Quenching of Spontaneous Emission Coefficients in Plasmas

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Abstract

We have observed changing Einstein coefficients of spontaneous emission as a function of electron density in CO₂ laser-produced plasmas. These measurements are based on the intensity branching ratio of CIV lines 5801-5812 Å and 312.41-312.46 Å which share a common upper level. Similar observations for CIII lines are also discussed.

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The coefficient of spontaneous emission, A , also called the Einstein coefficient, is considered to be one of the basic constants of a given transition in an atom or an ion. Therefore, the spectral line intensity I_{nm} (in photons) for $n \rightarrow m$ transition is always expressed as being proportional to the spontaneous emission coefficient A_{nm} (in sec^{-1}) for an optically thin plasma (e.g., see Ref. 1):

$$I_{nm} = k_n(r) N_n(N_e, T_e) A_{nm}, \quad (1)$$

where $k_n(r)$ is a geometrical factor, N_n (in cm^{-3}) is the population density of the upper level n , and N_e and T_e are electron density and temperature, respectively. It is always assumed that the plasma electron density has no effect on the A_{nm} coefficients and determines N_n through collisional processes. This has led to the development of models for the calculation of level populations, particularly collisional-radiative models (e.g., see Ref. 2). Also, as a consequence of this assumption, the so-called branching ratio technique was developed for absolute intensity measurements of spectral lines in the vacuum ultraviolet (VUV) wavelength region.³ From Eq. (1), it follows immediately that for optically thin lines with a common upper level [and therefore the same N_n and $k_n(r)$] and different lower levels, the ratio of line intensities is equal to the ratio of the corresponding A -coefficients, that is,

$$R = I_{nm}/I_{nk} = A_{nm}/A_{nk}. \quad (2)$$

In this way, if the ratio, R , of A -coefficients is known for a pair of lines and if the intensity of one line (e.g., for wavelength $\lambda > 2000 \text{ \AA}$) is measured, one may obtain the intensity of the second line (e.g., in VUV). Of course, this technique can be reversed, i.e., from measurements of the intensity ratio of two lines in an optically thin plasma, the ratio R and thus either of the corresponding spontaneous emission coefficients, A , can be deduced.

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Approximately three years ago, during the intensity calibration of VUV spectrometers by the branching ratio technique for X-ray laser experiments, we observed an unexplainable discrepancy between calibrations using a CO₂ laser and those using a spark discharge for producing plasma. One of us (S.S.) came to the hypothesis that in higher density plasmas (in our case laser-produced plasmas) spontaneous emission coefficients may decrease as a result of electrons interacting with atoms and ions. Such an effect should be larger for transitions with a longer lifetime (smaller A), in contrast to the effect of self-absorption. In this case it was expected that the branching ratio should change very significantly with increasing electron density.

We decided to study experimentally the quenching of spontaneous emission coefficients in plasmas using CIV lines with a common upper level 3p ²P (denoted here as level 3) and transitions to levels 3s ²S (5801.5 Å and 5812.1 Å lines) and 2s ²S (312.42 Å and 312.46 Å lines) which are denoted as levels 2 and 1, respectively. We chose these lines because in our experiment, radiation from Li-like CIV ions was intense in the plasma created by a relatively low power CO₂ laser (~ 10 J, 150 nsec pulse and 500 μ diameter focal spot) and therefore the CIV lines in the visible region of the spectra were easy to monitor. The coefficients of spontaneous emission for these transitions are $A_{32} = 0.32 \times 10^8 \text{ sec}^{-1}$ and $A_{31} = 45.6 \times 10^8 \text{ sec}^{-1}$, respectively⁴ and the statistical weights are $g = 4$ and 2 for the sublevels of level 3.

