Thermal Plasma Diagnostics Using Tunable Dye Laser (Report I)†

— Resonance Fluorescence in a High Power Microwave Discharge —

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Abstract

Laser-induced fluorescence method (LIFM) was applied to a nearly thermal high density plasma ($n_e = 10^{13} - 10^{15}$ cm^{-3}) of hygrogen gas. Resonance scattering was measured on the H_α line and the saturation curves of the fluorescence were obtained for various electron densities. The power broadening of the scattered signal was clearly observed at fully saturated conditions. From the analysis of this phenomenon, it was possible to select out the contribution of the homogeneous broadening such as Stark one in the emission spectrum without laser pumping, even when the spectrum was governed by the Doppler (inhomogeneous) broadening.

KEY WORDS: (Thermal Plasma) (Tunable Dye Laser) (Resonance Fluorescence) (Power Broadening) (Saturation Power) (Stark Broadening)

1. Introduction

With the invention of laser it had inherently a great possibility to be applied to many kinds of diagnostic usages. With the first practical solid laser (such as Ruby or YAG laser), Rayleigh scattering, Thomson scattering and interferometic measurement were greatly developed in the spectroscopic application. By the later development of tunable dye-laser on the commercial base, further tricky diagnostic methods have been applicable in various research fields. They include near-resonant Rayleigh scattering[1], coherent anti-Stokes Raman scattering (CARS)[2], laser-induced fluorescence method (LIFM)[3] and so on. From LIFM Doppler-free spectroscopy[4] and Doppler-free polarization spectroscopy[5] have also been developed and studied. Among these methods LIFM is very simple and easy to diverse to other methods. This method is based on the measurement of the fluorescence emitted from sample or object by pumping atoms at a resonant frequency with a tunable laser. Not only population density of ground and excited states of atoms but also its velocity distribution can essentially be measured directly with a very high sensitivity. It is usually applied in the chemical analysis to determine a small bit of composition included in the sample[6]. From the life-time measurement of the fluorescence, various transition processes between excited levels are also studied repeatedly[7]. In the study of a high temperature plasma for nuclear fusion, it has also become important[8] to make clear the behavior of impurities in the edge rigion of the plasma and the solid wall.

Plasma properties typically used in the welding research is said to be nearly in the local thermodynamic equilibrium (LTE) state with high electron and neutral particle densities. Application of LIFM to this type of thermal plasma is quite rare[9] to our knowledge. To make clear if LIFM can be a favorable diagnostic method to a high density nearly thermal plasma, we began to study a hydrogen plasma near around atmospheric pressure, by pumping atoms at the H_α line with dye laser. In a high density plasma such as an arc or HF plasma, temporal decay of the fluorescence after pumping is so rapid that it is usually hard to obtain correct information from the decay curve of the fluorescence because of the limited time response of the detection system. However, informations on the decay process may also be included in the spectral broadening of the fluorescence and we made our study devoted to the measurement of spectral profile by changing the laser wavelength. Moreover it is well accepted that the scattered spectrum is influenced by

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the intensity of the pumping laser, which is studied as "power broadening" in the gain characteristics of laser oscillation and amplification[10]. We have expected here to make use of this broadening mechanism to estimate the width of a homogeneous broadening such as Stark one in a plasma, even when emitted line spectrum from the plasma is originally governed by Doppler (inhomogeneous) broadening. It will make us know the electron density by measuring a widely broadened spectral profile of a line by the strong laser pumping.

In the first report experimental results are given of the resonance fluorescence of the H_α line in a high density hydrogen plasma, produced by a high power microwave around atmospheric pressure. Widely broadened spectral profile is obtained with a high accuracy and the estimation of the homogeneous half-width of the line without laser pumping was confirmed to be promising.

