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Undriven periodic plasma oscillation in electron cyclotron resonance Ar plasma

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We report experimental observation of periodic oscillation in a steady state electron cyclotron resonance argon plasma that is not driven by extra periodic forces. We interpret the oscillation according to the predator-prey model, which is a nonlinear plasma-neutral coupling in the plasma production region. The oscillation is observed in a narrow plasma parameter window and is evidence for neutral density depletion in a high density plasma system. © *1996 American Institute of Physics.* [S0003-6951(96)02440-0]

In recent years, there has been a growing interest in the investigation and quantitative characterization of determistic chaos in plasma. A plasma is a typical nonlinear dynamic system with a large number of degrees of freedom. The chaotic behavior in a DC discharge plasma is always associated with the occurrence of hysteresis and negative differential resistance in the current-voltage characteristics of the DC discharge.[1] Chaotic behavior in a high density plasma system, such an electron cyclotron resonance (ECR) plasma has not been reported, but the intense ionization of the system results in depleted gas density.[2,3] The plasma condition is often unstable because of the nonlinear coupling among neutral density, wave propagation, power absorption, and charge density profile.[4] Therefore, it is very important to study the neutral depletion and chaotic phenomena in the high density plasma system.

The purpose of this letter is to describe a theoretical and experimental investigation of self-driven plasma parameter oscillation in an Ar ECR plasma. Theoretically, we study the plasma and neutral density oscillation that is self-driven by neutral density depletion through nonlinear plasma-neutral coupling in the plasma production region. Experimentally, the floating potential measurement and the plasma emission measurement method are used.

Experiments are performed by using an ECR reactor (see Fig. 1).[5] This apparatus has three parts: a microwave power source and transmission section (circular TE_{11} mode), a plasma discharge vessel (quartz bell jar) 15 cm in diameter and a stainless steel reaction chamber 32 cm in diameter, and magnetic coils for static magnetic configuration (divergent type). A gas ring 15 cm in diameter is placed at the extraction window located midpoint of the plasma discharge vessel and the reaction chamber. A stainless steel substrate holder 14 cm in diameter without a wafer is placed at the reaction chamber. The distance between the substrate holder and the extraction window is 5 cm. The floating potential of a Langmuir probe and the plasma emission are monitored by using a digital oscilloscope and a photon-multiplier tube. The electron temperature and electron density are obtained through analysis of the Langmuir probe current-voltage curve.

The continuity equations of ions and neutrals in the plasma discharge vessel can be approximately written as follows:

$$\frac{\partial n_n}{\partial t}+\nabla\cdot\Gamma_n=-hn_nn_i, \tag{1}$$

$$\frac{\partial n_i}{\partial t}+\nabla\cdot\Gamma_i=hn_nn_i, \tag{2}$$

where $\Gamma_n=-D_n\nabla n_n$ is the neutral diffusion flux, $\Gamma_i=-D_a\nabla n_i$ the ion diffusion flux, h the ionization rate coefficient, n_n the neutral density, and n_i the plasma density. D_n and D_a are the neutral and ambipolar diffusion coefficients, respectively. The ionization rate coefficient h is given by[6]

$$h=2.5\times10^{-8}\sqrt{T_e}e^{-\frac{E_o}{T_e}}, \tag{3}$$

where T_e is the electron temperature, and E_o is the ionization threshold energy. If we replace the neutral and plasma density gradients with the characteristic system length L, then the continuity equations are written as