The experimental arrangement is presented in Figs. 1a and 1b. The CO₂ laser beam is focused onto a carbon target by a NaCl lens of focal length $f = 50 \text{ cm}$. Because near the target ($d = 0\text{--}20 \text{ μm}$) the plasma electron density is close to that of the solid state ($\sim 10^{23} \text{ cm}^{-3}$) and decreases very rapidly (due to expansion) with distance d to the critical electron density for the CO₂ laser, $N_e \approx 10^{19} \text{ cm}^{-3}$, and below it with further increasing d , the most significant changes of spontaneous emission were expected close to the target. Therefore,

in order to observe only a limited region of the plasma at a chosen distance from the target, a slit $S = 200 \mu$ was placed at a distance of 15 mm from the plasma.

The size of the slit was chosen as a trade-off between spatial resolution and detected signal strength. The slit S , being attached to the vacuum chamber, was unmovable and only the target could be moved by a micrometric vacuum feedthrough perpendicular to the laser beam and parallel to the slit S . The target had several steps and by moving the target the laser beam could initiate plasma at different distances d from the viewing path through slit S (see insert for Fig. 1a), thereby providing plasmas with a range of electron densities. The interaction of the CO_2 laser beam with the target was practically the same for different distances, d , as the depth of focus of the laser beam was much larger than the maximum change of d .

In order to provide simultaneous measurements of line intensities in VUV and Visible-UV spectra from exactly the same region of the plasma, a grazing incidence 1.5 m vacuum spectrometer, McPherson 252, was significantly modified and combined with a 0.5 m Ebert-Fastie type Jarrell-Ash air spectrometer ($\lambda \geq 2000 \text{ \AA}$). In this new type of visible-UV/VUV spectrometer, the plasma radiation, after passing the entrance slit, was partially diffracted and partially reflected (zeroth-order) by the VUV spectrometer's grating (1200 gr/mm). The diffracted beam was focused on the Rowland circle, whereas the zeroth-order beam was directed by the mirror into the Visible-UV spectrometer with its own normal incidence grating (1200 gr/mm). Signals in Visible-UV and in VUV regions were monitored by multichannel detectors (1024 pixels each).

In Figs. 2a and 2b are shown typical computer outputs for spectra in the vicinity of (a) CIV 312 $\text{\AA}$ and (b) 5801-5812 $\text{\AA}$ lines with the target surface at distances $d = 0.2 \text{ mm}$ and 1.0 mm from the farther edge of the slit S . The width of the target measured along the line of sight, w , was 1 mm. The results of the measurements of the intensity branching ratio $R = I_{32}/I_{31}$ as a function of distance, d , from target (hence as a function of electron

density N_e) for different target widths ($w = 0.35, 0.42, 1.0$, and 4.1 mm) are presented in Fig. 3. The electron density was measured from Stark broadening of the CIV 5801 Å line using the quasi-static approximation $\Delta\lambda_s = \alpha \times N_e^{2/3}$ (Ref. 1). Instrumental broadening $\Delta\lambda_{ins} = 1.6$ Å was measured with a low pressure Hg-lamp. The coefficient α was determined by the measured value of $\Delta\lambda_s = 6$ Å at $1.5 \times 10^{18} \text{ cm}^{-3}$ in Ref. 5. For a narrow target, $w = 0.35$ mm, and $d \geq 0.5$ mm, where the effect of self-absorption is negligible, the intensity branching ratio, R , is equal to the spontaneous emission coefficients ratio A_{32}/A_{31} (A_{VIS}/A_{VUV}). The intensities $I_{31}(\text{VUV})$ and $I_{32}(\text{VIS})$ were obtained from the multichannel detector signals (see Fig. 2) by integrating over the profile of the 312.42-312.46 Å and 5801.5-5812.1 Å lines after subtracting respective backgrounds. The intensities of 312 Å lines increased with decreasing d , as expected, due to the increasing abundance of CIV and rise of N_e , whereas the visible lines 5801-5812 Å increased significantly less and close to the surface of the target ($d \leq 0.2$ -0.3 mm) started to decrease. The intensity ratio of the two lines I_{5801}/I_{5812} was practically the same for all distances and close to its theoretical value 2:1, indicating that these lines are optically thin.

