High-speed and precise laser interferometry developed for plasma electron-density diagnostics

レーザー干渉計によるプラズマ電子密度計測の高速・高精度化

Keiichiro URABE
占部 継一郎

Laser interferometry is an optical method typically utilized to measure refractive index and distance. It is also known that the interferometry can detect electron density in plasmas by detecting the refractive-index variation induced by electron generation. Measurement technologies of the electron density are necessary for development of plasma material processes including those used in semiconductor device fabrication, since electron collision reactions such as ionization, excitation, and dissociation are keys to control plasma-assisted deposition and etching processes. The author has studied the laser interferometry to achieve more precise electron-density measurements by developing new interferometry arrangements. Detection speed of the refractive-index variation and elimination of gas-number density effects on the total measured signal are the main topics of this study.

レーザー干渉計は物質の屈折率や2点間の距離を精密に測定する光学的手法であり，プラズマ発生に伴う屈折率変動を捉えることで電子密度診断が可能であることが知られている。半導体製造に関わる薄膜堆積，エッチングプロセスに用いられる低圧熱非平衡プラズマ中の化学反応機構では，電子衝突による電離・励起・分子解離反応が重要な位置を占め，電子密度の計測技術はプラズマプロセス技術の発展に欠かせない。筆者は，従来のレーザー干渉計の装置構成を発展させ，屈折率計測の高速化やガス密度変動の影響除去などを実証し，より正確な電子密度測定を目指した研究を進めている。

Introduction

In various thin-film deposition and etching processes for semiconductor device fabrication, plasmas, which is ionized gaseous media possessing electrons, ions, and radicals, are widely utilized to drive chemical and physical reactions. In the deposition and etching processes, “thermal non-equilibrium” (also called as “low-temperature”) plasmas generated at low gas pressures in a range of 1~100 Pa are typically used. The “thermal non-equilibrium” is a category of plasmas where electron energies (electron temperatures) are significantly higher than translational motion energies of other particles (gas temperatures). The electron temperature is a temperature value defined when the electron energy distribution function is Maxwell-Boltzmann distribution. Typical range of the electron and gas temperatures in the plasma processes are over 10,000 K for electrons and from room temperature to 1,000 K for gases.

This thermal non-equilibrium feature of the plasmas enables us to drive chemical reactions which are necessary in the thin-film deposition and etching processes, under conditions that the gas temperatures are lower than the temperatures needed to trigger the reactions in thermal equilibrium processes. Also, it is possible to fabricate materials those cannot be stable in thermal equilibrium states, by the thermal non-equilibrium plasma processes. In the semiconductor device fabrication, the thermal non-equilibrium plasmas are utilized in various processes such as in PE-CVD (Plasma-Enhanced Chemical Vapor Deposition) of amorphous silicon (a-Si) and silicon nitride (SiN) thin films, magnetron sputtering deposition of metal thin films, and anisotropic etching forming nanometer scale device structures. To achieve the anisotropic etching processes, reactive ion etching utilizing synergy of reactive chemical species and high-energy ion irradiation both from the plasmas is necessary.[1]

In the plasma-enhanced deposition and etching processes, it is indispensable to understand chemical reaction mechanisms in and around the plasmas, and to control the reactions especially on the surface of processed materials precisely. Among the various parameters in plasmas, density and energy distribution of electrons are crucial, since reactive species in the plasmas generated in low-pressure atmosphere are mainly generated through ionization, excitation, and dissociation by electron-neutral collisions. There are some methods to measure the electron density such as Langmuir probe, optical emission spectroscopy (Stark broadening of hydrogen atomic emission), and laser Thomson scattering.[2] Laser interferometry, the main topic of this article, is also a method to measure the electron density in plasmas.

Interferometry is a broadly-used optical method to measure refractive indices and distances, not only in plasma electron-density diagnostics.[3] Almost measurements of the interferometry detect phase shifts of the probing light wave, usually by comparing phases between probing (transmit through or reflect on the measured sample) and reference (not interact with the sample) lights. One hears the keywords of “semiconductor” and “interferometry” may imagine a measurement tool of thin-film thickness conventionally used in in-situ monitoring of deposition and etching processes. The thickness monitoring tool is a broadband interferometry using a lamp light source, not a laser. The laser interferometry uses a single-wavelength laser beam as the probing light source, that can be seen in application areas for instance fluid dynamics observation and precise distance measurement.