2. Experimental apparatus and methods

Figure 1 shows schematic diagram of plasma apparatus[11]. It is composed of a closed gas circulating system which, after evacuation below 1 Pa, is filled with H_2 and He gas mixture at the prescribed pressure (0.1-1 atm). The working gas is circulated by the compressor C through a flow meter FM. The plasma is produced in a quartz pipe Q of 40 mm in diameter. The axially symmetric vortex gas flows in this pipe and the steady-state plasma is produced axially by a CW-microwave ($P_i \lesssim 20$ kW, $f = 915$ MHz) from a rectangular waveguide WG. The heated gas is cooled by the heat exchanger HE and again compressed by C.

Typically plasma diameter D_p is 2-5 mm and we can obtain electron density n_e of 10^{13}—10^{15} cm^{-3} by changing the gas pressure p_0 and/or the mixing ratio γ of H_2 and He. Plasma temperature T is measured to be 7000-8000 K and the plasma is nearly in LTE state[11].

Figure 2 shows schematic diagram of diagnostic system. We choose the H_α line at λ=6563 A for LIFM. Scattered signal or fluorescence light from the plasma enters into the monochromator through the observation window at the central part of the plasma. The observation is performed perpendicularly to the incident axial laser beam.

We use two monochromators; the one has 25 cm in focal length and the other 1 m. **Table 1** shows their specifications. The former is used mainly for the measurement of the total intensity of the scattered light and the latter of spectral profile of spontaneously emitted light from the plasma without laser pumping.

The dye laser (FL-2000, Lambda Physik) is excited by XeCl Excimer laser (EMG-100, Lambda Physik), and the dye used is DCM. The laser beam with the spectrum

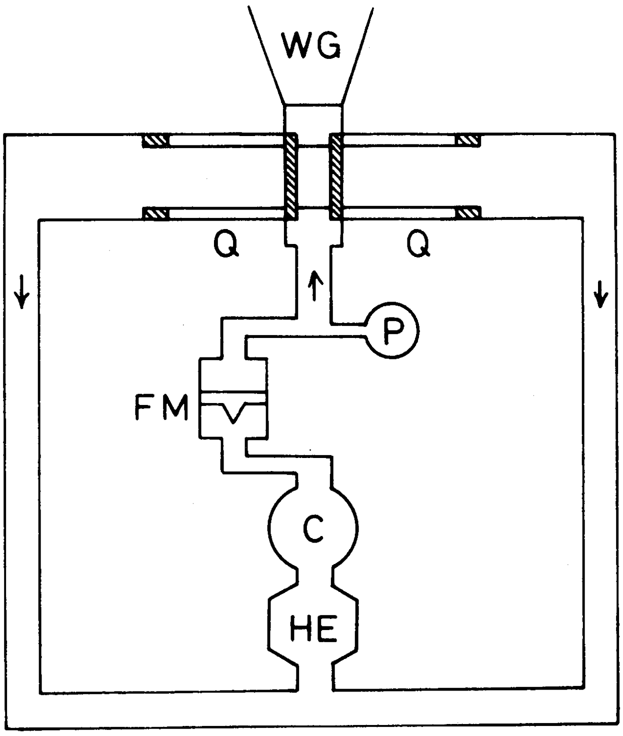

Fig. 1 Schematic diagram of plasma apparatus.

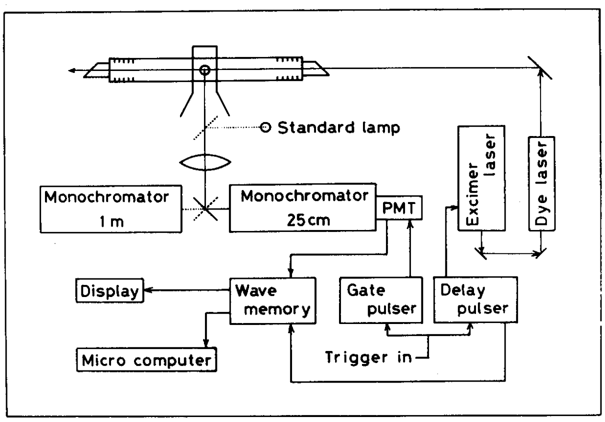

Fig. 2 Schematic diagram of diagnostic system.

