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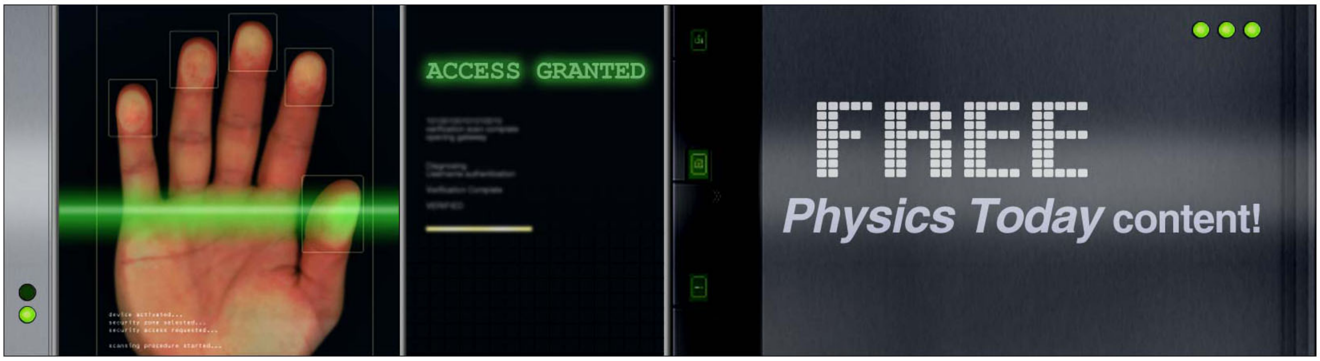

APPLIED PHYSICS LETTERS **102**, 134104 (2013)

# On anomalous temporal evolution of gas pressure in inductively coupled plasma

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The temporal measurement of gas pressure in inductive coupled plasma revealed that there is an interesting anomalous evolution of gas pressure in the early stage of plasma ignition and extinction: a sudden gas pressure change and its relaxation of which time scales are about a few seconds and a few tens of second, respectively, were observed after plasma ignition and extinction. This phenomenon can be understood as a combined result between the neutral heating effect induced by plasma and the pressure relaxation effect for new gas temperature. The temporal measurement of gas temperature by laser Rayleigh scattering and the time dependant calculations for the neutral heating and pressure relaxation are in good agreement with our experimental results. This result and physics behind are expected to provide a new operational perspective of the recent plasma processes of which time is very short, such as a plasma enhanced atomic layer deposition/etching, a soft etch for disposal of residual by-products on wafer, and light oxidation process in semiconductor manufacturing. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4798587]

The neutral gas pressure has been believed to be one of the most important of external plasma parameters in the plasma processing since the gas pressure influenced on key plasma elements such as a mean free path of electron-neutral related to the discharge kinetic property of local and non-local,[1–5] a residence time of gas and radicals related to the radical life time in the processes,[6,7] and an electron heating and its efficiency which is an inevitable element for plasma generation and sustaining.[8–10] Because of its importance, there have been lots of studies on the evolution of plasma characteristics depending on the gas pressure in the past decades.[11]

In low pressure glow discharge like an inductively coupled plasmas (ICP), normally, the gas pressure has been considered to be constant during the whole operation time including plasma ignition and extinction because the ionization fraction of the neutrals in normal glow discharge is very small (less than 1%) unless we considered for strong neutral depletion normally shown in almost fully ionized plasma like a Helicon source. In a past decade, however, as some papers reported the significant change of neutral gas density even in partially ionized plasmas like an ICP after plasma ignition, which can be explained in terms of a plasma pumping, a gas heating, and an electron-neutral pressure balance,[12–18] the change of gas pressure has been expected during the plasma operation. Most of these studies have focused on the neutral density depletion related to the gas pressure after a good while of plasma ignition meaning most of transient behavior of gas pressure was gone, but there have not been any studies for the temporal evolution of gas pressure during the whole plasma operation including plasma on and off, it has just been briefly mentioned in Ref. 19 without any experimental verification. However, recently the interest of the gas pressure in the plasma after a while of plasma ignition (a few seconds or less) has been greatly increased because of recent advent of plasma processes of which maximum time is less than 15 s, such as a soft etch for disposal of residual by-products on wafer and light oxidation process in semiconductor manufacturing. A study for the temporal behavior of gas pressure in the early stage of plasma after plasma ignition and its physical analysis is strongly needed.[20–22]