In an application of the laser interferometry to the diagnostics of electron densities, typical setup arrangement is so-called CO_2-laser heterodyne interferometry, explained in the next section. Features of the laser interferometry, in comparison with the other electron-density measurement methods, are (1) measurement is not influenced by gas composition and reaction mechanisms in plasmas and (2) it continuously outputs electron-density information as the phase shift linearly correlating to the electron density. These features suggest that the interferometry is appropriate to continuous monitoring and fault detection of plasma generation and stability. It is expected to contribute for achieving better control of the plasma-enhanced deposition and etching processes.

Laser interferometry for plasma electron-density diagnostics

This chapter explains fundamentals of the electron-density measurement by the laser interferometry and its potential issues in application to the plasma process monitoring. Fundamental scheme of the electron-density measurement by the interferometry is similar to other interferometry that measures variation of the refractive index (or optical length) by detecting phase shifts of the probing laser beam. The electron densities are calculated from the refractive-index variation induced by electron generation and the optical thickness of the plasma along the probing laser path. The refractive index of a plasma is generally expressed by a Drude model shown in Equation 1. And Equation 2 shows a relationship between the phase shift $\Delta\theta$ and the electron density n_e using the plasma refractive index Equation 1.[2]

$$N^2 = 1 - \left(\frac{\omega_{pe}}{\omega}\right)^2 \frac{1}{1 - i\left({\nu_m}/{\omega}\right)} \quad \left(\omega_{pe} = \sqrt{\frac{n_e e^2}{m_e s_0}}\right) \cdots\cdots \text{(1)}$$

$$\Delta\theta = \frac{\omega}{c}\int (N-1)\,dl \approx \frac{\omega}{c}\int\left(-\frac{1}{2}\left(\frac{\omega_{pe}}{\omega}\right)^2\right)dl = -\frac{e^2\lambda}{4\pi s_0 m_e c^2}\int n_e dl \quad \cdots\cdots \text{(2)}$$

Here, ω_{pe} is the electron plasma frequency, ν_m is the electron collision frequency, ω and λ are the angular frequency and the wavelength of the probing laser beam. It should be noted that calculation at central “≈” in Equation 2 has assumption that the electron plasma frequency and the electron collision frequency can be ignored in comparison with the probing laser frequency.[4]

The Equation 2 indicates that longer wavelength (lower frequency) of the probing laser beam decreases a minimum detection limit of the electron density. Dependence of the plasma refractive index on the wavelength (dispersion) causes this feature, and it is a reason why the CO_2 lasers with a wavelength in mid infrared 10.6 µm are typically used in the electron-density diagnostics. The CO_2 laser is a kind of gas laser making reverse distribution of CO_2 vibration excitation states by discharge and its oscillation wavelengths are 9.6 or 10.6 µm.

For sensitive detection of the phase shift, a Mach-Zhender optical arrangement and heterodyne phase detection (Figure 1) are typically used in the interferometry setup. The Mach-Zhender interferometry has optics splitting the laser beam into two axes, transmitting one axis to target materials as a probe laser, and merging two lasers axes again. The interference generated in the merged laser beam includes information of the phase shift of the probing laser beam. The heterodyne phase detection is that frequency modulation is applied to one laser axis (probe or reference) in the interferometry. Beat signal at a frequency of difference between probe and reference arm is generated in the merged laser beam. The phase shift of the measured beat signal is the same as the phase shift of

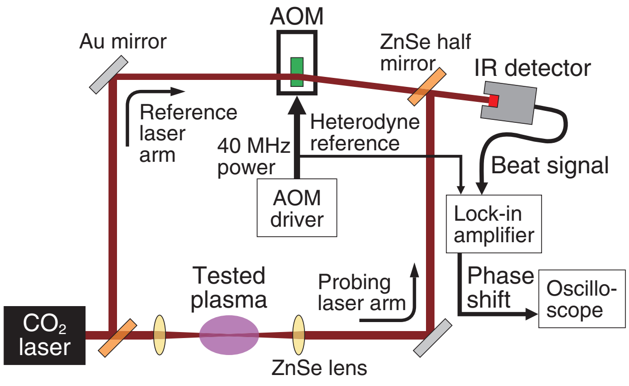

Figure 1 Schematic diagram of CO_2-laser heterodyne interferometry for plasma electron-density diagnostics. "AOM" is Acousto-Optical Modulator that applys frequency modulation to reference laser arm, to generate beat signal utilized in heterodyne phase detection.