Table 1 Specifications of monochromators.

Type	Focal length (cm)	1st order dispersion (A/mm)	Blaze (A)	Grating (grooves/mm)
JE-25E	25	32	5000	1180
SG-10BS	100	4.8	4500	2000

centered around the H_α line is injected into the plasma column in the axial direction. On both ends of the plasma region are set beam dampers of 6 mm in inner-diameter and 24 cm in length. The laser pulse lasts for about 20 nsec, and its dimension is limited by a rectangular slit of 4 mm x 6 mm before entering into the plasma region.

Scattered signal is detected and displayed by the wave memory (DM-901, Iwatsu) through the monochromater (JE-25E, NJA) and a photomultiplier tube (R955, Hamamatsu TV). The wave memory has a minimum sampling time of 10 nsec. The time resolution of the diagnostic system is limited to be 10 nsec by the sampling time of the wave memory.

3. Result and Discussion

Figure 3 shows a typical time variation of the scattered signal. It is detected with the 25 cm monochromator (JE-25E). The entrance slit of the monochromator is 500 μm in width, and the exit is selected to be 2 mm in width to obtain total intensity of the scattered signal. Clearly we can obtain the fluorescence with a good S/N ratio, but it is difficult to estimate its temporal behavior by the limited time resolution of the detection system and by strong collisional quenching caused by the high electron density of the plasma.

We here checked the spectral width of the dye laser around λ = 6563A with the 1 m monochromator (SG-10BS, Mizojiri). Entrance and exit slit widths were fixed to be 20 μm and the wavelength reading was fixed at the central wavelength of the H_α line from a commercial hydrogen discharge lamp (HSL-H-1, Toshiba). The laser was incident on to the monochromator directly after strong attenuation in the intensity with a ND-filter. By changing the wavelength of the dye laser, the spectral profile was recorded on an X-Y recorder. The result is shown in **Fig. 4**. The spectral profile is broadened widely with three peaks, and the spectral resolution of the monochromator is measured to be 0.1 A with a Cd-lamp (SL-Cd -1, Toshiba). Since spectral width of the dye laser is specified to be 0.4 cm^{-1} (0.1A around 6563A), we should think that the broad spectrum obtained is made by any misalignment of the laser system from its optimum condition. In this study we have performed experiments with this characteristics uncorrected.

Figure 5 shows saturation curves of the scattering intensity at various gas pressures and the laser wavelength

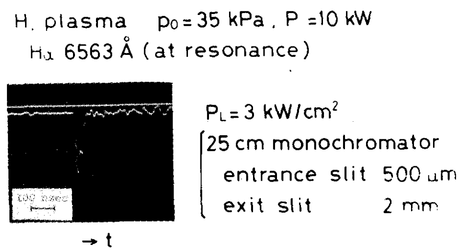

Fig. 3 Typical time variation of the scattered signal.

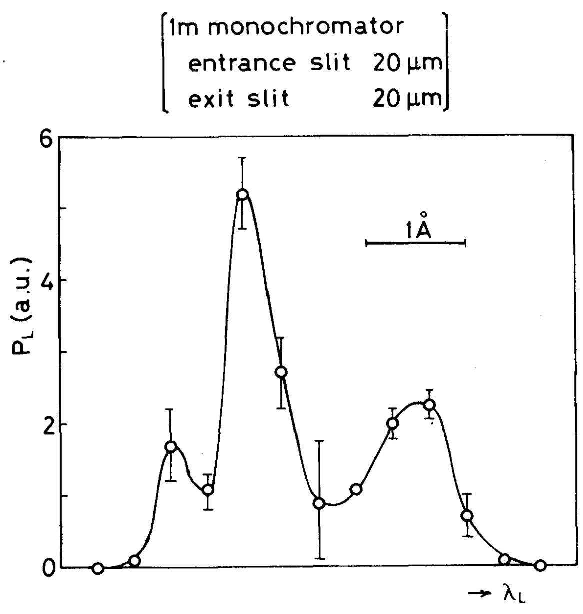

Fig. 4 The spectral profile of the dye laser around the H_α line obtained by changing the laser wavelength.