$$\frac{dn_n}{dt}-\frac{D_n}{L^2}n_n=-hn_in_n, \tag{4}$$

$$\frac{dn_i}{dt}+\frac{D_a}{L^2}n_i=hn_in_n. \tag{5}$$

We rewrite the plasma and neutral continuity equations as

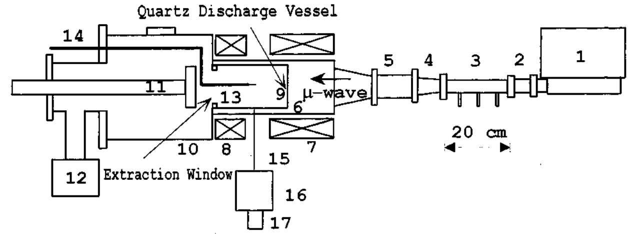

FIG. 1. A schematic diagram of an electron cyclotron resonance plasma machine. 1: magnetron, 2: directional coupler, 3: three-stub tuner, 4, 5, 6: waveguide, 7, 8: magnetic coil, 9: plasma discharge vessel (quartz bell jar), 10: stainless steel reaction chamber, 11: substrate holder, 12: turbomolecular pump, 13: gas ring, 14: Langmuir probe, 15: optical fiber, 16: monochrometer, 17: photo-multiplier (PM) tube. The pressure gauge is a capacitance manometer and is located at the reaction chamber.

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TABLE I. The data for calculation of plasma oscillation and critical point.

Gas	Ar
Mass (amu)	40
Ionization threshold energy (eV)	15.8
Operating gas pressure p (mTorr)	1.2
Measured electron temperature (eV)	6
Measured plasma density n_i (10^{11} cm^{-3})	2.2
Ionization rate constant h for T_e=6 eV (10^{-9} cm^{-3})	4.3
Ion temperature (eV)	0.3
Gas temperature (eV)	0.025
Neutral density n_n(10^{13} cm^{-3})	3.3p
Characteristic length L (cm)	10
Neutral diffusion coefficient D_n(10^5 cm^2/sec)	$7.8\sqrt{T_n}/p$
Ambipolar diffusion coefficient D_a(10^6 cm^2/sec)	$1.3\frac{T_e}{\sqrt{T_i}p}$
Predicted critical plasma density n_i^c(10^{11} cm^{-3})	2.4
Predicted critical neutral density n_n^c(10^{13} cm^{-3})	2.9
Predicted oscillation frequency (kHz)	1.7
Measured oscillating frequency (kHz)	1.1

$$\frac{dH}{dt}=aH-\alpha HP=H(a-\alpha P), \tag{6}$$

$$\frac{dP}{dt}=-cP+\gamma HP=P(-c+\gamma H), \tag{7}$$

where $H=n_n$, $P=n_i$, $a=D_n/L^2,\alpha=h,c=D_a/L^2$, and $\gamma=h$. The above equations are well-known as the predator-prey (P–H) (Lotka-Volterra[7]) equations. P and H are the populations of predator and prey in ecology (electron and neutral in plasma), respectively; a is the birthrate of prey H; c is the deathrate of predator P; and α and γ are interaction rate coefficients of predator-prey, respectively. The critical points of the Lotka-Volterra equations are

$$H(a-\alpha P)=0, \quad P(-c+\gamma H)=0. \tag{8}$$

The solutions are

$$H=0, \quad P=0,$$

and

$$H=c/\gamma, \quad P=a/\alpha. \tag{9}$$

The critical point ($H=0$, $P=0$) is an unstable saddle point. To study the critical point ($H=c/\gamma,P=a/\alpha$), we use the linear perturbation theory. The solutions are then given by

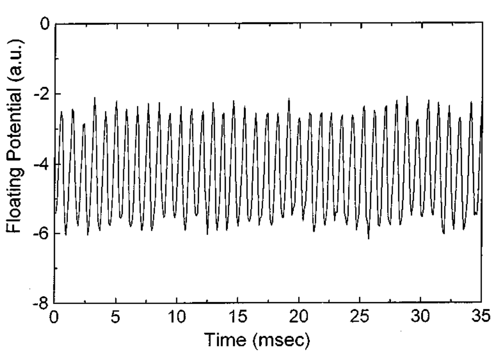

FIG. 2. An oscillating floating potential for 1.2 mTorr of Ar pressure and 200 W of microwave input power. The oscillation frequency is 1.1 kHz.

