Development of high resolution LIF spectroscopy with saturated absorption spectrum

Shuzo ETOH, Mitsutoshi ARAMAKI [1)], Kohei OGIWARA, Shinji YOSHIMURA[2)] and Masayoshi Y. TANAKA[3)]

Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Kasuga, Fukuoka 816-8580, Japan
[1)]*Department of Electrical Engineering and Computer Science, Nagoya University, Chikusa-ku, Nagoya 464-8603, Japan*
[2)]*National Institute for Fusion Science, Toki 509-5292, Japan*
[3)]*Department of High Energy Engineering Science, Kyushu University, Kasuga, Fukuoka 816-8580, Japan*

(Received: 18 September 2008 / Accepted: 26 December 2008)

High resolution flow velocity measurement of neutrals has been performed by using a laser induced fluorescence (LIF) system installed with a saturated absorption spectroscopy unit. The improvement of accuracy is achieved by utilizing Lamb dip as the reference of laser wavelength. The newly-developed LIF system can determine neutral flow velocity with an accuracy of 2 m/s, which is one order of magnitude higher than the velocity resolution of LIF systems with ordinary calibration technique utilizing iodine absorption spectrum.

Keywords: saturated absorption spectroscopy, laser induced fluorescence, laser wavelength calibration, neutral flow, high resolution velocity measurement, ECR plasma.

1. Introduction

Recently, it has been found that neutral particles play an important role in phenomena of high density plasmas. Neutral heating and neutral depletion have been observed in a high density plasma for chemical vapor deposition, and affect the characteristic behavior of the plasma. [1, 2] It is also found that a vortex structure, which rotates in the opposite direction to $\boldsymbol{E} \times \boldsymbol{B}$ drift, occurs in a high density argon plasma produced by electron cyclotron resonance (ECR) heating. These phenomena are considered to be caused by momentum transfer through charge exchange interaction between the neutral particles and ions. [3, 4] The measurement of neutral flow velocity is becoming an important issue and demands a high velocity resolution. Although the role of neutral flow on dynamical behavior of ions commonly has been attracting much attention, the flow velocity measurement of neutrals has not been carried out so far except for a fast flowing case. [5]

Laser induced fluorescence (LIF) method [6] is usually adopted to measure ion flow velocities of the order of several km/s. For neutral flow measurement, on the other hand, a high resolution LIF system is needed to measure slow neutral flow velocities of the order of 10 m/s, which has not been developed so far because system stability and precise calibration method are needed to achieve very high performance in velocity resolution.

We have developed a high resolution LIF spectroscopy system utilizing saturated absorption spectroscopy and measured a neutral flow velocity with an accuracy of 2 m/s, demonstrating that the newly-developed LIF spectroscopy system is a very powerful tool for studying the interaction of ion and neutral flow.

In Sec. 2, the experimental apparatus and the newly-developed LIF system are shown. The results of system performance and LIF measurements are presented in Sec. 3. The concluding remarks are given in Sec. 4.

2. Experimental setup and LIF system

The experiments have been performed in the high density plasma experiment (HYPER-I) device at National Institute for Fusion Science, which is shown in Fig. 1. [7] The vacuum vessel is 30 cm in diameter and 200 cm in axial length. The magnetic field line is weakly diverging and ECR point is located 120 cm away from the open end of the device connected to the waveguide. The plasma is produced by electron cyclotron resonance heating using a microwave of 2.45 GHz frequency. The filling gases pressure is 10 mTorr (Ar) and input microwave power is 250 W. The typical electron density is 4.8×10^{10} cm^{-3} and the electron temperature is 3 eV. The position of neutral flow measurement is located more than 20 cm away from the ECR region.

