Modification of Einstein $A$ coefficient in dissipative gas medium

Cao Chang-qi
Department of Physics, Peking University, Beijing 100871

Cao Hui (H.Cao)
Edward L. Ginzton Laboratory, Stanford University, Stanford, CA 94305, USA

Qin Ke-cheng
Department of Physics, Peking University, Beijing 100871.

Abstract

Spontaneous radiation in dissipative gas medium such as plasmas is investigated by Langevin equations and the modified Weisskopf-Wigner approximation. Since the refractive index of gas medium is expected to be nearly unity, we shall first neglect the medium polarization effect. We show that absorption in plasmas may in certain case modify the Einstein $A$ coefficient significantly and cause a pit in the $A$ coefficient-density curves for relatively low temperature plasmas and also a pit in the $A$ coefficient-temperature curves.

In the next, the effect of medium polarization is taken into account in addition. To our surprise, its effect in certain case is quite significant. The dispersive curves show different behaviours in different region of parameters.

1 Introduction

This work is motivated by an effect called by the original authors as “quenching of Einstein $A$ coefficient”: the ratio of two line intensities $I(5805\,\AA)$ and $I(312\,\AA)$ from a common upper level of $C_{1V}$ is reduced by an order of magnitude when the electron density $N_e$ changes from $10^{18}/\text{cm}^3$ to $10^{19}/\text{cm}^3$. But there is some dispute about the interpretation of their experiments. Our work is to see whether this effect is possible theoretically.

Physically, spontaneous emission is resulted from atom (ion) with “vacuum” electromagnetic field, therefore Einstein $A$ coefficient is a characteristic parameter of the total atom-field system, not just of atom itself. Its value may be different for different environment, such as cavity, or, in our case, the dissipative medium.

Our approach is based on Langevin equations. We don’t use Fermi golden rule to calculate the emission rate as did by Barnett et al, because the radiating ion and radiated field do not
make up a closed system, their states both before and after the emission process are not stationary states.

From Langevin equations an emission rate operator is defined, and a part of it, the spontaneous emission rate operator may be separated out.

Since the refractive index $n(\omega)$ of gas medium is expected to be nearly 1, we shall first, in Sec II, neglect the effect of medium polarization, which will be taken into consideration in Sec III.

2 The effect of medium absorption on Einstein $A$ coefficient

Since in this section we take $n(\omega) = 1$, both the frequencies and wave functions of light modes are the same as in vacuum. The reduction of Einstein $A$ coefficient is therefore not by the alteration of modes of e.m. field but is caused by alteration of dynamics: the relevant operators now do not obey the Heisenberg equations but obey Langevin equations instead. We shall see that the frequency dependence of photon decay parameter will play an importance role in the present case.

We have argued that the three-level ion problem may be reduced to two-level ion problem, the only exception is the calculation of population numbers among the levels, in which all the levels involved must be taken into account.

In considering the emission of a particular ion, the whole plasmas other than the radiating ion will be regarded as a reservoir, its effects are described by the damping and fluctuation terms in the Langevin equations:

$$\frac{d}{dt} \hat{a}_{kj}(t) = -\kappa(\omega) \hat{a}_{kj}^\dagger(t) + ig_{kj}\hat{S}_+(t)e^{-i(\omega-\omega_0)t} + \hat{F}_{kj}(t),$$

$$\frac{d}{dt} \hat{S}_+(t) = -\frac{\Gamma}{2} \hat{S}_+(t) - 2i \sum_{kj} \hat{S}_3(t)\hat{a}_{kj}^\dagger(t)e^{-i(\omega-\omega_0)t} + \hat{\Sigma}_+(t),$$

$$\frac{d}{dt} \hat{S}_3(t) = -\Gamma \hat{S}_3(t) - S_{30} + \sum_{kj} [ig_{kj}^*\hat{a}_{kj}^\dagger(t)\hat{S}_3(t)e^{-i(\omega-\omega_0)t} + h.c.] + \hat{\Sigma}_3(t).$$

where $\hat{S}_{\pm}$ are usual atom level-changing operators, $\hat{S}_3$ is the half of population difference operator: $\hat{S}_3 = \frac{1}{2}(\hat{P}_2 - \hat{P}_1)$, $\hat{a}_{kj}$ is the photon absorption operator of mode $(k,j)$, $j$ is photon polarization index. The free-varying phase factors have been separated out from the photon operators and atom level-changing operators.