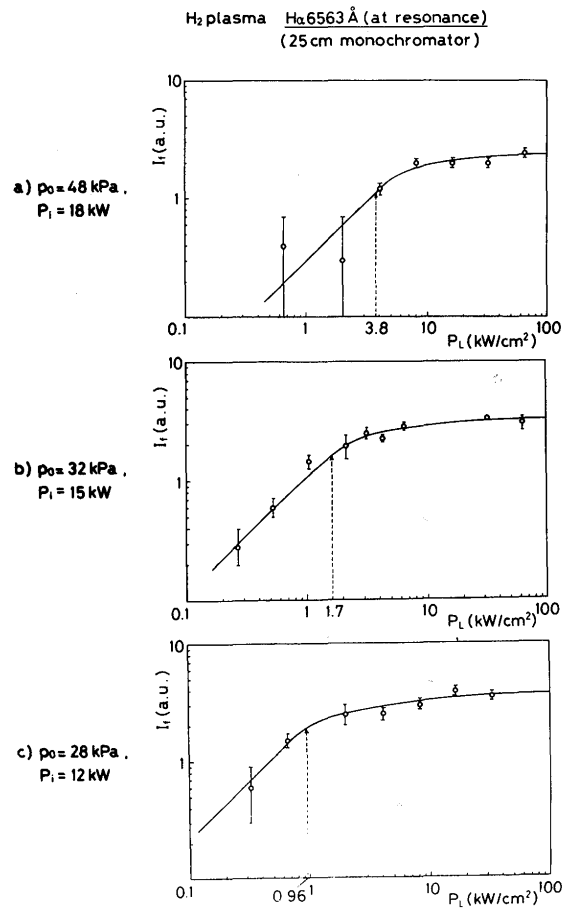

Fig. 5 Saturation curves of the scattered intensity at various gas pressures.

is the same as the one in Fig. 3 and the laser wavelength is fixed at $\lambda = 6563A$. Although the intensity is represented by the peak value in the time variation shown in Fig. 3, we have made certain that the time integrated signal also shows the same tendency as in Fig. 5. The saturation power density P_s of the laser is here defind[10] as the value at which the intensity becomes 1/2 of I_m which is the constant value at a very strong laser intensity. It is clear that the saturation power density P_s decreases with the gas pressure p_0. This tendency reflects the variation of electron density with pressure. For example we have $P_s = 3.8$ kW/cm^2 in a) of the figure and in this case the electron density n_e is decided to be 1.2×10^{15} cm^{-3} from Stark broadening of the H_γ line emission ($\lambda = 4340A$). Burges et al. have calculated the dependence of P_s on n_e for the H_α in a hydrogen plasma[9] and P_s was estimated to be 2.5 kW/cm^2 at this electron density. In their calculation the spectral line width of incident resonant laser is assumed to be the one of the background H_α line. In this study, the laser has a spectral width of about 3 A as stated earlier, and the background H_α line has about 1 A in its width. So that on-third of P_s in Fig. 5 a) should correspond to the exact saturation power density. Thus we obtain the actual P_s is $3.8/3 = 1.3$ kW/cm^2 which agrees roughly with the result of Burges et al. The difference of a factor 2 will come from the uncertainty in the evaluation of the laser spectral width.

Figure 6 shows spectral variations of the scattered intensity obtained by changing the laser wavelength λ_L at the same condition with Fig. 5 c). They become wider with the increase in the laser power density P_L. Here we should remark, that at $P_L \gtrsim 8$ kW/cm^2 flat region appears in the central part of the profiles, and also even at a very small value of P_L below saturation the spectral width remains to be 3A. Indeed this is because the dye laser has a spectral width about 3A as already described and the flat region of 3A in width remains when the saturation condition is fulfilled. So that below we pay attention to the spectral profiles at the fully saturated conditions subtracting the flat part of the profile.