In this letter, we investigated the time resolved measurement of gas pressure in ICP. The result showed that there is an interesting anomalous evolution of gas pressure in the early stage of plasma ignition and extinction: a sudden gas pressure change and its relaxation of which time scales are about a few seconds and a few tens of second, respectively, were observed after plasma ignition and extinction. This phenomenon can be understood as a combined result between the neutral heating effect induced by plasma and the pressure relaxation effect for new gas temperature. The temporal measurement of gas temperature by laser Rayleigh scattering (LRS) and the time dependant calculations for the neutral heating and pressure relaxation are in good agreement with our experimental results.

Figure 1 is a schematic arrangement of the experimental apparatus where our experiment was performed. The cylindrical ICP chamber of which inner diameter and height are 400 mm is covered by a cylindrical ceramic window of which diameter and thickness are 350 mm and 15 mm, respectively. The discharge is ignited and sustained by 13.56 MHz rf current delivered by the RF power supply (100 W to 300 W) through a coaxial cable and L-type matching network (Path Finder, PLASMART Inc.). The background base pressure in the vacuum chamber is in the $1 \times 10^{-6}$ Torr range, and operating pressure in the range of $3 \times 10^{-1}$ Torr is kept during the discharge operation by flowing the Ar gas (20 SCCM–100 SCCM) through the discharge chamber.

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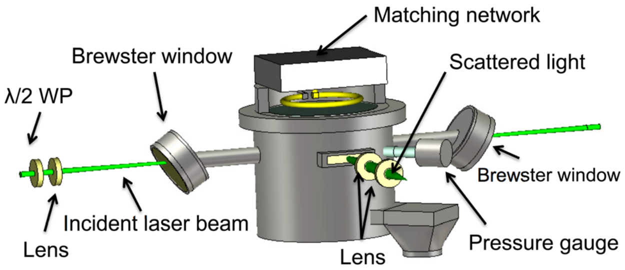

FIG. 1. A schematic arrangement of the experimental apparatus.

The temporal measurement of gas pressure was performed with the capacitor diaphragm gauge (MKS.INC, 627B, response time is about 20 ms) installed 10 cm below from the chamber cover and serial communicated with a computer. To remove the complexities originated from the response time of feedback control system between the pressure gauge and throttle valve, a manual mode of throttle valve which the open ratio of throttle valve is fixed was used instead of the auto mode. Although the fast time response of the pressure gauge, we can display the gas pressure in every 200 ms because of communication and process delays for our data acquisition system.

To convince that our result of temporal behavior of gas pressure is related to the neutral heating which is expected result in previous papers, the measurement of LRS was conducted simultaneously with the temporal gas pressure measurement. For the measurement of LRS, a frequency doubled Nd:YAG laser (Continuum Inc., Powerlite 9010) working at 10 Hz with a pulse energy of 230 mJ at 532 nm was used. The details for the alignment of optics and the data process are presented in the previous paper.[23] The temporal measurement of LRS light signal with the time resolution of 1 s was made by displaying 1 data point averaged over 10 laser shots, and its measurement was performed at the position of 10 cm below from the chamber cover (same height as that of pressure gauge). For theoretical investigation about our experimental result, the energy balance equations for neutrals and ions are solved simultaneously by coupling with the equations of relation between throughput and pressure difference[26] ($Q = d(PV)/dt$, $Q = C\Delta P$, where V and Q are a chamber volume and a net throughput on the chamber volume V, and C is the conductance of the system for neutral gas). The initial values used in the differential equations are given by the measurement of the single Langmuir probe (SLP2000, PLASMART Inc.) positioned at same position of laser focused point. The details for single Langmuir probe are available in previous reports.[24] The initial values of ions and neutral temperatures which cannot be measured by the probe are assumed to be room the temperature (300 K) for simplicity.