probing laser beam; therefore, we can decrease the measurement wave frequency from optical light to manageable modulation frequency. The heterodyne phase detection can avoid influence of temporal fluctuation of the probing laser intensity leading to better phase measurement resolution.

In the author's past studies on the electron-density diagnostics using the CO_2-laser heterodyne interferometer shown in Figure 1, a detection speed of the phase shift $\Delta\theta$ (output of the lock-in amplifier) was slow, in ms order, mainly due to a conventional lock-in amplifier was used to detect the phase shift. Also, sensitive detection of the electron density was difficult when gas densities (temperatures) in and around the plasma were varied at the same timing as the electron generation. The variation of gas number density changes the refractive index detected with the electron signal in the interferometer.[5, 6] It cannot be avoided in general since it is the principle of a laser-interferometry application to gas-flow dynamics observation. When the phase shift induced by the gas-density variation becomes comparable to the phase shift by the electron-density variation, the total measured phase shift will be $\Delta\theta = \Delta\theta_{\text{electron}} + \Delta\theta_{\text{gas density}}$. In this case, we have to carefully separate the two phase-shift components before the calculation of the electron density using Equation 2. The separation is often done using a difference of general time constants of two phenomena: electron generation/extinction and gas heating/cooling. From these experimental evaluation and discussions, the conventional CO_2 laser heterodyne interferometer cannot have enough detection speed and/or sensitivity of the electron density in cases of short pulsed plasmas and plasmas with the large gas-density variation.

Spatiotemporally resolved electron-density diagnostics by near-infrared diode laser interferometry

In order to improve detection speed of the electron density in the laser interferometry, one method is to increase the modulation frequency of the heterodyne system and decrease the time duration to acquire enough cycles to measure the phase shift. In this study, we used a near-infrared (NIR) external-cavity diode laser as the probing light source, an AOM with a modulation frequency at 110 MHz, and a high-speed phase sensitive detection (PSD) device. The designed output speed of the phase detection system was 10 MHz (~100 ns time resolution).[7] In addition to the high-speed phase detection, there is a wide variety of conventional optical devices prepared for the NIR range, so that we could try a microscopic interferometer with a polarization-controlled reflection system as in Figure 2. We note that a range of the electron density inside the tested plasma source should be carefully estimated since the minimum detection limit of the electron density becomes higher when the probing wavelength is shifted from middle to near infrared range as one can see in Equation 2.

