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QUENCHING OF EINSTEIN A-COEFFICIENTS IN PLASMAS AND LASERS

BY

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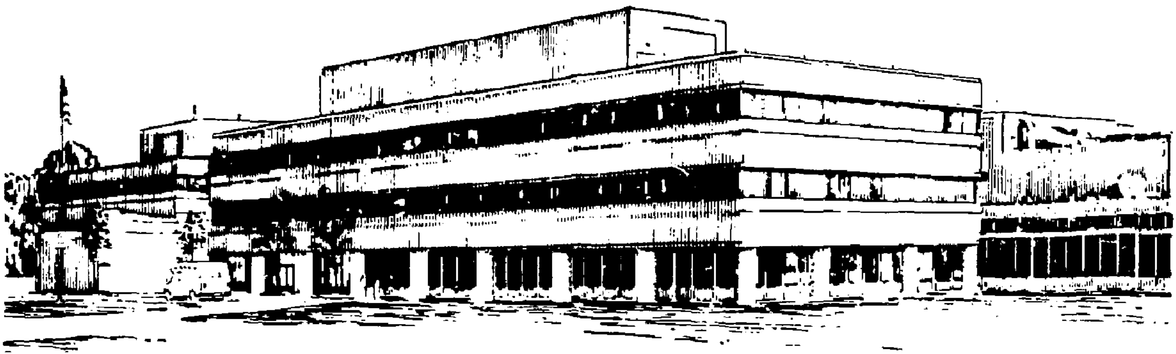

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Quenching of Einstein A-Coefficients in Plasmas and Lasers *

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Abstract

The coefficient of spontaneous emission (Einstein A-coefficient) is considered to be one of the basic constants of a given transition in an atom or ion. The formula for the Einstein A-coefficient was derived in the pioneering works of Weisskopf and Wigner (WW) based on Dirac's theory of light. More recently, however, it was noted in several papers that the rate of spontaneous radiative decay can deviate significantly from the WW expression in certain conditions, for example in a laser cavity.

A different type of change in A-coefficients was inferred from measurements of changes in the intensity branching ratio of spectral lines in a plasma. A change of branching ratio of up to a factor of 10 was observed in CIV for 3p-3s (580.1-581.2nm) and 3p-2s (31.2nm) transitions when the electron density changed from approximately $N_e \approx 1\text{x}10^{18}$ to $5\text{x}10^{18}$ cm^{-3}. This effect was also observed in CIII and NV. An initial theoretical approach to the problem based on the integration of the Schrödinger equation with the ion Coulomb potential modified by the electron cloud within the Debye radius was unsuccessful in predicting the experimental observations.

The effect of quenching of spontaneous emission coefficients was observed also in an Ar-ion laser as a function of the intracavity power density (photon density) for lines originating from the same upper level as the lasing line. Measurements of these line profiles absorption for different lasing conditions and related discussions are also presented.

I. Introduction

For many years we believed, based on work by Weisshopf and Wigner[1], that spontaneous emission coefficients (Einstein's A-coefficients) are constant for quite a wide range of conditions in plasmas and lasers. However a number of recent experiments indicated that atoms in laser cavities may enhance or inhibit their spontaneous emission (see e.g. Ref. 2).

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It is believed that this effect is related to the decoupling of the radiating atom from the vacuum field as the dimension of the cavity becomes comparable with the radiation wavelength (see e.g. Ref. 3).

Another observation of changes in spontaneous emission of ions in plasmas and lasers, which seems to be caused by a different effect than above mentioned, were reported in several papers[4-7]. In the present paper we present some major points related to changes of spontaneous emission which emerged from several years of measurements of the ratios of line intensities originating from the a upper level (branching ratios). Up to now we have found that such changes of branching ratios in relatively high density plasmas[4,5] ($N_e \approx 10^{18} - 10^{19} cm^{-3}$; N_e is the electron density) and in CW Ar-ion lasers[6,7] is most likely caused by the quenching of spontaneous emission coefficients (Einstein's A-coefficients). We have considered a number of other effects which could lead to the change of line intensity branching ratios. For experiments in plasmas we have examined: (a) self absorption, (b) cut-off of the visible light in a region of high electron density where the plasma frequency is higher than visible light frequency, (c) refraction, (d) stimulated emission of XUV light, and (e) errors in measurements. All these (a)-(e) points were eliminated as a cause of the change of branching ratios in plasmas by careful experimental and theoretical analysis.

Similarly, before concluding that Einstein A-coefficients are quenched in Ar-ion lasers due to photons interacting with ions, a number of experimental points were examined such as: (a) a nonlinear response of detection system, (b) mis-identification of spectral lines, (c) broadening or shifting of lines, (d) independence of the effect from angle of polarimeter set up in front of the monochrometer, (e) the independence of transmissivity and reflectivity of all applied optics from the laser beam power, and most important, (f) no absorption change for spontaneous lines of interest with and without lasing action at constant laser discharge conditions. A number of experiments and calculations were especially dedicated to this last (f) point and it will be discussed in more detail in Section 3, whereas questions related to measurements of branching ratios in plasma will be addressed in Section 2.

