DIAGRAM METHOD IN A LASER THEORY

by A. V. TOULOUB, *Department of Physics and Astrophysics, University of Delhi, Delhi 7, India*

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The calculation of nonlinear macroscopic polarization is being described with the help of diagram method formalism.

One of the main problems in a laser theory is the calculation of nonlinear macroscopic polarization of the medium. In order to calculate this function we need to investigate a perturbation expansion of one particle density matrix for those electronic states which are active in the laser radiation. In some problems the contribution of the third order terms to the medium polarization may be of great importance. In those cases it would also be necessary to take into account a next order perturbation expression. In connection with this problem it seems to be reasonable to standardize our computation labour in the sense of introduction of diagram technique formalism. We shall use, following Lamb (1964), a density matrix describing an ensemble of atoms which arrive at $\vec{r}$ with velocity $\vec{v}$ at time t regardless of their position $\vec{r}_0$ at time t_0 or of the initial condition j of excitation

$$\rho_{m\mu}\left(\vec{r}, \vec{v}, t\right) = \sum_{m,\mu} \int_{-\infty}^{t} dt_0 \int d\vec{r}_0 \Lambda_j\left(\vec{r}_0, t_0\right) \rho_{m\mu}\left(j, \vec{r}_0, t_0, \vec{v}, t\right) \delta\left(\vec{r} - \vec{r}_0 - \vec{v}t + \vec{v}t_0\right) \quad .. \quad (1)$$

In expression (1), the indices m and μ belong correspondingly to the excited and to the ground states of each individual atom and $\Lambda_j\left(\vec{r}_0, t_0\right)$ gives the number of atoms excited to state j per unit volume and time. For using the standard perturbation method formalism it is useful to introduce a_m, α_μ and a_m^+, α_μ^+, the annihilation and creation operators respectively, for the excited and for the ground states. In terms of these operators the density matrix $\rho_{m\mu}\left(j, \vec{r}_0, t_0, \vec{v}, t\right)$ in (1) can be written in the form *

$$\rho_{m\mu}(j, t) = < j \left| \alpha_{(t)}^+ a_{(t)} \right| j > \quad .. \quad .. \quad .. \quad (2)$$

where the operators are taken in Heisenberg representation and $|j> = \psi_j$ is the

* We drop here all other variables.

initial wave function for the pure state at time $t = t_0$. The interaction energy operator is

$$H_I(t) = \sum_{m,\mu} \left\{ < \mu \,|\, U \,|\, m > \alpha_\mu^+ a_m e^{-i\omega_{m\mu} t} + \text{h.c.} \right\} \quad .. \quad .. \quad (3)$$

where

$$< \mu \,|\, U \,|\, m > = - \sum_{\xi = 0, \pm 1} (-1)^\xi < \mu \,|\, d_{-\xi} \,|\, m > E_\xi \quad .. \quad .. \quad (4)$$

E_ξ being the spherical components of the electrical field. For the case of linear polarization of radiation, the electrical field can be expanded in normal mode eigenfunctions (Lamb 1964).

$$E_z = E_0 = \sum_n \sin k_n z \, \mathcal{E}_n(t) \cos (\nu_n t + \phi_n(t)) \quad .. \quad .. \quad (5a)$$

and for circular polarized radiation

$$E_+ = -\sqrt{2}\, E_1 = \sum_n \sin k_n z \left\{ \mathcal{E}_{nI}(t) e^{i(\nu_{nI} t + \phi_{nI}(t))} + \mathcal{E}_{nII}(t) e^{-i(\nu_{nII} t + \phi_{nII}(t))} \right\}$$

$$E_{-1} = -E_1^*, \quad .. \quad .. \quad .. \quad .. \quad (5b)$$

where ν_{nI}, ν_{nII} are respectively the frequencies and $\mathcal{E}_{nI}$, $\mathcal{E}_{nII}$ the amplitudes for the left and the right circularly polarized lights. *

