# Effect of <br> Nonlinear Level Crossing on the Output of a Gas Laser 

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#### Abstract

A generalized expression for electromagnetic polarization under level crossing in a single-mode gas laser is derived up to the third order in the field strength. This is made possible by the use of the probability integral of complex arguments to avoid the Doppler limit approximation. The level crossing effect is seen to appear in the third order, and its linewidth is shown to be asymmetric. It is also broader than the Lorentzian shape usually obtained under the Doppler limit. The dimensionless intensity parameter of the laser field is seen to lose its symmetry about the resonance value in the presence of level crossing. This is attributed to incoherence among different transitions. Further, the other terms like those describing the two-photon process in the third-order value are briefly discussed.


## Introduction

The phenomenon of level crossing, well known in the investigation of the fine structure of the atom (Colegrove et al. 1959; Rose and Carovillano 1961), occurs when energy levels overlap within their linewidths. Excitation and hence scattering take place coherently, resulting in a change in angular distribution of scattered radiation, though the total resonance scattering rate is unaffected.

More recently, Dumont and Decomps (1968) have given a semiclassical theory for the level crossing effect on spontaneous emission in a laser. Luntz et al. (1969) observed the level crossing effect in nonlinear absorption in a Stark-tuned $\mathrm{CH}_{4}$ cell placed inside a laser cavity. Shimoda (1972) has described theoretically level crossing in a two-level system in which both the upper and the lower levels consist of a bunch of nearly degenerate levels. His theory brings out the influence of level crossing on laser performance, but the usefulness of his work is restricted because of the Doppler limit (where the transverse relaxation parameter $\gamma_{a b}$ is assumed to be extremely small compared with the Doppler parameter $k u$ ). The purpose of this paper is to describe the effect of level crossing on the output of a gas laser when the Doppler limit is lifted, so that the theory of level crossing is generalized to laser operation away from resonance and to any ratio of $\gamma_{a b}$ to $k u$.

## Formulation of Problem

We follow Lamb (1964) in formulating the problem. A cavity of length $L$ operating at a single frequency is considered. The field inside the cavity is given by

$$
\begin{equation*}
E(z, t)=E(t) U(z) \cos (v t+\phi) \tag{1}
\end{equation*}
$$

where the frequency $v$ is nearly equal to the laser output frequency, and $U(z)$ is the single normal mode of the cavity and may be taken to be equal to $\sin (k z)$. The field $E(z, t)$ is a scalar representing plane polarized light. We consider a two-level atom, each level consisting of a set of nearly degenerate sublevels, with the upper level $a$ having sublevels $a, a^{\prime}, \ldots$ and the lower level $b$ having sublevels $b, b^{\prime}, \ldots$. The major levels are separated by

$$
\begin{equation*}
E_{a}-E_{b}=\hbar \omega_{a b} \tag{2}
\end{equation*}
$$

where $\omega_{a b}$ is the resonant frequency. The state of the system is represented by a density matrix $\rho$ which obeys the equation

$$
\mathrm{i} \hbar \dot{\rho}=[H, \rho], \quad \text { with } \quad H=H_{0}+H_{1},
$$

where $H$ is the total Hamiltonian, $H_{0}$ is the unperturbed Hamiltonian and $H_{1}$ is the time-dependent perturbation. In the dipole approximation, $H_{1}$ is given by

$$
H_{1}=-(\mathscr{P} / \hbar) E(z, t),
$$

where $\mathscr{P}$ is the dipole moment operator. The matrix for $H$ contains a large number of rows and columns, depending on the number of sublevels of the lasing levels. For example, the Hamiltonian for an atom whose levels are doubly degenerate has the form

$$
H=\left[\begin{array}{llll}
E_{a} & 0 & H_{a b} & H_{a b^{\prime}}  \tag{3}\\
0 & E_{a^{\prime}} & H_{a^{\prime} b} & H_{a^{\prime} b^{\prime}} \\
H_{b a} & H_{b a^{\prime}} & E_{b} & 0 \\
H_{b^{\prime} a} & H_{b^{\prime} a^{\prime}} & 0 & E_{b^{\prime}}
\end{array}\right],
$$

where $H_{i j}=-\left(\mathscr{P}_{i j} / \hbar\right) E(z, t)$ is the interaction Hamiltonian between the states $i$ and $j$. We have neglected the interaction Hamiltonians $H_{a a^{\prime}}, H_{b b^{\prime}}, \ldots$, assuming them to be extremely feeble.

At this stage, the relaxation parameters $\gamma_{a b}, \gamma_{a a^{\prime}}, \gamma_{b b^{\prime}}, \gamma_{a}$ and $\gamma_{b}$ are introduced phenomenologically to account for the damping of the corresponding levels when the perturbation is switched off. The equations of motion for different elements of the density matrix are given by