Figure 2 shows the result of time resolved measurement of the gas pressure (black line) and laser Rayleigh scattering signal (blue line) at the condition of 300 W and 300 mTorr during the whole discharge operation period including plasma discharge ignition and extinction. As shown in the Figure 2, there is a non-monotonic evolution of gas pressure in the early stage of plasma ignition and extinction: a sudden gas pressure change and its relaxation of which time scales are about a few seconds and a few tens of second, respectively, were observed after plasma ignition and extinction. This is quite interesting result because of the following reasons: in ICP discharge, normally, the gas pressure has been considered to be constant during the whole operation time including plasma ignition and extinction because the ionization fraction of the neutrals in normal glow discharge is very small (less than 1%). Although some papers reported the significant change of neutral gas density even in ICP discharge after plasma ignition and the increase of gas pressure induced by the neutral heating might be expected in the steady state plasma after plasma ignition,[12–18] such a transient result of gas pressure, a non-monotonic temporal evolution of the gas pressure, is the first observation in the low pressure discharge experiments. From Figure 2 (see black line), we can also convince that the temporal change of gas pressure, especially a sudden change of gas pressure when plasma turns on and off, is related to the neutral heating which is expected result in previous papers, because the decrease of laser Rayleigh scattering signal reflects the rarefied gas state which can be induced by gas heating as previous report denoted.

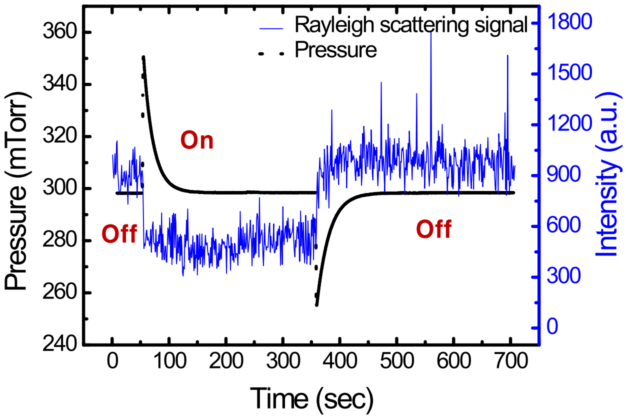

FIG. 2. Temporal measurement of the gas pressure (black line) and laser Rayleigh scattering signal (blue line) at 300 mTorr, 50 sccm, and 300 W (on: plasma turns on, off: plasma turns off).

This temporal evolution of the gas pressure can be understood as a combined result between the neutral heating effect induced by plasma and the pressure relaxation effect for new gas temperature as follows: Under the circumstance of the fixed inlet flow rate controlled by mass flow controller (MFC) and fixed outlet conductance controlled by throttle valve (manual mode), the action of plasma ignition can increase the (neutral) gas pressure with the increasing gas temperature ($P \propto T_n$) due to neutral heating.[12–18] After a characteristic time related to the neutral heating, the gas pressure can decrease (or relax) to a certain gas pressure which is a new equilibrium of the gas heated, because the outlet throughput also increases with the pressure increase (due to the relation $Q_{outlet} = C\Delta p = C(p - p_{out})$), where $p$ and $p_{out}$ are gas pressures in the front and back sides of throttle valve of conductance $C$, respectively. Because of these two processes, the gas pressure suddenly changed by the neutral heating starts to relax slowly approaching the new equilibrium condition of gas pressure as shown in Figure 2. The gas pressure for new equilibrium condition turns out to be the initial gas pressure which is the pressure before plasma turns on, because the (steady state) equilibrium can be made only when the throughput of inlet equals to that of outlet ($Q_{inlet} = Q_{outlet} = C(p - p_{out})$) and the gas pressure ($p$) to meet the condition is uniquely determined as an initial equilibrium pressure value before plasma turns on as shown in Figure 2 ($p_{out}$ which is the gas pressure in a backside of the throttle valve is normally small $p \gg p_{out}$, thus we set $p_{out} = 0$ for simplicity). Although these two processes (the sudden pressure change and pressure relaxation) take place at the same time, the sudden pressure increase is observed first just after plasma ignition and then the pressure relaxation is observed in a rapid succession as shown in Figure 2, because normally the difference between the two characteristic time is huge (a order of millisecond or second for neutral heating and its related processes, a order of ten second for the pressure relaxation, which will be shown later). When the plasma turns off, a similar event but opposite direction of pressure change can take place due to the same reason: action of plasma extinction can decrease the (neutral) gas pressure due to the loss of heating source of neutrals, the plasma. After a characteristic time related to the neutral cooling, the gas pressure can increase (or relax) to a certain gas pressure which is a new equilibrium of gas pressure, the initial gas pressure. As in neutral heating, normally, the time difference between the two characteristic time is huge, the sudden pressure decrease is observed first just after plasma ignition, and then the pressure relaxation is observed in a rapid succession as shown in Figure 2.