The Doppler effect can be taken into account by considering the electrical field at time t to be (Lamb 1964)

$$E\left(\vec{r}, t\right) = E\left(\vec{r_0} + \vec{v}(t - t_0), t\right) \quad .. \quad .. \quad .. \quad .. \quad (5c)$$

where $\vec{v}$ is the velocity of an atom. †

In the interaction representation, the expression (2) can be rewritten as

$$\rho_{m\mu}(j, t) = < j \,|\, U^+(t_1 t_0) \alpha_\mu^+(t) a_m(t) U(t, t_0) \,|\, j > \quad .. \quad .. \quad (6)$$

where $U(t, t_0)$ means the well-known expansion

$$U(t, t_0) = 1 + \sum_{n=1}^{\infty} (-i)^n \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \ldots \int_{t_0}^{t_{n-1}} dt_n H_I(t_1) H_I(t_2) \ldots H_I(t_n). \quad (7)$$

The effect of the external field is usually described in diagrams by some kind of a line, going from infinity to some point at the electron line. But, for the case of the density matrix, we need to introduce two electron lines for the one-particle density matrix (Konstantinov and Perel 1960). At the upper line, which begins at point t_0 and ends at point t, points t_1, t_2, ...

* This type of field expansion is used, for instance, in the theory of Zeeman effect for the case of an axial magnetic field.

† The most important case is that one, where only an axial dependence of electrical field is being considered.

indicate the actions of operators $H_I(t_1)$, $H_I(t_2)$, ... on the initial wave function. Points $t_1', t_2', \ldots$ at the bottom line would similarly indicate the actions of operators in the expression for $U^+(t, t_0)$. It should be mentioned, however, that the position of operators at the bottom line is reversed in time with respect to those on the upper line: the operator whose time variable is greater acts first on the wave function. Vertical slits in the diagram give relative values of times $t_1, t_2, \ldots$ and $t_1', t_2', \ldots$, or, in other words, the limits of integration. As an example, in Fig. 1 are shown four diagrams for density matrix $< \psi_a \mid \alpha^+(t)a(t) \mid \psi_a >$ in the third order perturbation theory corresponding to the Lamb approximation (Lamb 1964). The first diagram indicates that the limits of integration are

$$\int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{t_2} dt_3 \quad \text{where } t_3 \equiv t'.$$

In fact, the limits of integration for times $t_1, t_2, \ldots$ are independent of the limits $t_1', t_2', \ldots$ on the bottom line. In order to obtain this independence, it would be necessary to rearrange all the bottom points in all possible positions with respect to the points on the upper line. An example of such a rearrangement is given in Fig. 1.

So the first rule can be formulated as follows:

Graph of the nth order contains n-external lines on both the electron lines. All possible diagrams for the given order of perturbation theory can be obtained from the initial one by means of rearrangement of relative positions of points on the two electron lines. The limits of time integration are indicated by vertical slits.

Now it is necessary to point out a one-to-one correspondence between graphs and definite mathematical expressions. In order to solve the equation of motion for the density matrix it is necessary to fix the initial condition at the time $t = t_0$. If the direction of time growth is chosen from left to right, then we shall write at the left side of each electron line the letter m corresponding to the initial wave function $|m> = a_m^+ \Phi_0$.

The initial state can also be chosen in a more complicated manner in the form of a coherent mixture. In this case, more general matrix element $< m_j | \alpha_\mu^+ a_m | m_i >$ would occur in the theory. It can be described by letters m_i and m_j at the left side for the upper and the bottom electron lines respectively. For those lines on the right side we shall write the letters m and μ respectively for the symbols for annihilation and creation operators. Indices of intermediate states should be written between the external lines.

Each vertex point has a factor $\exp(-i\omega_{ij}t)$ which corresponds to the transition $i \to j$ along the electron line or, in other words, from the left to the right for the upper line and from the right to the left for the bottom line.