Figure 7 shows a full-logarithmic plot of a scattered intensity versus laser wavelength λ_L in a fully saturated condition. It agrees well with a typical Lorentzian line shape drawn in the figure, and the mechanism of the spectral broadening by the laser power is considered to be the well known power broadening in quantum electronics.

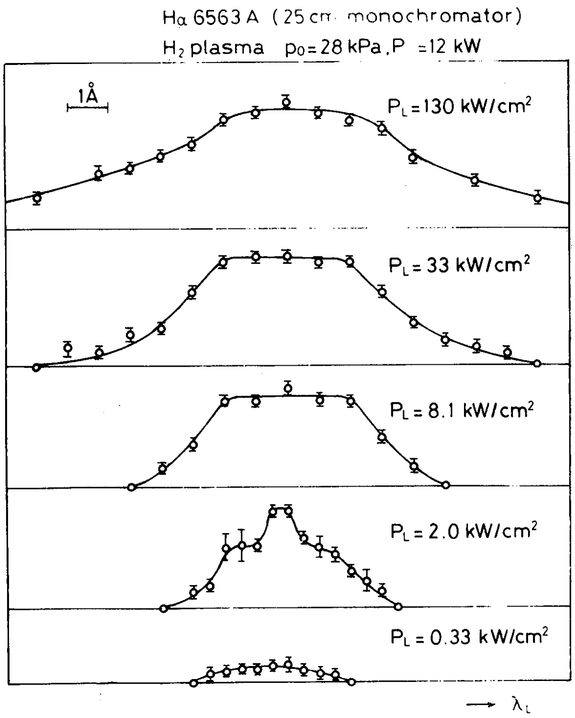

Fig. 6 Spectral profiles of the scattered intensity by changing the laser wavelength.

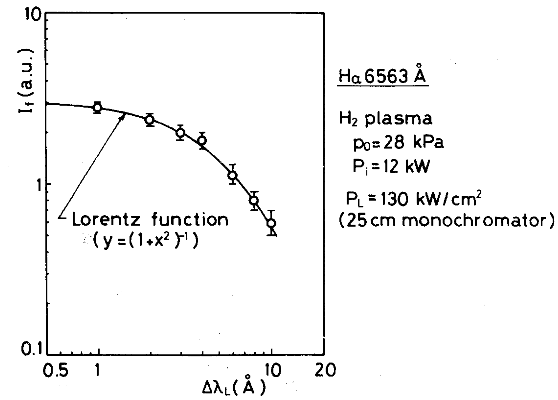

Fig. 7 Full-logarithmic plot of scattered intensity versus laser wavelength in a fully saturated condition.

As stated by Carlsten et al.[11], scattered signal at resonance or near-resonance from a highly collisional plasma and/or gas is mainly composed of two parts. The one is from near-resonant Rayleigh scattering, and the other is from collision-induced fluorescence. Since the former is usually a poralized radiation[1], we have examined the contribution of near-resonant Rayleigh scattering to our experimental data. By rotating polarization direction of the incident dye laser with $\lambda/4$ plate and Glan-Thompson prism, we measured the spectral profile as shown in **Fig. 8**. As we can clearly see the rotation gives negligible influence on the intensity and the shape of the scattered signal. So that we may conclude that detected signal in our experiment is composed of only collision-induced fluorescence because of a strong collisional process by electrons and/or neutral particles.

