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QUENCHING OF EINSTEIN'S A-COEFFICIENTS BY PHOTONS

BY

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Quenching of Einstein's A-Coefficients by Photons

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Abstract

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We present evidence for the quenching of Einstein A - coefficients in an Ar-ion laser discharge due to the presence of a high intensity laser flux. The reduction in spontaneous emission intensity when lasing occurs was found to be dependent on the Einstein A - coefficient for transitions originating from the same upper level.

In two recent papers^{1,2} measurements of a change in branching ratio of spontaneous emission coefficients (Einstein's A-coefficients) in a plasma as a function of electron density were presented. A change of branching ratio of up to a factor of 10 was observed in C IV for 3p-3s (580.1 - 581.2 nm) and 3p-2s (31.2 nm) transitions¹ when the electron density changed from approximately $N_e \approx 10^{18} \text{ cm}^{-3}$ to 10^{19} cm^{-3} . This effect was also observed in C III for the 3d-3p (569.6 nm) and 3d-2p (57.4 nm) transitions and in NV for the 3p-3s (460.3 - 462.3 nm) and 3p-2s (20.9 nm) transitions.

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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.^{1,2} However, the simple hypothesis that the spontaneous emission of an atom may depend rather on dynamic effects like collisions could qualitatively (but not, of course, quantitatively) explain the observable effects. If electron collisions with atoms or ions have a significant disruptive effect on their spontaneous emission, the change of emissivity should depend on the ratio of collisional frequency ν_{coll} to the frequency of spontaneous emission from level n : $\nu_n \approx A_n = \sum_{k < n} A_{nk}$, where subscript nk indicates transitions between level n and levels $k < n$. Indeed, for our plasma with $T_e \approx 5$ eV and $N_e \approx 10^{19} \text{ cm}^{-3}$ the frequency, ν_{coll} , is approximately 10^3 times larger than ν_n for the 3p level of CIV. According to the above hypothesis, one would expect a larger disruptive effect for weaker (slower) transitions, therefore, a larger change in A-value should occur for the 3p - 3s than for the 3p - 2s transition. This leads to observable changes in the branching ratio.

Support for this hypothesis can be found in papers by Robinson³ and Deng and Eberly.⁴ The disappearance of forbidden lines in tokamak plasmas at higher electron densities would also be in accordance with this hypothesis. The iron forbidden line Fe XX 266.5 nm was well observable at $N_e = 2 \times 10^{13} \text{ cm}^{-3}$ (Ref. 5) and was not observable at $N_e = 10^{14} \text{ cm}^{-3}$ (Ref. 6). It has an A-value $\approx 10^2 \text{ s}^{-1}$, which is 10^5 times smaller than the A-value of the 3p-3s transition in C IV. Therefore, a simple scaling of collision frequency, according to our hypothesis, would lead one to expect strong quenching of this forbidden line at $N_e \sim 10^{14} \text{ cm}^{-3}$, as experiments indicate.

A much more important implication of the model is that collisions of photons with atoms or ions, should also provide quenching of Einstein coefficients if the frequency of such collisions is sufficiently high.

We have investigated a gaseous lasing medium inside a laser cavity in order to detect any quenching of A-coefficients by photons. The laser is tuned to a particular lasing line by changing the position of the 100% reflectivity mirror and the angle of the prism in front of the mirror. In this way it is possible to control the photon density in the discharge over a very large range and, therefore, change the photon-ion collision frequency without any significant change of the gas or plasma parameters in the discharge. The chosen lasing line has a common upper level with at least two other, non-lasing lines, with significantly different Einstein's A-coefficients. The larger the difference in A-values is, the larger the change in the branching ratio of non-lasing lines.

For the experiment we choose a CW Ar-ion laser which has a number of lasing lines and appropriate non-lasing lines in a convenient (visible) spectral range. For example, the upper level $4p\ ^4D_{5/2}^0$ of the Ar II 514.53 nm lasing line is a common upper level for seven non-lasing transitions^{7,8} (see Fig. 1) whose A-coefficients differ by up to a factor of 50 (Refs. 7,8). Similar conditions hold for the Ar II 487.99 nm ($4p\ ^2D_{5/2}^0 \rightarrow 4s\ ^2P_{3/2}$) lasing line. Both lasing transitions share a common lower level.⁹ This lower lasing level decays very rapidly by spontaneous emission to the ground level $3p^5\ ^2P$ with a rate $A = 2.7 \times 10^9\ s^{-1}$. The fast decay of the $4s\ ^2P_{3/2}$ level (resulting in the low population of this level) is the primary cause of the creation of the population inversion rather than a selective population of upper lasing level $4p\ ^4D_{5/2}^0$. In addition, the transitions between the $4s\ ^2P_{3/2}$ level and lower levels of non-lasing transitions ($4s\ ^4P$, $3d\ ^4D$ and $3d\ ^4F$) are intercombination transitions and, therefore, have quite low radiative transition probability.

