Three point method to characterize low-pressure electronegative discharges using electrostatic probe

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Electrostatic probe measurements for low-pressure inductively coupled SF_6 plasmas are performed. From the current-voltage (*I-V*) curves of probe, the saturation currents of the positive ions and electrons and the electron temperature are measured. The electronegativity and the negative ion density are deduced by using the ratios of these parameters at three adjacent pressure points. The positive ion density is calculated by the orbital-motion-limited theory, and the electron temperatures are given either by the slope of the *I-V* curves or by the electron energy distribution function with the second derivative of *I-V* curves. The variations in the charged species density with pressure and power are investigated. © *2009 American Institute of Physics.* [DOI: 10.1063/1.3065089]

I. INTRODUCTION

Electronegative gases, such as SF_6, oxygen, chlorine, and fluorocarbons, are used extensively in discharges for various applications of plasma processing. The presence of negative ions modifies the transport properties of the discharge. Negative-ion sources can be applied to charge-free ion implantation in semiconductor manufacture, and negative-ion assisted etching is found to reduce the charging of substrates.[1]

There has been considerable scientific and technological interest in electronegative plasmas[2–5] and there have been various methods in the determination of negative ion density by using lasers and electrostatic probes.[6–9] Electrostatic probes,[10–13] laser photodetachment (LPD),[6,7] and laser Thomson scattering (LTS)[14] are diagnostic tools for investigating negative ions in plasma, among which electrostatic probe technique is simple and inexpensive and provides the spatial resolution of plasma parameters.

In previous works,[15–17] we estimated the positive ion density for inductively coupled oxygen discharges directly from the positive ion saturation current assuming that the positive ions have the normal Bohm velocity. The interpretation of current-voltage (*I-V*) characteristics of the electrostatic probe has been performed by different methods. If there is no negative (charged) ions and no magnetic field, electron density can be obtained either by measuring the electron saturation current or by integrating electron energy distribution function (EEDF) integrals, but it is better to use the ion saturation current with quasineutrality. However, a recent study demonstrates that if a right theory is used, measurements of electron and ion density from electron and ion saturation current agree.[18] If plasma is magnetized with respect to the electric probe (i.e., probe size is larger than the ion gyroradius), it would be better to use the ion saturation current density. If there are negative ions without magnetic field, it is proper to use the positive and negative ions for the electron density, if both ion densities are known. However, unless both the density and the temperature of negative ion are very large compared with the density and temperature of electron, electron density could be calculated from the electron saturation current approximately due to huge mass ratio of negative ions to electrons,

$$N_e \approx \frac{4I_{se}}{eA_p(8kT_e/\pi m_e)^{0.5}}, \tag{1}$$

where I_{se} is the electron saturation current, e is the electron charge, A_p is the probe area, m_e is mass of electrons, k is the Boltzmann constant, and T_e is the electron temperature. This would hold if

$$\zeta \equiv I_{se}/I_{sm} = (0.25A_p eN_e v_e)/(0.6AeN_m v_i) = 0.4(A_p/A) \times (N_e/N_m)(M_-/m_e)^{0.5} \gg 1, \tag{2}$$

where I_{sm} is the saturation current of negative ions, N_m is the negative ion density, A is the sheath area for negative ion collection and M_- is the mass of negative ion. Here negative ions are assumed to be collected by the same way as the positive ions, i.e., they are passing the sheath with Bohm speed. If negative ions are to be governed by the Boltzmann relation as electrons, ζ becomes $(N_e/N_m)(M_-/m_e)^{0.5}$, which is much larger than unity for most cases. As for the electron temperature, either the logarithmic slope of *I-V* curve or EEDF can be used by assuming the Boltzmann electrons or isotropic electron distribution.

In order to deduce the density of negative ions, practical methods to compare the ratios of ion and electron saturation currents were introduced by Amemiya,[19] Cooney *et al.*,[20] and Shindo *et al.*[21] Chabert and co-workers[22,23] introduced a two-probe method to deduce the ratio of negative ion density to electron density. However, these methods need many other information: temperatures of positive and negative ions, sheath potential, sheath area for positive ion collection, and effective mass of positive ions. The negative ion temperature can be known accurately from other methods.[14,24,25] In this work, a simple probe method to determine the electronegativity is introduced and tested for inductively coupled SF_6 plasmas.