II. Discussion of Branching Ratio Measurements in Plasmas

Here we will discuss some questions related to the branching ratio measurements in plasmas generated by lasers, and primarily by CO_2 lasers. We will start our discussion with branching ratio $R = I_{vis}/I_{vuv}$ measurements for 3p-3s (wavelength 5801-5812Å) and 3p-2s (wavelength 312.42-312.46Å) transition intensities in CIV ions (I_{vis}, I_{vuv} are intensities of visible and vuv lines). These transitions were chosen due to large difference in the value of its spontaneous emission coefficients of non-perturbed ions which are $0.38 \times 10^8 sec^{-1}$ and $45.6 \times 10^8 sec^{-1}$ for visible and VUV transitions, respectively[8]. The lines from these transitions are also very suitable for measurements due to the high intensities and easy accessible spectral regions. The experimental set up and details of measurements are described in Refs. 4 and 5.

The first problem which we will consider is related to the effect of background on intensity ratio measurements. In Fig. 1 from Ref. 4, one can see that with decreasing distance to the target electron density N_e is increasing. Electron density in Fig. 1 was estimated from Stark broadening of visible lines of interest using quasi-static approximation[9] $\Delta\lambda \sim N_e^{2/3}$. For our experimental conditions a more appropriate approximation might be electron-ion collision broadening leading to a linear approximation[9] $\Delta\lambda \sim N_e$, which would change the density scale in Fig. 1 (e.g. lg N_e would change from 17.32 to 17.6 at d = 1.2mm, and from 18.76 to 18.57 at 0.2mm). These changes are not very important for the general picture of A-coefficients behaviour. However an important fact is that the intensity branching ratio R decreases by almost an order of magnitude for 3p-3s/3p-2s transitions of CIV as N_e increases from approximately 1×10^{18} cm^{-3} (at d = 0.6 mm from target surface) up to approximately 4-5 x 10^{18} cm^{-3} (at d = 0.2mm). At $N_e \approx 4 \times 10^{18}$ cm^{-3} the CIV 5801Å and 5812Å line broadening reaches (FWHM) $\Delta\lambda_s \approx$ 15Å. There is a strong continuum in the visible part of spectra at these densities, whereas continuum intensity is rather smaller in the VUV as can be seen in Fig. 2a, b from Ref. 5. The visible continuum is rapidly decreasing with increasing distance from the target (Fig. 2b). In measurements of line intensities the level of background, which was subtracted,was determined from extrapolation of both wings of the line into longer and shorter wavelengths up to 5-7.5 FWHM in each wavelength direction from the line center. Because in the calculation of branching ratios we integrated the experimental line profiles, determination of the proper background level and related boundary of integration can have significant effect on accuracy of R measurement. If the line is very broad or the background is noisy the boundary of the line profile to be integrated becomes less distinct and the problem of choosing limits of the integration becomes quite difficult. However the effect of branching ratio changes usually also become very large (for properly chosen pair of lines) so integration error becomes less important. In Fig. 3 there are shown three cases of integration of measured line profiles with different limit points and a comparison with integrated Lorenzian (theoretical) line profiles for the same limit points and the same backgrounds (dashed lines connecting limit points, in upper part of Fig. 3)[5]. There is a good agreement between the integrated experimental profile of CIV 5801-5812Å lines and its approximation by Lorenzian profiles. This figure also shows that the error in estimating the integral intensity for 6xFWHM (50Å) integration limit (corresponding background is dashed line 3) in comparison with integral intensity for 15xFWHM (120Å) integration limit (background:

dashed line 1) is significantly smaller (~40%) then the observed change in branching ratio R (~300%) for these conditions.

In two recent papers[10,11] there are attempts to explain the observed changes in branching ratios as being due to blending a large part of the visible line radiation into background continuum rather than due to changes of A-coefficients. Y . Chen and J. Lebowitz[10] divided the radiating ions into two classes: those with weakly trapped plasma electrons and those without. The second class is the "regular" case with the usual quasi-static Stark and impact collisional broadening of up to 10-20Å. It is the first class of ions which, the authors made "responsible" for the measured changes in the intensity branching ratio. For these ions weakly trapped electrons ("spectator electrons") induce very large red shifts of the lines which dissolve into the background as a very broad (several hundred Å) continuum. Because this part of the line radiation would be missed in the integral intensity measurement, this would lead to a decrease in the ratio $R = I_{vis}/I_{vuv}$ for Li-like CIV ions. While this is very interesting theoretical work, the electron density in the experiment with the CO_2 laser created plasma was probably too low to justify such explanation. In addition, Chen and Lebowitz[10] model cannot account for measured changes in the branching ratios of spontaneous line intensities in Ar-ion lasers, which we believe is a manifestation of the same effect (see next Section).