So far widely broadened spectral profile obtained in this experiment was considered to be due to the power

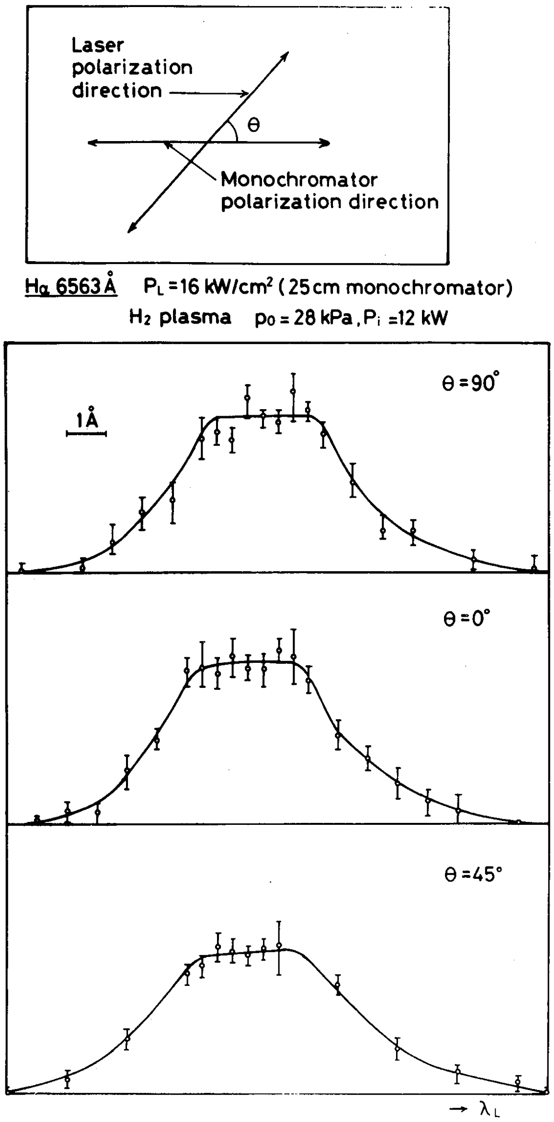

Fig. 8 Spectral profiles of the scattered intensity at various laser polarization directions.

broadening. The spectral width by this broadening mechanism is usually given by[10)]

$$(\Delta\lambda/\Delta\lambda_0) = (1 + P_L/P_s)^{1/2}, \qquad (1)$$

where $\Delta\lambda$ is the power-broadened; half-width (FWHM), and $\Delta\lambda_0$ is the homogeneously broadened half-width (FWHM) before laser pumping. From this equation it is possible to estimate $\Delta\lambda_0$ by measuring $\Delta\lambda$ and if $\Delta\lambda_0$ is given by a known function of electron density n_e, we can estimate n_e through $\Delta\lambda$, P_L, and P_s.

Figure 9 shows experimental result on the relation of $\Delta\lambda$ with $\Delta\lambda_0$. In a) of the figure the gas pressure p_o is 48 kPa and it corresponds to that in Fig. 5 a). The data is curve-fitted to the function $y = \sqrt{1+x}$, where x and y are $(\Delta\lambda/\Delta\lambda_0)$ and (P_L/P_s), respectively. Experimental result

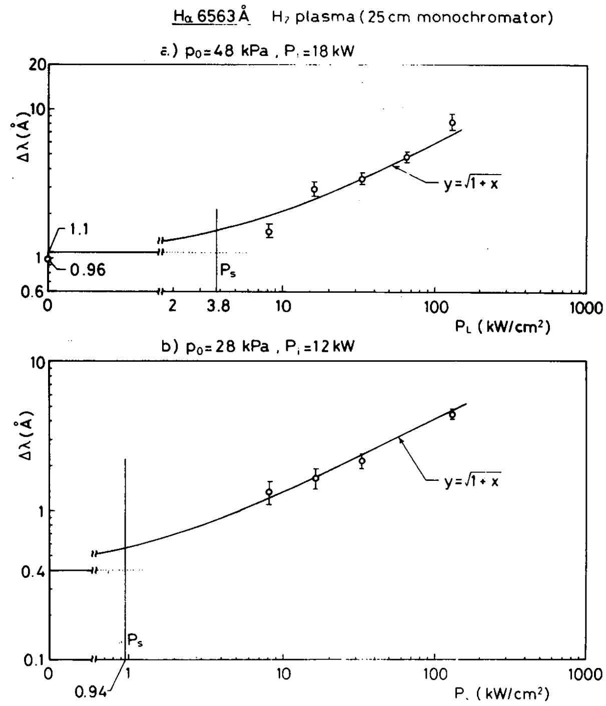

Fig. 9 Dependence of power-broadened half-width Δλ on the laser power density P_L.