$$
\begin{align*}
& \dot{\rho}_{a b}=-\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \rho_{a b}-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{a^{\prime}} \rho_{a a^{\prime}} \mathscr{P}_{a^{\prime} b}-\sum_{b^{\prime}} \mathscr{P}_{a b^{\prime}} \rho_{b^{\prime} b}\right),  \tag{4a}\\
& \dot{\rho}_{a a^{\prime}}=-\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right) \rho_{a a^{\prime}}-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{b} \rho_{a b} \mathscr{P}_{b a^{\prime}}-\sum_{b} \mathscr{P}_{a b} \rho_{b a^{\prime}}\right),  \tag{4b}\\
& \dot{\rho}_{b b^{\prime}}=-\left(\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b b^{\prime}}\right) \rho_{b b^{\prime}}-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{a} \rho_{b a} \mathscr{P}_{a b^{\prime}}-\sum_{a} \mathscr{P}_{b a} \rho_{a b^{\prime}}\right),  \tag{4c}\\
& \dot{\rho}_{a a}=-\gamma_{a} \rho_{a a}-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{b} \rho_{a b} \mathscr{P}_{b a}-\sum_{b} \mathscr{P}_{a b} \rho_{b a}\right),  \tag{4d}\\
& \dot{\rho}_{b b}=-\gamma_{b} \rho_{b b}-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{a} \rho_{b a} \mathscr{P}_{a b}-\sum_{a} \mathscr{P}_{b a} \rho_{a b}\right) . \tag{4e}
\end{align*}
$$

In the zeroth order, we have

$$
\begin{gather*}
\rho_{a a}^{(0)}\left(a, z_{0}, t_{0}, v, t\right)=\exp \left\{-\gamma_{a}\left(t-t_{0}\right)\right\},  \tag{5a}\\
\rho_{a b}^{(0)}=\rho_{a a^{\prime}}^{(0)}=\rho_{b b^{\prime}}^{(0)}=0,  \tag{5b}\\
\rho_{b b}^{(0)}\left(b, z_{0}, t_{0}, v, t\right)=\exp \left\{-\gamma_{b}\left(t-t_{0}\right)\right\} . \tag{5c}
\end{gather*}
$$

Thus the equation of motion for $\rho_{a b}$ in the first order is given by

$$
\begin{equation*}
\dot{\rho}_{a b}^{(1)}+\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \rho_{a b}^{(1)}=-\{\mathrm{i} E(z, t) / \hbar\} \mathscr{P}_{a b}\left(\rho_{a a}^{(0)}-\rho_{b b}^{(0)}\right) . \tag{6}
\end{equation*}
$$

As this does not show any level crossing effects, we need not discuss them further in this approximation.

## Calculation of Third-order Polarization

In the iterative procedure, the equation of motion for the third-order off-diagonal element is given by

$$
\begin{equation*}
\dot{\rho}_{a b}^{(3)}+\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \rho_{a b}^{(3)}=-\{\mathrm{i} E(z, t) / \hbar\}\left(\sum_{a^{\prime}} \rho_{a a^{\prime}}^{(2)} \mathscr{P}_{a^{\prime} b}-\sum_{b^{\prime}} \mathscr{P}_{a b^{\prime}} \rho_{b^{\prime} b}^{(2)}\right) . \tag{7}
\end{equation*}
$$

The solution for $\rho_{a a^{\prime}}^{(2)}, \ldots$ can be obtained from

$$
\begin{equation*}
\dot{\rho}_{a a^{\prime}}^{(2)}+\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right) \rho_{a a^{\prime}}^{(2)}=-\{\mathrm{i} E(z, t) / \hbar\} \sum_{b}\left\{\rho_{a b}^{(1)} \mathscr{P}_{b a^{\prime}}-\mathscr{P}_{a b} \rho_{b a^{\prime}}^{(1)}\right\} \tag{8}
\end{equation*}
$$

by substituting the expression for $\rho_{a b}^{(1)}$, obtained from equation (6), to give

$$
\begin{align*}
& \rho_{a a^{\prime}}^{(2)}\left(z_{0}, t_{0}, z, v, t\right) \\
&=-\sum_{b}\left\{\mathscr{P}_{a b} \mathscr{P}_{b a^{\prime}} / \hbar^{2}\right\} \int_{t_{0}}^{t} \mathrm{~d} t^{\prime} \int_{t_{0}}^{t^{\prime}} \mathrm{d} t^{\prime \prime} E\left(z, t^{\prime}\right) E\left(z, t^{\prime \prime}\right) \\
& \times \exp \left\{\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right)\left(t^{\prime \prime}-t^{\prime}\right)+\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right)\left(t^{\prime}-t\right)\right\} \\
& \times\left[\exp \left\{\gamma_{a}\left(t_{0}-t^{\prime \prime}\right)\right\}-\exp \left\{\gamma_{b}\left(t_{0}-t^{\prime \prime}\right)\right\}\right] \\
&-\sum_{b}\left\{\mathscr{P}_{a b} \mathscr{P}_{b^{\prime}} / \hbar^{2}\right\} \int_{t_{0}}^{t} \mathrm{~d} t^{\prime} \int_{t_{0}}^{t^{\prime}} \mathrm{d} t^{\prime \prime} \\
& \times \exp \left\{\left(\gamma_{a^{\prime} b}-\mathrm{i} \omega_{a^{\prime} b}\right)\left(t^{\prime \prime}-t^{\prime}\right)+\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right)\left(t^{\prime}-t\right)\right\} \\
& \times\left[\exp \left\{\gamma_{a^{\prime}}\left(t_{0}-t^{\prime \prime}\right)\right\}-\exp \left\{\gamma_{b}\left(t_{0}-t^{\prime \prime}\right)\right\}\right] \tag{9}
\end{align*}
$$