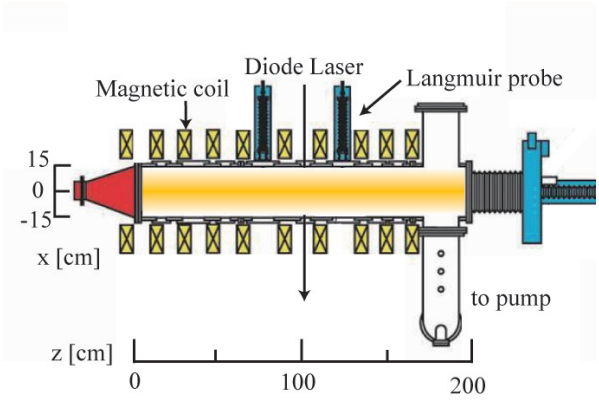

Fig. 1 Schematic of HYPER-I device.

author's e-mail: syu-eto@aees.kyushu-u.ac.jp

The LIF spectroscopy system used in the present experiment is shown in Fig. 2. A narrowband tunable diode laser (696.73 nm) is used to pump Ar metastable atoms ($4s[3/2]^o_2$) to an upper energy level ($4p'[1/2]_1$). The fluorescence photons of 826.67 nm in the $4s'[1/2]_1 - 4p'[1/2]_1$ transition are collected by a lens, detected by a photo multiplier tube (PMT), and amplified by a lock-in amplifier. The similar excitation scheme of Ar ($4s[3/2]^o_2 - 4p[3/2]_1$) is possible using a 772.59 nm diode laser. [8] The wavelength of the fluorescence photons is 810.59 nm ($4s[3/2]^o_2 - 4p[3/2]_1$) and is very close to another Ar I line ($4s[3/2]^o_1 - 4p[3/2]_1$), located at 811.75 nm. [9] The distinction between these two lines is difficult when the fluorescence photons are detected by a PMT. On the other hand, in the scheme adopted in the present experiment, the fluorescence photons are emitted in a wavelength of 826.67 nm, around which there is no Ar I line. [10] As a result, the present LIF scheme will provide a higher signal to noise ratio.

The spectral bandwidth of the laser is 5 fm and the maximum output power is 15 mW. The spatial measurement is performed by moving the collection optics along the radial direction. A narrow filter with the center frequency of 825 ± 5 nm is set in front of the PMT. Since the LIF spectrum is proportional to the velocity distribution function of neutral particles, the neutral flow velocity can be determined by the Doppler shift of the LIF spectrum. To obtain the exact value of Doppler shift, the laser wavelength must be precisely calibrated. For this purpose, the absolute laser wavelength is determined by using a Fabry Perot resonator and saturated absorption spectrum. The laser beam is divided into two sub beams, one is introduced into the Fabry Perot resonator. The output fringes, which consist of series of pulse with constant frequency interval (Δf = 294 MHz), is used for the reference of wavelength scale when scanning the laser frequency. Another beam is introduced to the vacuum vessel and the transmitted beam is reflected by a mirror. The reflected laser beam reentered along the same optical path and is detected to obtain the saturated absorption spectrum.

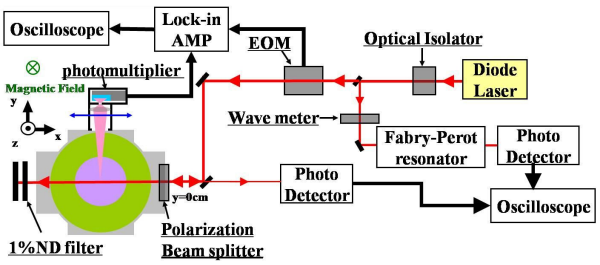

Fig. 2 LIF spectroscopy system.

The electron density profile is measured by a Langmuir probe. The diameter and length of the tip are 1.5 mm and 0.1 mm, respectively. The probe is movable in the radial direction (-15 cm < x < 15 cm).