II. THEORY

The EEDF integral method has been used to obtain plasma parameters for many processing plasmas utilizing molecular gases.[26,27] The effective electron temperature can be calculated with the measured EEDF as follows:

$$T_{\text{eff}} = \frac{2}{3kn_e}\int_0^{\epsilon_{\max}} \epsilon f(\epsilon)d\epsilon, \tag{3}$$

where $\epsilon_{\max}$ is determined by the dynamic range of the EEDF measurement, and the EEDF $f(\epsilon)$ is given as

$$f(\epsilon) = \frac{2m_e}{e^2A_p}\left(\frac{2eV}{m_e}\right)^{1/2}\frac{d^2I}{dV^2}, \tag{4}$$

where V is the probe potential referenced to the plasma potential (space potential) and electron energy ϵ is measured in units of eV. The electron temperature can also be determined from the slope of logarithmic I-V curve in the exponential region when corrected for positive ion current. We observe that the both methods yield almost same values of the electron temperature, indicating one-temperature electron and confirming the Boltzmann electrons for our experimental conditions.

As for the positive and negative ion densities, one can modify the method of Shindo *et al.*,[21] as Chung *et al.*[28] did for the planar probe, or for the cylindrical and planar probes.[9] The positive ion saturation current in electronegative plasma at pressures p_1, p_2, and p_3, which is adjacent to each other, is described as

$$I_{s+}(X_j) = eA_jN_+(X_j)n_s(X_j)v_s(X_j)\left[\frac{kT_e(X_j)}{M_+(X_j)}\right]^{1/2}, \tag{5}$$

where X_j and A_j $(j=1,2,3)$ are the three adjacent pressure points and sheath areas for the collection of ions at those points, N_+ is the positive ion density, M_+ is an effective mass of positive ions, and n_s and v_s are the normalized density and velocity of positive ions at the sheath edge, respectively, which can be given as,

$$n_s = (1-\alpha)e^{-\eta_s} + \alpha e^{-\gamma_- \eta_s}, \quad v_s = \sqrt{2\left(\frac{1}{\gamma_+}+\eta_s\right)}, \tag{6}$$

where $\alpha = N_m/N_+$ is the ratio of negative ions to positive ions, $\gamma_\pm = T_e/T_\pm$, $\eta_s = eV_{\text{sh}}/kT_e$. The temperatures of positive ions and negative ions are, T_+, and T_-, respectively, and V_{sh} is the sheath edge potential with respect to the plasma potential. From the quasineutrality, the following should be satisfied:

$$N_+(X_{1,2,3}) = N_m(X_{1,2,3}) + N_e(X_{1,2,3}). \tag{7}$$

The electron saturation current is given by

$$I_{es}(X_{1,2,3}) \propto N_e(X_{1,2,3})A_p\sqrt{kT_e(X_{1,2,3})/m_e}. \tag{8}$$

The combination of Eqs. (5) and (8) leads to the simpler form as

$$\frac{i_2}{\Omega_2} = \frac{N_{+2}}{N_{+1}}\sqrt{\frac{\tau_2}{\mu_2}}, \quad \varepsilon_2 = \frac{N_{e2}}{N_{e1}}\sqrt{\tau_2}, \quad \frac{i_3}{\Omega_3} = \frac{N_{+3}}{N_{+1}}\sqrt{\frac{\tau_3}{\mu_3}}, \quad \varepsilon_3 = \frac{N_{e3}}{N_{e1}}\sqrt{\tau_3}, \tag{9}$$

where the dimensionless parameters are defined as

$$i_2 = \frac{I_{+s2}}{I_{+s1}}, \quad \varepsilon_2 = \frac{I_{es2}}{I_{es1}}, \quad \tau_2 = \frac{T_{e2}}{T_{e1}}, \quad \mu_2 = \frac{M_{+2}}{M_{+1}},$$

$$\Omega_2 = \frac{A_2 n_{s2} v_{s2}}{A_1 n_{s1} v_{s1}},$$

$$i_3 = \frac{I_{+s3}}{I_{+s1}}, \quad \varepsilon_3 = \frac{I_{es3}}{I_{es1}}, \quad \tau_3 = \frac{T_{e3}}{T_{e1}}, \quad \mu_3 = \frac{M_{+3}}{M_{+1}},$$

$$\Omega_3 = \frac{A_3 n_{s3} v_{s3}}{A_1 n_{s1} v_{s1}}.$$

Another combination of Eqs. (7) and (9) results in the following simpler forms as

$$\frac{\sqrt{\mu_2}}{\Omega_2}i_2 = \varepsilon_2(1-\alpha_1) + \alpha_2\sqrt{\tau_2},$$

$$\frac{\sqrt{\mu_3}}{\Omega_3}i_3 = \varepsilon_3(1-\alpha_2) + \alpha_3\sqrt{\tau_3}. \tag{10}$$