These expressions are for the atoms excited at the space-time point $z_{0}, t_{0}$. We are considering the effects at the space-time point $z, t$. The relation between these two space-time points is given by

$$
\begin{equation*}
z=z_{0}+v\left(t-t_{0}\right), \tag{10}
\end{equation*}
$$

where $v$ is the velocity of the atom. The value of $\rho_{a b}^{(3)}$ from equations (7) and (9) is

$$
\begin{array}{rl}
\rho_{a b}^{(3)}\left(z_{0}, t_{0}, z, v, t\right)=-\mathrm{i} \hbar^{-1} \int_{t_{0}}^{t} & \mathrm{~d} t^{\prime} \\
& E\left(z, t^{\prime}\right) \\
& \times\left(\sum_{a^{\prime}} \rho_{a a^{\prime}}^{(2)} \mathscr{P}_{a^{\prime} b}-\sum_{b^{\prime}} \mathscr{P}_{a b^{\prime}} \rho_{b^{\prime} b}^{(2)}\right) \exp \left\{\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right)\left(t^{\prime}-t_{0}\right)\right\}
\end{array}
$$

The summation over $z_{0}, t_{0}$ gives

$$
\begin{array}{rl}
\rho_{a b}^{(3)}(z, v, t)=-\mathrm{i} \hbar^{-1} \int_{-\infty}^{t} & \mathrm{~d} t_{0} \int_{t_{0}}^{t} \mathrm{~d} t^{\prime} E\left(z, t^{\prime}\right) \\
& \times\left(\sum_{a^{\prime}} \rho_{a a^{\prime}}^{(2)} \mathscr{P}_{a^{\prime} b}-\sum_{b^{\prime}} \mathscr{P}_{a b^{\prime}} \rho_{b^{\prime} b}^{(2)}\right) \exp \left\{\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right)\left(t^{\prime}-t_{0}\right)\right\} . \tag{11}
\end{array}
$$

To account for the excitation of atoms to the lasing levels, the rate of excitation $\lambda_{\alpha}\left(z_{0}, t_{0}, v\right)$ of atoms to the state $\alpha$ per unit time and unit volume is introduced. Assuming $\lambda_{\alpha}\left(z_{0}, t_{0}, v\right)$ to be a slowly varying function of $z_{0}, t_{0}$, we may replace it by $\lambda_{\alpha}(z, t, v)$. Thus, after substitution of equations (9) and (1) into (11), and changing the order of integration as follows:

$$
\begin{equation*}
\int_{-\infty}^{t} \mathrm{~d} t_{0} \int_{t_{0}}^{t} \mathrm{~d} t^{\prime} \int_{t_{0}}^{t^{\prime}} \mathrm{d} t^{\prime \prime} \int_{t_{0}}^{t^{\prime \prime}} \mathrm{d} t^{\prime \prime \prime} \rightarrow \int_{-\infty}^{t} \mathrm{~d} t^{\prime} \int_{-\infty}^{t^{\prime}} \mathrm{d} t^{\prime \prime} \int_{-\infty}^{t^{\prime \prime}} \mathrm{d} t^{\prime \prime \prime} \int_{-\infty}^{t^{\prime \prime \prime}} \mathrm{d} t_{0} \tag{12}
\end{equation*}
$$

with

$$
\tau^{\prime}=t-t^{\prime}, \quad \tau^{\prime \prime}=t^{\prime}-t^{\prime \prime} \quad \text { and } \quad \tau^{\prime \prime \prime}=t^{\prime \prime}-t^{\prime \prime \prime}
$$

equation (11) becomes

$$
\begin{align*}
& \rho_{a b}^{(3)}(z, v, t) \\
& =\sum_{a^{\prime}} \sum_{b^{\prime}} \mathrm{i} \mathscr{P}_{a b^{\prime}} \mathscr{P}_{b^{\prime} a^{\prime}} \mathscr{P}_{a^{\prime} b} \hbar^{-3} E^{3} \int_{0}^{\infty} \mathrm{d} \tau^{\prime} \int_{0}^{\infty} \mathrm{d} \tau^{\prime \prime} \int_{0}^{\infty} \mathrm{d} \tau^{\prime \prime \prime} \\
& \times U\left(z-v \tau^{\prime}\right) U\left(z-v\left(\tau^{\prime}+\tau^{\prime \prime}\right)\right) U\left(z-v\left(\tau^{\prime}+\tau^{\prime \prime}+\tau^{\prime \prime \prime}\right)\right) \cos \left(v t^{\prime}+\phi\right) \cos \left(v t^{\prime \prime}+\phi\right) \cos \left(v t^{\prime \prime \prime}+\phi\right) \\
& \times\left\{\left(\lambda_{a} \gamma_{a}^{-1}-\lambda_{b^{\prime}} \gamma_{b^{\prime}}^{-1}\right) \exp \left(-\left(\gamma_{a b^{\prime}}+\mathrm{i} \omega_{a b^{\prime}}\right) \tau^{\prime \prime \prime}-\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right) \tau^{\prime \prime}-\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \tau^{\prime}\right)\right. \\
& \quad+\left(\lambda_{a^{\prime}} \gamma_{a^{\prime}}^{-1}-\lambda_{b^{\prime}} \gamma_{b^{\prime}}^{-1}\right) \exp \left(-\left(\gamma_{a^{\prime} b^{\prime}}-\mathrm{i} \omega_{\left.a^{\prime} b^{\prime}\right)}\right) \tau^{\prime \prime \prime}-\left(\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}\right) \tau^{\prime \prime}-\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \tau^{\prime}\right) \\
& \quad+\left(\lambda_{a^{\prime}} \gamma_{a^{\prime}}^{-1}-\lambda_{b} \gamma_{b}^{-1}\right) \exp \left(-\left(\gamma_{a^{\prime} b}+\mathrm{i} \omega_{a^{\prime} b}\right) \tau^{\prime \prime \prime}-\left(\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}\right) \tau^{\prime \prime}-\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \tau^{\prime}\right) \\
& \left.\quad+\left(\lambda_{a^{\prime}} \gamma_{a^{\prime}}^{-1}-\lambda_{b^{\prime}} \gamma_{b^{\prime}}^{-1}\right) \exp \left(-\left(\gamma_{a^{\prime} b^{\prime}}-\mathrm{i} \omega_{a^{\prime} b^{\prime}}\right) \tau^{\prime \prime \prime}-\left(\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}^{\prime}\right) \tau^{\prime \prime}-\left(\gamma_{a b}+\mathrm{i} \omega_{a b}\right) \tau^{\prime}\right)\right\} . \tag{13}
\end{align*}
$$