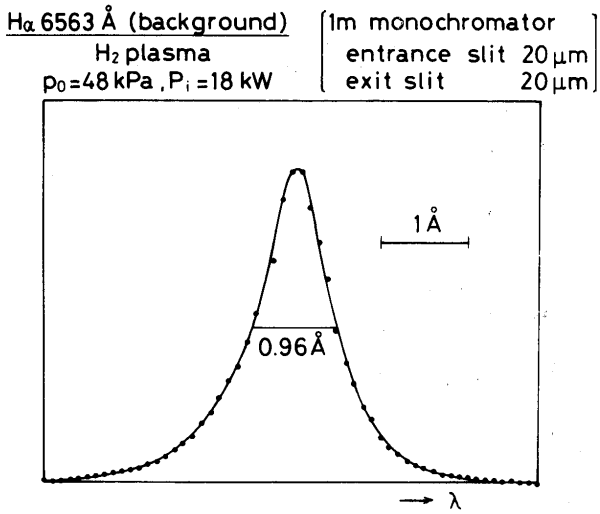

Fig. 10 Spectral profile of the H_α line without laser pumping.

is fitted with theoretical curve and the homogeneously broadened half-width $\Delta\lambda_0$ is estimated to be 1.1 A at $(P_L/P_s)\to 0$. While spontaneous H_α line emission from the background plasma has a typical Stark (homogeneous) broadening as shown in Fig. 10. Its half-width is 0.96 A and it agrees rather well to the one estimated from the curve-fitting, although the convolution of the Stark and the Doppler profiles should be checked further. In case of p_0 = 28 kPa corresponding to the data in Fig. 5 c) and Fig. 6, we obtain Fig. 9 b). In this case, the background

H_α line spectral profile is measured to have a Gaussian shape of Doppler (inhomogeneous) broadening, but from the curve-fitting in the figure we can estimate the contribution of the homogeneous broadening to be 0.4A.

It is known that the saturation power density P_s in Eq. (1) varies in accordance with the half-width $\Delta\lambda_0$ before pumping in a high density colision-dominant plasma[9]. Besides, under the condition $P_L >> P_s$, the equation has a relation $(\Delta\lambda/\Delta\lambda_0)^2 \propto (P_L/P_s)$. When we fix P_L, $(\Delta\lambda/\Delta\lambda_0)^2 \propto (1/P_s)$ is obtained. So that

$$(\Delta\lambda/\Delta\lambda_0)^2 \propto (1/\Delta\lambda_0) \qquad (2)$$

is satisfied. We have performed further experiment to confirm this relation.

In **Fig. 11** is shown the dependence of $\Delta\lambda_0$ on H_2 and He mixing ratio γ at p_0 = 100kPa. $\Delta\lambda_0$ was estimated as the half-width in arbitrary unit of the Stark broadening from the electron density in ref. 11. Also shown is the power broadened half-width $\Delta\lambda$ obtained in this study at p_0 = 28 kPa. Although the gas pressure is different in the estimation of $\Delta\lambda_0$ with the present experiment, it is made certain that n_e increases nearly proportionally to the gas pressure. So that relative variation of $\Delta\lambda_0$ with γ can be considered to be the same even at p_0 = 28 kPa and we may use these $\Delta\lambda_0$ and $\Delta\lambda$ to check the validity of Eq. (2). The result is given in **Fig. 12** as the full-logarithmic plot of the relation between $(\Delta\lambda/\Delta\lambda_0)^2$ and $\Delta\lambda_0$. The data points coincide appropriately with a line of negative slope whose gradient is −1, confirming the relation of Eq. (2). Thus we can conclude that the spectral

