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Development of vacuum ultraviolet absorption spectroscopy system for wide measurement range of number density using a dual-tube inductively coupled plasma light source

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A vacuum ultraviolet absorption spectroscopy system for a wide measurement range of atomic number densities is developed. Dual-tube inductively coupled plasma was used as a light source. The probe beam profile was optimized for the target number density range by changing the mass flow rate of the inner and outer tubes. This system was verified using cold xenon gas. As a result, the measurement number density range was extended from the conventional two orders to five orders of magnitude. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4770118>]

I. INTRODUCTION

Atomic species in plasma are important in several fields such as the oxidation of materials,¹ chamber cleaning,² organic polymer etching,³ space propulsion,⁴ and thermal protection systems for entry vehicles.⁵ In these varied fields, however, no established method is yet available to measure these number densities. Intrusive methods such as mass spectroscopy, titration, and the use of a quartz crystal microbalance (QCM) are difficult to apply to reactive plasma diagnostics because of the disturbance they introduce, as well as their limited thermal tolerance and bad spatial resolution. Non-intrusive forms of spectroscopy have also been used for such plasma diagnostics.^{6–8} Since the resonance lines of many atomic ground states belong to the vacuum ultraviolet region, two-photon absorption laser-induced fluorescence spectroscopy (TALIF) is a unique method for accessing a ground state directly, with enough wavelength resolution to obtain the line profiles.⁹ However, a theoretical two-photon absorption cross section is so complex and unreliable that it creates inaccuracy as to the absolute number density measurement.¹⁰ Recently, vacuum ultraviolet absorption spectroscopy (VUVAS) that uses a plasma light source has been developed as an approach for direct number density measurement to the ground state.^{11,12} Absorption spectroscopy gives an absolute number density from the fractional absorption without any calibration, and it is theoretically applicable to any number density target.¹³ However, the measurement error increases drastically for fractional absorption that is smaller than 1% or larger than 99%. In addition, the practical measurement number density range is usually limited to around two orders of magnitude.

Since fractional absorption depends not only on the number density and absorption length, but also on the relationship between the probe beam and the target absorption profiles, we developed a variable beam profile light source that can be used for in the measurement of a wide number density range.

This system, useful for VUVAS, was validated and evaluated using cold xenon gas.

II. VACUUM ULTRAVIOLET ABSORPTION SPECTROSCOPY

A. Number density measurement

Since the wavelength resolution of a commercial VUV spectrometer is much larger than the absorption line width, the measured fractional transmitted beam intensity I_t/I_0 is given as a frequency integrated value as

$$\frac{I_t}{I_0} = \frac{\int f_{\text{probe}}(\nu) \exp\{-Kl f_{\text{target}}(\nu)\} d\nu}{\int f_{\text{probe}}(\nu) d\nu}, \quad (1)$$

where I_t , I_0 , $f_{\text{probe}}(\nu)$, $f_{\text{target}}(\nu)$, K , and l are the incident beam intensity, transmitted beam intensity, normalized probe beam and absorption profiles, integrated absorption coefficient, and absorption length, respectively. K is related to the number density of the absorption state, n_i as

$$n_i = \frac{8\pi}{\lambda^2 A_{ji}} \frac{g_i}{g_j} K, \quad (2)$$

where A , i , and j are the Einstein coefficient, absorption state, and excited state, respectively. Since the proportional constant in Eq. (2) depends on the absorption line, in this paper, the discussion is focused not on n_i but on Kl .

Under low-pressure conditions, the beam and absorption profiles are Gaussian, with a Doppler width of $\Delta\nu_D$. These profiles are then expressed as¹³

$$f(\nu) = \frac{1}{\sqrt{\pi} \Delta\nu_D} \exp\left[-\left(\frac{\nu - \nu_0}{\Delta\nu_D}\right)^2\right], \quad (3)$$

$$\Delta\nu_D = \frac{\nu_0}{c} \sqrt{\frac{2k_B T}{M_A}}, \quad (4)$$

where ν_0 , M_A , k_B , and T are the center frequency, atomic weight, Boltzmann constant and translational temperature,

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FIG. 1. KI and KI error as a function of I_t/I_0 for variations in $\Delta(I_t/I_0)$.

respectively. Equations (1)–(4) show that I_t/I_0 depends not only on KI , but also on T of the light source and the target.

B. Measurement error and measurable range

As shown in Eq. (1), any KI gives I_t/I_0 in the range of 0 to 1, where an infinite KI and KI equal to 0 correspond to I_t/I_0 of 0 and 1, respectively. Theoretically then, there is no measurement limit for KI . In practice, however, there is measurement error $\Delta(I_t/I_0)$. Figure 1 shows KI and the KI error as functions of I_t/I_0 for several $\Delta(I_t/I_0)$. Here, T_{probe} and T_{target} are set to 800 K and 300 K, respectively. As can be seen in the figure, $\Delta(I_t/I_0)$ causes a drastically large KI error as I_t/I_0 approaches 0 and 1. The, reliable measurement region of KI is then limited to about two orders of magnitude, from 10^9 to 10^{11} for I_t/I_0 from 1% to 99%.

One approach to extending the measurement region of KI is to vary the inductively coupled plasma (ICP) temperature T_{probe} in Eq. (1). However, it is difficult to control T_{probe} more than one order of magnitude. Therefore, in this study, the probe beam profile is varied by controlling the absorption in an ICP light source.