Similarly H. Griem and co-workers[11] propose an explanation for the changes in the CIV branching ratio by a blending in part of the visible line radiation with continuum, which was based on their own experiment. In the experiment[11] a 4J, 20 ns ruby laser was used to produce a blow-off plasma from polyethylene cylindrical or 0.5mm thin carbon blade targets. The plasma was observed transversely from opposite directions using grazing incidence and visible monochrometers. (In such experimental set up there is a danger that instruments might "look" at slightly different directions and "see" a significantly different plasma due to a large gradients near the target surface). Near the target surface, the authors measured very different electron densities N_e, from the Stark broadening of visible (5801-5812Å) and VUV (312Å) lines of CIV. For example, for a distance d= 0.2 mm from the target visible lines broadening corresponded to $N_e \approx 6\text{x}10^{18}$ cm^{-3}, while broadening of the VUV lines corresponded to $N_e \approx 0.5 - 1 \times 10^{20}$ cm^{-3}. For $N_e \approx 0.5 - 1\text{x}10^{20}$ cm^{-3} CIV 3p-3s visible lines would be very broad (200-400Å) and dissolve into the continuum, which the authors[11] took as an explanation for the observed changes in the branching ratio of CIV ion. Of course, if this explanation is correct, one should see a similar effect for any branching ratios independent of the ratio of A-coefficients. However, in measured the branching ratios in CIII such changes were not observed (Ref. 11), although

CIII ions should be located in the plasma, close to CIV ions, due to similar ionization potentials for both ions.

In order to check if, in our major experiments with a CO_2 laser generated plasma, broadening of visible lines correspond to the same or different electron density than one of the VUV lines, we have analyzed the broadening of the 312Å and 289Å (4d-2p) lines of CIV. The entrance slit of the VUV spectrometer was 100m and both lines were measured in second order. In Fig. 4 spectra in vicinity of 600Å are shown for d=0.2 mm and d = 0.8 mm from the target surface. These spectra were obtained for conditions very similar to that in Fig. 2 and with similar change of branching ratio for CIV 3p-3s/3p-2s transitions (change by factor ~3.5).

From Fig. 4a one might see broadening of CIV 4d-2p line (second order of 289Å). At a distance d = 0.8 mm the electron density is below 10^{18} cm^{-3} and the VUV line profiles in Fig. 4b practically represent instrumental broadening with a FWHM $\Delta\lambda_{ins} \approx 0.8$Å. After subtracting $\Delta\ell_{ins}$ (Lorenzian profile) we obtained, for the second order of CIV 289Å line, a FWHM ≈ 0.41Å which corresponds to $\Delta\lambda_s \approx 0.205$Å. For the second order of CIV 312Å, the FWHM did not exceed 0.1Å ($\Delta\lambda_s \leq 0.05$Å). Using the R. Lee code[12] for calculating line profiles, we could conclude from the measured $\Delta\lambda_s$ and profile for 289Å line that this line (hence 312Å line as well) was emitted from a plasma with $N_e \leq 4\text{x}10^{18}$ cm^{-3}, which agreed quite well with $N_e \approx 2 - 3\text{x}10^{18}$ cm^{-3} estimated from visible line broadening for the same d = 0.2 mm assuming relation[11] $\Delta\lambda \sim N_e$.

The last point of the discussion in this Section is related to the measurement of the branching ratio for CIII 3d-3p (5696.0Å) and 3d-2p (574.3Å) line intensities. It was pointed out[11] that in Fig. 7 of our paper[5], the visible CIII line at 5696Å is strongly blended by an intense Aℓ III line (second component of the doublet is at 5722Å) which leads to apparent changes of the CIII branching ratio as N_e changes. Therefore we analyzed another set of data in which Aℓ-lines were absent (like e.g. in case of Fig. 2). VUV lines of CIII 574Å and second order of CIV 312Å lines (see Fig. 4) were measured simultaneously. The changes of the CIII and CIV branching ratios R = I_{vis}/I_{vuv} with increasing N_e were similar with R decreasing by factor of 3 and 4, respectively, at a distance d = 0.2 mm from the target ($N_e \approx 3\text{x}10^{18}$ cm^{-3} estimated from visible line broadening.)

III. Discussion of Branching Ratio Measurements in Ar-Ion Lasers

The observation of changes in the branching ratios in plasmas stimulated analogous experiments in the Ar-ion laser[6,7]. Assuming that changes in the A-coefficients in a plasma were caused by the disruptive effect of electron-ion collisions on spontaneous emission, it was expected that similar changes of branching ratios in lasers could be induced by photon-ion interactions. By extrapolating plasma results, one could expect about a 10% quenching effect of the ratio of A-coefficients for ArII $4p^4D_{5/2}$ - $3d^4D_{3/2}$ (399.21 nm) and $4p^4D^o_{5/2}$ - 4s $^4P_{3/2}$ (442.60 nm) lines at 5kW/cm^2 intracavity power density in an Ar-ion laser (the A-coefficient is ~50 times larger for the 442 nm line than for the 399 nm line[13]). In earlier measurements[6] the maximum change of the branching ratio for these two lines was ~7% and in more recent measurements this branching ratio change exceeds 10%, shown in Fig. 5 from Ref. 7 (Experimental arrangements are presented in Refs. 6 and 7).