The polarization $P$ of the medium at $z, t$ is given by

$$
\begin{equation*}
P(z, t)=\sum_{a} \sum_{b} \int_{-\infty}^{+\infty}\left\{\mathscr{P}_{a b} \rho_{b a}(z, v, t)+\mathscr{P}_{b a} \rho_{a b}(z, v, t)\right\} \mathrm{d} v . \tag{14}
\end{equation*}
$$

The Fourier component of the polarization is

$$
\begin{equation*}
P(t)=2 L^{-1} \int_{0}^{L} P(z, t) \sin (k z) \mathrm{d} z . \tag{15}
\end{equation*}
$$

We separate the time dependence of $\lambda_{\alpha}$ by means of

$$
\begin{equation*}
\lambda_{\alpha}(z, v, t)=\Lambda_{\alpha}(z, t) W(v), \tag{16}
\end{equation*}
$$

where $W(v)$ is the velocity distribution function for the lasing atoms. We take $W(v)$ to be Maxwellian, i.e.

$$
\begin{equation*}
W(v)=\pi^{-\frac{1}{2}} u^{-1} \exp \left(-v^{2} / u^{2}\right), \tag{17}
\end{equation*}
$$

where $u$ is the speed parameter given by $u^{2}=2 k_{\mathrm{B}} T / M$, with $T$ the temperature of the medium. The product of the $U$ 's and $\sin (k z)$ from equation (15) give different cosine functions. We pick the term $\cos \left(k v\left(\tau^{\prime}-\tau^{\prime \prime \prime}\right)\right)$ from among them, as this is the only one having a peak within the limits of integration. In the rotatingwave approximation, we get the third-order polarization as

$$
\begin{align*}
P^{(3)}(z, t)= & \sum_{a} \sum_{a^{\prime}} \sum_{b} \sum_{b^{\prime}} \mathrm{i} \frac{\mathscr{P}_{b a} \mathscr{P}_{a b^{\prime}} \mathscr{P}_{b^{\prime} a^{\prime}} \mathscr{P}_{a^{\prime} b}}{32 \hbar^{3} \pi^{\frac{1}{2}} u} E^{3} \exp (-\mathrm{i}(v t+\phi)) \\
& \times\left(A \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\}^{2}+(k v)^{2}}+B \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left\{\gamma_{a b^{\prime}}+\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)\right\}^{2}+(k v)^{2}}\right. \\
& \left.+C \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left\{\gamma_{a^{\prime} b}+\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)\right\}^{2}+(k v)^{2}}+D \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left\{\gamma_{a^{\prime} b^{\prime}}+\mathrm{i}\left(\omega_{a^{\prime} b^{\prime}}-v\right)\right\}^{2}+(k v)^{2}}\right) . \tag{18}
\end{align*}
$$

The coefficients $A, B, C$ and $D$ in terms of

$$
\begin{equation*}
\bar{N}_{\alpha \beta}=L^{-1} \int_{0}^{L} N_{\alpha \beta}(z) \mathrm{d} z, \quad \text { with } \quad N_{\alpha \beta}=\Lambda_{\alpha} / \gamma_{\alpha}-\Lambda_{\beta} / \gamma_{\beta}, \tag{19}
\end{equation*}
$$

