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Comparison of pressure dependence of electron energy distributions in oxygen capacitively and inductively coupled plasmas

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Electron energy distribution functions (EEDFs) were measured with increasing gas pressure in oxygen capacitively and inductively coupled plasmas. It was found that, in the capacitive discharge, abnormally low-energy electrons became highly populated and the EEDF evolved to a more distinct bi-Maxwellian distribution as the gas pressure was increased. This pressure dependence of the EEDF in the oxygen capacitive discharge is contrary to argon capacitively coupled plasma, where—at high gas pressure—low-energy electrons are significantly reduced due to collisional heating and the EEDF evolves to the Maxwellian. The highly populated low-energy electrons at high gas pressure, which was not observed in inductively coupled oxygen plasma, show that collisional heating is very inefficient in terms of the oxygen capacitive discharge. It appears that this inefficient collisional heating seems to be attributed to a low electric field strength at the center of the oxygen capacitive plasma.

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I. INTRODUCTION

Oxygen is a simple diatomic gas. Owing to this relative simplicity of oxygen in comparison with other electronegative processing gases and to numerous databases of reaction rate constants [1,2], much research has been studied on the characteristics of oxygen plasma itself or on the characteristics of argon-oxygen mixture plasma both by theoretical [2–12] and experimental [12–19] methods. Oxygen plasmas have been used in numerous applications in plasma processing such as photoresist ashing, chemical vapor deposition, and oxidation. There have been much experimental investigations on the oxygen plasma in an inductive discharge [12,14–19]. Tuszewski reported on an instability of an electronegative inductive discharge [17], and Barnes *et al.* and Schwabedissen *et al.* measured the electron energy distribution function (EEDF) and the electron density in an oxygen inductive discharge [18,19]. Gudmundsson *et al.* also measured EEDF as a function of the gas pressure and the radial position in an inductively coupled oxygen discharge [12]. However, there have not been as many experimental investigations of oxygen plasmas in a capacitive discharge as studies in an inductive discharge, although the plasma characteristics of oxygen plasmas in a capacitive discharge are possibly different from those in an inductive discharge.

It is well known mainly by investigations in argon plasma that, at low gas pressure, electron heating is mainly done by collisionless heating and the EEDF often has a bi-Maxwellian distribution [20]. Low-energy electrons of bi-Maxwellian distribution are produced by ionizations provided from energetic electrons. These low-energy electrons are confined in dc ambipolar potential and are unable to gain energy from an oscillating sheath. However, as the gas pressure increases, collisional heating becomes an important energy-transfer mechanism, and the low-energy electrons can gain energy through momentum-transfer collisions. Due to this collisional heating of low-energy electrons at high gas pressure, the EEDF evolves from a bi-Maxwellian distribution to a Maxwellian distribution as the gas pressure increases. This is a generally well-known scenario of the evolution of the EEDF with a change in the gas pressure. However, in our experiments, we observed that, in a capacitively coupled oxygen plasma, more low-energy electrons were populated and the EEDF became more distinct bi-Maxwellian as the gas pressure was increased. This result is contrary to argon capacitive plasma where the EEDF becomes Maxwellized with increasing the gas pressure. In sequential experiments, the EEDFs in oxygen inductively coupled plasma showed a Maxwellian distribution at wide range of gas pressures, even though the nearly same electron densities with the capacitive oxygen discharge. Therefore, the highly populated low-energy electrons at a high-pressure oxygen discharge are a characteristic of a capacitive discharge. The measured results in our experiments seem to indicate that the collisional heating in an oxygen capacitive discharge is very inefficient even at high gas pressures. The following experiments show that this inefficient collisional heating seems to originate from a weak electric field at the center of the oxygen capacitive discharge.

