# Application of Mollow's theory to Lamb-dip line shape

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By introducing the Doppler shift into Mollow's theory of saturated absorption and by performing the average over the velocity distribution, a Lamb-dip line shape is obtained. Analytical expressions and numerical results are presented. In some cases, these results do not agree with the results of the existing theory.

Mollow[1] has developed a theory for the absorption of radiation from a weak probe field by a two-level atom pumped by a strong near-resonant field. Absorption line shape was calculated for radiative and collisional relaxation, in the absence of Doppler broadening. Experimental measurements, using atomic beam techniques, verified the theory.[2] In such experiments Doppler shift and collisional effects are essentially eliminated. It is straightforward to incorporate the Doppler shift into this theory. Since the theory is valid for arbitrary pump field strength, it should be interesting to apply it to the Lamb-dip line shape in saturation spectroscopy,[3,4] and to compare the results with the existing theory for the case of no velocity changing collisions.[5]

In this work we derive the appropriate expressions and present the numerical results for several cases of interest. To keep the comparison simple, we also neglect the velocity dependence of the collision rates.[5] For the weak pump field case, our expression reduces to the standard result. However, for higher field strength, our results are quite different, compared with the results obtained in Ref. 5.

We consider a dilute gas of two-level atoms (radiators with lower state $|a\rangle$ and upper state $|b\rangle$) perturbed by foreign gas collisions. The absorption rate from the weak probe field of frequency $\omega'$ is determined by the imaginary part of the dielectric susceptibility. For an electric dipole transition one can write

$$\chi(\omega')=\hbar^{-1}|d_{ba}^{\lambda'}|^2F(\omega') \ , \tag{1}$$

where $d_{ba}^{\lambda'}$ is the component of the electric dipole matrix element for the transition $|a\rangle\rightarrow|b\rangle$ along the polarization vector $\vec{e}'_\lambda$ of the probe field. In the absence of the pump field, $F\rightarrow F_0$, given by the following expression (see, e.g., Ref. 6):

$$F_0(\omega')=\int\frac{d\vec{v}\,[n_b^{(0)}(\vec{v})-n_a^{(0)}(\vec{v})]}{\omega'-\omega_0-\vec{v}\cdot\vec{k}'+i\gamma_0/2} \ , \tag{2}$$

where $n_i^{(0)}(\vec{v})$ represents the thermal velocity distribution of the atoms in the two states $(i=a,b)$, $\omega_0$ is the transition frequency, $\vec{k}'$ is the propagation vector of the probe field, and $\gamma_0=\gamma_c+\gamma_N$, where $\gamma_c$ ($\gamma_N$) denote the collisional (natural) damping rate constant. Note that $\vec{v}\cdot\vec{k}'$ is the Doppler shift, and $k'=\omega'/c$, where $c$ is the velocity of light. Here we have neglected the collisional shift, the velocity changing collisions,[3,6] and the resonant collisions.[7] We have also made the rotating wave approximation, which is valid for the near-resonance case. If $\hbar\omega_0/KT>>1$ ($K$ is the Boltzmann constant and $T$ is the temperature), then $n_b^{(0)}<<n_a^{(0)}$ and, for radiators of mass $m$,

$$n_a^{(0)}(\vec{v})=n\left(\frac{m}{2\pi KT}\right)^{3/2}\exp\left(\frac{-mv^2}{2KT}\right) \ , \tag{3}$$

where $n$ is the number density of the radiators. Equations (2) and (3) give the well-known Voigt profile if $\gamma_c$ is independent of $\vec{v}$.

For a weak pump field, $n_b^{(0)}-n_a^{(0)}$ in (2) can be replaced by the modified population difference $n_b-n_a$, but the line-shape function can be left unchanged. For example, introducing Doppler shift into Eq. (2.11b) of Ref. 1, we obtain

$$n_b-n_a=\frac{(n_b^{(0)}-n_a^{(0)})(\Delta\omega^2+\gamma_0^2/4)}{(\gamma_0/2\gamma)\Omega^2+\Delta\omega^2+\gamma_0^2/4} \ , \tag{4}$$

$$\Delta\omega=\omega-\omega_0-\vec{v}\cdot\vec{k} \ , \tag{5}$$

where $\Omega$ is the Rabi frequency at resonance, $\vec{k}$ is the propagation vector of the pump field, and $\gamma=\gamma_N+\gamma_I$, where $\gamma_I$ is the inelastic collision rate. Now (2) is replaced by the following (our first approximation which is valid for the weak pump field case):

$$F_1(\omega')=\int\frac{d\vec{v}\,[n_b(\vec{v})-n_a(\vec{v})]}{\omega'-\omega_0-\vec{v}\cdot\vec{k}'+i\gamma_0/2} \ . \tag{6}$$

The modified absorption line shape is given by $\mathrm{Im}F_1$, whereas $\mathrm{Im}F_0$ represents the background. The Lamb-dip line shape is given by the difference between these two functions.