are given by

$$
\begin{align*}
\begin{aligned}
A= & \left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} \\
& \times\left\{\frac{1}{\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}} \frac{\bar{N}_{a b^{\prime}}}{\gamma_{a b^{\prime}}+\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}\right. \\
& \quad+\frac{\bar{N}_{a^{\prime} b^{\prime}}}{\gamma_{a^{\prime} b^{\prime}}-\mathrm{i}\left(\omega_{a^{\prime} b^{\prime}}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}\left(\frac{1}{\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}}+\frac{1}{\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}}\right) \\
& \left.\quad+\frac{1}{\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}} \frac{\bar{N}_{a^{\prime} b}}{\gamma_{a^{\prime} b}+\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}\right\}, \\
B= & \frac{\bar{N}_{a b^{\prime}}}{\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}} \frac{\gamma_{a b^{\prime}}+\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)}{\gamma_{a b^{\prime}}+\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}, \\
C= & \frac{\bar{N}_{a^{\prime} b}}{\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}} \frac{\gamma_{a^{\prime} b}+\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)}{\gamma_{a^{\prime} b}+\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}, \\
D= & \frac{\bar{N}_{a^{\prime} b^{\prime}}\left\{\gamma_{a^{\prime} b^{\prime}}-\mathrm{i}\left(\omega_{a^{\prime} b^{\prime}}-v\right)\right\}}{\gamma_{a^{\prime} b^{\prime}}-\mathrm{i}\left(\omega_{a^{\prime} b^{\prime}}-v\right)+\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)}\left(\frac{1}{\gamma_{a a^{\prime}}+\mathrm{i} \omega_{a a^{\prime}}}+\frac{1}{\gamma_{b b^{\prime}}+\mathrm{i} \omega_{b^{\prime} b}}\right) .
\end{aligned}
\end{align*}
$$

The crossed levels should satisfy the usual selection rules, that is, $\Delta m=1$ or 2 , where $m$ is the magnetic quantum number of the sublevels and $\Delta m$ is the difference in the magnetic quantum numbers of the crossing levels. In the present notation, the condition for level crossing (for two sublevels in each level) can be represented as:

$$
\begin{array}{lll}
E_{a}=E_{a} & \text { and } & E_{b}=E_{b^{\prime}} \\
E_{a} \neq E_{a^{\prime}} & \text { and } & E_{b}=E_{b^{\prime}} \\
E_{a}=E_{a^{\prime}} & \text { and } & E_{b} \neq E_{b^{\prime}} . \tag{21c}
\end{array}
$$

Under the condition $a=a^{\prime}$ and $b=b^{\prime}$, equation (18) reduces to the form of Lamb's (1964) expression (Mohanty and Nayak, to be published). As the relaxation parameters are independent of sublevels in many cases, we assume the relations

$$
\begin{align*}
& \gamma_{a a^{\prime}}=\gamma_{a^{\prime} a}=\gamma_{a^{\prime}}=\gamma_{a},  \tag{22a}\\
& \gamma_{b b^{\prime}}=\gamma_{b^{\prime} b}=\gamma_{b^{\prime}}=\gamma_{b},  \tag{22b}\\
& \gamma_{a b^{\prime}}=\gamma_{a^{\prime} b}=\gamma_{a^{\prime} b^{\prime}}=\gamma_{a b} . \tag{22c}
\end{align*}
$$

On keeping the terms in equation (18) which obey the selection rules (21), we obtain after some rearrangements

$$
\begin{equation*}
P^{(3)}(t)=\sum_{a, b} \mathrm{i}\left|\mathscr{P}_{a b}\right|^{2} \frac{1}{32} \hbar^{-3} E^{3} \exp \{-\mathrm{i}(v t+\phi)\}\left\{L_{\mathrm{D}}+L_{2 a}+L_{2 b}+L_{\mathrm{DX}}+L_{a \mathrm{X}}+L_{b \mathrm{X}}\right\}+\mathrm{c.c} \tag{23}
\end{equation*}
$$

where c.c. denotes the complex conjugate. The various $L$ terms appearing in equation (23) are:

$$
\begin{align*}
L_{\mathrm{D}}= & \left|\mathscr{P}_{a b}\right|^{2} \bar{N}_{a b}\left[2 \gamma^{-1} I_{a b}+\gamma^{-1} \gamma_{a b}^{-1}\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}\right. \\
& \left.+\gamma^{-1} \gamma_{a b}^{-1}\left\{\gamma_{a b}-\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}^{*}\right],  \tag{24a}\\
L_{2 a}= & \left|\mathscr{P}_{a^{\prime} b}\right|^{2} \bar{N}_{a^{\prime} b} \gamma_{b}^{-1}\left\{2 \gamma_{a b}+\mathrm{i}\left(\omega_{a^{\prime} b}+\omega_{a b}-2 v\right)\right\}^{-1} \\
& \times\left[\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}+\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)\right\} I_{a^{\prime} b}\right],  \tag{24b}\\
L_{2 b}= & \left|\mathscr{P}_{a b^{\prime}}\right|^{2} \bar{N}_{a b^{\prime}} \gamma_{a}^{-1}\left\{2 \gamma_{a b}+\mathrm{i}\left(\omega_{a b}+\omega_{a b^{\prime}}-2 v\right)\right\}^{-1} \\
& \times\left[\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}+\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)\right\} I_{a b^{\prime}}\right],  \tag{24c}\\
L_{\mathrm{DX}}= & \bar{N}_{a b}\left\{\left|\mathscr{P}_{a^{\prime} b}\right|^{2}\left(\gamma_{a}+\mathrm{i} \omega_{a^{\prime} b}\right)^{-1}+\left|\mathscr{P}_{a b^{\prime}}\right|^{2}\left(\gamma_{b}+\mathrm{i} \omega_{b^{\prime} b}\right)^{-1}\right\},  \tag{24d}\\
L_{a \mathrm{X}}= & \left|\mathscr{P}_{a^{\prime} b}\right|^{2} \bar{N}_{a^{\prime} b} \gamma_{b}^{-1}\left(\frac{\gamma_{a}+\gamma_{b}+\mathrm{i} \omega_{a a^{\prime}}}{\left(\gamma_{a}+\mathrm{i} \omega_{a a^{\prime}}\right)\left\{2 \gamma_{a b}+\mathrm{i}\left(\omega_{a b}-\omega_{a^{\prime} b}\right)\right\}}\right) \\
& \times\left[\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}+\left\{\gamma_{a b}-\mathrm{i}\left(\omega_{a^{\prime} b}-v\right)\right\} I_{a^{\prime} b}^{*}\right],  \tag{24e}\\
L_{b \mathrm{X}}= & \left|\mathscr{P}_{a b^{\prime}}\right|^{2} \bar{N}_{a b^{\prime}} \gamma_{a}^{-1}\left(\frac{\gamma_{a}+\gamma_{b}+\mathrm{i} \omega_{b^{\prime} b}}{\left(\gamma_{b}+\mathrm{i} \omega_{b^{\prime} b}\right)\left\{2 \gamma_{a b}+\mathrm{i}\left(\omega_{a b}-\omega_{a b^{\prime}}\right)\right\}}\right) \\
& \times\left[\left\{\gamma_{a b}+\mathrm{i}\left(\omega_{a b}-v\right)\right\} I_{a b}+\left\{\gamma_{a b}-\mathrm{i}\left(\omega_{a b^{\prime}}-v\right)\right\} I_{a b^{\prime}}^{*}\right], \tag{24f}
\end{align*}
$$