II. EXPERIMENTAL SETUP

Figure 1 shows a schematic of the experimental setup. The measurements were taken in a cylindrical discharge reactor driven by oxygen gas. The dimensions of the reactor were an inner diameter (D) of 26 cm and a length (l) of 19 cm. The substrate had a radius of 8 cm and its thickness was 1 cm. The side and bottom walls of the reactor were made of electrically grounded stainless steel and the top wall was made of aluminum oxide, which is nonconducting material. An rf power at 12.5 MHz was delivered to the substrate through a matching network. Along with the substrate, a two-

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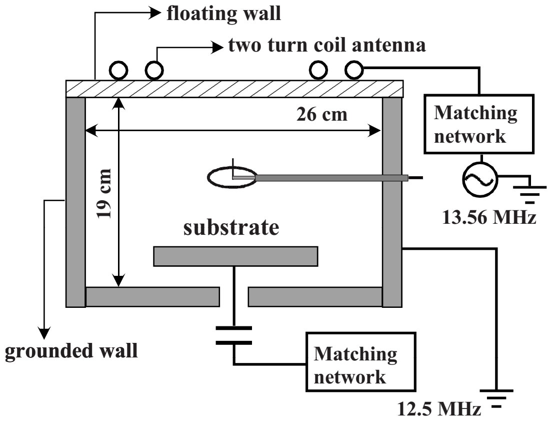

FIG. 1. Schematic of the experimental setup.

turn copper coil antenna for inductive coupling was placed on the top wall, and an rf power source at 13.56 MHz was delivered to the coil antenna.

An rf-compensated single Langmuir probe made from tungsten wire, 10 mm in length and 0.1 mm in diameter, was placed at the reactor, and all measurements were performed at the center of the discharge. The probe system contained a floating loop reference probe and 12.5 MHz resonance filters to reduce the rf distortion of the *I*-*V* characteristics. The second derivative of the probe current with respect to the probe potential (I_e''), which is proportional to the EEDF $g_e(\epsilon)$, was obtained by numerical differentiation and smoothing [21]. The measured second derivative I_e'' relates to the EEDF as

$$g_e(\epsilon) = \frac{2m}{e^2 A}\left(\frac{2\epsilon}{m}\right)^{1/2} I_e''(\epsilon), \tag{1}$$

where ϵ, e, m, and A are the electron energy, the electron charge, the electron mass, and the probe area, respectively. The electron density n_e and the electron temperature T_e were calculated from the EEDF $g_e(\epsilon)$ [22].

III. RESULTS AND DISCUSSION

The evolution of EEDFs at the center of the reactor with changing gas pressure in oxygen [Fig. 2(a)] and argon [Fig. 2(b)] capacitively coupled plasmas is presented in Fig. 2, in terms of electron energy probability functions (EEPFs), $g_p(\epsilon)$. The substrate of the reactor was only powered and a power was not delivered to the coil antenna in these experiments. The EEPF $g_p(\epsilon)$ is related to the EEDF $g_e(\epsilon)$ as follows:

$$g_p(\epsilon) = \epsilon^{-1/2} g_e(\epsilon). \tag{2}$$

The measurements were performed by fixing discharge conditions to 120 W of an rf power and 15 SCCM of the gas flow rate. In Fig. 2 the EEPFs of both the oxygen and argon plasmas had a bi-Maxwellian distribution at 3 mTorr of gas pressure. However, the changes in the EEPFs with an increase in the gas pressure were very different from each other. In the case of argon plasma, as the gas pressure was increased, the low-energy electrons heated up and the distribution evolved to being Druyvesteyn-like through a Maxwellian distribution. The bi-Maxwellian distribution at low gas pressure is, as is well known, a characteristic of the nonlocal kinetics [20,23]. The low-energy electrons generally originate from the ionization provided from the energetic electrons in the discharge. In the oxygen plasma, the production of low-energy electrons is enhanced by inelastic collisions such as rotational and vibrational excitations, dissociation of oxygen molecule, detachment reactions, and electronic excitation [6]. There are many more inelastic reactions in an oxygen plasma when compared with an argon plasma, and therefore electrons in an oxygen plasma are likely to lose their energy and to fall into the low-energy group. These low-energy electrons are confined within the plasma bulk due to the dc ambipolar potential and unable to reach an oscillating sheath where collisionless heating occurs. The momentum-transfer collision frequency of low-energy electrons is very small, resulting in a little gain of energy for the low-energy electrons from either collisional or collisionless heating. However, as the gas pressure is increased, the low-energy electrons are able to collide with neutral species more frequently gaining energy through collisional heating. As a result, these electrons can overcome the dc ambipolar potential and participate in the collisionless heating at the oscillating sheath. In such a situation, the EEPF evolves from a bi-Maxwellian to a Maxwellian distribution as shown in Fig. 2(b). The increase in the electron density with the gas pressure promotes electron-electron collisions, which also results in the Maxwellian EEPF.