Now we specialize to the case of two counterpropagating beams, where $\vec{k}'=-\vec{k}$ and $\omega'=\omega$. Neglecting the velocity dependence of the collision rates, the integration over the two components of $\vec{v}$ which are perpendicular to $\vec{k}$ can be readily performed. Introducing a different set of the variables, we can write the result, obtained from (1)–(6), in the following form:

$$\Delta\chi_1(\eta)=\hbar^{-1}|d_{ba}^{\lambda'}|^2\mathrm{Im}(F_1-F_0)$$
$$=A\ \mathrm{Im}\int_{-\infty}^{+\infty}\frac{dV\exp(-V^2/\gamma_D^2)(\gamma_0/2\gamma)\Omega^2}{[(\eta-V)^2+g^2](\eta+V+i\gamma_0/2)]} \ , \tag{7}$$

 

where

$$A = n\,|d_{ba}^{\lambda'}|^2(\sqrt{\pi}\hbar\gamma_D)^{-1}\ , \tag{8}$$

$$\eta = (\omega' + \omega)/2 - \omega_0 = \omega - \omega_0\ , \tag{9}$$

$$g^2 = (\gamma_0/2\gamma)\Omega^2 + \gamma_0^2/4;\quad V = v_z k\ , \tag{10}$$

where $\gamma_D = k(m/2KT)^{-1/2}$, the Doppler width. This result is equivalent to Eq. (15) of Ref. 5. In the large Doppler width approximation, the exponential factor can be taken outside, and the integration can be done by completing the contour in the upper half of the complex $V$ plane. The result is

$$\Delta\chi_1(\eta) = A\,\exp(-\eta^2/\gamma_D^2)\phi_1(\eta)\ , \tag{11}$$

$$\phi_1(\eta) = \frac{\pi}{g}\,\frac{(\gamma_0/2\gamma)\Omega^2(g+\gamma_0/2)}{(2\eta)^2 + (g+\gamma_0/2)^2}\ . \tag{12}$$

$\phi_1(\eta)$ gives the standard result for the Lamb-dip line shape in the weak field and infinite Doppler width approximation.[5]

Next we generalize (7) for arbitrary pump field strength by using the complete absorption line-shape function obtained by Mollow.[1] In place of (6) we now use the following function obtained from Eq. (3.8) of Ref. 1, on introducing the Doppler shift:

$$F(\omega') = \int d\vec{v}[n_b(\vec{v}) - n_a(\vec{v})]f_a(\omega')\ , \tag{13}$$

$$f_a(\omega') = \frac{(\omega\omega' + \Delta\omega - i\gamma_0/2)(\omega\omega' - i\gamma) + (\Delta\omega - i\gamma_0/2)^{-1}\omega\omega'\Omega^2/2}{(\omega\omega' + \Delta\omega - i\gamma_0/2)(\omega\omega' - \Delta\omega - i\gamma_0/2)(\omega\omega' - i\gamma) - \Omega^2(\omega\omega' - i\gamma_0/2)}\ , \tag{14}$$

$$\omega\omega' = \omega - \omega' - \vec{v}\cdot(\vec{k} - \vec{k}')\ . \tag{15}$$

Again subtracting the background, we obtain from (1)–(4) and (13) the following result for the case $\vec{k}' = -\vec{k}$ and $\omega' = \omega$:

$$\Delta\chi(\eta) = A\,\mathrm{Im}\int_{-\infty}^{+\infty} dV\,\exp(-\eta^2/\gamma_D^2)\left\{\frac{1}{(\eta + V + i\gamma_0/2)} - \frac{(\eta - V)^2 + \gamma_0^2/4}{(\eta - V)^2 + g^2}\,\frac{(\eta - 3V - i\gamma_0/2)(2V + i\gamma) + (\eta - V - i\gamma_0/2)^{-1}V\Omega^2}{(\eta + V + i\gamma_0/2)(\eta - 3V - i\gamma_0/2)(2V + i\gamma) + \Omega^2(2V + i\gamma_0/2)}\right\}\ . \tag{16}$$

This result generalizes (7) for arbitrary pump field strength, assuming the fields are not so strong as to influence the collision rates.[8] In the large Doppler width approximation, the integral over $V$ can be evaluated as before, in the complex $V$ plane. [Note that all the poles of $f_a(V)$ are in the lower half plane.] The result is

$$\Delta\chi(\eta) = A\,\exp(-\eta^2/\gamma_D^2)\phi(\eta)\ , \tag{17}$$

$$\phi(\eta) = (\pi\Omega^2\gamma_0/\gamma g)\,\mathrm{Im}(B/D)\ , \tag{18}$$

$$B = (2g + \gamma_0)(4\eta + i6g + i\gamma_0) \times (2\eta + i2g + i\gamma) - i4\Omega^2(\eta + ig)\ , \tag{19}$$

$$D = (2g + \gamma_0)[(4\eta + i2g + i\gamma_0)(4\eta + i6g + i\gamma_0) \times (2\eta + i2g + i\gamma) - 2\Omega^2(4\eta + i4g + i\gamma_0)]\ . \tag{20}$$