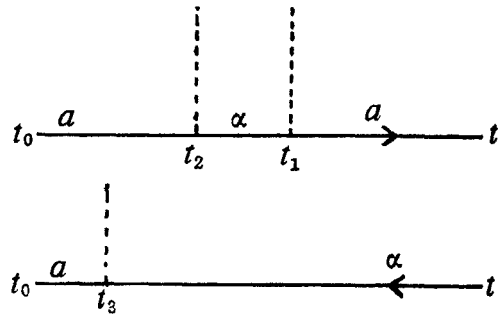

FIG. 1a.

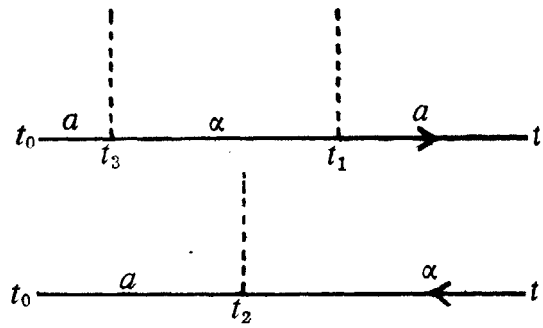

FIG. 1b.

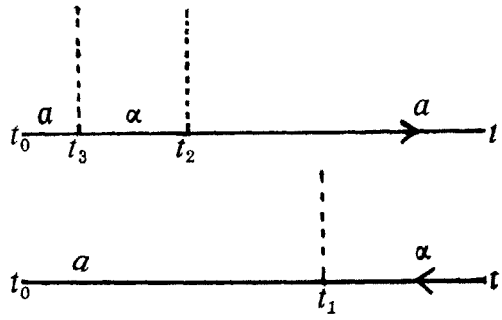

FIG. 1c.

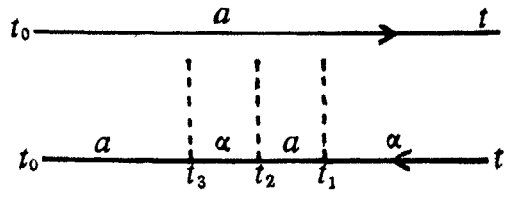

FIG. 1d.

It is known that the product of all these factors can be rewritten with the help of the transformation $\omega_{ij} = \omega_{ik} - \omega_{jk}$ in a form which contains only the time differences $\tau_1 = t - t_1$, $\tau_2 = t_1 - t_2$, ... $\tau_0 = t_n - t_0$, namely,

$$\exp\left\{-i\sum_{k=0}^{n}\omega_{st}\tau_k\right\},$$

where ω_{st} stands for the frequency difference between the upper and the bottom electron lines (Lamb 1964).

We shall now discuss in a more detailed form the effect of the external fields (5*a*) and (5*b*) in resonance case. If the electrical field in a laser tube has a linear polarization (*see* eqn. 5*a*), then the dependence would be a trivial one. At every vertical slit we shall write, instead of the factor $\exp\{-i\omega_{st}\tau_k\}$, the factor $\exp\{-i(\omega_{st}\pm\nu)\tau_k\}$.* Let us consider now the case of circular polarized radiation. At each vertex point t_k would be a factor

$$\exp\{-i(\omega_{ij}-\nu_{ij})t_k\},$$

which corresponds to the transition $i \to j$ and the frequency would be $\nu_{ij} = \nu_I$ if $\Delta m_{ij} = 1$ and $\nu_{ij} = \nu_{II}$ if $\Delta m_{ij} = -1$. These are the usual selection rules. The product of all these functions should now be written as a function of all (τ_k). Then to every vertical slit between $[t_{k-1}, t_k]$ would correspond a factor