If the constant of linearity η is defined as the following by assuming that the negative ion density is linearly dependent on the small change in pressures,

$$\eta \equiv \frac{p_3-p_1}{p_2-p_1} = \frac{N_{m3}-N_{m1}}{N_{m2}-N_{m1}} = \frac{\alpha_3-\alpha_1}{\alpha_2-\alpha_1}, \tag{11}$$

where

$$\alpha_1 = \frac{N_{m1}}{N_{+1}}, \quad \alpha_2 = \frac{N_{m2}}{N_{+1}}, \quad \alpha_3 = \frac{N_{m3}}{N_{+1}}. \tag{12}$$

This linearity was confirmed by some earlier works on the probe I-V measurements utilizing low-pressure electronegative gases.[17,21] Then electronegativity (the ratio of negative ion density to positive ion density) is obtained as

$$\alpha_1 = \left[1 - \frac{\sqrt{\mu_3}}{\Omega_3}\frac{i_3}{\varepsilon_3} + \eta\frac{\sqrt{\mu_2}}{\Omega_2}\frac{i_2}{\varepsilon_3}\sqrt{\frac{\tau_3}{\tau_2}} - \eta\frac{\varepsilon_2}{\varepsilon_3}\sqrt{\frac{\tau_3}{\tau_2}}\right]\Bigg/\left[1 - \frac{\sqrt{\tau_3}}{\varepsilon_3}\left(1-\eta+\eta\frac{\varepsilon_2}{\sqrt{\tau_2}}\right)\right],$$

$$\alpha_2 = \left(\alpha_1 + \frac{\sqrt{\mu_2}}{\Omega_2}\frac{i_2}{\varepsilon_2} - 1\right)\frac{\varepsilon_2}{\sqrt{\tau_2}},$$

$$\alpha_3 = \alpha_1(1-\eta) + \eta\alpha_2. \tag{13}$$

The values of i, ε, and τ can be given from measurement of three I-V curves at three adjacent pressures. Yet the ratios of sheath factors $(\Omega_{2,3})$ and ratios of reduced masses $(\mu_{2,3})$ are required for the deduction in α. It turned out that one can set $\Omega_{2,3}$ and $\mu_{2,3}$ as unity for the parameter region considered in this study. Once α is obtained, the negative ion density is calculated from α and N_e.

$$N_{m1} = \frac{\alpha_1}{(1-\alpha_1)} N_{e1}. \tag{14}$$

Using quasineutrality given by $N_e + N_m = N_+$, we can determine the positive ion density N_{+1}, then N_{m2}, N_{m3} are evaluated from Eq. (12). To determine the positive ion density from another way, the orbital motion-limited (OML) theory can be used for the low density plasmas with the thick sheath, and the ion positive current (I_i) is expressed as

$$I_i = N_+ e A_p \sqrt{\frac{kT_e}{2\pi M_+}} \frac{2}{\sqrt{\pi}} \left[\frac{e(V_p - V_{\mathrm{pr}})}{kT_e} \right]^{1/2}, \tag{15}$$

where V_{pr} is the probe voltage and V_p is the plasma potential. In the positive ion saturation region where the probe current is virtually all positive ion current, we can plot $I_i^2 = AV_{\mathrm{pr}}^2 + B$. From the slope, we can determine the positive ion density.

III. EXPERIMENT

In this study, an inductively coupled SF_6 rf plasma is employed as an example of electronegative discharge since SF_6 plasmas have found numerous applications in plasma processing such as dry etching of silicon. The plasma generation chamber consists of a stainless steel cylinder with a diameter 28 cm and a length of 30 cm. A 1.9 cm thick by 27 cm diameter tempered glass plate mounted on the one end separates the planar one-turn induction coil from the plasma. The induction coil is made of copper (with water cooling) and connected to an L-type capacitive matching network and a rf power generator. The details of the apparatus are found in Ref. 29.

The plasma chamber is evacuated by a diffusion pump, roughly pumped by a rotary pump, giving a base pressure of 9×10^{-6} Torr. The operating gas pressure is controlled by adjusting the mass flow controller. The SF_6 gas pressure is varied in the range of 0.4–1.0 mTorr. A 13.56 MHz generator with a power output of 100–500 W drives rf current in a flat one-turn coil through the rf power generator and matching network. A rf-compensated cylindrical electrostatic probe (SLP-2000, Plasmart) with a tungsten tip with diameter of 0.1 mm and length of 10 mm is used to measure the plasma parameters. The probe tip is located on the axis of the cylinder at 14 cm below the tempered glass plate.