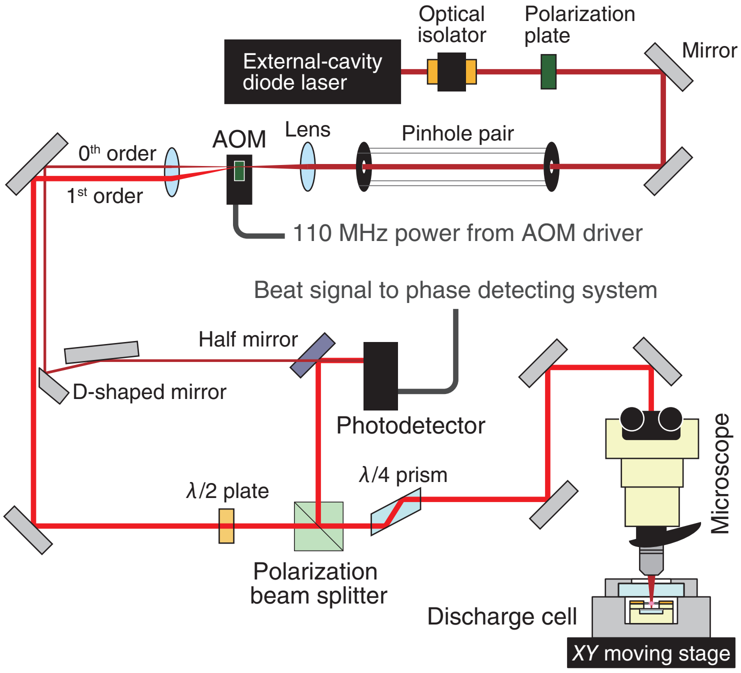

Figure 2 Near-infrared diode-laser heterodyne interferometry with microscope and reflection optical systems. This is a Mach-Zehnder arrangement using both zero (reference arm) and first (probe) order lights from the AOM. λ/2 plate, polarization beam splitter, λ/4 prism are placed to separate forward and back lights reflected at a mirror below the tested plasma inside the discharge cell under the microscope.

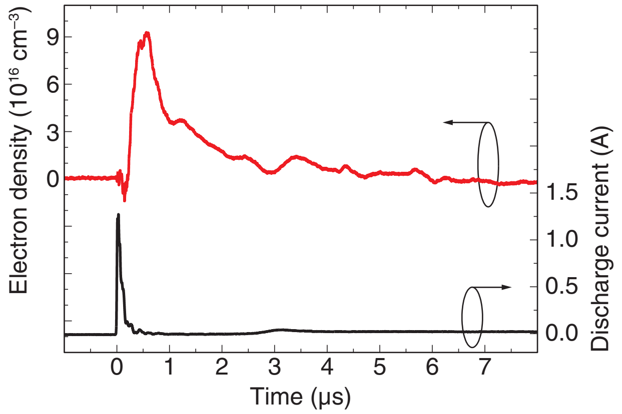

Figure 3 An example of measured temporal evolution of electron density generated in small-scale pulsed plasma source. Black line (below) is the discharge current pulse with a pulse width approximately 200 ns. Red line (above) is the measured electron density using the NIR heterodyne interferometer (Figure 2).

Measurements of a short-pulsed plasma source was done using the NIR interferometer having the feature of high-speed phase detection. The example measured data of temporal change of the electron density with a discharge current waveform is shown in Figure 3. The measurement was done with a ~0.1 mm short gap discharge put in a small gas cell under a microscope (Figure 2). In the measured electron density synchronized with the discharge current, there was a fast increase for 350 ns just after the discharge current impulse (~200 ns width), and a decay for 5 μs. The time difference between the current impulse and the rise timing of the electron density is a signal delay time in our phase detection system. This measurement result is an evidence of the improvement in temporal and spatial resolutions of the electron-density diagnostics by the developed NIR interferometer. Among advanced materials processes using plasmas those are related to the semiconductor device fabrication, the developed NIR interferometer is expected to show its potential effectively in application to investigation of plasma dynamics and electron-density monitoring of short-pulsed plasma processes such as thin-film deposition using HiPIMS (High-Power Impulse Magnetron Sputtering) with a typical pulse width of 20 μs.

Sensitive detection of electron density by dispersion interferometry

Dispersion interferometry is a kind of two-color interferometry system measuring “dispersion” of the refractive index by using two probing lasers with different wavelengths. A feature of the dispersion interferometry is to generate a second probing laser by harmonic generation from a fundamental laser beam (Use one laser source for two probing lasers). This type of interferometry has been developed for application to electron-density monitoring in nuclear fusion plasma reactors, such as in National Institute of Fusion Science (NIFS) in Japan and International Thermonuclear Experiment Reactor (ITER).[8, 9] Fusion plasmas are thermal equilibrium and their plasma parameters significantly different from those in thermal non-equilibrium plasmas used in the semiconductor device fabrication. The author found in this study that the dispersion interferometry originally designed for the fusion plasmas also has a good features in application to measure the electron density in the plasma material/surface processes. In this chapter, performance of the dispersion interferometer in the thermal non-equilibrium plasma diagnostics and its potentials to be utilized in process monitoring of the semiconductor device fabrication are introduced.