As previous studies reported,[12,25] this study also measured the decrease of LRS signal when the plasma is on, which reflects the rarefied gas density ($n$) induced by gas heating. Although both measurement results show the neutral heating, the detailed scenarios for our measurement result are different from the previous one as follows: as in the previous studies, if we consider the condition that the gas pressure reaches the steady-state after the plasma ignition in fixed volume, meaning $pV = p_0V_0 =$ constant, where the subscript 0 represents the initial state before plasma ignition, the total number of particles ($N$) or density ($n = N/V$) of neutrals is inversely proportional to the neutral temperature ($T_n$), $n \propto 1/T_n$ as in previous studies.[12,15–18] Therefore, the previous studies showed that the decrease of LRS signal means the rarefied gas density ($n$) induced by neutral gas heating. However, as in our experiment, if we consider the condition that the pressure does not reach the steady-state after the plasma ignition, meaning $pV \neq$ constant, the effect of neutral heating results in a complicated scenario, not only the LRS signal but also the gas pressure change drastically after plasma ignition and extinction as shown in the Figure 2. The condition for previous study may correspond to the gas pressure state after the gas pressure relaxation is made (see pressure after 150 s in Figure 2).

There is an interesting finding in this measurement of LRS signal (relaxation time of LRS signal): When the plasma is ignited, the gas pressure starts to rise with the neutral temperature because the total particle number ($N$) and the volume are not changed that much in the period. If the gas temperature reaches the steady state, the gas pressure starts to decrease because the total particle number ($N$) in the vacuum chamber is decreased by the throughput increase of throttle valve $Q_{outlet} = C\Delta p = C(p - p_{out})$. Because the LRS signal is to reflect the neutral gas density, the LRS signal is expected to reach the steady-state by following the gas pressure curve in Figure 2. However, the LRS signal reaches the steady-state so earlier than gas pressure. This problematic result can be solved by considering the situation that the plasma does not fill the whole volume of the vacuum chamber (this situation is frequently observed at intermediate or high pressure discharge where the electron relaxation length is shorter than the chamber length as in 300 mTorr in our vacuum chamber (the electron energy relaxation length: $\sim$ a few cm)):[31] when the plasma turns on occupying small volume in the chamber volume, the neutral gases in the space where the plasma is filled are heated and pressure in the region increases.

In order to confirm these arguments on the experiment, we divided the chamber into two part, part 1(subscript 1) is the plasma filled region and part 2(subscript 2) is non-plasma region for simplicity of calculations. Total pressure is expressed as follows:

$$P = \frac{V_1}{V_1 + V_2} P_1 + \frac{V_2}{V_1 + V_2} P_2 \tag{1}$$

$$= \frac{N_1}{V} k_B T_1 + \frac{N_2}{k_B} k_B T_2 \tag{2}$$

$$= \frac{N}{V} k_B \left( \frac{N_1 T_1 + N_2 T_2}{N} \right) \tag{3}$$

$$= \frac{N}{V} k_B T_{ave}, \tag{4}$$

where $P_1 = \frac{N_1}{V_1} k_B T_1$, $P_2 = \frac{N_2}{V_2} k_B T_2$ and $T_{ave}$ is averaged temperature of $T_1$ and $T_2$. We theoretically investigated the pressure evolution by considering the total differential equation of the gas pressure as follows:

$$\frac{dP}{dt} = \frac{\partial P}{\partial t} + \frac{\partial P}{\partial T_1}\frac{dT_1}{dt} + \frac{\partial P}{\partial T_2}\frac{dT_2}{dt} + \frac{\partial P}{\partial N}\frac{dN}{dt} + \frac{\partial P}{\partial V}\frac{dV}{dt}. \tag{5}$$