Saturated absorption spectrum is a Doppler broadened spectrum superposed with a natural broadened spectrum. [11] So-called Lamb dip appears on the absorption spectrum when the laser frequency is exactly the same as the resonant frequency of the medium at rest. When the laser frequency ω is not equal to resonance frequency ω_0, the incident wave is absorbed by the atoms with a velocity $v = (\omega - \omega_0)/k$, and the reflected wave by the atoms $v = -(\omega - \omega_0)/k$, where k is wavenumber of the laser beam. When $\omega = \omega_0$, the atoms with $v = 0$ are excited for both the incident and reflected beams. Since the lower level atoms is excited to an upper level by the incident laser, "burnout" of lower level atoms takes place for the reflected beam (probe beam) to be absorbed. Then, the absorption coefficient reduces for the reflected beam, indicating a sharp peak at frequency $\omega = \omega_0$, which is called Lamb dip. The saturated absorption coefficient $\alpha_s(\omega)$ is given by

$$\frac{\alpha_s(\omega)}{\alpha(\omega)} = \left[1 - \frac{S}{2}\left(1 + \frac{(1+S)(\gamma/2)^2}{(\omega-\omega_0)^2 + (1+S)(\gamma/2)^2}\right)\right], \tag{1}$$

where $\alpha(\omega), S, \gamma$ are the absorption coefficient, the saturation parameter and the natural linewidth, respectively. The profile of saturated absorption spectrum depends on laser intensity since S is a function of laser intensity and resonance frequency. The quantity $\alpha(\omega)$ is a Maxwellian distribution function corresponding Doppler broadening due to thermal motion.

When the intensity of reflected beam is weak ($S \ll 1$), $\alpha_s(\omega)$ profile becomes

$$\frac{\alpha_s(\omega)}{\alpha(\omega)} = \left[1 - \frac{S_0}{2}\frac{(\gamma_s/2)^2}{(\omega-\omega_0)^2 + ((\gamma_s+\gamma)/4)^2}\right], \tag{2}$$

where γ_s is given by $\gamma_s = \gamma\sqrt{1+S}$. [12] The $\alpha_s(\omega)$ profile is approximated by a convolution of Maxwellian distribution function and Lorentz distribution function. The full width at half maximum (FWHM) of Lamb dip is much narrower than that of Doppler broadening. Therefore, Lamb dip is an useful reference for calibrating the laser wavelength. An absorption line of iodine is frequently used as the wavelength reference, however, it is subjected to Doppler broadening due to iodine temperature, which is about two order of magnitude wider than the width of Lamb dip (~ 0.1 pm). The reflected laser beam is attenuated to 1 % of incident beam by a neutral density (ND) filter in the present experiment.

3. Experimental Results

A typical example of LIF and saturated absorption spectra are shown in Fig. 3. There are three sharp peaks in the absorption spectrum (Lamb dip). The FWHM of absorption spectrum is much larger than that of Lamb dip. The stark broadening, pressure broadening and power broadening are considered to be negligible. [13] The width of Lamb dip is due to natural broadening. The three components of Lamb dip are attributable to the anomalous Zeeman Effect. It is emphasized that the Lamb dip in the absorption spectrum occurs at the fixed wavelength corresponding the resonant frequency (ω_0) in the rest frame regardless of the experimental conditions, and is highly suitable as a wavelength standard. On the other hand, the LIF spectrum is shifted due to average motion of neutrals (Doppler shift), from which the flow velocity is determined. The LIF spectrum exhibits a Maxwellian distribution with a temperature of 0.033 eV.

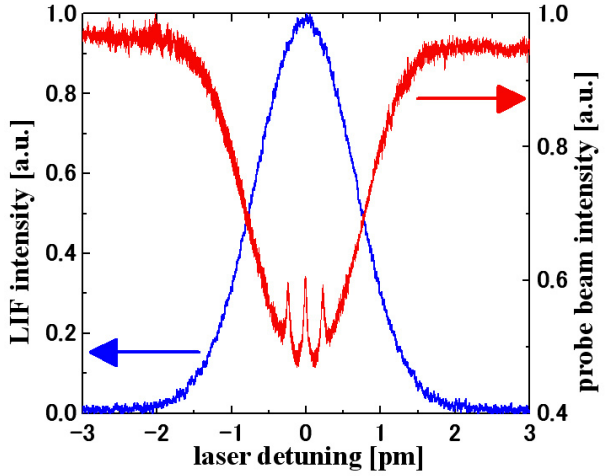

Fig. 3 LIF and saturated absorption spectra. There are three Lamb dips at the resonant wavelength (bottom of the absorption spectrum).