IV. RESULTS AND DISCUSSION

Figure 1 shows the electron temperature obtained by two different methods as a function of pressure of SF_6: one is by the slope of *I-V* curves and the other is by EEDF. The electron temperature decreases with pressure at a low pressure range of 0.4–1 mTorr. The electron temperatures obtained by these two different methods seem to agree well with each other, which indicate that the electrons behave as isothermal with one temperature even with negative ions.

In these experiments, a quadrupole mass analyzer (QMS 200F2, Pfeiffer Vacuum) was mounted on the main chamber and mass spectrometry was performed. As shown in Fig. 2, the positive ions present in appreciable concentrations were SF_5^+ (127 amu), SF_4^+ (108 amu), SF_3^+ (89 amu), SF_2^+ (70 amu), SF^+ (51 amu), F^+ (19 amu), and S^+ (32 amu). The gas ionizer also produces three doubly ionized species SF^{++}, SF_2^{++}, and SF_4^{++}, which are seen at 25.5, 35, and 54 amu. The peaks at 18 and 28 are mostly due to H_2O^+ and CO_2^+ impurities, respectively. Small peaks of SiO^+, SO^+, SOF^+, and SiF_3^+ are due to the tempered glass etching and observed at 44, 48, 67, and 85 amu. All these peaks being considered, the effective mass of the positive ions was determined as 59.3 amu. The negative ion species have not been identified because it is difficult to attract them in a mass analyzer. Negative ions such as SF_6^-, SF_5^-, SF_3^-, and F^- have been observed in inductively coupled SF_6 discharges with similar operating conditions to those in this study.[30]

Figure 3(a) shows the negative ion density as a function of pressure obtained from the three pressure method (TPM). In a probe *I-V* curve for electronegative plasmas, it is usually difficult to determine the plasma potential (and thus electron saturation current). Therefore, instead of using I_{se} [Eq. (1)], the electron density was calculated by fitting the formula

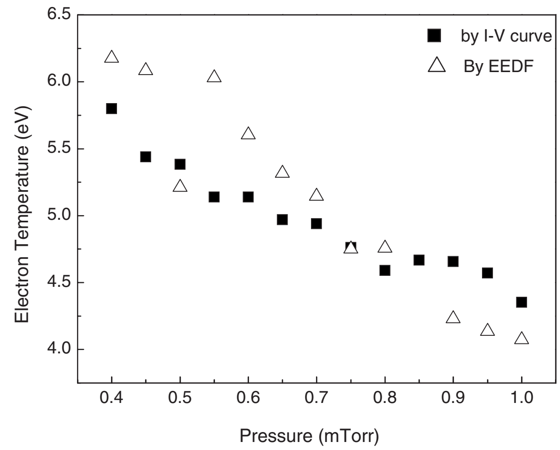

FIG. 1. Electron temperature as a function of pressure obtained by two different methods for $P=300$ W. The pressure range is from 0.4 to 1.0 mTorr. (All axes suppress zero point).

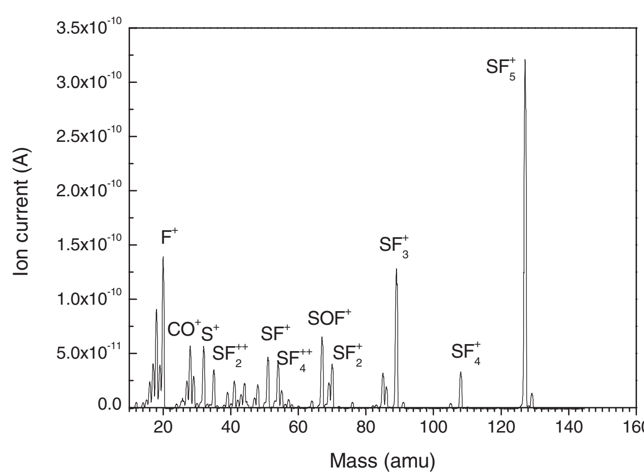

FIG. 2. Mass spectrum of SF_6 plasma at $P=300$ W and $p=1.0$ mTorr. (The *x*-axis suppresses zero point).