Eq. (5) shows that the gas pressure is changed by time (t), neutral gas temperature ($T_1$ and $T_2$), particle number ($N = N_1 + N_2$), and volume where plasma is filled in ($V = V_1 + V_2$). If we focus our interest on the pressure evolution depending on the neutral temperature and particle number and assume that the pressure is not changed explicitly with time and the volume of the plasma is not changed, the equation can be simplified with help of gas state equation

$$\frac{dP}{dt} \approx \frac{\partial P}{\partial T_1}\frac{dT_1}{dt} + \frac{\partial P}{\partial N}\frac{dN}{dt} \tag{6}$$

$$= \frac{N_1 k_B}{V_1}\frac{dT_1}{dt} + \frac{k_B T_{ave}}{V}\frac{dN}{dt}. \tag{7}$$

We assumed that $P_1$ and $P_2$ are independent of $T_2$ and $T_1$, respectively, and $T_2$ is isothermal set to 300 K for simplicity. After substituting a terms of particle number change by pumping loss into Eq. (7), $(\frac{dN}{dt}) = \frac{1}{k_B T_{ave}}$ ($Q_{in}$ $-c(p - p_{out})$), the equation can be reduced as follows (see supplementary material[34] for detailed derivation of each terms):

$$\frac{dP}{dt} = n_1 k_B \frac{dT_1}{dt} + \frac{1}{V}(Q_{in} - c(P - P_{out}), \qquad (8)$$

where $n_1 = N_1/V_1$ is number density of neutrals in the plasma region and we set $n_1 = 0.18 n_g$ by fitting with experimental results. We can know that $n_1$ is much small compared with $n_g$ because Rayleigh scattering decreases to level of stray light. For simplicity of the calculation of Eq. (8), we set $V = 0.137\ \mathrm{m}^{-3}$, $Q_{in} = 0.0338,\ 0.0845,\ 0.169\ \mathrm{Pam}^3/\mathrm{s}$, and $C = 0.0027,\ 0.0067,\ 0.013\ \mathrm{m}^3/\mathrm{s}$ for each flow rate 20, 50, 100 SCCM, respectively. The needed information of the gas temperature $(T, \frac{dT}{dt})$ to solve the equation are given by the neutral heating equation in Refs. 16, 27–30 (see supplementary material[34]). Now we can inspect the pressure evolution induced by both neutral heating and pressure relaxation via the equations (a differential form of a gas law including the neutral heating and throughput-pressure difference relation).

Figure 3 shows the calculation result of Eq. (8) in the same condition of Figure 2. As shown in Figures 2 and 3, the calculation result reproduces the anomalous temporal evolution of the gas pressure properly and the result is in a good agreement with experimental result, qualitatively and quantitatively. In the sense of extension for the application of our physical model, time resolved measurement of gas pressure at different conditions of gas flows and gas pressures from Figure 2 and the calculation at the same conditions were performed, and the results were presented in Figures 4(a) and 4(b), respectively. As shown in Figure 4, our model can reproduce the temporal evolution of gas pressure at various discharge conditions. For the characteristics time for each pressure sudden change (jump) and relaxation, the experiments showed that the rising time of the pressure jump is around a second whether the calculation showed that the rising time of the pressure jump is around a few ms. The reason for the discrepancy between calculation and experiment in pressure sudden change induced by neutral heating might be originated by overlooking the neutral heating time which has diffusion effect in the chamber between heating zone and non-heating zone in the calculation. One of the possible reason for the longer raising time compared with theoretical examination is that the plasma impedance matching used in our experiment has slow matching time 0.5 s. However, for the pressure relaxation, the calculation result is in a quit good agrees with experimental result quantitatively and qualitatively. Fortunately, because the overall characteristic of temporal behavior of gas pressure evolution depends on the process of which characteristic time is more longer, our physical modeling considering the both neutral heating and pressure relation can explain successfully the anomalous evolution of the gas pressure.