Hence, one should not expect to see any significant change of population of these $4s\ ^4P$, $3d\ ^4D$, and $3d\ ^4F$ levels due to lasing action to the $4s\ ^2P_{3/2}$ level. This is a very important and unique feature of the Ar-ion laser which makes this laser an almost ideal one for our experiment.

The Ar-ion laser (Coherent CR-8 UV) had a maximum output beam power of about 4 W at the 514.53 nm lasing line and about 2W at 487.99 nm. The length of the discharge tube was $L = 1.2$ m, its effective diameter was $d = 1.4$ mm, and the output mirror had a transmissivity of 5% at 514.53 nm. Therefore, a maximum intra-cavity power density of approximately 5 kW/cm^2 for the 514.53 nm lasing line (electric field strength of about 1 kV/cm) could be achieved. A powerful laser is required in order to provide a large density of photons inside the cavity. A further increase of the intra-cavity intensity is planned by the use of a lower transmissivity output mirror.

In our experimental setup the Ar - ion laser light was directed by a set of mirrors towards the entrance slit of a 0.5 m Czerny-Turner monochromator (SPEX - 1870) with a 2400 gr/mm holographic grating (for lines with a wavelength $\lambda > 600$ nm a 1200 gr/mm grating was used). A dichroic filter with low transmission at the lasing wavelengths and relatively high transmission elsewhere was utilized to block the laser lines. A small portion of the laser output beam was sampled by a silicon photodiode to constantly monitor the laser power.

By tuning the cavity to obtain lasing action at 514.53 nm, we noticed that a change in the $4p\ ^4D_{5/2}$ upper level population resulted in a decrease of the intensity of the spontaneous emission lines to the $4s\ ^4P$, $3d\ ^4D$ and $3d\ ^4F$ levels (see Fig. 1). Since these intensities are proportional to the upper level population, all emission lines should exhibit the same decrease. However, in our measurements each line yields a different "apparent" depopulation of the $4p\ ^4D_{5/2}$ upper level, as can be seen from Fig. 2.

In this figure the ratio of emission intensities at the maximum achievable intra-cavity power density to emission intensities at zero intra-cavity power density for different spontaneous emission lines is shown. Since the volume averaged $4p\ ^4D_{5/2}$ population can only have a single value, our data indicate a monotonic dependence on Einstein A-coefficients of the respective transitions. Obviously there is no smooth dependence on wavelength. The largest discrepancy in apparent depopulation ($14.7\% \pm 0.8$ and $20.8\% \pm 1.0$) was found between the strongest (442.6 nm) and weakest (399.2nm) transition from this upper level.

However, before concluding that quenching of Einstein coefficients due to collisions with photons was occurring, several other possible explanations were checked:

- A nonlinear response of our photon detection system (photomultiplier tube and amplifier) at low and high signal intensities was ruled out quite easily.
- The identification of the different lines was checked very carefully (the resolution of our monochromator was 0.035 nm FWHM). The background signal from residual lasing radiation inside the monochromator was found to be negligible due to the dichroic filter used to block the lasing line.
- No shift in wavelength or line broadening (which in principle could explain the results shown in Fig. 2) could be observed when tuning the laser cavity to 514.53 nm. Measuring both the line integrals and peak heights yielded identical results.
- The apparent depopulation did not depend on the polarization of the spontaneous emission lines as was checked experimentally by inserting a Glan-Taylor prism polarizer into the beam path and rotating it.
- Investigations also showed that the wavelength - dependent transmissivities of the dichroic filter (used to block the lasing line) and the cavity mirrors did not change with the laser beam power change.