Of course, the observed effect in Ar-ion lasers might not be related to the effect observed in plasmas and the above correlation may be accidental. This is the position of H. Griem who tries to explain the decrease in the branching ratios of ArII spontaneous emission lines in the Ar-ion laser by a decrease in absorption near the peak of the line intensities due to Rabi oscillations[14]. Because the Rabi frequency shift is larger for stronger lines, the author predicted a larger decrease of absorption (e.g. 399/442 the branching ratio would decrease with increasing intracavity power density due to a decrease in absorption and hence an increase in the intensity, primarily, of the 442 nm line- according to H. Griem). However there was no evidence in the any of our experiments which would support such an explanation.

First of all, we would like to stress that in our experiments, when we compare line intensities with and without lasing action, the laser discharge current, fill pressure, etc., all stay constant. Hence, only a change in the absorption with the lasing action can affect our results. This could be caused by changes in the line profile due to Rabi splitting or by collisional population of the lower levels of the spontaneous emission lines from the lower lasing level, $4s^2P_{3/2}$. However this level decays very rapidly to the ArII ground state $3p^5\ ^2P$, and its population should therefore always be small[7]. In addition, the radiative transition rates between $4s^2P_{3/2}$ level and lower non-lasing levels will be small due to the intercombination character of these transitions. Furthermore, no change in the non-lasing line intensity from the upper lasing level $4p^4D^o_{5/2}$ could be observed when tuning the laser to the 487.99 nm line, which has the same $4s^2P_{3/2}$ lower level as the 514.53 nm line. Moreover, we observed similar changes in the 399 nm/442 nm branching ratio for three Ar-ion lasers with different tube lengths of 0.9m, 1.2m and 1.6m. While the intracavity power density was the same, the opacity of the 442nm line changed from 45% for the 0.9m tube to 85% for the 1.6m tube.

Independent measurements of absorption for the 399 nm and 442 nm lines with and without lasing at 514 nm were provided in two different ways. Initial measurements were done using probing radiation from a tunable dye laser. No difference in the absorption of spontaneous emission lines was observed in the 1.6 m Ar-ion laser for on and off lasing conditions. More precise measurements (error ≈ ± 0.6%) with probing spontaneous emission from a second Ar-ion laser discharge tube (without lasing) have confirmed initial measurements that there is no change in absorption of spontaneous emission lines as a function of intracavity power density (the power density was changed by tuning the output mirror of the laser without changing any other laser parameters). In Fig. 6 are shown transmission signals for 399 nm and 442 nm lines for lasing on and off[7]. One can see that even for the largest change of our laser beam power between 0 and 10 W (10 W output power corresponds $5.5 kW/cm^2$ intracavity power density) there is no observable change in transmissivity, hence absorption, for these two lines in the 1.6m long laser discharge.

Our final remarks are related to the effect of Rabi oscillations on changes in absorption of the spontaneous emission lines. The Rabi oscillations cause a reduction in the intensity of all the spontaneous emission lines from the upper lasing level. This reduction, however, is constant for all lines and cannot in itself account for the A-value dependence of the reduction factors. The Rabi oscillations also cause splitting of the emission line profiles which can change the opacity. Since the 442 nm line is more strongly absorbed in the discharge than the 399 nm line, a reducdon in opacity could preferentially increase the emergent 442 nm line compared to the 399 nm line and account for the observed change of branching ratio without the need to postulate quenching of A-coefficients. As discussed above, we did not detected any change in integral line opacity due to Rabi splitting. Because the largest change in the line opacity would be expected in the line center, we performed high resolution line shape measurements using a piezo-electrically driven Fabry-Perot interferometer located behind the exit slit of the monochrometer. Measurements of the ArII 442.6 nm line profiles had shown that there is no difference in its shape for non-lasing and lasing of the 514.5 nm laser line. These measurements clearly demonstrated, that any change in the line profiles due to Rabi splitting are negligibly small, which should not change line opacity. Very recently, additional prove that the observed change in branching ratio of spontaneous emission lines is not related to the change of its absorption was obtained by conducting measurements in a very short (ℓ = 4.5 cm) discharge tube. This discharge tube was placed inside the Ar-ion laser cavity and by using system of splitters and mirrors it was possible to measure the intensity of the 399 nm and 442 nm lines on the axis of this tube without interference of radiation from the 1.6 m laser discharge. Discharge current and gas pressure were similar to those in an Ar-ion laser. Although absorption along the axis of the 4.5 cm discharge was negligible small, the quenching effect was similar to that observed in the Ar-ion laser at the same power density. (A more detailed description of this experiment will be published elsewhere).

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Figure Captions

Fig. 1 Branching ratio $R = I_{vis}/I_{VUV}$ for CIV 5801-5812Å and 312Å lines as a functions of distance d and electron density for target widths w = 0.35, 0.42, 1.0 and 4.1 mm.