We also considered the effect of a pressure fluctuation induced by turbulence because Rayleigh intensity looks like a pressure fluctuation, somehow indicating the existence of the turbulence. However, the turbulence effect is considered to be negligible because probability density function (PDF) of Rayleigh intensity is almost Gaussian,[32] the Reynolds number, Re, for our experimental condition is below turbulent flow condition ($\mathrm{Re} > 2100$ for turbulent and our result is below 1000),[33] and, consequently, the fluctuation of Rayleigh intensity is from laser energy fluctuation and the fluctuation of weak scattering signals due to the small cross section of Rayleigh scattering for Argon.

In conclusion, the time resolved measurement of gas pressure in ICP showed that there is an interesting anomalous evolution of gas pressure in the early stage of plasma ignition and extinction. Based on the temporal measurement of gas temperature by LRS and the time dependant calculations for the neutral heating and pressure relaxation, we can conclude that this phenomenon is a combined result between the neutral heating

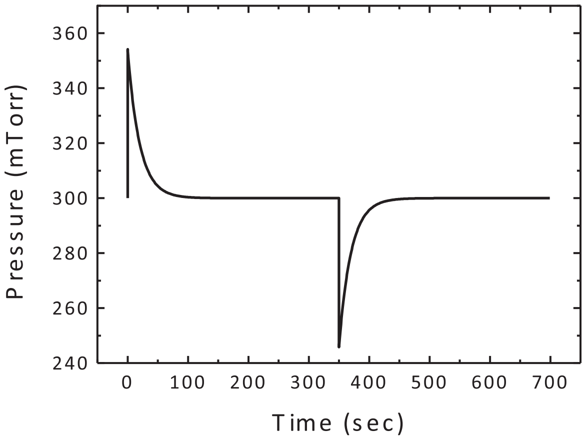

FIG. 3. A calculated result of Eq. (8) at the same condition of Fig. 2.

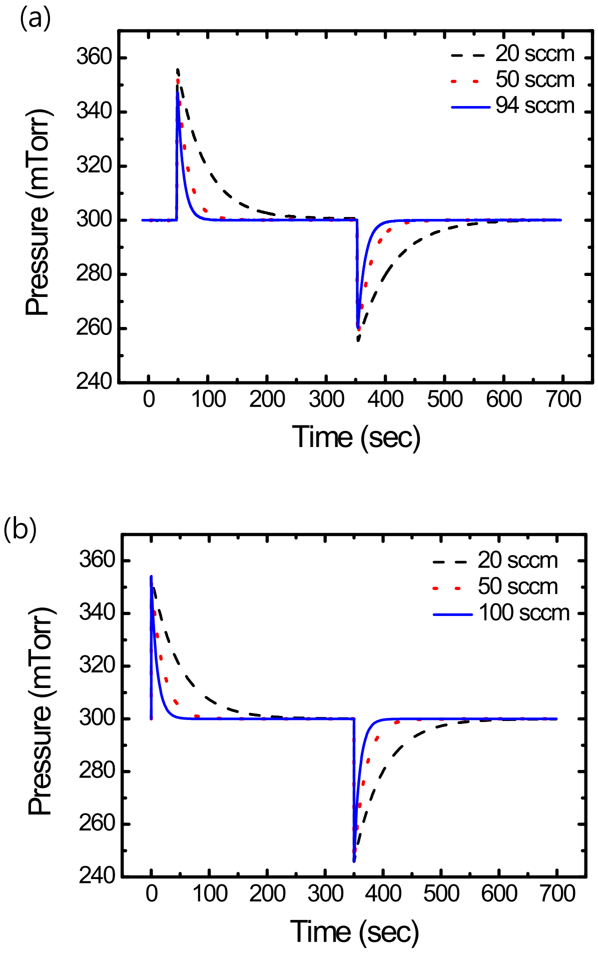

FIG. 4. (a) Temporal measurement of the gas pressure at various flow rates (300 mTorr, 300 W) and (b) calculated results at the same condition of (a).

effect induced by plasma and the pressure relaxation effect for new gas temperature. This result and physics behind is expected to provide a new operational perspective of the recent plasma processes of which time is very short, such as a plasma enhanced atomic layer deposition/etching, a soft etch for disposal of residual by-products on wafer, and light oxidation process in semiconductor manufacturing.

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[34] See supplementary material at http://dx.doi.org/10.1063/1.4798587 for equations for neutral heating and particle number change due to pumping loss.