All measurements of the ratio, R , were normalized to the theoretical value⁴ $R = 7 \times 10^{-3}$ for nonperturbed ions. For wider targets ($w = 1.0$ mm, 4.1 mm) normalization was done at a distance of $d = 1.5$ and 1.2 mm, respectively, in order to avoid the effect of self-absorption on the 312 Å lines. For narrower targets, normalization was done at a smaller distance, $d = 1$ mm.

The effect of self-absorption on the VUV lines is evident in the wider targets from the higher ratio, R , and its peak at $d = 0.6$ mm ($w = 1.0$ and 4.1 mm). For these targets, self-absorption increases with increasing N_e , therefore, only when the effect of quenching of the coefficient of spontaneous emission for visible lines becomes dominant at $d < 0.5$ mm ($N_e > 10^{18} \text{ cm}^{-3}$) can the rapid decrease of branching ratio, R , be observed. On the other hand, if the electron density and hence $R = A_{32}/A_{31}$ is kept constant ($d = \text{constant}$) and the

width, w , of the target is changed, the line intensity ratio I_{32}/I_{31} changes due to self-absorption of VUV lines mainly, as is demonstrated by the top curve in Fig. 3 (this intensity ratio in Fig. 3 is multiplied by 1.4 in order to separate the curves). These measurements were done for $d = 0.5$ mm using a wedge carbon target in which the width varied from $w = 0.2$ mm up to 1.3 mm.

Very recently, the effect of quenching spontaneous emission was also observed in CIII in measurements of the intensity ratio of $5696.0 \text{ \AA}$ ($3d \text{ } ^1D-3p \text{ } ^1P^0$) and $574.3 \text{ \AA}$ ($3d \text{ } ^1D-2p \text{ } ^1P^0$) lines.

We also describe briefly our initial experiment which was conducted for one year before the modification of VUV spectrometer. In that experiment the VUV spectrometer viewed plasma as in Fig. 1; however, the air monochromator was located on the other side of the plasma and viewed it through another slit S' , identical with the first one. Initially VUV and air monochromators with an electron multiplier and photomultiplier, respectively, monitored the time evolution of single spectral line intensities. The signals for the CIV 312 $\text{\AA}$ and 5801 $\text{\AA}$ lines were practically identical with FWHM $\sim 250\text{-}300$ nsec. Subsequently a multichannel detector was used in the VUV spectrometer (time integrated signal); hence, the signal from the air monochromator was integrated in time in order to compare it with the VUV signal. Extremely careful and precise alignment was required to ensure that both instruments viewed exactly the same part of the plasma. The results were similar to the later results with improved instrumentation presented in Fig. 3.

The effect described here may provide an explanation of the disappearance of lines from transitions between levels with principal quantum number 4 and 3 in XUV spectra of high density plasmas ($N_e \approx 10^{21}\text{-}10^{22} \text{ cm}^{-3}$) produced by the OMEGA laser (University of Rochester), in which lines from $4 \rightarrow 1$ transitions were very intense.⁶

The fact that the spontaneous emission coefficient is not constant as we used to think about it may be found in recent work on inhibition of spontaneous emission of an atom in a

cavity which has no modes at the transition frequency⁷ and in studies of cyclotron radiation of an electron confined in a Penning trap,⁸ and a similar effect predicted in solid state cavities.⁹

In order to explain the experimental results, the Schrödinger equation was solved numerically for CIV ions in the electric potential of surrounding plasma. This potential was approximated by using Debye radius $r_D \propto N_e^{-1/2}$, assuming T_e , the electron temperature, to be constant. The transitions of the valence electron were assumed to occur in the internal potential of two bound electrons ($1s^2$), approximated by a uniform cloud of radius R_0 and influenced by the external field. The potential $V(r)$ was therefore approximated by

$$V(r) = \begin{cases} [Z'e^2/2R_0 \{3-(r/R_0)^2\} - Ze^2/r] \exp(-r/r_D) & r \leq R_0 \\ -(Z-Z')e^2/r \exp(-r/r_D) & r > R_0, \end{cases} \quad (3a)$$

$$(3b)$$

where e is electron charge, r is distance from the center of the cloud, $Z' = 2$ and $Z = 6$ for CIV and r_D was taken to be much larger than R_0 .

