Diagnostics of VHF Argon Plasmas by Laser Thomson Scattering

Weiting CHEN, Kohei OGIWARA1), Koichiro KOGE, Kentaro TOMITA, Kiichiro UCHINO and Yoshinobu KAWAI

Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Kasuga, Fukuoka 816-8580, Japan
1)Graduate School of Information Science and Electrical Engineering, Kyushu University, Nishi-ku, Fukuoka 819-0395, Japan

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The laser Thomson scattering (LTS) method has been applied to measure the electron density \( n_e \) and electron temperature \( T_e \) of very-high-frequency (VHF) argon plasmas. When the probing laser wavelength was 532 nm and the laser power density was \( \sim 10^{15} \text{ W/m}^2 \), the Thomson scattering spectrum was obviously deformed by the effect of the photo-ionization of metastable argon atoms. The threshold laser power density at which the scattered light intensity from electrons in the plasma and that from electrons produced by photo-ionization are equivalent was found to be unexpectedly low \( (4 \times 10^{13} \text{ W/m}^2) \). To avoid the photo-ionization of metastable argon atoms, the laser power density was decreased to around \( 1 \times 10^{13} \text{ W/m}^2 \) by using a cylindrical lens as the focusing lens. Then, the \( n_e \) and \( T_e \) values measured by LTS and the probe method were compared for a VHF plasma using argon gas at a pressure of 100 mTorr. This comparison confirmed that the LTS method gave reasonable \( n_e \) and \( T_e \) values.

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Very-high-frequency (VHF) plasmas are widely used to fabricate microcrystalline silicon thin-film solar cells [1, 2]. However, the deposition rate of microcrystalline silicon is much lower than that required by industry. A high-pressure depletion method [3, 4] in which the VHF plasma is produced with a narrow discharge gap at high pressures was proposed recently to increase the deposition rate. However, faster deposition of microcrystalline silicon is needed to reduce the production cost of solar cells. The deposition rate is closely related to plasma characteristics. Thus, an important subject in the study of microcrystalline silicon thin-film solar cells is the measurement of the parameters of the VHF plasma produced by a narrow gap discharge.

There are several methods to measure plasma properties. The Langmuir probe method can be used to investigate the characteristics of VHF plasmas, such as the electron density \( n_e \) and the electron temperature \( T_e \). However, the parameters of a plasma produced by a narrow gap discharge will be seriously disturbed when the probe is inserted. In addition, the probe cannot be used at gas pressures higher than 10 Torr, because the ion mean-free path is much shorter than the sheath length, and ion collisions frequently occur in the sheath [5].

The laser Thomson scattering (LTS) method is a powerful diagnostic method under such conditions. Therefore, we have been applying LTS to various discharge plasmas. When LTS is applied to VHF plasmas, a high power-laser light source is required to secure an adequate signal-to-noise ratio of the Thomson scattering signal, because the electron densities of VHF plasmas are expected to be relatively low \( (< 10^{17} \text{ m}^{-3}) \). We must also consider the laser disturbance of the plasma. We used argon gas at pressures ranging from 0.1 to 1 Torr as the working gas, and the second harmonic (wavelength 532 nm) of a YAG laser as the light source. Under these conditions, metastable argon atoms in the plasma may be photo-ionized by two-photon absorption of laser photons. Yamamoto [6] recently reported that the Thomson scattering signal was affected by the photo-ionization of metastable xenon atoms when LTS experiments were performed with laser power densities of \( > 10^{15} \text{ W/m}^2 \). In this study, we examined the influence of the photo-ionization of metastable argon atoms on the Thomson scattering spectrum and the laser power density at which the photo-ionization becomes a problem for the LTS diagnostics of argon plasmas.

Figure 1 shows a schematic of the LTS system for the VHF plasma. The chamber (diameter 200 mm, length 400 mm) was specifically designed for LTS measurements. The chamber was equipped with baffles, two Brewster windows, a beam dump, and a triple grating spectrometer (TGS). The structure and function of the TGS have been reported in Ref. [7]. This TGS made it possible to detect the Thomson scattering spectrum at 1 nm away from the laser wavelength without the problem of stray light. Finally, the scattered light signals passing through the TGS were detected by an ICCD camera (Princeton Instruments, PI-MAX III). The quantum efficiency of the camera was \( \sim 50\% \) at \( \lambda = 532 \text{ nm} \). The photon counting method was
applied to enable reliable measurements of small scattered signals.

The 60-MHz VHF plasma was sustained between two parallel-plate electrodes (60 mm × 60 mm × 8 mm) made of stainless steel; the distance between them was always maintained at 10 mm. The working gas was argon; the gas pressures used in this study were 500 mTorr and 100 mTorr, and the VHF power ranged from 20 W to 80 W. In the conventional power feeding method, one electrode is connected to the power supply through the matching box, and another electrode is connected to the ground. This type of a power feeding system has been applied in many studies. However, abnormal discharges can be produced between the power feeding cable and the chamber wall in the VHF range. To avoid such abnormal discharges, we used a balanced power feeding method [8]. Thus, the plasma was mainly produced in the region between the two electrodes.

To examine the effect of the photo-ionization of metastable argon atoms, Thomson scattering experiments were performed under two conditions with greatly different laser power densities by using a plano-convex lens and a cylindrical lens as the focusing lens of the probing laser beam. Both lenses had focal lengths of 500 mm, and the probing laser energy was 300 mJ. When the plano-convex lens was used, the diameter of the laser spot at the measuring point was 160 μm (full width at half-maximum). The estimated laser power density for this case was $1.5 \times 10^{15}$ W/m². When the cylindrical lens was used, the laser spot seen from the detection optics side was 290 μm in height and 8 mm in depth. The laser power density in this case was $1.3 \times 10^{13}$ W/m². For these experiments, plasmas were produced at an argon gas pressure of 500 mTorr and a VHF power of 80 W.