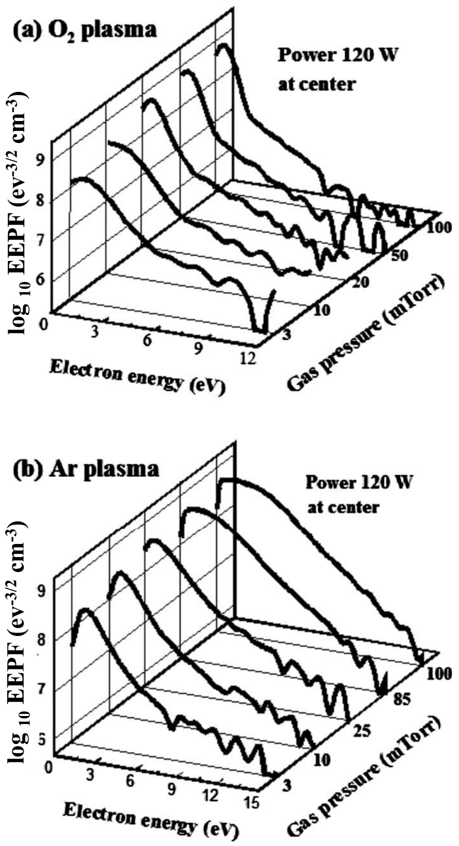

FIG. 2. An evolution of the EEPFs with the gas pressure in (a) oxygen and (b) argon plasmas in capacitive discharge.

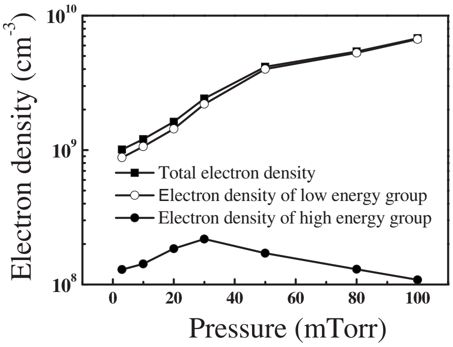

FIG. 3. Changes in total (solid square), low-energy group (open circle), and high-energy group (solid circle) electron densities with the gas pressure measured in capacitively coupled oxygen plasmas.

However, as seen in Fig. 2(a), the heating of the low-energy electrons at high gas pressure was not observed in oxygen capacitively coupled plasma. Contrary to the argon plasma, low-energy electrons were more populated and the EEPFs evolved to a more distinct bi-Maxwellian distribution as the gas pressure was increased. Figure 3 shows clearly this increase in the density of low-energy electrons (open circle) with the gas pressure in the oxygen capacitive discharge. The distribution function of the bi-Maxwellian takes a following form when the potential at the plasma center is set to zero:

$$f_e(\epsilon)=B[\alpha\exp(-\epsilon/T_1)+(1-\alpha)\exp(-\epsilon/T_2)]. \tag{3}$$