Numerically solving the radial part of Schrödinger equation with potential $V(r)$, the wave functions and hence coefficients A_{32} and A_{31} were obtained. However, changes in the branching ratio predicted in this model were an order of magnitude smaller than those observed in the experiment for $N_e \approx 5 \times 10^{18} \text{ cm}^{-3}$. Also, according to these calculations, significant shifts of spectral lines should occur, which were not observed. This indicates, in agreement with discussion in Ref. 12, that any significant modification of A-coefficients may be due to variation of the transition strengths and not to the transition energy.

We also considered another model which predicts quenching of the spontaneous emission coefficients by collisions. For an unperturbed atom or ion the average time between radiative transitions is given as $\tau_A = 1/A$. If the frequency of collisions of atoms or ions with, e.g., electrons, $\nu_{ei} = N_e S_{ei}$ (S_{ei} is the rate of collisions), is such that $\tau_{ei} =$

$\nu_{ei}^{-1} \ll \tau_A$, then collisions may disrupt the process of spontaneous emission and may cause a decrease of the A-coefficients due to change of the transition strength¹². In this case, the line intensity will become proportional to the new A-coefficient which depends on the collisional frequency. This is to be distinguished from the Lorentz model of line broadening in which collisions disturb the phase of the radiation but do not change the transition probability. In a pair of transitions, a stronger effect is expected for the transition with the smaller A-coefficient. Of course, population of the upper level is dependent on collisional processes; hence, to observe the effect of collisional quenching the branching ratio has to be measured as in our experiment.

Some indication of collisional quenching of spontaneous emission may be seen in observations of long wavelength forbidden lines in the ground configuration of highly ionized ions.¹⁰ The spontaneous emission coefficients for such forbidden lines with $\lambda > 2000 \text{ \AA}$ are usually in the range of $A = 10^2\text{-}10^3 \text{ sec}^{-1}$. Therefore, in this model, collisional quenching for these lines at $N_e = 10^{14} \text{ cm}^{-3}$ should be similar to lines with $A = 10^7\text{-}10^8 \text{ sec}^{-1}$ at $N_e = 10^{19} \text{ cm}^{-3}$. Indeed it was noticed¹¹ that for $N_e \geq 10^{14} \text{ cm}^{-3}$ it was impossible to observe the forbidden line FeXX 2665 $\text{\AA}$ ($A = 5.7 \times 10^2 \text{ sec}^{-1}$) in a tokamak plasma due to its very low intensity (this line was easily observed¹⁰ at a lower density $N_e = 2 \times 10^{13} \text{ cm}^{-3}$). Some difficulties in the observation of forbidden lines in higher density plasmas are related to the higher radiation background; however, the rise of the background is not sufficient to explain the disappearance of these lines.

The measurements of a variety of lines in a broad range of plasma parameters should help in the construction of the rigorous theoretical model for the prediction of quenching of spontaneous emission coefficients in plasmas.

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Figure Captions

Fig. 1 Experimental setup.

Fig. 2 Example of spectra in the vicinity of (a) CIV 312 Å and (b) CIV 5801 - 5812 Å lines at distance from target $d = 0.2$ mm and 1.0 mm.

Fig. 3 Branching ratio I_{VIS}/I_{UV} for CIV 5801-5812 Å and 312 Å lines as a function of distance d and electron density for target width $w = 0.35, 0.42, 1.0$, and 4.1 mm. The top curve corresponds to I_{VIS}/I_{UV} (multiplied by 1.4) as a function of target width w (approximately the width of the plasma) for $d = 0.5$ mm.

Fig. 1

FIG. 2

Fig. 3

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