This result should be compared with Eq. (30) of Ref. 5; the two results are different. To see this difference more clearly, let us consider the value of the Lamb-dip profile, at $\eta = 0$. (The profile is symmetrical about $\eta = 0$.) From (17) one obtains

$$\Delta\chi(0) = \frac{A\pi\gamma_0\Omega^2}{\gamma g(2g + \gamma_0)} \times \frac{(2g + \gamma_0)(6g + \gamma_0)(2g + \gamma) - 4\Omega^2 g}{(2g + \gamma_0)(6g + \gamma_0)(2g + \gamma) + 2\Omega^2(4g + \gamma_0)}\ . \tag{21}$$

The comparison of this result with Eq. (31) of Ref. 5 is shown in Fig. 1. (In our notation $\Omega^2 = 4\beta^2$, $\gamma_0 = 2\gamma_{ab}$, and $\gamma = \tau^{-1}$.) Dimensionless variables ($\Omega' = \Omega/\gamma$, $\gamma_0' = \gamma_0/\gamma$, $\eta' = \eta/\gamma$, and $\gamma_D' = \gamma_D/\gamma$) are used.

By a direct numerical integration of (16) it was found that the approximation (17) is quite accurate, at least for the case $\omega' = \omega$ and large Doppler width. Therefore (17) can be directly compared with the weak-field approximation (11).

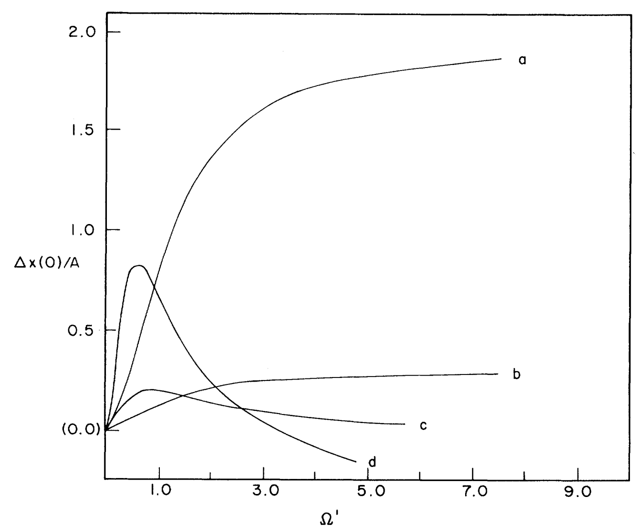


FIG. 1. Comparison of Eq. (21) of this work with Eq. (31) of Ref. 5. Curves a and d represent Eq. (21) of this work with $\gamma_0' = 2.0$ and $\gamma_0' = 0.2$, respectively; curves b and c represent Eq. (31) of Ref. 5 for the same parameters. ($\gamma_D' = 50$ in all cases.) Amplification is visible in curves c and d for large $\Omega'$. ($\Omega' = \Omega/\gamma$, etc.)

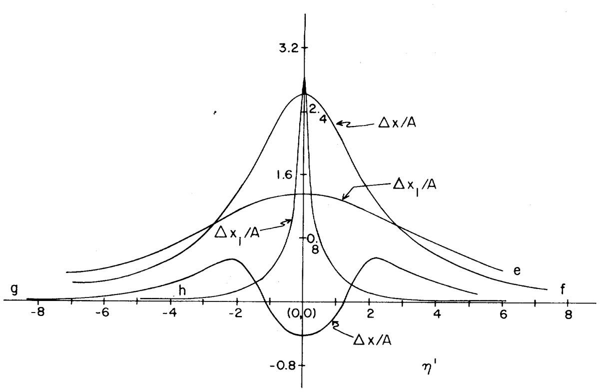


FIG. 2. Comparison of Eqs. (17) and (11) of this work for $\omega = \omega'$ and $\gamma_D' = 50$. Curves e and g represent Eq. (17) with $\gamma_0' = 1.0$, $\Omega' = 5.0$ and $\gamma_0' - 0.1$, $\Omega' = 3.0$, respectively. Curves f and h represent Eq. (11) for the same parameters. Amplification is visible in curve g. ($\Omega' = \Omega/\gamma$, etc.)

The results are as shown in Fig. 2. As expected, the difference between the two results increases as $\Omega'$ increases, and (17) exhibits amplification for large $\Omega'$. For low $\Omega'$, Eqs. (17) and (11) agree quite well.

It appears that the nature of the approximations involved in the two theories is different. Mollow's theory is based on Markov approximation and quantum regression theorem. The nature of the approximations in Ref. 5 is not so clear. For example, in addition to the impact approximation and the neglect of memory effects, the correlations between successive photon events are also neglected in this work.

The other approximations, mentioned earlier, are related to the collision model. For example, velocity changing collisions are of considerable interest.[3,6,8,9] However, the equations of motion cannot be solved for arbitrary $\Omega$, if such collisions are retained; perturbative solutions, to third order in $\Omega$, have been obtained.[10] Further work on this problem is in progress.

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