Here, B is a normalization constant; T_1 and T_2 are the low and high electron temperatures in eV, respectively; $\alpha(=n_1/n_0)$ is the population ratio of the low-temperature group electrons to total electrons; and n_0 and n_1 are the total and low-temperature group electron densities, respectively. Figure 4 shows the change in an electron temperature of the low-energy group, T_1, and α with the gas pressure measured in the oxygen and argon capacitively coupled plasmas. In argon plasma, the population ratio of the low-energy group electrons, α, was continuously reduced and its temperature (T_1) was increased with the gas pressure due to the enhancement of the collisional heating and electron-electron collisions. α at 3 mTorr was 0.84 and was reduced to 0.62 at 50 mTorr. The EEPF of the argon plasma was almost turned into a Maxwellian distribution above 50 mTorr; therefore, α is plotted up to 50 mTorr in Fig. 4. However, in the oxygen plasma, the dependence of T_1 and α were on the contrary. The temperature of low-energy group decreased continuously while their population ratio increased with the gas pressure. T_1 and α at 3 mTorr were 0.71 eV and 0.87, respectively, and they changed to 0.21 eV and 0.98 at 100 mTorr gas pressure. The heating of the low-energy electrons was hardly observed even up to 100 mTorr gas pressure, and the EEPF became more distinctly bi-Maxwellian as seen in Fig. 2(a). This occurred in spite of an increase in the electron density. These results indicate that collisional heating is very inefficient in oxygen capacitively coupled plasma compared to argon plasma.

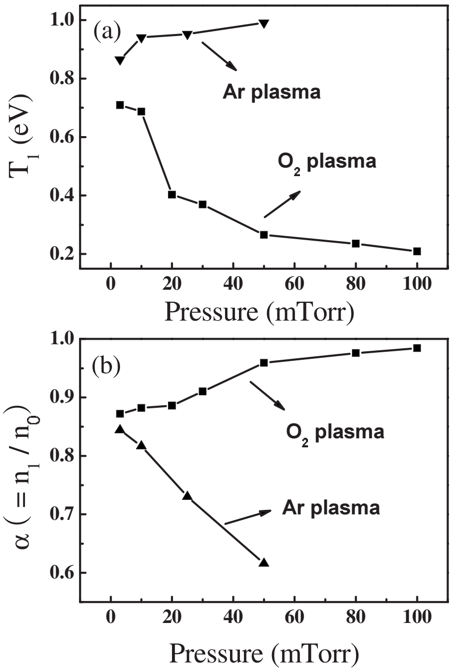

FIG. 4. Changes in (a) low-energy group temperature (T_1) and (b) density proportion of energy electrons to the total electron density (α) with gas pressure in capacitively coupled oxygen plasmas.

Despite that there are a few reports on EEPFs in an oxygen inductive discharge [12,18,19], there have not been as many experimental studies of the EEPF evolution in a capacitive discharge. The pressure dependence of the EEPF in an oxygen inductive discharge shows no low-energy peak and has a Maxwellian distribution for wide rage of the gas pressures [18], which is unlike the capacitive discharge. This Maxwellian EEPF in inductive oxygen discharge may be attributed to the relatively higher electron density or a penetration of inductive fields to the plasma core. Simulated EEPFs by Lee *et al.* in an oxygen capacitive discharge (Fig. 4(c) in Ref. [5]) also show the low-energy peak and two temperature EEPFs like our results. However, in their simulations, the population of low-energy electrons was not elevated with the gas pressure, which is contrary to our experimental results.

The power transferred to electrons per unit volume by the collisional heating is given in following equation [22]:

$$p_c=\frac{1}{2}|E|^2\sigma_{dc}\frac{\nu_m^2}{\omega_{rf}^2+\nu_m^2}. \tag{4}$$

Here, E is an electric field strength, $\sigma_{dc}=e^2n_e/m\nu_m$ is the dc plasma conductivity, ν_m is the momentum-transfer collision frequency, and ω_{rf} is the angular frequency of the rf power. Equation (4) shows that, for a given electric field strength, the transferred power by the collisional heating becomes its maximum when the momentum-transfer collision frequency coincides with the rf power frequency, $\nu_m=\omega_{rf}$. Cross sections for the momentum-transfer collisions, calculated ν_m, and ω_{rf} for argon and oxygen plasmas at 10 and 100 mTorr gas pressures are presented in Fig. 5 [24,25]. For low-energy electrons ($0.1<U<1.0$ eV) and at a fixed rf power frequency (12.5 MHz in our experiments), the momentum-transfer cross section in oxygen plasma is much higher than the cross section in argon plasma as shown in Fig. 5. The electrons with their energies of $0.1<U<1.0$ eV correspond