C. Effect of absorption in ICP light source

Considering the absorption between plasma and an incident window in an ICP tube, the effective incident beam intensity I_0' is a value that is absorbed value to a certain degree. Equation (1) is then rewritten as the effective fractional transmitted beam intensity I_t'/I_0' , which is expressed as

$$\frac{I_t'}{I_0'} = \frac{\int f_{\text{probe}}(\nu) \exp\{-K_{\text{tube}} l_{\text{tube}} f_{\text{tube}}(\nu) - KI f_{\text{target}}(\nu)\} d\nu}{\int f_{\text{probe}}(\nu) \exp\{-K_{\text{tube}} l_{\text{tube}} f_{\text{tube}}(\nu)\} d\nu}, \quad (5)$$

where I_t' , $f_{\text{tube}}(\nu)$, K_{tube} , and l_{tube} are the effective transmitted beam intensity, absorption profile, integrated absorption coefficient, and absorption length between the plasma and the incident window, respectively. Figure 2 shows I_t'/I_0' as a function $K_{\text{tube}} l_{\text{tube}}$ for several KI . Here, the temperature T_{tube}

FIG. 2. Relations between I_t'/I_0' and $K_{\text{tube}} l_{\text{tube}}$ for variations in KI .

is set to be the same as the supplied gas temperature of 300 K. As can be seen in the figure, I_t'/I_0' increases with the increase in $K_{\text{tube}} l_{\text{tube}}$. Thus, even for a large target KI , I_t'/I_0' becomes detectable by increasing $K_{\text{tube}} l_{\text{tube}}$. In this study, $K_{\text{tube}} l_{\text{tube}}$ is varied by changing the mass flow rate ratio of a dual-tube ICP source.

III. EXPERIMENTAL EQUIPMENT

A schematic of the VUVAS system is shown in Fig. 3. This system employs a dual-tube ICP source for XeI 146.9 nm beam generation. The diameters of the outer and inner tubes are 50 mm and 22 mm, respectively. The xenon gas is supplied from the outer tube by swirl injection in order to stabilize the ICP. For the inner tube, argon is supplied in order to control the partial pressure of the xenon between the plasma and the window, which makes it possible for there to be variable $K_{\text{tube}} l_{\text{tube}}$. The ICP is operated by a 13.56 MHz rf power source (OWM-12, ENI Inc.) with a five-turn copper coil. The net input power is 60 W and the operation pressure ranges from 10^{-1} to 10^3 Pa. Magnesium fluoride (MgF_2) is used as a window material that can transmit the VUV beam. The ICP temperature T_{probe} is measured by a conventional laser absorption spectroscopy (LAS) using a XeI 823.1 nm line. The details of the LAS measurement system are given in Refs. 14 and 15. The transition data of the xenon lines are tabulated in Table I.¹⁶

The VUV beam is led to the VUV spectrometer through a test chamber, which is separated from both the ICP source and the spectrometer by two MgF_2 windows. The distance

TABLE I. Transition data for the target absorption lines.

	Xe line	E_i (eV)	E_f (eV)	g_i	g_f	A_{ji} (10^7 s^{-1})
VUVAS	146.9 nm	0	8.44	1	3	28.1
LAS	823.1 nm	8.32	9.82	5	5	2.45

FIG. 3. Schematic of VUVAS system.

between the two MgF_2 windows is 160 mm, which corresponds to the absorption length. The chamber pressure is measured by three kinds of vacuum gauges: a Pirani gauge (WP-01, ULVAC Inc.), Penning gauge (WI-PA, ULVAC Inc.), and ionization gauge (WIT-G1, ULVAC Inc.). The transmitted beam intensity is measured by a corrected holographic type VUV spectrometer (234/302, McPherson Inc.) with a photo multiplier tube (R8486, Hamamatsu Photonics K.K.). The wavelength resolution is 0.1 nm. The ICP, test chamber, and VUV spectrometer are independently evacuated using a mechanic booster pump (YM-300, ULVAC Inc.), diffusion pump (ESV-4C, Shibaura Eletec Corp.), and turbo molecular pump (PT-300, Shimadzu Corp.), respectively.

IV. RESULTS AND DISCUSSION

The xenon pressure of the test chamber was varied from 10^{-3} Pa to 10 Pa by changing the mass flow rate and pumping speed. The operation conditions of the ICP are listed in Table II. Three conditions were tested. Case 1 was a pure xenon operation, and corresponds to a conventional single-tube ICP light source. In cases 2 and 3, argon was supplied from the inner tube in order to reduce the xenon partial pressure between the ICP and the incident window.

Figure 4 shows the measured I_t/I_0 and the fitted curves of Eq. (5). Here, the horizontal axis Kl was estimated from the chamber pressure using an equation of state, assuming a temperature of 300 K. The measured T_{probe} by LAS was 830 K and $K_{\text{tube}}l_{\text{tube}}$ was set as a fitting parameter. As can be seen in the figure, the measured and fitted curves show good agreement in every case. The measurable region is found to shift to a higher Kl region with a decrease in the argon mass flow rate. The reason for this is that $K_{\text{tube}}l_{\text{tube}}$ increases with the decrease in non-absorbent argon. As a result, the measurable

TABLE II. Operating conditions.

Case no.	Inner : Ar (sccm)	Outer: Xe (sccm)
1	0	10
2	10	0.05
3	50	0.05

FIG. 4. Measured I_t/I_0 and fitted Eq. (5) with $K_{\text{tube}}l_{\text{tube}}$ as a parameter.

number density range can be extended from the conventional two orders to five orders of magnitude. With this method, a higher $K_{\text{tube}}l_{\text{tube}}$ permits a higher Kl measurement. However, for the lower region of Kl of less than 10^8 , modulation or cavity techniques are useful.^{17,18}

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