where

$$
\gamma^{-1}=\frac{1}{2}\left(\gamma_{a}^{-1}+\gamma_{b}^{-1}\right)
$$

The quantities $I_{i j}$ and $I_{i j}^{*}$ are defined as

$$
\begin{align*}
I_{i j} & =\frac{1}{\pi^{\frac{1}{2}} u} \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left.\gamma_{i j}+\mathrm{i}\left(\omega_{i j}-v\right)\right\}^{2}+(k v)^{2}} \\
& =\frac{\pi^{\frac{1}{2}}}{k u} \frac{\gamma_{i j}-\mathrm{i}\left(\omega_{i j}-v\right)}{\gamma_{i j}^{2}+\left(\omega_{i j}-v\right)^{2}}\{U(x, y)+\mathrm{i} V(x, y)\} \tag{25a}
\end{align*}
$$

and

$$
\begin{align*}
I_{i j}^{*} & =\frac{1}{\pi^{\frac{1}{2}} u} \int_{-\infty}^{+\infty} \frac{\exp \left(-v^{2} / u^{2}\right) \mathrm{d} v}{\left.\gamma_{i j}-\mathrm{i}\left(\omega_{i j}+v\right)\right\}^{2}+(k v)^{2}} \\
& =\frac{\pi^{\frac{1}{2}}}{k u} \frac{\gamma_{i j}+\mathrm{i}\left(\omega_{i j}-v\right)}{\gamma_{i j}^{2}+\left(\omega_{i j}-v\right)^{2}}\{U(x, y)+\mathrm{i} V(x, y)\}, \tag{25b}
\end{align*}
$$

where $x=\left(v-\omega_{i j}\right) / k u, y=\gamma_{i j} / k u$, and $U$ and $V$ are the real and imaginary parts of the complex probability integral whose numerical values are widely tabulated (Faddeyeva and Terent'ev 1961).


Fig. 1. Comparison of the present expression (full curve) for the real part of $L_{a \mathrm{X}}$ as a function of $\omega_{a a^{\prime}} / k u$ with the expression (dashed curve) obtained by Shimoda (1972). (See text for assumptions.)

The term $L_{\mathrm{D}}$ shows a Lamb dip at transition frequencies $\omega_{a b}, \omega_{a^{\prime} b}, \ldots$ The halfwidths of these transition lines are not affected by level crossing. The term $L_{\mathrm{DX}}$ represents the effect of level crossing on the Lamb dip. The terms $L_{a \mathrm{X}}$ and $L_{b \mathrm{X}}$ show level crossing between the levels $a$ and $a^{\prime}$ and between $b$ and $b^{\prime}$ respectively. The real part of $L_{a \mathrm{X}}$ is plotted (full curve) against $\omega_{a a^{\prime}} / \gamma_{a}$ in Fig. 1, where we have assumed that $\gamma_{a b}=\gamma_{a}=\gamma_{b}$ holds so as to simplify the plotting. We have also assumed that $\gamma_{a b} / k u=0 \cdot 1$ and $\left(v-\omega_{a b}\right) / k u=0 \cdot 1$. It can be seen that the line is noticeably asymmetric about $\omega_{a a^{\prime}}=0$. The halfwidth of $2.08 \gamma_{a}$ is broader than that of the Shimoda (1972) line shape (dashed curve in Fig. 1) which is symmetric about the $\omega_{a a^{\prime}}=0$ line. The expression for $L_{a x}$ reduces to Shimoda's result at the Doppler limit very near to resonance. Finally, the term $L_{2 a}$ represents a two-photon transition of frequency $\frac{1}{2}\left(\omega_{a b}+\omega_{a^{\prime} b}\right)$. It may be emphasized here that the present results are free from any approximations except for those encountered in the numerical
evaluation of $I_{i j}$, and so the present theory is more general than Shimoda's, and is free from the Doppler limit restriction in particular.

