac{3}{2} n_e T_e \right) = P_{\text{abs}} - P_{\text{loss}},$$

where

$$P_{\text{loss}} = K_{iz} n_e n_0 \eta_{iz} + K_{att} n_e n_0 \eta_{att} + \Gamma_e (\Phi + 2T_e) A_{\text{eff}} / V + \Gamma_- \Phi A_{\text{eff}} / V$$

is the energy-loss per unit volume. The surface terms in Eq. (8), proportional to $\Phi$, give the energy losses for positive ions that fall across the sheath potential, and the term proportional to $2T_e$ gives the electron kinetic energy carried to the walls; the negative ion flux to the walls yields a negligible energy loss because $T_- < T_e$.

In Eq. (7), $P_{\text{abs}} = P_{\text{ind}} + P_{\text{cap}}$ is the power absorbed per unit volume. Inductive power is transferred to electrons within a skin layer of thickness $\delta$ near the coil–plasma surface. For a fixed coil current $I_{\text{df}}$, the power transfer depends on $n_e$ and is low at low densities, where the weakly conducting plasma loop acts as an open circuit, and is low at high densities, where the highly conducting loop acts as a short circuit.

$$P_{\text{ind}} = I_{\text{df}}^2 R_{\text{ind}} \frac{n_e n_0}{n_e + n_0} \frac{1}{V},$$

where $R_{\text{ind}}$ depends on the power deposition volume, and $n_0$ gives the density for maximum power; i.e., where $\delta \sim V/A_{\text{eff}}$.

Electron heating is also produced by capacitive coupling of the coil voltage across the dielectric window to the plasma. The voltage is divided across the window and sheath capacitances. To include the capacitively coupled power, we use the form

$$P_{\text{cap}} = \frac{I_{\text{rf}}^2 R_{\text{cap}}}{2} \frac{n_e}{n_e + n_0} \frac{1}{V},$$

where $I_{\text{rf}}^2 R_{\text{cap}}$ gives the capacitive power per volume coupled to the plasma at low densities ($n_0 \ll n_e$) and $n_e$ estimates the density at which the sheath thickness is comparable to the window thickness. For this study, we arbitrarily vary the parameters $R_{\text{cap}}$ and $n_e$ to vary, respectively, the ratio of peak capacitive to peak inductive power, and the ratio of the plasma density at which the capacitive power peaks to the density at which the inductive power peaks.

Differential Eqs. (1), (2), and (7) can be integrated, together with the subsidiary conditions, to produce the dynamical behavior. The basic dynamics leading to relaxation oscillation behavior are: (1) in the inductive mode the electron density builds up in the discharge reaching a quasi-equilibrium before the negative ion density has fully built up; (2) the negative ion density continues to build up, leading to loss of the quasi-equilibrium; (3) electrons are lost rapidly and the discharge decays to a capacitive state; and (4) negative ion density decays slowly until the discharge can reestablish the inductive mode. In Fig. 2, results of the time integrations are given for $n_e$, $n_-$ in the inductive mode, and $T_e$ and $\Gamma_-$ for a typical case with SF$_6$ parameters and a ratio of peak capacitive/peak inductive power $2R_{\text{cap}}/R_{\text{ind}}$ of approximately 2%. The density oscillations have similar frequencies to the optical emission oscillation in Fig. 1. The electron modulation in Fig. 2 is so deep that the potential collapses and negative ions escape to the walls. Langmuir probe measurements have also been made, for somewhat different parameters than in Fig. 1, for both electron and ion saturation, giving shapes similar to those for $n_e$ and $n_-$ in Fig. 2. Detailed comparisons have not yet been made.

To understand the physics, we obtain a reduced set of equations by noting that, typically, there are three time scales. The fastest time scale is for changes in power (7), the next fastest is for changes in electron density (1), and the slowest is for changes in negative ion density (2). Using this ordering, we first set $d(n_e T_e)/dt = 0$ to eliminate the highly temperature-sensitive term $K_{iz}$ in Eq. (7) to obtain
Substituting Eq. (11) into Eq. (1), we find
\[
\frac{dn_e}{dt} = P_{\text{abs}} - K_{\text{att}} n_e \varepsilon_{iz} - K_{\text{att}} n_e \varepsilon_{att} - [\Gamma_e (\Phi + 2T_e) + \Gamma_{-} \Phi] A_{\text{eff}} / V.
\]
(11)

Setting \( \frac{dn_e}{dt} = 0 \) in Eq. (12) yields an equation for \( n_-(n_e) \), where \( \Gamma_e \) and \( \Gamma_{-} \) are substituted from Eq. (5), and \( T_e \) and \( \Phi \) are obtained from Eq. (11) and flux conservation, using Eqs. (5) and (6). The results (dashed lines) in the \( n_- \) vs \( n_e \) phase plane are shown for two different cases in Fig. 3; \( \frac{dn_-}{dn_e} = \frac{dn_e}{dn_-} \) is positive “below” these lines. The abrupt increase in the quasiequilibrium \( n_- \) density from Eq. (12) at a small value of \( n_e \) is due to the collapse of the plasma potential, where the loss of electrons can no longer match the positive ion loss, so that negative ions must also escape to preserve the charge equilibrium. In the same figure we also plot (dot-dashed lines) the equation for \( n_-(n_e) \) obtained by setting \( \frac{dn_-}{dt} = 0 \) from Eq. (2); \( \frac{dn_-}{dn_e} \) is positive “below” these lines. The crossing of the two \( n_-(n_e) \) curves obtained from Eqs. (12) and (2) gives the equilibrium. Examining a small departure from the equilibrium will easily convince the reader that the equilibrium is unstable if both slopes \( \frac{dn_-}{dn_e} \) are positive at the crossing, with the slope from Eq. (2) steeper than the slope from Eq. (12). All other crossings are stable. We have chosen the coil current \( I_t \) in Fig. 3 to illustrate (a) the instability onset shortly after the Hopf bifurcation\(^{11} \) (4% capacitive/inductive power ratio) and (b) a strong oscillation between inductive and capacitive modes with collapse of the potential (2% capacitive/inductive power ratio). Superimposed on these figures are the actual trajectories in the phase plane, for which the complete time-dependent equations are solved with no ordering. In Fig. 3(a) the oscillations are weakly modulated. In Fig. 3(b), corresponding to the time variations in Fig. 2, fully developed relaxation oscillations are seen similar to those observed in our planar coil experiment.

According to our model, below 5 mTorr in SF\(_6\) the potential no longer collapses as \( n_e \) decays. In this case, a relaxation oscillation can still occur between a low \( n_e \) near-extinguished state and a high \( n_e \) inductive state; reignition of the low \( n_e \) state occurs when \( n_- \) decays to a sufficiently low value that electropositive ignition occurs.

In conclusion, we have shown that low-pressure inductive discharges with attaching gases are subject to instabilities if the parameters lie within a certain range. Such relaxation oscillations have been observed both with a cylindrical coil using 0.46 MHz driving power and with a planar coil using 13.56 MHz driving power in our experiments.

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