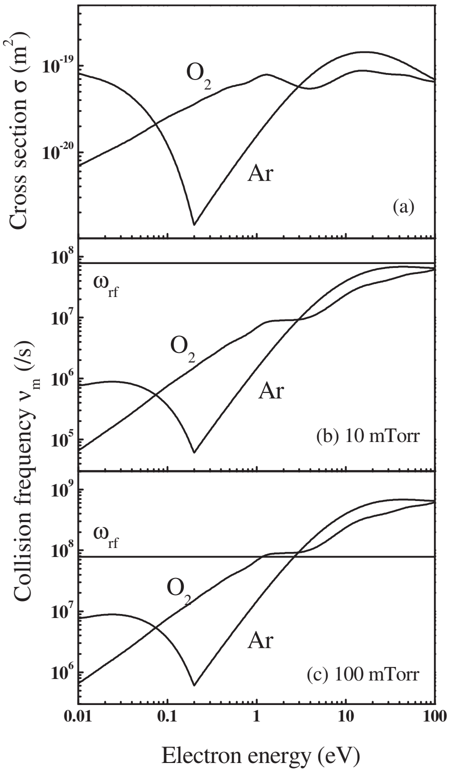

FIG. 5. (a) Cross sections for momentum-transfer collisions of argon atom [24] and O$_2$ molecule [25] and calculated ν_m at (b) 10 and (c) 100 mTorr gas pressures; $\omega_{rf}/2\pi = 12.5$ MHz.

to the low-energy group electrons in argon and oxygen plasmas, as shown in Fig. 4(a). The higher cross section for low-energy electrons in oxygen plasma, which is mainly due to the Ramsauer minimum of the argon gas, results in the higher momentum-transfer collision frequency than in argon plasma, as shown in Fig. 5. Therefore, Figs. 2 and 5 show that the collisional heating of the low-energy electrons in the oxygen capacitive discharge is very inefficient when compared with argon discharge even though the collision cross section of the momentum-transfer collisions in oxygen plasma is larger.

One of the possible reasons for the inefficient collisional heating in the oxygen capacitive discharge would be a weak electric field strength at the plasma core. There are two experimental results showing that the weak electric field is responsible for the observed inefficient heating of low-energy electrons at the core of the oxygen capacitive discharge. One of them is EEDF measured in an inductively coupled oxygen plasma. Figure 6 shows EEPFs measured at the center of the reactor with changes in the gas pressure in an oxygen inductive discharge. The experiment was performed in the same reactor as in Fig. 1 with only coil antenna being powered for inductive coupling. The inductive power was set to 120 W and the gas flow rate was 15 SCCM. Notice that the low-energy peak, which was observed and became obvious at high gas pressure in the capacitively coupled discharge, was not observed in the inductive discharge even at 100 mTorr gas pressure. The measured EEPFs have a single Maxwellian in the pressure region of $3 \leq p \leq 100$ mTorr except for the depletion of the high-energy region at high gas pressure (100 mTorr), which was due to enhanced inelastic collisions.

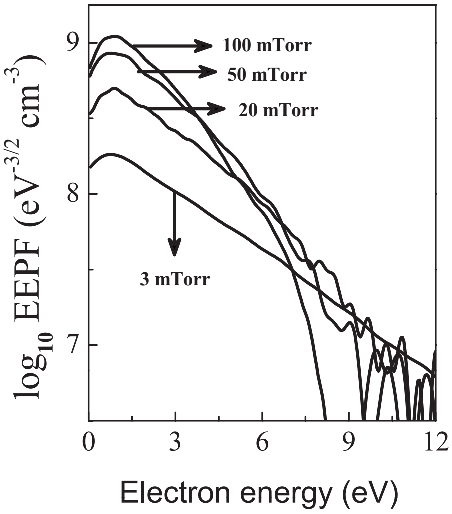

FIG. 6. Measured EEPFs with changing the gas pressure in inductively coupled oxygen plasma. The inductive power was 120 W and a gas flow rate was 15 SCCM.