Figure 4 shows the magnified view of central Lamb dip component. The profile of saturated absorption spectrum is well fitted by eq (2), as seen in Fig. 4(a). The FWHM of Lamb dip is 0.056 pm and exhibits the natural line width. The π transitions are not shifted from the central wavelength of an absorption spectrum and are linearly polarized along the external magnetic field. The Doppler broadening and the natural line width are 1.3 pm and 0.075 pm in the experimental condition, respectively. [14, 15]

The schematic of the excitation diagram is shown in Fig. 5. Since the polarization of the laser beam is parallel to the external magnetic field, only the π component is considered in this scheme. As seen in the figure, the magnetic quantum number of lower and upper levels are 3/2 and 1/2, respectively, and thus the number of π component is three. Consequently, three Lamb dips (peaks) appear in the saturated absorption spectrum. We take the center peak as the wavelength reference to determine the absolute value

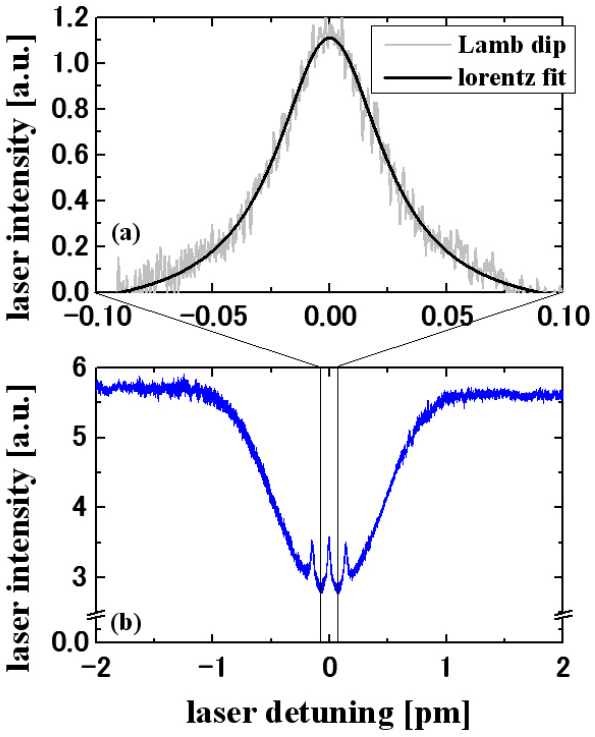

Fig. 4 Lamb dip component (a) and the saturated absorption spectrum (b).

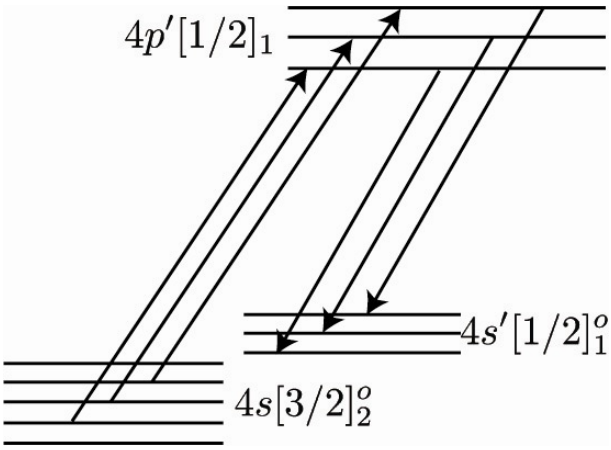

Fig. 5 Excitation diagram of $4s[3/2]^o_2$ π component including anomalous Zeeman effect.

of Doppler shift.