* Our most serious concerns were dedicated to the problem of self-absorption of the spontaneous emission lines. Of course, a 1.2m long Ar-ion laser plasma tube (gas fill pressure 300 mTorr) cannot be treated as optically thin, and self-absorption is expected especially for the strong transitions. Since in our experiment we compare only intensities with and without lasing action while the laser discharge current, fill pressure, etc. stay constant, only a change in absorption with lasing action can affect our results. Such a change might be caused by a collisional interaction of the lower lasing level $4s\ 2P_{3/2}$ (whose population should increase during lasing at 514 nm) with the lower levels of the spontaneous emission lines. However, as was indicated earlier, this level decays to the Ar ion ground state $3p^5\ 2P$ with a large transition rate, hence, its population should always be small. Transition rates from the $4s\ 2P_{3/2}$ level to lower non-lasing levels are small due to the intercombination character of these transitions. Furthermore, no change in spontaneous emission line intensity could be observed when tuning the laser to the 487.99 nm lasing line, which has the same $4s\ 2P_{3/2}$ lower level as a 514.53 nm line.

In addition, we repeated our measurements using a second Ar-ion laser with a shorter discharge tube (0.90 m). Within our experimental errors both lasers yielded the same apparent depopulation when compared at the same intra-cavity intensities. We also used this second laser to measure absorption of the spontaneous emission lines in the plasma column. This was achieved by injecting the light from the second laser into the first laser cavity via reflection at the prism surface. A lock-in amplifier and chopper was used to distinguish the light from each laser. Considerable absorption (80%) was found for the strong transition at 442.6 nm, whereas absorption was small (13%) for the weakest transition at 399.2 nm. Within our experimental errors of about 4% we could not observe any change in absorption when the front mirror was brought into alignment for lasing at 514.53 nm.

We conclude, therefore, that the differences in apparent depopulation shown in Fig. 2 can only be explained by a change in Einstein A-coefficients for spontaneous emission. This quenching of A-coefficients is larger for lines with small transition probabilities and, therefore, decreases the branching ratio between weak and strong transitions. An example shown in Fig. 3 is the change in branching ratio between the 399.2 nm and the 442.6 nm transitions (whose A-coefficients differ by a factor of 54) plotted as a function of laser power density (full symbols).

In continuation of our work, we most recently repeated parts of our experiments by using a more powerful Ar-ion laser (Coherent Innova 100-20). Intra-cavity power densities exceeding those of the previous laser by more than a factor of three could be reached due to a higher current density and a longer discharge tube. Open symbols in Fig. 3 achieved by using this laser demonstrate that the signal ratio $S_{399.2\text{nm}}/S_{442.6\text{nm}}$ continues to decrease, as the intra cavity power density is further increased. We also remeasured the apparent depopulation for the spontaneous emission lines to the $4s\ ^4P$ and $3d\ ^4D$ levels. At about 16 kW/cm^2 the trend shown in fig.2 remains unchanged, however, due to the larger depopulation of the upper level and the larger quenching effect at this intra-cavity power density, the signal ratios $S(P=P_{\text{max}})/S(P=0)$ dropped considerably, e.g. to 0.56 ± 0.015 for the 442.6 nm transition and to 0.46 ± 0.015 for the 399.2 nm transition.

One final remark: with a changed A-coefficient we should also observe a change in the stimulated absorption B-coefficient, otherwise the thermodynamic relation between the A and B Einstein coefficients would be violated. Such a change would be of the order of 1% (7% effect of quenching for the 399 nm line with 13% absorption). This is significantly lower than the present uncertainty in our measurements (~4%) and we were unable to detect

a change in the 399.2 nm and 442.6 nm line absorptions due to lasing action at 514 nm. Absorption measurements could be feasible for longer laser tubes (hence larger absorption) with higher intra-cavity power density (larger quenching effect) and sufficiently bright probing ArII radiation for a good signal to noise ratio.

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Figure Captions

- Fig. 1. Partial Ar II energy level diagram with transition probabilities in units of 10^7 s^{-1} .
- Fig 2. Ratio of measured spontaneous emission intensities at full (approx. 5 kW/cm^2 at 514.53 nm) intra-cavity power density $S(P=P_{\text{max}})$ to emission intensities at zero intra-cavity power density $S(P=0)$ as a function of Einstein A-Coefficient A_{ij} of respective transitions from the common ArII $4p \text{ } ^4D^o_{5/2}$ upper level.
- Fig 3. Ratio of the spontaneous emission intensities of the 399.2 nm to the 442.6 nm transition as a function of intra-cavity (514.53 nm) laser power density P . (The signal ratio has been normalized to 1 at $P=0$.) Open and full symbols refer to two different Ar-ion lasers (cf. text).

Fig. 1

Fig. 2

Fig. 3

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