## Discussion

For a laser operating in the vicinity of the transition frequency $\omega_{a b}$, the two terms $L_{2 a}$ and $L_{2 b}$ representing the two-photon transition need not be considered, as they are of no importance. The terms $L_{a \mathrm{X}}$ and $L_{b \mathrm{x}}$ have the same sign. As a result, they have the same effect on the intensity parameter and their contributions to it accordingly add. So, for simplicity, we take only the upper lasing level $a$ to be degenerate,


Fig. 2. Dependence of the dimensionless intensity parameter $\mathscr{I}$ on $\left(v-\omega_{a b}\right) / k u$ for $\omega_{a a^{\prime}}=0.5 \gamma_{a}$ (full curve) and $\omega_{a a^{\prime}}=0 \cdot 1 \gamma_{a}$ (dashed curve). (See text for assumptions.)
that is, we assume $b=b^{\prime}$. Further, as the dipole moment operators are nearly equal to each other, we assume that $\mathscr{P}_{a b}=\mathscr{P}_{a^{\prime} b}=\mathscr{P}_{a^{\prime} a}=\ldots=\mathscr{P}$. For convenience, the transverse relaxation term $\gamma_{a b}$ is taken equal to $\frac{1}{2}\left(\gamma_{a}+\gamma_{b}\right)$. Under these conditions, the amplitude equation (Lamb 1964) $\dot{E}=\alpha E-\beta E^{3}$ becomes at steady state $E^{2}=\alpha / \beta$, where

$$
\alpha=-\frac{1}{2} v Q^{-1}\left\{1-\bar{N}_{a b} U(x, y) / U(0, y)\right\}
$$

and

$$
\begin{aligned}
& \beta=v Q^{-1} \sum_{a} \mathscr{P}^{2}\left\{32 \hbar^{2} U(0, y) \gamma_{a} \gamma_{b}\right\}^{-1} \\
& \times \quad \bar{N}_{a b}\left\{1+\left(v-\omega_{a b}\right)^{2} \gamma_{a b}^{-2}\right\}^{-1} k u \gamma_{a b}^{-1} \\
& \times\left(4\left\{\gamma_{a b}(k u)^{-1} U(x, y)-\left(v-\omega_{a b}\right)(k u)^{-1} V(x, y)\right\}\right. \\
&\left.\quad+2\left(v-\omega_{a b}\right) \gamma_{a b}^{-1}\left\{\left(v-\omega_{a b}\right)(k u)^{-1} U(x, y)+\gamma_{a b}(k u)^{-1} V(x, y)\right\}\right) \\
&+ \bar{N}_{a^{\prime} b}\left(1+\omega_{a a^{\prime}}^{2} \gamma_{a}^{-2}\right)^{-1} \\
& \times\left(\left\{U(x, y)+U\left(x^{\prime}, y\right)\right\}+\omega_{a a^{\prime}} \gamma_{a}^{-1}\left\{V(x, y)-V\left(x^{\prime}, y\right)\right\}\right) \\
&+ 2 \bar{N}_{a b} U(x, y) \\
&+ \bar{N}_{a b}\left\{1+\left(v-\omega_{a b}\right)^{2} \gamma_{a b}^{-2}\right\}^{-1} k u \gamma_{a b}^{-1} \\
& \times\left(\left[\gamma_{b} \gamma_{a b}^{-1}\left\{1+\left(\omega_{a a^{\prime}} \gamma_{a}^{-1}\right)^{2}\right\}^{-1}+\gamma_{a} \gamma_{a b}^{-1}\right]\right. \\
& \times\left\{\gamma_{a b}(k u)^{-1} U(x, y)-\left(v-\omega_{a b}\right)(k u)^{-1} V(x, y)\right\} \\
&+\left(1+\omega_{a a^{\prime}}^{2} \gamma_{a}^{-2}\right)^{-1}\left(\omega_{a a^{\prime}} \gamma_{b}\right)\left(\gamma_{a} \gamma_{a b}\right)^{-1} \\
&\left.\left.\times\left\{\left(v-\omega_{a b}\right)(k u)^{-1} U(x, y)+\gamma_{a b}(k u)^{-1} V(x, y)\right\}\right)\right\} .
\end{aligned}
$$

The dimensionless intensity parameter $\mathscr{I}$ is given by

$$
\begin{align*}
\mathscr{I} & =\frac{1}{2}(\mathscr{P} E)^{2} / \hbar^{2} \gamma_{a} \gamma_{b} \\
& =8\left\{U(x, y)-\bar{N}_{a b}^{-1} U(0, y)\right\} / \mathscr{J}, \tag{26a}
\end{align*}
$$