To realize high resolution Doppler spectroscopy, it is also important to achieve stability of the system. We have repeated the same LIF measurements under the constant experimental conditions and confirm the reproducibility of LIF spectrum. Figure 6 shows the LIF peak positions in wavelength measured during 5 hours. The magnitude of deviation means the stability of the LIF spectroscopy system. The shadowed area in Fig. 6 shows the laser line width and is the limit of accuracy of the system. The LIF peak deviation determined by iodine absorption line (696.74 nm) is plotted as reference data to be compared

with. An iodine cell, which was heated up to 90 degrees Celsius by silicon rubber heater, was used in the experiment.The LIF peak deviation corresponds to the total accuracy of the system, because the deviation is the sum of fluctuations of the LIF spectrum and the error of reference of laser wavelength. From Fig. 6(a), the LIF peak deviation is 5 fm for saturated absorption spectrum and 20 fm for iodine absorption spectrum. These deviations correspond to velocity resolution of 2 m/s and 8 m/s, respectively. The results show that the accuracy of the present system is 2 m/s and the system performance has been substantially improved by using the saturated absorption spectroscopy.

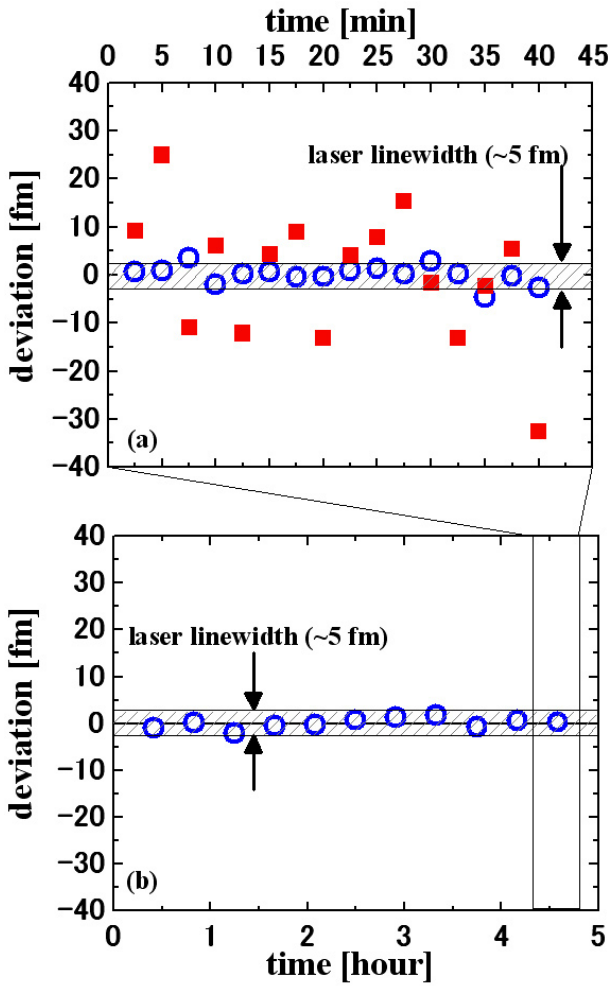

Fig. 6 Deviation of LIF peak position in wavelength for the repeated measurements under the same experimental condition. (a): short term; (b): long term. The reference of laser wavelength is Lamb dip (open circles) or iodine absorption spectrum (closed square).

The iodine absorption spectrum exhibits a complicated pattern because it consists of several hyper fine spectra within the single absorption line. Furthermore, the peak of iodine absorption spectrum changes depending on the iodine cell temperature since each hyperfine structure in the absorption spectrum broadens due to thermal motion. It is also emphasized that convection of iodine vapor also