$$\exp\{-i(\omega_{st}-\nu_{st})\tau_k\},$$

where the frequency ν_{st} should be chosen according to the following rules. If, at every vertical slit, indices s and t correspond to different levels then for the case $\Delta m_{st} = +1$, the frequency ν_{st} would be equal to $\nu_{st} = \nu_I$ and for $\Delta m_{st} = -1$ equal to $\nu_{st} = \nu_{II}$, the frequency of a right polarized light. At some vertical slits there can also arise Zeeman sub-levels belonging to the same principal quantum number. In the latter case, the frequency ν_{st} is given by $\nu_{st} = \Delta\nu \equiv \nu_I - \nu_{II}$, if $\Delta m_{st} > 0$ and by $\nu_{st} = -\Delta\nu$, if $\Delta m_{st} < 0$. So the second rule can be formulated as follows:

To every vertical slit corresponds a factor exp $\{-i(\omega_{st}-\nu_{st})\tau_k\}$, *where* ω_{st} *is the difference between upper and bottom frequencies, and* $\nu_{st} = \nu_I$ *for* $\Delta m_{st} = +1$, $\nu_{st} = \nu_{II}$ *for* $\Delta m_{st} = -1$, *if s and t are different levels, and* $\nu_{st} = \pm\Delta\nu$ *for* $\Delta m_{st} \gtrless 0$, *if s and t are Zeeman sub-levels.*

Let us now consider the self-energy diagrams. The most important terms in this expression arise from one photon approximation. This sequence of diagrams is shown in Fig. 2 for the upper line. If $A_m(+)$ is the wave

* For $\omega_{st} > 0$ negative sign should be taken, i.e. $(\omega_{st}-\nu)$ and for $\omega_{st} < 0$ positive sign, i.e. $(\omega_{st}+\nu)$.

function which includes all the above-mentioned diagrams, then it would satisfy the following integral equation (Fock and Tulub 1965):

$$A_m(t) = e^{-i\omega_m t} + \int_0^t dt_1 e^{-i\omega_m(t-t_1)} \int_0^{t_1} dt_2 \sum_m^{(2)} (t_1 - t_2) A_m(t_2) \quad .. \quad (8)$$

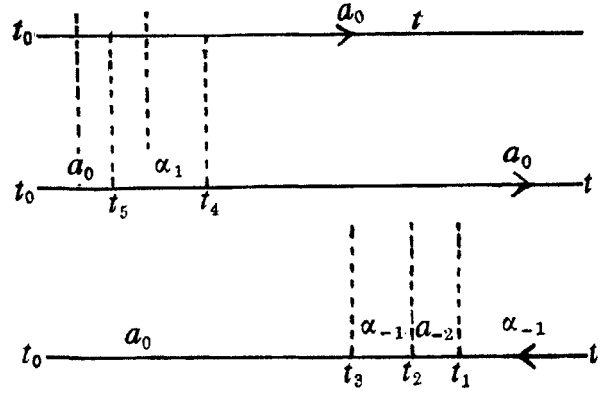

FIG. 2.

where we have put

$$A_m(0) = 1$$

and

$$\sum_m^{(2)} (\tau) = (-i)^2 \sum_{q,\lambda,\mu} |< \mu | V_{q\lambda} | m >|^2 e^{i(\omega_{m\mu} - \omega_q)\tau} \quad .. \quad .. \quad (9)$$

and $< \mu | V_{q\lambda} | m >$ is the interaction energy matrix element for the quantized electromagnetic field.

The solution of eqn. (8) can be easily obtained with the help of Laplace transformation (Fock and Tulub 1965):

$$A_m(t) = \frac{1}{2\pi i} \int_{\sigma - i\infty}^{\sigma + i\infty} \frac{e^{\gamma t}}{\gamma + i\omega_m - \sum_m^{(2)} (\gamma)} d\gamma . \quad .. \quad .. \quad (10)$$

From this expression can be derived in some approximation the usual formula

$$A_m(t) = e^{-i(\omega_m + \Delta\omega_m)t - \frac{\Gamma_m}{2} t} \quad .. \quad .. \quad .. \quad (11)$$

where Γ_m is the natural decay constant and $\Delta E_m = \hbar \Delta \omega_m$ is the Lamb shift in non-relativistic approximation. If the quantity $\Delta \omega_m$ is considered to be included in the energy of the upper and the bottom lines, then the effect of all the self-energy diagrams consists in the introduction of a factor exp $\left(-\frac{\Gamma_s}{2} t\right)$ for the upper and correspondingly exp $\left(-\frac{\Gamma_t}{2} t\right)$ for the bottom electron lines.