Fundamental scheme to measure the electron density by the dispersion interferometry is shown in Figure 4a. A laser beam from the light source firstly transmits a nonlinear crystal to generate the second harmonic beam. Here, there is no need to set the two-wavelength lasers on a same light axis, since the second harmonic beam is along the light axis of the fundamental beam automatically. Then, the two (fundamental and harmonic) beams

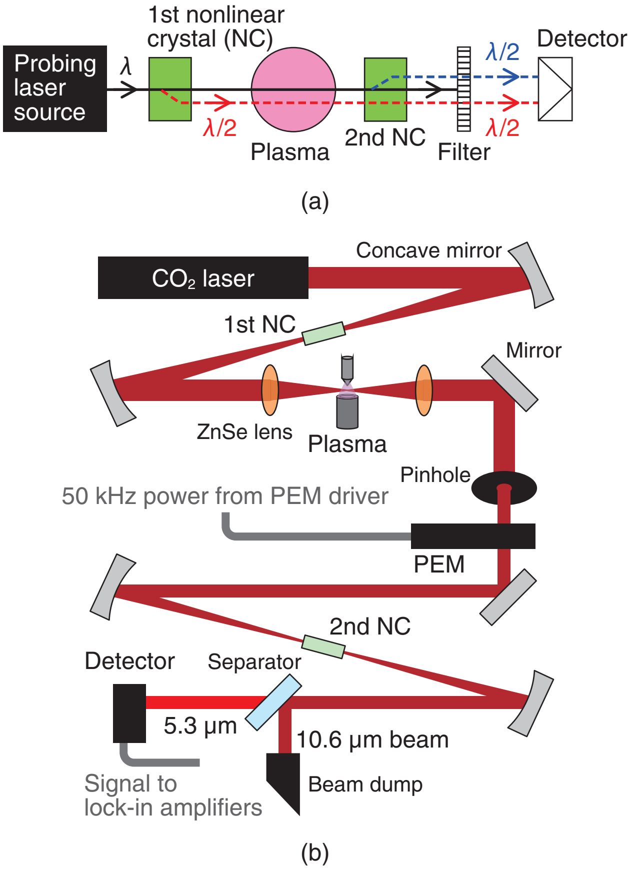

Figure 4 Fundamental scheme (a) and experimental optics setup (b) of dispersion interferometry. “NC” in the figures are the Nonlinear Crystals to generate second harmonic beam. “PEM” in (b) is the Photo-Elastic Modulator that modulates a phase of fundamental or harmonic beam. The optical devices located just before the detector [filter in (a), separator and beam dump in (b)] have the same function to eliminate fundamental laser beam from the light axis.

pass through the tested plasma, and the phases are shifted depending on variation of the refractive index in each wavelength. After passing through the plasma, another second harmonic beam is generated from the plasma-transmitted (phase-shifted) fundamental beam. Finally, a phase difference between the two harmonic beams is measured by the interference and it contains information of the refractive-index dispersion between fundamental and harmonic beam wavelengths. In the dispersion interferometry, there is no reference laser arm (laser arm not passing through tested materials), so that the laser light axis around the tested material is simpler than the normal interferometry.

The phase difference of two second harmonic beams $\Delta(\theta_1-\theta_2)$ and the electron density n_e are related as following.

$$\Delta(\theta_1-\theta_2) = -\frac{3e^2\lambda}{8\pi c^2 m_e s_0}\int n_e \mathrm{d}l \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (3)$$

The sensitivity to electron density in the dispersion interferometry that can be derived from Equation 3 is similar to that in the normal interferometry [Equation 2]. This similarity is due to a strong dispersion of the plasma refractive index [Equation 1]. On the other hand, dispersion of the refractive index is quite small in phenomena of variation of optical path length (zero in vacuum) and variation of gas number density (corresponding to gas temperature). Therefore, the output-signal amplitude of the dispersion interferometry for these phenomena becomes smaller than that for the electron-density variation compared to the normal interferometry. The change of optical path length is often due to vibration of optical devices (mirror, lens, etc.) because of plasma-generation mechanics such as vacuum pumps. These discussions suggest that the dispersion interferometry is a “robust” technique to measure the electron density in plasma sources.[10] We expected that it can show good performance in the electron-density measurement of thermal non-equilibrium plasmas with the gas-density (temperature) variation not only nuclear fusion reactors. Therefore, feasibility test experiments were done in collaboration of The University of Tokyo (Affiliation of the author at that point of time) and the NIFS.[11]