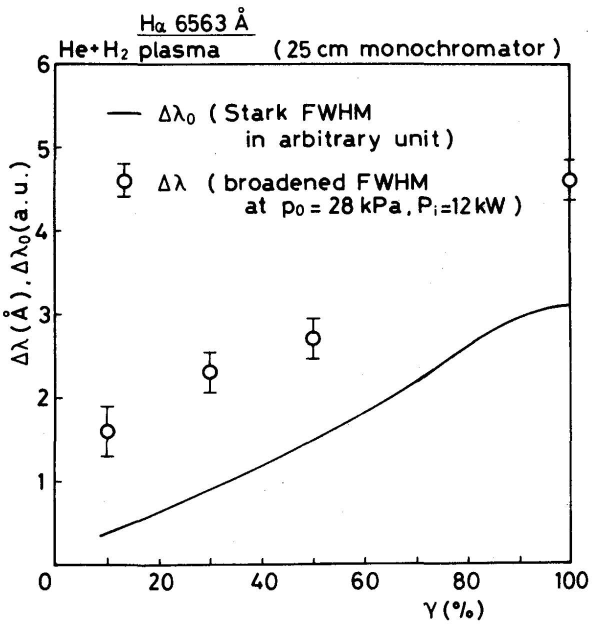

Fig. 11 Dependence of $\Delta\lambda_0$ from the Stark broadening and $\Delta\lambda$ by power broadening on H_2 and He mixing ratio γ.

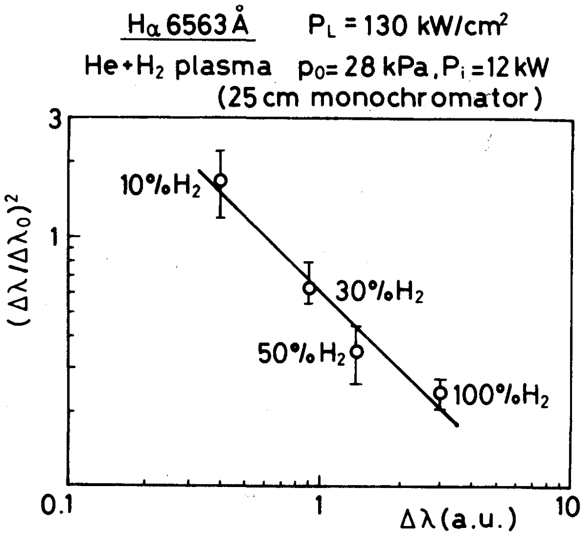

Fig. 12 REelation between $(\Delta\lambda/\Delta\lambda_0)^2$ and $\Delta\lambda_0$.

width by power broadening reflects well the one by Stark broadening in a high density plasma through Eqs. (1) and (2), and application of power broadening to the determination of n_e seems to be promising, even when the Stark broadening is smaller than the Doppler one.

4. Conclusion

By using a tunable dye laser, laser-induced fluorescence was measured on the H_α line (λ= 6563 A) from a steady-state hydrogen plasma produced by a high power microwave.

LIFM was made clear to be applicable to a thermal plasma diagnostics with n_e = 10^{13} -10^{15} cm^{-3} under experiment. The scattered signal had a strong collisional quenching caused by the high electron density of the plasma, and it was composed only of collision-induced fluorescence and near-resonant Rayleigh scattering was negligible.

The fluorescence intensity showed a saturation with the pumping laser power density and the saturation power density increased with the electron density as the theory predicted.

Widely broadened spectral profile of the fluorescence was obtained at a fully saturated condition and it had a typical Lorentzian shape. The broadening mechanism was the well known power broadening and it influenced on the homogeneous (Stark) profile of the spontaneous H_α line emission without laser pumping and the inhomogeneous (Doppler) profile was of no importance to the broadening mechanism.

By appropriately analyzing the power broadening the Stark width of the plasma could be estimated, even when the original line emission without laser pumping was governed by the Doppler effect.

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