In a laser theory decay constant has not any simple connection with the natural line width constant and in the expression $\exp\left(-\frac{\gamma}{2}t\right)$ quantity γ should be considered as some phenomenological parameter. We can now formulate the third rule:

To every electron line corresponds some decay factor $\exp\left(-\frac{\gamma}{2}t\right)$. *At every vertical slit decay constants should be added.*

Figure 3 gives an example of a fifth order non-diagonal density matrix for the states with the quantum numbers $j_m = 2$, $j_\mu = 1$. To this diagram

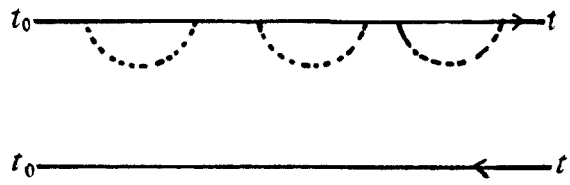

FIG. 3.

correspond the following expressions under the sign of time integral:

$$\exp(-i\nu_I t)\cdot\exp\{-i(\omega_{0,-1}-\nu_I)\tau_1-i(\omega_{0,-2}-\Delta\nu)\tau_2-i(\omega_{0,-1}-\nu_I)\tau_3$$
$$+i(\omega_{0,1}-\nu_{II})\tau_5\}\exp\{-\gamma_{a\alpha}\tau_1-\gamma_a\tau_2-\gamma_{a\alpha}\tau_3-\gamma_a\tau_4-\gamma_{a\alpha}\tau_5-\gamma_a\tau_0\},$$
$$\gamma_{a\alpha}=\tfrac{1}{2}(\gamma_a+\gamma_\alpha).$$

Let us now consider the integrals over space and time coordinates r_0 and t_0 in expression (1), assuming that the quantity $A_j(r, t)$ can be effectively replaced by the constant A_j. The effect of integration consists in replacing all the arguments $\vec{r}_0+\vec{v}(t_k-t_0)$ in the product $H_I(t_1)H_I(t_2)\ldots$ (*see* 5*c*) by the arguments

$$\vec{r}_0+\vec{v}(t_k-t_0)\to\vec{r}-\vec{v}(t-t_k).$$

The integration over the variable t_0 in (1) can be easily done according to the formula

$$\int_{-\infty}^{t}dt_0\int_{t_0}^{t}dt_1\ldots\int_{t_0}^{t_{n-1}}dt_n e^{-\gamma_j(t_n-t_0)}F(t_1,\ldots t_n)=$$
$$\frac{1}{\gamma_j}\int_{-\infty}^{t}dt_1\ldots\int_{-\infty}^{t_{n-1}}dt_n F(t_1,\ldots t_n). \qquad (12)$$

In other words we need to take into account in expression (1) for $\rho(z, v, t)$ only n vertical slits between the points $[t, t_1]$, $[t_1, t_2]$, $\ldots$, $[t_{n-1}, t_n]$ and then to

multiply the whole expression over the initial decay time γ_j^{-1}. After this the arguments $(t-t_k)$ can be written as functions of time differences,

$$t-t_k = \sum_{e=1}^{k} \tau_e . \quad .. \quad .. \quad .. \quad .. \quad (13)$$

We thus obtain the fourth rule:

The coefficients before the time integrals would be equal to $(-i)^k i^e d^n (\Lambda_j/\gamma_j)$ *exp* $(-i\nu_{m\mu}t)$ *where k and e mean the number of vertex points on the upper and on the bottom lines if the initial condition is chosen as 'j', and d— is the dipole matrix element. The frequency* $\nu_{m\mu}$ *is* $\nu_{m\mu} = \nu_I$ *for* $\Delta m = +1$ *and* $\nu_{m\mu} = \nu_{II}$ *for* $\Delta m = -1$.