In the experiment using a setup shown in Figure 4b, we inserted a tested small thermal non-equilibrium plasma source with a pair of ZnSe lenses focusing the beam on the plasma, into the light axis of the dispersion interferometer. The dispersion interferometer constructed in the NIFS has a system to apply phase modulation either fundamental or harmonic beam to eliminate the influence of temporal fluctuation of the CO_2 laser intensity. (Phase-Modulated Dispersion Interferometry: PMDI).[12] Figure 5 is a measurement result of a sqare-pulsed discharge. Left vertical axis is the output signal of the PMDI and it corresponds to the phase difference of two harmonic beams including the information of electron density. In the measurement, at starting and ending edge of the discharge pulse (light blue backgrounded regions), sharp increase and decrease of the output signal were observed. These are the signal indicating electron generation and extinction. Still there are the signals due to variation of the gas number density during and after the discharge pulse (light green backgrounded regions). However, the signal amplitudes for the gas-density variation in the PMDI was over 100 times smaller than that measured in the normal interferometer. The author’s group concluded that the PMDI can eliminate the influence of gas-density variation and leads to precise measurement of the electron density (in line-integrated density of 10^{12} cm^{-2}).

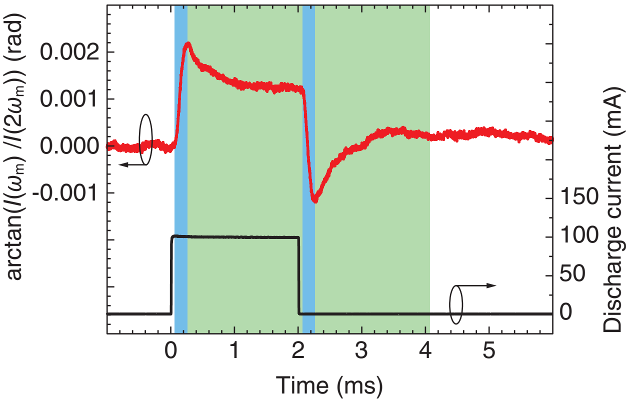

Figure 5 Measurement result of the PMDI applied to thermal non-equilibrium pulsed plasma source. Black line (below) is the discharge current, and red line (above) is the output signal of the PMDI including the information of phase difference between two harmonic beams. ω_m in the left vertical axis is the phase modulation frequency of PEM. The signal variation in the regions with blue background color is the electron density variation and that in green regions is the gas density signal.

Further development is ongoing to make the system more compact and to improve temporal resolution, for the application of the dispersion interferometry to wider variety of plasmas.[13] The dispersion interferometry has potential to achieve better electron-density diagnostics especially in plasmas possessing temporal change of gas pressure (density) and mechanical vibration. Among the processes for the semiconductor device fabrication, deposition and etching processes with cyclic gas feed and purge with plasma generation, for instance plasma-enhanced atomic layer deposition (PE-ALD) process, can be the application target.

Conclusions

The studies introduced in this article are on improvement in spatiotemporal resolution and detection sensitivity of plasma electron-density diagnostics using laser interferometry. Each technique is possible to be combined to reach a target performance if there will be demands in the future, especially for the application to plasma-enhanced thin-film process monitoring. Not only the introduced techniques, it should be pointed out that one-time spatial distribution measurement by multi-point or imaging interferometry and use of quantum cascade laser (QCL) covering wavelengths from near- to mid-infrared range have potentials to make further improvement in the laser interferometry for the electron-density diagnostics. The QCL's wavelength choice and compact package similar to diode lasers enable us to choose optimum wavelength of the probing laser beam and to construct the compact in-line process monitoring system of the laser interferometry.

The potential application areas of electron-density diagnostics using the laser interferometry is not limited to semiconductor device fabrication process discussed in this article. In all areas related to plasma technologies (e.g. electric propulsion and re-entry high temperature fluid in aerospace engineering), it is expected that laser interferometry will contribute better understanding and control of physical and chemical characteristics of plasmas through the electron-density diagnostics. The author would like to make continuous efforts on fundamental and application studies of the laser interferometry.

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Keiichiro URABE
占部 継一郎
Assistant Professor
Department of Aeronautics and Astronautics,
Graduate School of Engineering, Kyoto University
Ph. D.