The experimental results are shown in Fig. 2, which compares the spectra observed using the cylindrical and plano-convex lenses. Each data point represents signals accumulated over 40,000 laser shots. As noted above, the signals were detected by the ICCD camera. The 29 plotted data points are in the wavelength range from $\Delta \lambda = 1.204$ nm to $\Delta \lambda = 3.612$ nm with an interval of 0.086 nm. Here $\Delta \lambda$ is the wavelength difference between the laser wavelength and the measured wavelength.

The ordinate is the detected photon number on a logarithmic scale; the abscissa is $(\Delta \lambda)^2$. In this plot, a straight line indicates a Gaussian spectrum, and therefore a Maxwellian electron energy distribution function. As can be seen from Fig. 2, the data points for the cylindrical lens case fall on a single line. The line gives an electron density of $n_e = 1.6 \times 10^{17}$ m⁻³ and an electron temperature of $T_e = 3.1$ eV.

On the other hand, for the plano-convex lens case, the spectrum was fitted by two lines. The gradient of the line with the moderate slope agrees well with that of the line fitted for the cylindrical lens case. When the steeper spectrum is subtracted from the moderate slope spectrum, the resultant spectrum gives an electron temperature of 0.5 eV, assuming a Gaussian spectrum. Thus, the average energy of this low-energy component is around 0.75 eV.

Metastable argon atoms have two energy levels: 11.55 eV and 11.72 eV [9]. The energy obtained by two-photon absorption of the probing laser light should be
4.66 eV. On the other hand, the energies required to ionize argon to the metastable states are 4.22 eV and 4.03 eV. Therefore, the excess energies of 0.44 eV and 0.63 eV can be delivered to the free electrons that are released by two-photon ionization from the argon metastable states.

These facts show that the average energy of the low-energy component and the energies acquired by electrons produced by photo-ionization are comparable. In fact, the intensity of the low-energy component decreased when the probing laser energy was decreased. Therefore, it can be considered that the low-energy component was produced by the photo-electrons as a result of the photo-ionization of metastable argon atoms.

Then, we examined the dependence of the intensity of the low-energy component against the laser power density. The measurements were performed for various laser power densities. As a measure of the intensity of the low-energy component, signals in the wavelength range from $\Delta \lambda = 1.2 \text{ nm}$ to $\Delta \lambda = 1.6 \text{ nm}$ were integrated. The results are shown in Fig. 3. In this figure, both the signal count number (the ordinate) and the laser power density (the abscissa) are plotted on a logarithmic scale.

In Fig. 3, the solid line, which is the sum of a linear function (slope 1) and a cubic function (slope 3), is well fitted to the data points with laser power densities below $7 \times 10^{13} \text{ W/m}^2$. Above this range (> $10^{14} \text{ W/m}^2$), the signal intensity increases with a slope of 1.5 against the laser power density. This is interpreted as follows. At laser power densities below $1 \times 10^{13} \text{ W/m}^2$, the signal is proportional to the laser power density, and the effect of the photo-ionization of metastable argon atoms is negligible. The critical laser power density at which the real Thomson scattering signal is comparable to the effect of the photo-ionization of metastable argon atoms is around $4 \times 10^{13} \text{ W/m}^2$. In the second range of laser power density, between $4 \times 10^{13} \text{ W/m}^2$ and $7 \times 10^{13} \text{ W/m}^2$, the effect of the photo-ionization of metastable argon atoms dominates. In this range, the signal is determined by the two-photon ionization process and the Thomson scattering (one-photon process), and therefore is proportional to the third power of the laser power density. In the third region, where the laser power density is greater than $7 \times 10^{13} \text{ W/m}^2$, the signal is again almost linear with respect to the laser power density. This may be due to the depletion of metastable argon atoms, because most of them are ionized early in the strong laser pulse. However, the spatial profile of the laser power density has a tail to some extent; the signal increases with a slope of 1.5 against the laser power density.

Now we know that LTS can be applied to VHF argon plasmas when the laser power density is below $1 \times 10^{13} \text{ W/m}^2$. We compared the values of the electron density and electron temperature measured by LTS and the Langmuir probe. We selected an argon gas pressure of 100 mTorr so that the ion mean-free path could be comparable to the sheath length, and the results of the probe might be relatively reliable.

The results are shown in Fig. 4. The electron den-
sity values obtained by LTS are higher than those obtained by the probe, and the values of the electron temperature obtained by LTS are lower than those obtained by the probe. This tendency is in agreement with previous reports [10, 11]. Therefore, these results further support the validity of the results of LTS measurements.

In conclusion, we examined the applicability of LTS to the diagnostics of VHF argon plasmas. As a result, it was shown that the photo-ionization of metastable argon atoms affects the Thomson scattering spectrum when the probing laser power density is greater than $4 \times 10^{13}$ W/m$^2$. This can be attributed to the fact that the density of metastable argon atoms in VHF plasmas produced in argon gas at pressures above 100 mTorr is comparable to the electron density of the plasmas. The threshold laser power density may change slightly according to the ratio of the metastable argon density and the electron density. Unexpectedly, the threshold laser power density for argon gas was lower than that for xenon gas by a factor of 20.

As a result of this study, we can recognize the effect of the photo-ionization of metastable atoms on the Thomson scattering spectrum by comparing the spectra obtained with a plano-convex lens and a cylindrical lens. We can remove the influence of the photo-ionization of metastable argon atoms on the Thomson scattering spectrum of the VHF argon plasma by reducing the power density of the YAG laser below $1 \times 10^{13}$ W/m$^2$.