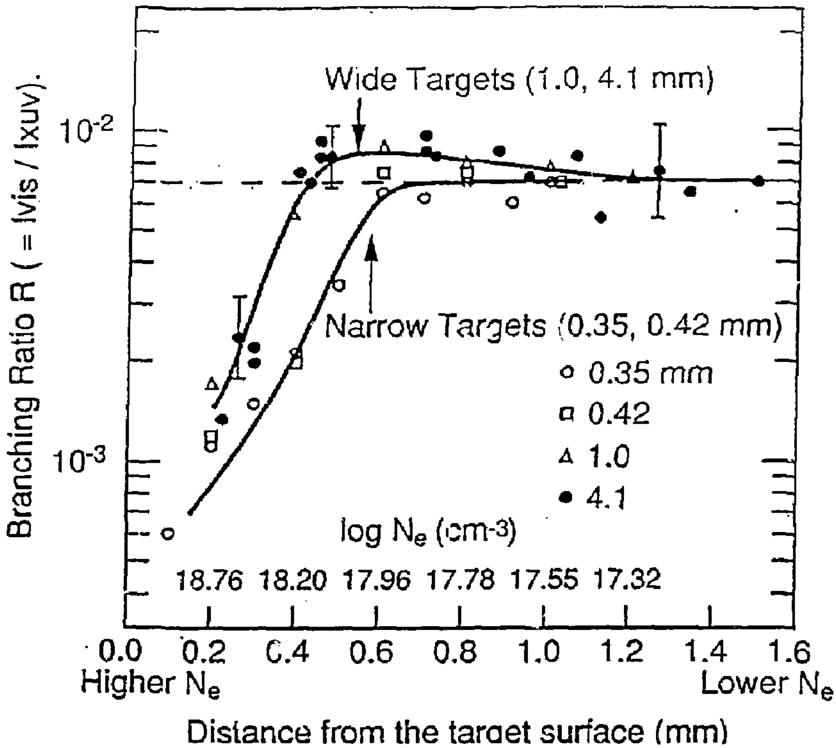
Fig. 2 VUV and visible spectra of CIV taken simultaneously at distances of (a) d = 0.2 mm and (b) d = 0.4 mm from the target surfaces. The upper ones are VUV spectra and the lower ones are visible spectra.

Fig. 3 Estimation of error in integrating the line profile of CIV 5801-12 Å from Fig. 2a.

Fig. 4 VUV spectra in vicinity of second order of CIV 289 Å and 312Å lines at distance (a) d = 0.2 mm and (b) d = 0.8 mm.

Fig. 5 Ratio of the spontaneous emission intensities of the 399.2 nm to the 442.6 nm transition as a function of intra-cavity laser (514.53 nm) power density P (The signal ratio has been normalized to 1 at P =0).

Fig. 6 Transmitted fraction of the probing radiation as a function of time for (a) 399.2 nm and (b) 442.6 nm lines. No change in signal is notable when the argon ion laser is tuned to lasing action at 514.53 nm. [in (a) the two straight lines correspond to a ± 1% region around the average value].



Wide Targets (1.0, 4.1 mm)
Narrow Targets (0.35, 0.42 mm)
0.35 mm
0.42
1.0
4.1
log Ne (cm-3)
18.76 18.20 17.96 17.78 17.55 17.32
Branching Ratio R (= Ivis / Ixuv).
10-2
10-3
0.0 0.2 0.4 0.6 0.8 1.0 1.2 1.4 1.6
Higher Ne
Lower Ne
Distance from the target surface (mm)

Fig. 1

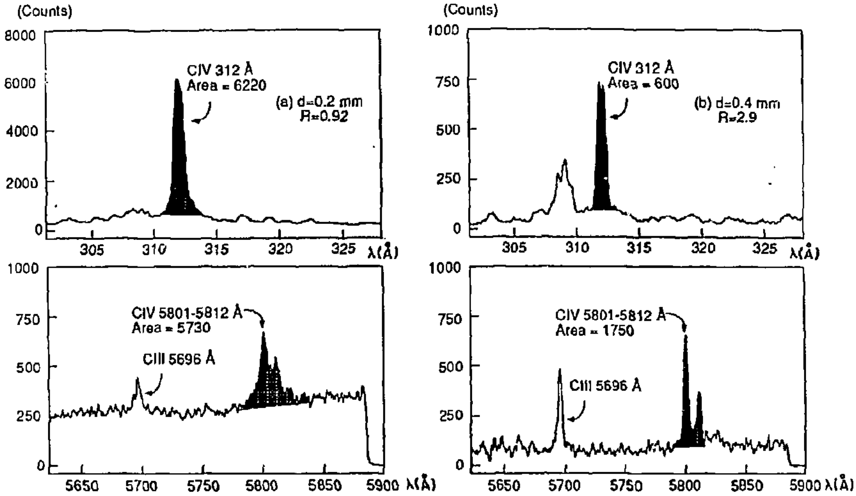

(Counts)
CIV 312 Å
Area = 6220
(a) d=0.2 mm
R=0.92
CIV 312 Å
Area = 600
(b) d=0.4 mm
R=2.9
CIV 5801-5812 Å
Area = 5730
CIII 5696 Å
CIV 5801-5812 Å
Area = 1750
CIII 5696 Å
λ(Å)