where the denominator $\mathscr{J}$ is given by

$$
\begin{align*}
\mathscr{J}= & k u \gamma_{a b}^{-1}\left\{1+\left(v-\omega_{a b}\right)^{2} \gamma_{a b}^{-2}\right\}^{-1}\left[4\left\{\gamma_{a b}(k u)^{-1} U(x, y)-\left(v-\omega_{a b}\right)(k u)^{-1} V(x, y)\right\}\right. \\
& \left.+2\left(v-\omega_{a b}\right) \gamma_{\mathrm{ab}}^{-1}\left\{\left(v-\omega_{a b}\right)(k u)^{-1} U(x, y)+\gamma_{a b}(k u)^{-1} V(x, y)\right\}\right] \\
+ & \bar{N}_{a^{\prime} b} \bar{N}_{a b}^{-1}\left(1+\omega_{a a^{\prime}}^{2} \gamma_{a}^{-2}\right)^{-1} \\
& \times\left[U(x, y)+U\left(x^{\prime}, y\right)+\omega_{a a^{\prime}} \gamma_{a}^{-1}\left\{V(x, y)-V\left(x^{\prime}, y\right)\right\}\right] \\
+ & 2 U(x, y)+k u \gamma_{a b}^{-1}\left\{1+\left(v-\omega_{a b}\right)^{2} \gamma_{a b}^{-2}\right\}^{-1} \\
& \times\left[\left\{\gamma_{b} \gamma_{a b}^{-1}\left(1+\omega_{a a^{\prime}}^{2} \gamma_{a}^{-2}\right)^{-1}+\gamma_{a} \gamma_{a b}^{-1}\right\}\right. \\
& \times\left\{\gamma_{a b}(k u)^{-1} U(x, y)-\left(v-\omega_{a b}\right)(k u)^{-1} V(x, y)\right\} \\
& +\left(\omega_{a a^{\prime}} \gamma_{b} \gamma_{a}^{-1} \gamma_{a b}^{-1}\right)\left(1+\omega_{a a^{\prime}}^{2} \gamma_{a}^{-2}\right)^{-1} \\
& \left.\quad \times\left\{\left(v-\omega_{a b}\right)(k u)^{-1} U(x, y)+\gamma_{a b}(k u)^{-1} V(x, y)\right\}\right] \tag{26b}
\end{align*}
$$

with $x^{\prime}=x-\omega_{a^{\prime} a}(k u)^{-1}$. It should be noted that the expression for $\mathscr{I}$ (equations (26)) does not reduce to the generalized Lamb (1964) expression on substitution of $a=a^{\prime}$ and $b=b^{\prime}$, as all the terms are not included. The value of $\mathscr{I}$, which is a result of an iteration calculation, is valid for $\bar{N}_{a^{\prime} b}<2$ and $\bar{N}_{a b}<2$ (Mohanty and Nayak, to be published).

The variation of $\mathscr{I}$ with detuning, namely $\mathscr{I}\left(\left(\nu-\omega_{a b}\right) / k u\right)$ for $\omega_{a a^{\prime}}=0 \cdot 5 \gamma_{a}$ (full curve) and $\omega_{a a^{\prime}}=0 \cdot 1 \gamma_{a}$ (dashed curve), with the relation $\gamma_{a}=10 \gamma_{b}$, is shown in Fig. 2. We have also assumed that $k u / \gamma_{a b}=10, \bar{N}_{a b}=1.5$ and $\bar{N}_{a^{\prime} b}=1.4$. The linewidth is seen to increase with increasing $\omega_{a a^{\prime}}$ as expected. One striking feature is that the lineshape becomes asymmetric about the resonance line. This is a consequence of incoherence between different transitions owing to the finite width of the upper lasing level. The existence of this incoherence is clear from the form of the equations (26) and can also be demonstrated through a correlation analysis of the field $E(z, t)$. It is consistent with the facts that the output of a gas laser is coherent and the lineshape is symmetric about the resonance line only when both levels are nondegenerate. For degeneracy in any of the levels, the field is more chaotic, and its fluctuations have a higher average value due to the nonlinearity of the medium (Shen 1969). Thus the peak value for $\mathscr{I}$ increases as $\omega_{a a^{\prime}}$ increases, which is demonstrated in Fig. 2.

The lineshapes for the two-photon transition terms $L_{2 a}$ and $L_{2 b}$ are not pure Lorentzians, as can be seen from equations (24b) and (24c). These lines are broader than a Lorentzian line. In the Doppler limit $(y \gg 1)$ very near to resonance, $L_{2 a}$ becomes symmetric and is given by

$$
\begin{equation*}
L_{2 a}=\left|\mathscr{P}_{a^{\prime} b}\right|^{2} \bar{N}_{a^{\prime} b} \gamma_{b}^{-1} 2 \pi^{\frac{1}{2}}(k u)^{-1} /\left\{2 \gamma_{a b}+\mathrm{i}\left(\omega_{a^{\prime} b}+\omega_{a b}-2 v\right)\right\}, \tag{27}
\end{equation*}
$$

which is the same as Shimoda's (1972) result.

If we take collisions between the atoms into account then, for the physically realizable condition $\gamma_{a} \gg \gamma_{b}$, the level crossing term $L_{a \mathrm{X}}$ becomes

$$
\begin{equation*}
L_{a \mathrm{X}}=\left|\mathscr{P}_{a^{\prime} b}\right|^{2} \bar{N}_{a^{\prime} b} \gamma_{b}^{-1} \pi^{\frac{1}{2}}(k u)^{-1}\left\{U(x, y)+U\left(x^{\prime}, y\right)\right\} /\left(2 \gamma_{a b}+\mathrm{i} \omega_{a a^{\prime}}\right), \tag{28}
\end{equation*}
$$

which shows that collisions broaden the line shape for $L_{a \mathrm{X}}$. The same effect is seen in $L_{2 a}$ and $L_{2 b}$. Here again we note that our results reduce to those of Shimoda (1972) very near resonance under the Doppler limit.

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