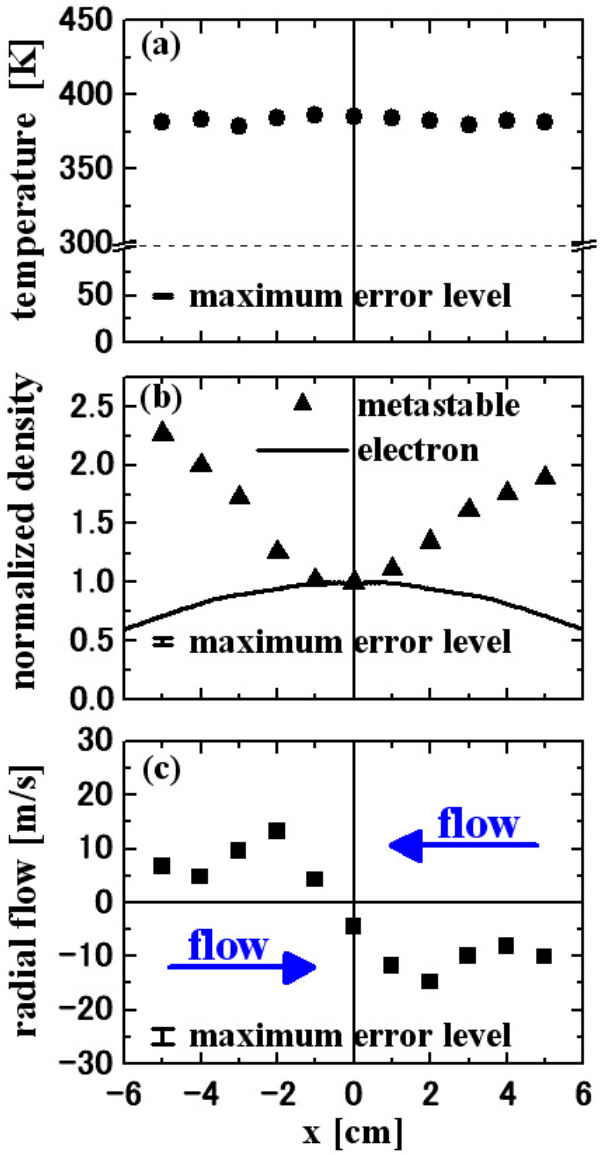

Fig. 7 Metastable temperature, density, radial flow and electron density profile. The flow direction of metastable (neutral) particles is inward. The metastable and electron density are normalized at x = 0, respectively. The electron density at x = 0 is about 4.8×10^{10}cm^{-3}

causes Doppler shift of absorption spectrum, which is expected to be of the order of 1 m/s and is the same as the velocity resolution of the system. On the other hand, the Lamb dip in saturated absorption spectrum remains unchanged even when the experimental condition has been changed. The LIF peak deviation during 5 hours is within 5 fm, indicating long term stability (Fig. 6(b)).

The temperature, density and radial flow have been measured with the newly-developed LIF system (Fig. 7). Since the LIF spectrum is well fitted by a Maxwellian distribution, the temperature, density n^* and flow are evaluated from FWHM, amplitude of the fitted Maxwellian distribution function and Doppler shift, respectively. In the present experimental condition, the component of ionizing

plasma is much larger than that of recombining component. [16] Thus, the plasma is considered to be in corona equilibrium, in which almost all of the metastable atoms are excited from the ground state level. [17] Therefore, the simple evaluation of neutral density is possible by dividing the LIF intensity by the electron density, since the metastable atoms is mainly excited by electron-neutral (ground state) collisions;

$$I \propto n^* \propto < \sigma v_{en} > n_n n_e, \tag{3}$$

where I, σ, v_{en} are LIF intensity, electron-neutral collision cross section and relative velocity of electrons to neutral particles, respectively.

There is a depletion in neutral density, and an inward going flow due to the density gradient is present in the plasma (Fig. 7(b),(c)). The neutral temperature is slightly higher than the room temperature and is about 400 K. The experiment measurement been done 5 times at each radial position. The error levels in Fig. 7 show the maximum standard deviation and are small. The reason is that the newly-developed LIF system has high stability and the reproducibility of HYPER-I plasma is also very good. In addition, the signal to noise ratio is high enough so that the LIF spectrum is well fitted by Maxwellian distribution, because there is no spectral line in the vicinity of the observing fluorescence (826.67 nm).

4. Conclusions

We have developed a high resolution neutral flow measurement system utilizing LIF and saturated absorption spectroscopy. The accuracy of this system is about 2 m/s and is one order of magnitude higher than that with iodine absorption spectrum. This result shows that the LIF Doppler velocimetry with saturated absorption spectroscopy is a very powerful tool for measuring slow neutral flow.