FIG. 2

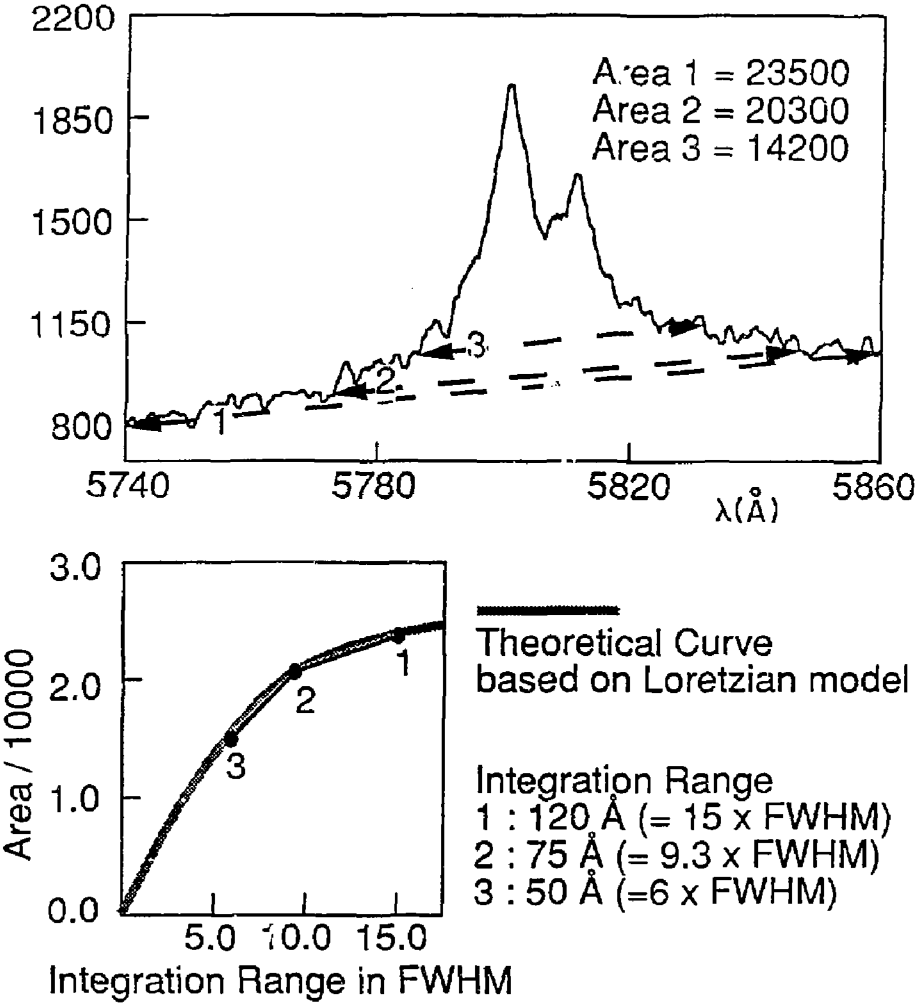

2200
1850
1500
1150
800
Area 1 = 23500
Area 2 = 20300
Area 3 = 14200
1
2
3
5740
5780
5820
5860
λ(Å)
3.0
2.0
1.0
0.0
Area / 10000
5.0
10.0
15.0
Integration Range in FWHM
Theoretical Curve
based on Loretzian model
Integration Range
1 : 120 Å (= 15 x FWHM)
2 : 75 Å (= 9.3 x FWHM)
3 : 50 Å (=6 x FWHM)