We have defined an emission rate operator $\hat{I}(t)$ as that part of $-\frac{d}{dt}\hat{a}^\dagger$ which is caused by the interaction with photons, with the result

$$\hat{I}(t) = \sum_{kj} ig_{kj}^*\hat{a}_{kj}^\dagger(t)\hat{S}_3(t)e^{-i(\omega-\omega_0)t} + h.c..$$

Solving eq.(1.1) to get $\hat{a}^\dagger_{kj}(t)$ and substituting it into eq.(2), one may separate out the spontaneous emission rate operator $\hat{I}_{sp}(t)$ from $\hat{I}(t)$:

$$\hat{I}_{sp}(t) = \sum_{kj} [g_{kj}]^2 \int_0^t \hat{S}_3(t')\hat{S}_+(t)e^{-i(\omega-\omega_0)(t-t')-\kappa(\omega)(t-t')}dt' + h.c..$$
The Weisskopf-Wigner approximation now takes the modified form\[2\]
\[\sum_{k_j} |g_{k_j}|^2 e^{-i(\omega_0)(t-t')-\kappa(\omega)(t-t')} = (\bar{\gamma} + 2i\delta\tilde{\omega})\delta(t-t'), \text{ for } t-t' \geq 0. \tag{4}\]

Substituting it into eq.(3), we get immediately
\[I_{sp} = \bar{\gamma}\hat{S}_+(t)\hat{S}_-(t) = \bar{\gamma}\hat{P}_2(t). \tag{5}\]

Eq.(5), after taking the expecting value, is just the Einstein formula for spontaneous emission with \(\bar{\gamma}\) as the new \(A\) coefficient.

Taking real part of eq.(4) and integrating it over \(t\) from \(-\infty\) to \(t\), we get\[2\]
\[\bar{\gamma}/\gamma = \frac{1}{\pi\omega_0} \int_{\omega_p}^{\omega_{\text{max}}} \frac{\omega\kappa_1(\omega)}{(\omega - \omega_0)^2 + \kappa^2(\omega)} d\omega \tag{6}\]

where \(\gamma\) is the usual Einstein \(A\) coefficient in vacuum, \(\omega_p\) is the plasma frequency and \(\omega_{\text{max}}\) denotes a cut-off frequency representing the limit of dipole approximation.

We see that \(\bar{\gamma}/\gamma\) is determined solely by photon decay parameter \(\kappa(\omega)\), with no direct reference to atomic parameter. This is contrary to the conjecture of Aumayr et al\[5\].

When \(\kappa\) is small and independent of \(\omega\), the right hand side of eq.(6) will reduce to 1.

Actually, in plasmas, \(\kappa\) is contributed by the inverse Bremstrahlung of free electrons and selfabsorption of ions of the same kind as the radiator. Both are \(\omega\) dependent, but only the latter is important unless at very high plasma electron density (\(\sim 10^{21}/\text{cm}^3\)). So we shall only consider the latter, which will be denoted by \(\kappa_f(\omega)\), in the following.

\(\kappa_f(\omega)\) is expressed by
\[\kappa_f(\omega) = \frac{\pi e^3 N_f \gamma (P_1 - P_2)}{4\omega_0^2} \frac{\Gamma_T}{(\omega - \omega_0)^2 + \frac{1}{4}\Gamma_T^2} \tag{7}\]

where \(N_f\) represents the ion density, \(\Gamma_T\) is the total width of the spectral line, \(P_1\) and \(P_2\) are the average populations of the lower and upper levels. We have approximated the line profile by a Lorentzian shape in eq.(7), actually the profile is convolution of a homogeneous broadening (Stark broadening) and an inhomogeneous broadening (Doppler broadening).

The frequency behavior of \(\kappa_f(\omega)\) is characterized by a peak at \(\omega = \omega_0\). It is this behavior which may cause significant reduction of \(\bar{\gamma}/\gamma\).

To explain this, we simulate the variation of \(\kappa_f\) with \(\omega\) by a simple stepwise function
\[\kappa_f(\omega) = \begin{cases} \kappa_0 & \omega_0 < \omega_c \\ \kappa_0' & \omega > \omega_c \end{cases} \tag{8}\]

with \(\kappa_0 > \kappa_0'\).

If in the whole range \(\kappa_f(\omega)\) either equals \(\kappa_0\) or equals \(\kappa_0'\), the integral in the eq.(6) will amount to \(\pi\omega_0\). The curve \(\frac{\omega_0}{(\omega - \omega_0)^2 + \kappa_0^2}\) has a larger height but a smaller width as compared with that of \(\frac{\omega_0}{(\omega - \omega_0)^2 + \kappa_0'^2}\), so both gives the same integral value when substituted in eq.(6). But the situation of stepwise function eq.(8) is different, comparing it with the above mentioned constant cases, it is not difficult to understand why the stepwise function will reduce the integral value.
One may use $\kappa(\omega_0) \approx \kappa_f(\omega_0)$ to scale the integration interval of important contribution in eq.(6). If $\kappa_f(\omega)$ drops down rapidly in this region, then the reduction will be large. Since $\Gamma_T/2$ scales the interval of significant variation of $\kappa_f(\omega)$, it follows that the ratio of these two scales $\Gamma_T/2\kappa_f(\omega_0)$ determines the amount of reduction of $\dot{\gamma}/\gamma$. The smaller the value of $\Gamma_T/2\kappa_f(\omega_0)$, the heavier will be the reduction.