If we take into account both types of initial states at the moment $t = t_0$, then the non-diagonal matrix element would have a typical factor $(\Lambda_m/\gamma_m - \Lambda_\mu/\gamma_\mu)$.

With the help of diagram technique it is relatively easy to investigate high order perturbation expressions. As an example we consider two level systems with excited state a and the ground state α. It can be shown, after a simple rearrangement of points on the upper and the bottom lines, that the perturbation expansion in the rotation field approximation has the form*

$$\rho_{a\alpha}(a, z, t) = i\frac{d \cdot \mathcal{E}}{2} \sum_{n=0}^{\infty} (-1)^n \left(\frac{d^2\mathcal{E}^2}{4}\right)^n \int_0^t dt_1 \ldots \int_0^{t_{2n}} dt_{2n+1} \phi(a, z, \tau_1, \ldots \tau_{2n+1}) \quad .. \quad (14)$$

where

$$\phi(a, z, \tau_1, \ldots \tau_{2n+1}) = \prod_{e=1}^{2n+1} (\sin kz_e) \exp\{[-i(\omega_0-\nu)-\gamma_{a\alpha}]\tau_1\}$$

$$u(\tau_2)v(\tau_3) \ldots u(\tau_{2n})v(\tau_{2n+1}) \exp(-\gamma_a t_{2n+1}) \quad .. \quad .. \quad (14a)$$

and functions $u(\tau)$ and $v(\tau)$ are equal

$$u(\tau) = \exp(-\gamma_a\tau) + \exp(-\gamma_\alpha\tau)$$

$$v(\tau) = 2\cos(\omega_0-\nu)\tau \quad .. \quad .. \quad .. \quad .. \quad .. \quad (15)$$

where

$$\omega_0 = \omega_{a\alpha}, \quad z_e = z - v(t-t_e).$$

The complicated dependence of the arguments z_e on the τ variables makes the summation difficult in a general case. But if we put the velocity $v = 0$, the sum (14) can be easily calculated with the help of Laplace transformation:

$$\rho_{a\alpha}(a, z, t) = \frac{1}{2\pi i} \int_{\sigma-i\infty}^{\sigma+i\infty} \frac{\left(\frac{\vec{id} \cdot \vec{\mathcal{E}}}{2} \sin kz\right) e^{\gamma t} d\gamma}{[\gamma+\gamma_a][\gamma+\gamma_{a\alpha}+i(\omega_0-\nu)]\left[1+\frac{\mathcal{E}^2 d^2}{4} u(\gamma)v(\gamma)\right]} \quad .. \quad (16)$$

* Perturbation expansion contains in this case only odd number terms.

where $u(\gamma)$, $v(\gamma)$ are the Laplace transformations of $u(\tau)$ and $v(\tau)$. Integral (16) can be easily calculated. If we put $\gamma_a = \gamma_\alpha = 0$,

$$\rho_{a\alpha}(a, z, t) = i\frac{\vec{d}.\vec{\mathscr{E}}}{2\Lambda}\sin kz\left[\sin \Lambda t + i\frac{(\omega_0-\nu)}{\Lambda}\cos \Lambda t - i\frac{(\omega_0-\nu)}{\Lambda}\right] \quad .. \quad (17)$$

$$\Lambda = \sqrt{(\omega_0-\nu)^2+\vec{d}^2\vec{\mathscr{E}}^2}\sin^2 kz.$$

The latter expression can also be obtained directly from the equations of motion for density matrix.

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