FIG. 3

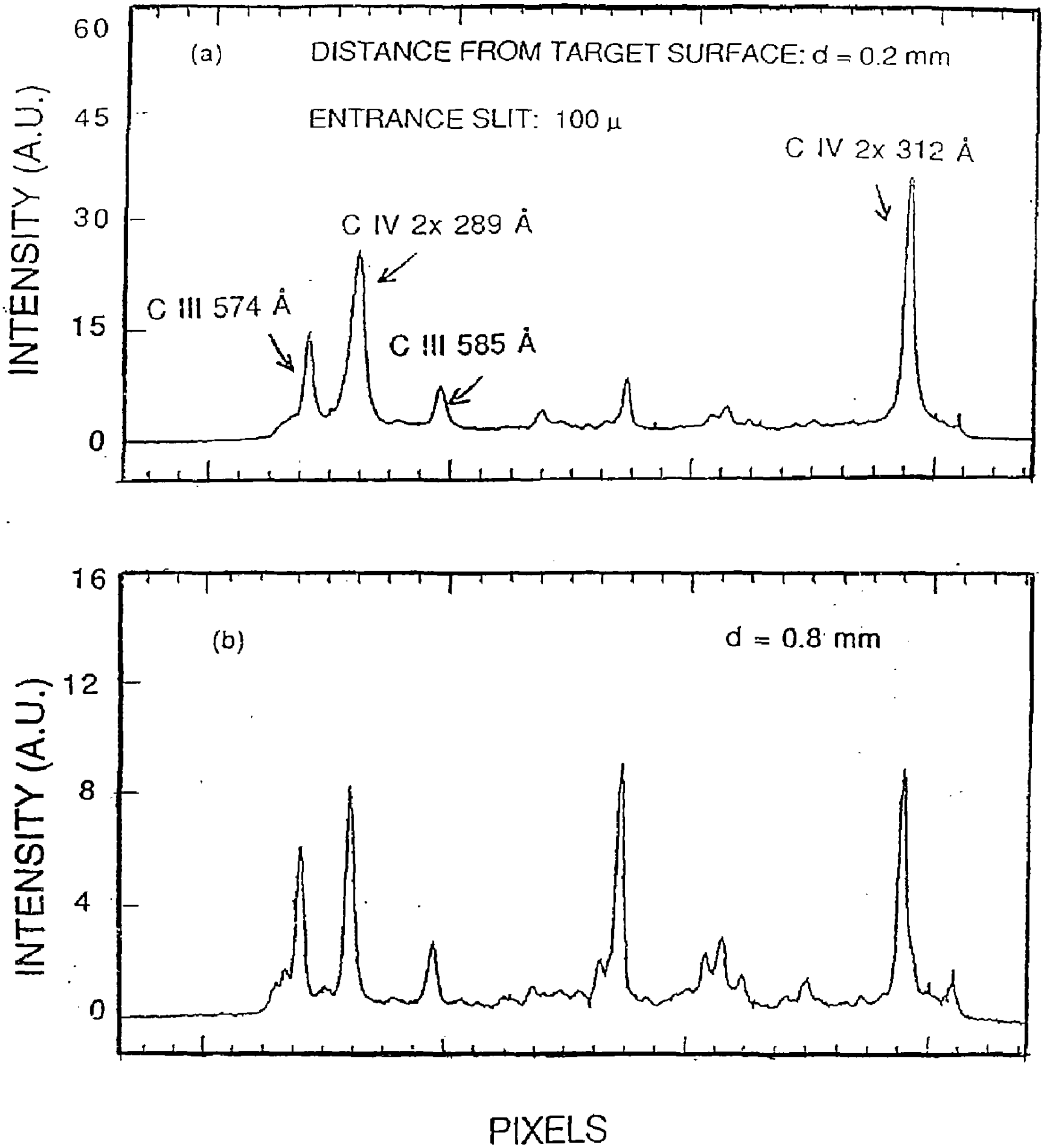
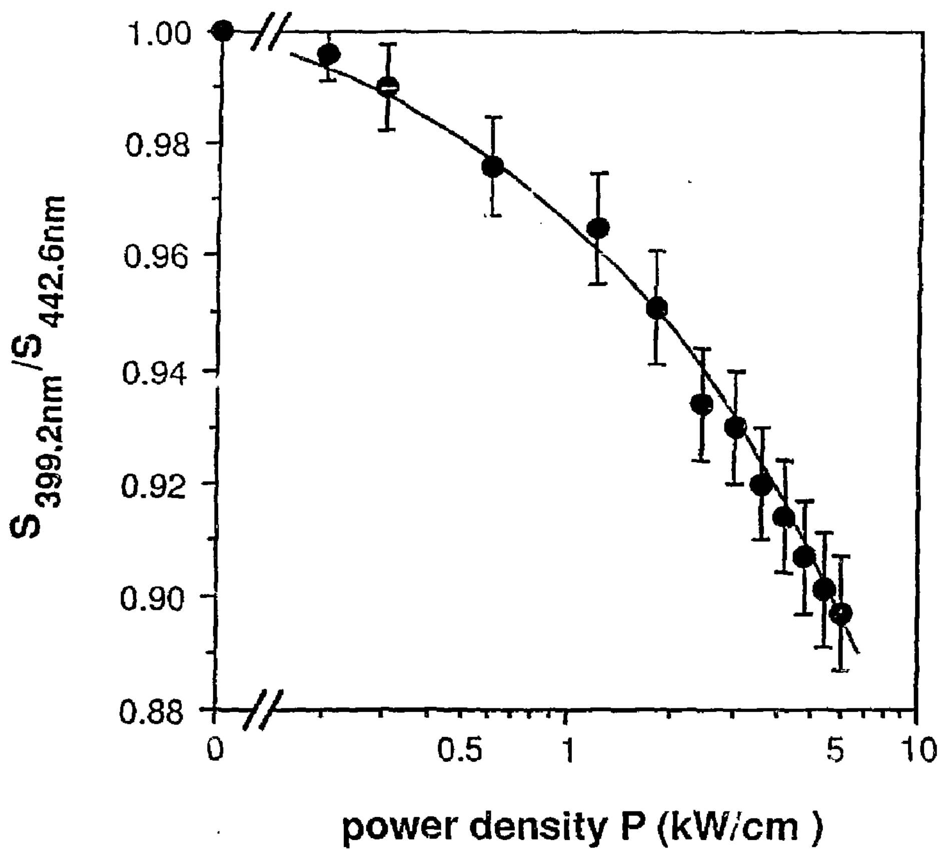
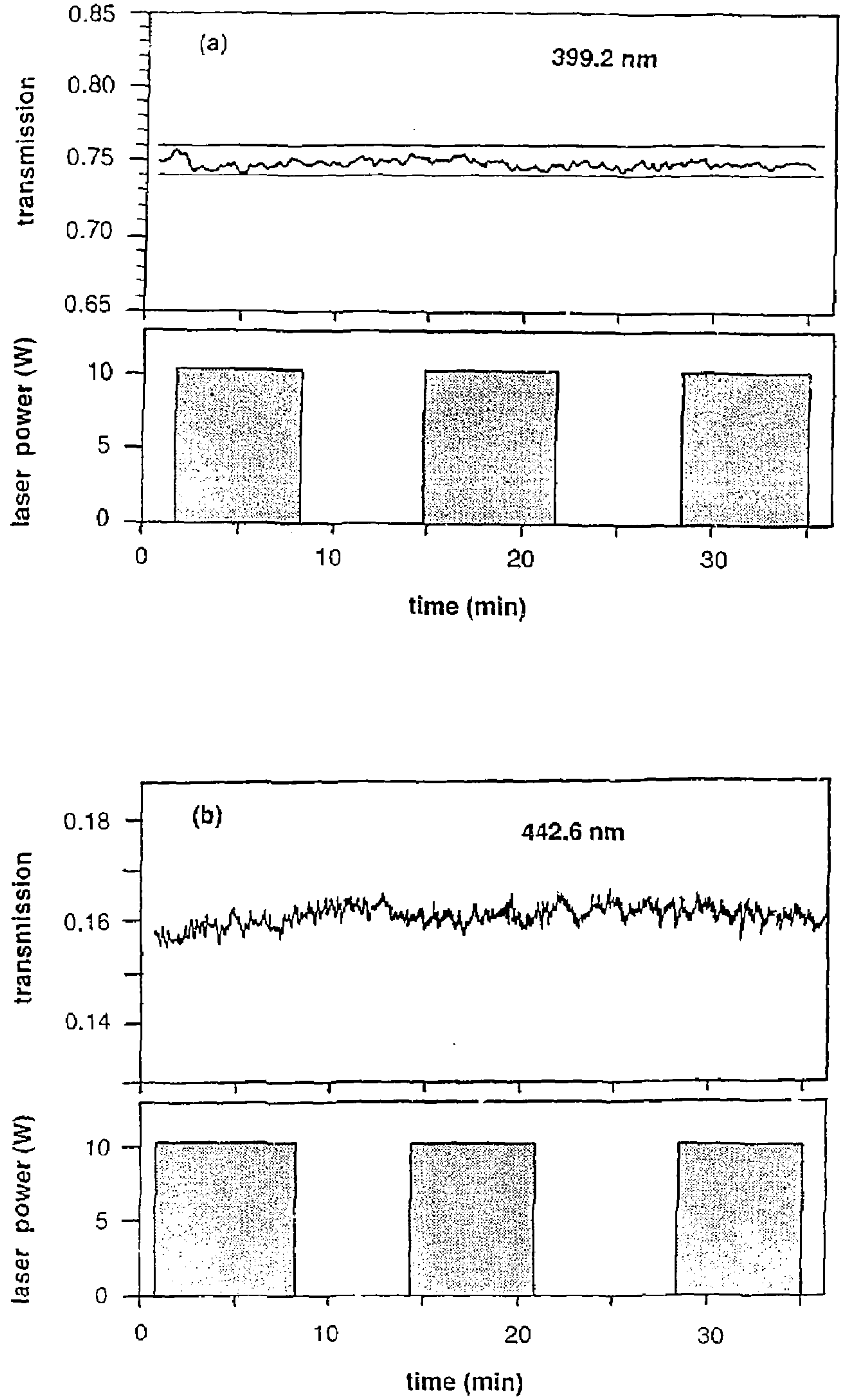

(a)
DISTANCE FROM TARGET SURFACE: d = 0.2 mm
ENTRANCE SLIT: 100 μ
C IV 2x 312 Å
C IV 2x 289 Å
C III 574 Å
C III 585 Å
INTENSITY (A.U.)
60
45
30
15
0
(b)
d = 0.8 mm
INTENSITY (A.U.)
16
12
8
4
0
PIXELS

Fig. 4



1.00
0.98
0.96
0.94
0.92
0.90
0.88
$S_{399.2nm}/S_{442.6nm}$
0
0.5
1
5
10
power density P (kW/cm)

Fig. 5



(a)
399.2 nm
transmission
0.85
0.80
0.75
0.70
0.65
laser power (W)
10
5
0
0
10
20
30
time (min)
(b)
442.6 nm
transmission
0.18
0.16
0.14
laser power (W)
10
5
0
0
10
20
30
time (min)

Fig. 6