The Maxwellian distribution in an oxygen inductive discharge, which has also been reported in previous experiments [12,16], indicates that, unlike a capacitive discharge, the low-energy electrons produced by inelastic collisions or by ionization from high-energy electrons were effectively heated up in an inductive discharge. Enhanced electron-electron collisions also can result in the Maxwellian distribution. However, as seen in Fig. 7, the electron densities in the inductive discharge were nearly the same as the densities in the capacitive discharge (Fig. 3). This indicates that the Maxwellian distribution in the inductive discharge was not due to the electron-electron collisions. Instead, inductive fields that penetrate into the reactor would be responsible for the Maxwellian EEPF in the inductive discharge. Figure 7 shows the electron densities and corresponding skin depths with respect to the gas pressure measured in the oxygen inductive discharge. The collisionless skin depth δ_p is given as

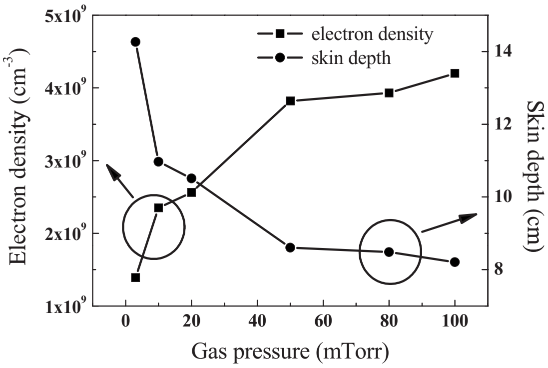

FIG. 7. Changes in an electron density and a skin depth with a gas pressure measured in oxygen inductive plasma. The inductive power was 120 W.

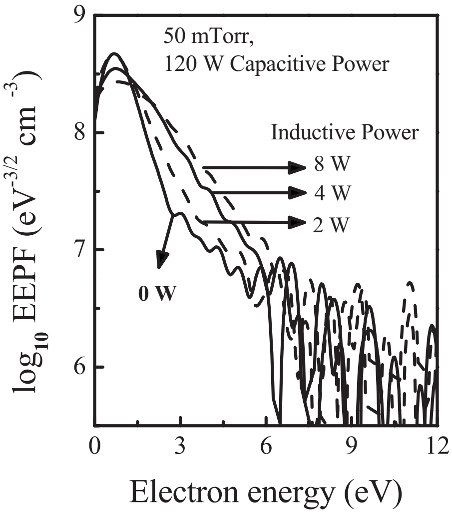

FIG. 8. An evolution of the EEPFs when adding a small inductive power to the capacitively coupled oxygen plasmas. The pressure was 50 mTorr, capacitive power was 120, and flow rate was 15 SCCM.

$$\delta_p = \left(\frac{m}{e^2 \mu_0 n_e} \right), \tag{5}$$

where μ_0 is the permeability of the vacuum. The skin depths in our experiment were larger than the half-length $(l/2)$ of our reactor, and therefore the inductive fields are able to penetrate into the center of the reactor. In such a situation, low-energy electrons in the discharge center are able to be heated up by the fields, and they are not confined in an ambipolar potential. Therefore, the low-energy peak is not observed in an oxygen inductive discharge. The results show that EEPFs of an inductively coupled oxygen discharge, where the inductive fields can penetrate into the center of the reactor, are Maxwellian, while EEPFs measured at the center of the oxygen capacitive discharge have a bi-Maxwellian distribution, even though the electron densities of the two (inductive and capacitive) discharges are nearly the same. This may show that a weak electric field strength at the discharge center could be a possible reason for the inefficient collisional heating of the low-energy electrons in the capacitively coupled oxygen plasmas.