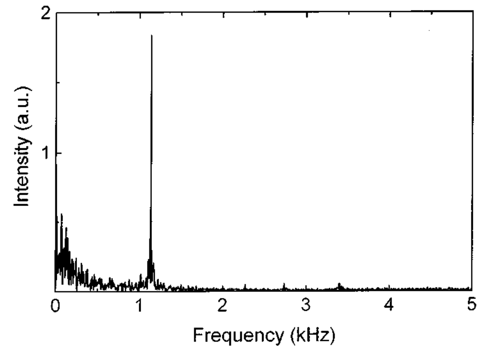

FIG. 3. The Fourier transformed frequency spectrum of the oscillating plasma emission for Fig. 2 conditions. The observed spectral line is 763.5 nm.

$$H(t)=\frac{c}{\gamma}+\frac{c}{\gamma}K\cos(\sqrt{\mathrm{act}}), \tag{10}$$

$$P(t)=\frac{a}{\alpha}+\frac{a}{\alpha}\sqrt{\frac{c}{a}}K\sin(\sqrt{\mathrm{act}}+\phi), \tag{11}$$

where the constants K and ϕ are determined by the initial conditions. These equations are valid for the elliptical trajectories close to the critical point ($H=c/\gamma,P=a/\alpha$). The oscillation frequency is independent of the initial conditions, and the amplitude of the oscillation depends on the initial conditions as well as on the parameters of the plasma.

Table I shows the predicted critical point and the oscillation frequency of the Ar discharge for the specific plasma parameters. The neutral diffusion coefficient D_n is given by $kT_n/m\nu_{nn}$,[8] where T_n is the neutral temperature, m the neutral mass, and ν_{nn} the neutral–neutral collision frequency.[8] The ambipolar diffusion coefficient D_a is given by $D_a\approx D_{||i}T_e/T_i$,[8] where $D_{\|i}$ is the diffusion coefficient along the magnetic field, T_i the ion temperature, and T_e the electron temperature. $D_{\|i}$ is given by $kT_i/m\nu_{in}$, where ν_{in} is the ion-neutral collision frequency.[8] The critical point ($H_c=c/\gamma$, $P_c=a/\alpha$) is given by ($D_a/(L^2h)$, $D_n/(L^2h)$). The critical neutral density n_n^c (H_c) and the plasma density n_e^c (P_c) for Ar 1.2 mTorr, 6.5 eV of T_e, 0.3 eV of T_i,[9] and 0.025 eV of T_n are 2.4×10^{11} cm^{-3} and 2.9×10^{13} cm^{-3}, respectively. The predicted oscillation frequency for the above conditions is given by $\sqrt{D_aD_n}/L^2$ (1.7 kHz).

Figure 2 shows the floating potential oscillation at 1.2 mTorr and 200 W power. The observed frequency is 1.1 kHz. Figure 3 shows the oscillating plasma emission Fourier transformation frequency spectrum at 1.2 mTorr and 200 W power. The plasma density oscillation is nearly in phase with the plasma emission oscillation. If we assume that the electron temperature does not vary in time, the plasma emission intensity (I) is given by

$$\begin{aligned}I&=An_in_n\\&=A\frac{ca}{\gamma\alpha}\Bigg[1+\sqrt{\frac{c}{a}}K\sin(\sqrt{\mathrm{act}}+\phi)+K\cos(\sqrt{\mathrm{act}}+\phi)\\&\quad+\sqrt{\frac{c}{a}}K^2\sin(\sqrt{\mathrm{act}}+\phi)\cos(\sqrt{\mathrm{act}}+\phi)\Bigg],\end{aligned} \tag{12}$$

where A is a constant. Since the second term is $\sqrt{c/a}$ (≈ 10) times larger than the third term, the plasma emission is in phase with the plasma density. We obtain K(0.025) from the oscillation amplitude of the negative biased Langmuir probe.

The hollow radial emission profile was reported by Rhoades and Gorbatkin in an ECR Ar plasma, and it was interpreted as the neutral depletion.[10] Hardrich *et al.* measured the hollow neutral density profile by using anti-stokes Raman spectroscopy method.[3] The plasma parameter window for the periodic oscillation is very narrow because the periodic oscillation is only valid near the critical point in $P-H$ phase space. This oscillation is also evidence for the neutral density depletion.

In conclusion, we observe the oscillating floating potential not driven by extra periodic forces in the Ar ECR discharge. We interpret the oscillation according to the predator–prey model. The predicted oscillation frequency and critical point are in accord with the experimental results. This oscillation gives evidence for the neutral depletion of the high density plasma source.

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