The temperature, density and radial flow have been measured by using the newly developed LIF spectroscopy system. The profile of neutral density is estimated from the LIF intensity and the electron density. It is found that there is a depletion in neutral density at the center of the plasma, and also found that there is a radially inward flow of neutrals induced by the density gradient. A slow neutral velocity has been measured for the first time by using the LIF system with saturated absorption spectroscopy. The considerations of other excitation and deexcitation processes are needed for the accurate evaluation of the neutral density more accurately. The measurement of absolute neutral density is left for future work.

It is worth mentioning that the error of velocity measurement may increase in a plasma under high input power cases, in which the electron density increases and the resultant collisional deexcitation reduces the number of metastable atoms, substantially. In these cases, further improvement of collection optics would be needed. We have observed the LIF signal in HYPER-I device up to 10 kW input cases.

The relationship between ion temperature and neutral temperature is important for estimating the effect of charge exchange collisions because the ions transfer their thermal energy to the neutral particles through ion-neutral collisions. The measurement of ion temperature is now undergoing.

Acknowledgment

The work is supported by the Ministry of Education, Science, Sports and Culture, Grant-in-Aid for Scientific Research (A), 19204057 and collaboration programs with NIFS (NIFS06KOAP016). The author (S. E) would like to thank ECW group members for the useful discussions and technical advices.

[1] M. Hori, M. D. Bowden, K. Uchino, K. Muraoka and M. Maeda, J. Vac. Sci. Technol. A, **14**, 144(1996).
[2] H. Abada, P. Chabert, J. P. Booth and J. Robiche, J. Appl. Phys., **92**, 4223(2002).
[3] A. Okamoto, K. Hara, K. Nagaoka, S. Yshimura, J. Vranjes, M. Kono and M. Y. Tanaka, Phys. Plasmas, **10**, 2211(2003).
[4] J. Vranjes, A. Okamoto, S. Yoshimura, S. Poedts, M. Kono and M. Y. Tanaka, Phys. Rev. Lett, **89**, 265002(2002).
[5] R. Engeln, S. Mazouffre, P. Vankan, I. Bakker and D. C. Schram, Plasma Sources Sci. Technol. **11**, 595-605(2001).
[6] R. A. Stern and J. A. Johnson Ⅲ, Phys. Rev. Lett., **34**, 1548-1551(1975).
[7] M. Y. Tanaka, R. Nishimoto, S. Higashi, N. Harada, T. Ohi, A. Komori and Y. Kawai, J. Phys. Soc. Jpn., **60**, 1600-1607(1991).
[8] D. O' Connell, T. Gans, D. L. Crintea, U. Czarnetzki and N. Sadeghi, J. Phys. D: Appl. Phys, **41**, 035208(2008).
[9] J. E. Chilton, J. B. Boffard, R. S. Schappe and C. C. Lin, Phys. Rev. A, **57**, 267-277(1998).
[10] J. B. Boffard, G. A. Piech, M. F. Gehrke, L. W. Anderson Phys. Rev. A, **59**, 2749-2763(1998).
[11] W. Demtröder: *"Laser Spectroscopy: basic concepts and instrumentation"* (Berlin, Springer-Verlag, 1996) p.484-500.
[12] V. S. Letokhov and V.P. Chebotayev : *"Nonlinear Laser Spectroscopy"*, Springer Ser. Opt. Sci., **4** (Springer, Berlin, Heidelberg 1997).
[13] V. Bakshi and R. J. Kearney, J. Quant. Spectrosc. Radiat. Transfer., **42**, 405-413(1989).
[14] H. R. Griem, Phys. Rev., **128**, 515-522(1962).
[15] G. Marr, *"Plasma spectroscopy"* (Elsevier, New York, 1968), p.246.
[16] T. Fujimoto, J. Phys. Soc. Jpn., **47**, 273(1979).
[17] H. R. Griem, *"Principles of Plasma Spectroscopy"* (McGraw-Verlag, berlin, 1981), p.158.