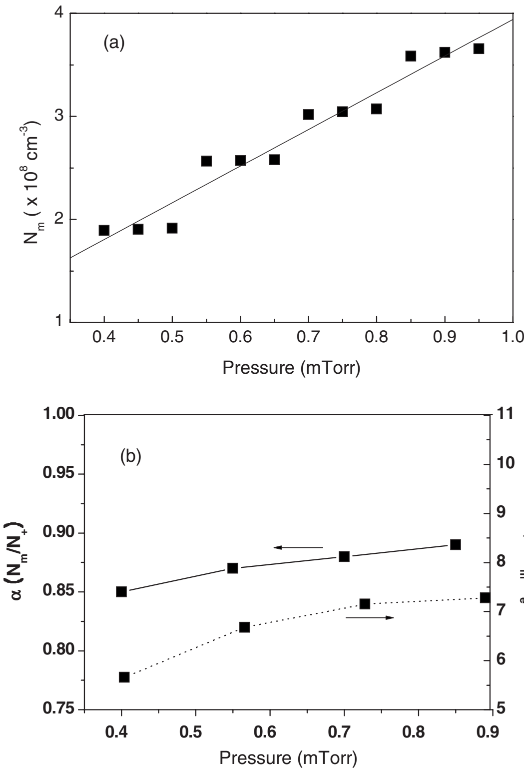

FIG. 3. (a) Negative ion density as a function of pressure at $P=300$ W. (b) Electronegativity ($\alpha \equiv N_m/N_+$) and the ratio of negative ion to electron density ($\beta \equiv N_m/N_e$) as a function of pressure at $P=300$ W. The pressure range is from 0.4 mTorr to 1.0 mTorr. (All axes suppress zero point).

$$I_e = eA_pN_e\sqrt{\frac{kT_e}{2\pi m_e}}a\left[b+\frac{e(V_{\rm pr}-V_p)}{kT_e}\right]^c, \tag{16}$$

where a, b, and c are fitting constants. The result verifies the assumption that the negative ion density increases linearly with pressure. According to Eq. (13), the values of α_1, α_2, and α_3 vary at every pressure point. These values determine the slope at that point. This slope is meaningful only for three closely spaced pressure points. This does not have a direct relation to the slope of the overall variation in the negative ion density as a function of pressure (in a broader range). Physically, one of the three α contributes to the overall variation in the negative ion density with pressure. Figure 3(b) shows the electronegativity ($\alpha \equiv N_m/N_+$) and the ratio of negative ion to electron density ($\beta \equiv N_m/N_e$), which increase slightly with pressure. This behavior of β is qualitatively consistent with the experimental results for other electronegative discharges measured by Tuszewski[31] and Gudmundsson.[32]

Figure 4 shows the positive ion densities deduced both by the TPM (using $N_+=N_e+N_m$) and by the OML theory. Since the electron density is very low in the parameter region of this study, the Debye length (and the sheath thickness) is much larger than the probe radius. Therefore, the OML theory can be applicable in determining the positive ion density. The results obtained by these two different methods are overall compared well with each other at low pressures. Since the OML theory could have been widely used for the determination of positive ion density for low pressure discharges, the TPM suggested in this work can be used to determine the negative ion density. The difference between the values by TPM and those by OML may be due to the application of TPM for the planar probe to the cylindrical probe, although the relative ratios of sheath areas and effective masses are almost same for the small changes in adjacent pressures.

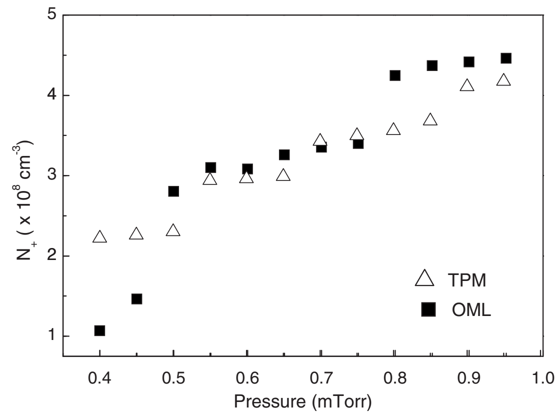

FIG. 4. Comparison of the positive ion density obtained by the TPM and the OML theory. The pressure range is from 0.4 to 1.0 mTorr. (All axes suppress zero point).

V. CONCLUSION

In conclusion, the electronegative inductively coupled SF_6 discharges have been studied based on electric probe measurements. The electron temperature is obtained based on two different methods, EEDF integral method and *I-V* curve graphic method. The positive ion density obtained by TPM and OML theory are overall compared well with each other at low pressures. This indicates that TPM can be used to determine the negative ion density, although detailed correction should be made by laser diagnostics such as LPD for N_m, LTS for N_e, and laser induced fluorescence (LIF) for N_+. From the deduced data of electron temperature and density, the ratio of the sheath thickness (or the Debye length) to the probe radius is much larger than the unity, which justifies the use of OML for the positive ion density.

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