In this way, we see that the atomic parameters do affect the value of Einstein $A$ coefficient, but in an indirect way.

The numerical results are as follows. For line $\lambda = 5805\AA$, $\dot{\gamma}/\gamma$ is almost kept to be nearly 1 in the whole range from $N_e = 10^{15}/cm^3$ to $N_e = 10^{20}/cm^3$, because $\Gamma_T/2\kappa_f(\omega_0)$ is large, since the factor $P_1 - P_2$ in eq.(7) is very small in this case. The result for line $\lambda = 312\AA$ is shown in Fig 1. In low density region, $\Gamma_T$ is mainly contributed by Doppler broadening so that it is independent of $N_e$, and this in turn makes $\kappa_f(\omega_0)$ proportional to $N_e$. Therefore when $N_e$ increases, $\Gamma_T/2\kappa_f(\omega_0)$ becomes smaller, leading to the drop of $\dot{\gamma}/\gamma$. But when $N_e$ goes beyond a value about $1.5 \times 10^{18}/cm^3$, $\dot{\gamma}/\gamma$ turns up, because $\Gamma_T$ is gradually dominated by Stark broadening, so that it is proportional to $N_e$, which in turn leads to $N_e$-independence of $\kappa_f(\omega_0)$. As the result, there is a pit or hollow in the curve $\dot{\gamma}/\gamma-N_e$.

The dependence of $\dot{\gamma}/\gamma$ on temperature is also interesting. By a similar analysis, we have argued\cite{14} that there will be also a pit or hollow in the curve $\dot{\gamma}/\gamma-T$.

3 Einstein $A$ coefficient with index of refraction taken into account

We begin our discussion by deriving the plasmas’ refractive index from microscopic equations. First we omit the contribution of free electrons which is less important.

There are numerous ions in the plasmas, the effective coupling constant for ion $l$ and e.m. field has a position-dependent factor $e^{ik\cdot x(t)}$.

In gas, the orientation of atomic dipole $\langle d \rangle_{21}$ is random, yielding $|g_{kj}|^2$ independent of $j$,

$$|g_{kj}|^2 = G_k^2, \quad G_k^2 = \frac{\pi c^3 \gamma}{2\omega_0^3 V},$$

where

$$\omega_k = kc.$$

We define a collective atomic operator as usual

$$\hat{\mathcal{S}}_{+,kj}(t) = \sum_l e^{ikx(t)} \hat{\mathcal{S}}_{+,l}^{(t)}(t),$$

the equation for $\hat{a}_{kj}^\dagger(t)$ will be of the form

$$\frac{d}{dt} \hat{a}_{kj}^\dagger(t) = i\omega_k \hat{a}_{kj}^\dagger(t) + iG_k \hat{\mathcal{S}}_{+,kj}(t).$$

In the case of steady-state plasmas, we may approximate the $\hat{\mathcal{S}}_{3}^{(t)}$ in the equation of $\hat{\mathcal{S}}_{+,kj}$ by its average value $\frac{1}{2}(P_2 - P_1)$ plus a fluctuation term. This leads to

$$\frac{d}{dt} \hat{S}_{+,kj}(t) = (i\omega_0 - \frac{1}{2}\Gamma) \hat{S}_{+,kj}(t) + iG_k N_I V (P_1 - P_2) \hat{a}_{kj}^\dagger(t) + \text{fluctuation term}.$$
We see from eqs. (11) and (12) that \( \hat{a}^\dagger_{kj} \) and \( \hat{S}_{+,kj} \) turn into each other.

To derive the expression for index of refraction \( n \), we may assume, as usually does in classical electrodynamics, that \( \hat{S}_{+,kj} \) will do forced vibration following \( \hat{a}^\dagger_{kj} \) except a fluctuation term \[^8\]. By this, we get an expression for \( \hat{S}_{+,kj} \) from eq. (12). Substituting it into eq. (11), yields

\[
\frac{d}{dt} \hat{a}^\dagger_{kj} = i \left[ \omega_k + \frac{\pi c^3 \gamma N_I(P_1 - P_2)}{2\omega_0^2 (\omega - \omega_0 - \frac{1}{2}i\Gamma)} \right] \hat{a}^\dagger_{kj} + \hat{F}^\dagger_{kj}(t) \tag{13}
\]

where \( \omega \) denotes the actual frequency of \( \hat{a}^\dagger_{kj} \).