Stronger experimental evidence showing that the inefficient collisional heating of the low-energy electrons at the center of an oxygen capacitive discharge seems to originate from the weak field strength there is presented in Fig. 8. In this experiment, the discharge was mainly sustained by capacitive power (120 W) delivered through the substrate, and we added a very small inductive power through the coil antenna. The measurements were performed at the discharge center and under the conditions of 50 mTorr oxygen gas pressure and 15 SCCM gas flow rate. When the inductive power was not supplied and the discharge was solely sustained by the capacitive power, the EEPF had a bi-Maxwellian distribution like in Fig. 2(a). The electron temperatures of low- and high-energy groups were $T_1 = 0.29$ eV and $T_2 = 2.03$ eV, respectively, and α was 0.93. However, when a very small amount of inductive power was added, low-energy electrons were significantly reduced and the EEPF evolved toward the Maxwellian distribution. As shown in Fig. 8, the distribution almost turned into Maxwellian at even 4 W of inductive power. In this experiment, the electron density was low enough that the skin depth was much larger than the half-length of the discharge. When 4 W of the inductive power was applied, the electron density was 1.03×10^9 cm^{-3} and the skin depth was 16.5 cm. Therefore, the inductive fields are able to penetrate into the discharge center and to heat up the low-energy electrons there. The inductive fields may not be sufficiently strong enough to heat up the low-energy electrons to become high-energy electrons because of the low inductive power. However, the fields supply the energy for low-energy electrons to overcome the dc ambipolar potential. As a result, these low-energy electrons are able to access an oscillating sheath where the collisionless heating takes place and are able to gain sufficient energy at the sheath to become high-energy electrons. The fact that low-energy electrons in the capacitive oxygen discharge are heated up effectively and the EEPF changes to a Maxwellian by supplying very small (inductive) electric fields may show that the highly populated low-energy electrons at the center of a high-pressure capacitive oxygen discharge are due to the weak electric fields there, resulting in insufficient collisional heating at the center of the capacitive oxygen discharge.

IV. CONCLUSIONS

In this study, the evolution of EEDFs with increasing gas pressure in a capacitively coupled oxygen plasma was measured. The result was compared with the EEDFs of an argon capacitive plasma and of oxygen inductive plasma. At low gas pressures, the EEDFs both in the oxygen and argon capacitively coupled plasmas had a same electron distribution of the bi-Maxwellian EEDF. However, the pressure dependences of the EEDFs of two gases were quite different. At the oxygen capacitive discharge, as the pressure was increased, more low-energy electrons were populated at the discharge center and the EEDFs evolved to be more distinctly bi-Maxwellian. This is contrary to the pressure dependence of an argon capacitive discharge, where low-energy electrons were reduced and the EEDF changed to Maxwellian as the gas pressure was increased. These results seem to indicate that the collisional heating of the low-energy electrons in the oxygen capacitive discharge is very inefficient when compared to the argon discharge. This inefficient collisional heating is not due to a small electron-neutral collisional frequency, because the momentum-transfer collision frequency of low-energy electrons with an oxygen molecule is much larger than the collision frequency with an argon atom.

The highly populated low-energy electrons at high-pressure oxygen plasma are a characteristic of a capacitive discharge and were not observed in an inductive discharge. In an inductively coupled plasma, the EEDFs had a Maxwellian distribution for wide range of gas pressures (3–100 mTorr), and low-energy peak was not observed even though the electron densities were nearly the same with the densities for the capacitive discharge. This Maxwellian EEDF in the inductive discharge seems to be attributed to a penetration of inductive fields into the center of the reactor owing to the relatively large skin depth. In a following experiment, it was found that when a very small inductive power (a few W) was added to a high-pressure oxygen capacitive discharge, low-energy electrons were effectively heated up and the EEDF evolved to the Maxwellian. This result shows that the relatively inefficient collisional heating of the capacitively coupled oxygen plasma seems to originate from the weak electric field strength at the discharge center.

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