According to the definition, the real part of what inside the square brackets is just equal to \( \omega \). Thus we get an equation for \( \omega \):

\[
\omega = \omega_k + \frac{\pi c^3 \gamma N_I(P_1 - P_2)}{2\omega_0^3 (\omega - \omega_0)^2 + \frac{1}{4}\Gamma^2} (\omega - \omega_0). \tag{14}
\]

The expression of index of refraction follows immediately,

\[
n \equiv \frac{\omega}{\omega_k} \cong 1 + \frac{\pi c^3 \gamma N_I(P_1 - P_2)}{2\omega_0^3 (\omega - \omega_0)^2 + \frac{1}{4}\Gamma^2} (\omega - \omega_0). \tag{15}
\]

The imaginary part of what inside the square bracket will be \( \kappa_I(\omega) \). The expression so obtained for \( \kappa_I(\omega) \) is the same as eq. (7), except \( \Gamma_T \) is replaced by \( \Gamma \). The difference lies in that \( \Gamma_T \) in eq. (7) already contains the contribution of Doppler broadening.

We may generalize the above result by including the contribution of plasma free electrons. Besides, since in the present case the wave number \( k \) is real, the operator \( \hat{a}^\dagger_{kj} \) decays with time, \( \hat{S}_{+,kj} \) must also do damped vibration accordingly. This means that \( \omega \) in eq. (14) must be analytically continued to complex value which will be denoted by \( \Omega \):

\[
\Omega = \omega_k + \frac{\pi c^3 \gamma N_I(P_1 - P_2)}{2\omega_0^3 (\Omega - \omega_0 - \frac{1}{2}i\Gamma_T)} + \frac{\omega_p^2}{2(\Omega - if)} \tag{16}
\]

in which \( \Gamma \) is also replaced by \( \Gamma_T \) as in eq. (7).

This is a third order equation for \( \Omega \), it allows us to derive \( \Omega \equiv \omega + i\kappa \) for every given real value of \( \omega_k (\omega_k = kc) \). We will choose the root whose real part is nearest to \( \omega_k \) for photon. In the case of \( \lambda = 5805 \AA \), no problem appears. The so obtained curve \( \omega - \omega_k \) is like the usual dispersion curve of gas. In this case the affection of medium polarization on \( \gamma \) is indeed very small.

In the case of \( \lambda = 312 \AA \), the situation is different. For certain range of \( N_e \), the dispersion curve for photon and for collective atomic dipole (\( \hat{S}_{+,kj} \)) are totally mixed in the resonance region. This means the quantum of the polarization field has coupled to photon to form polaritons as in solids [6, 7]. In this case, the affection of medium polarization is significant.

The occurrence of such situation depends on the relative strength of coupling and damping. To show explicitly, let us examine the simpler case without contribution of free electrons. Now \( \Omega \) satisfies a second order equation. The two roots at \( \omega_k = \omega_0 \) are

\[
\Omega = \omega_0 + \frac{i}{4} \Gamma_T \pm \sqrt{\alpha - \frac{1}{16}} \Gamma_T^2, \tag{17}
\]

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where
\[ \alpha = \frac{\pi c^3 \gamma}{2 \omega_0^2} N_I (P_1 - P_2), \] (18)

just the product of the two couple constants between \( \hat{a}_{kj}^\dagger \) and \( \hat{s}_{+,kj} \) in eqs. (11) and (12).

When \( \alpha < \frac{1}{16} \Gamma_T^2 \) (weak coupling), the real part of the two roots given by eq.(17) equal each other, corresponding to the usual case as \( \lambda = 5805 \text{Å} \). On the other hand when \( \alpha > \frac{1}{16} \Gamma_T^2 \) (strong coupling), the real parts of the two roots are different, corresponding to the situation of polariton formation. It is interesting to note that
\[ \Gamma_T^2 \frac{\Gamma_T}{2 \alpha} = \frac{\kappa_T(\omega_0)}{\kappa_I(\omega_0)}, \]

therefore the two conditions \( \Gamma_T^2 \frac{\Gamma_T}{2 \alpha} \ll 1 \) and \( \Gamma_T^2 \frac{\Gamma_T}{\alpha} \approx \frac{1}{16} \Gamma_T^2 \) almost correspond to each other. Namely the region of polariton formation will cover the region of the pit discussed in last section.

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References


2. Cao Chang-qi and Cao Hui (H. Cao), SPIE 1726, 124 (1992),


Figure Caption

1. Dependence of \( \tilde{\gamma} / \gamma \) on \( x \) for line 312Å of ion \( C_{IV} \ continume x = N_e/10^{19} \text{cm}^{-3}, T = 5 \text{eV.} \)
Fig. 1