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Lowest order second harmonic conversion in a two-level gaseous system

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This paper deals with second harmonic conversion in a TLS which has the property of a gas. The ensemble consisting of TLS is assumed to obey the Maxwell velocity distribution. In solving the problem use has been made of the probability function of a complex argument to discuss operation away from Doppler limit. The influence of the relaxation constant γ_{ab} and the Doppler parameter ku on various physical quantities like second harmonic field and quality factor are brought out.

1. Introduction

In a recent publication (Mohanty 1974), (hereafter referred to as I) one of the authors has discussed the process of lowest order optical second-harmonic conversion in a two-level system (TLS), which was assumed to be an ideal solid. Expressions for the second-harmonic electric polarisation $P_{2\nu}$, electric field $E_{2\nu}$ and the threshold condition were derived. This paper discuss some aspects of the same problem when the material system is closer in its physical properties to a gas than to a solid. Although the magnitude of $P_{2\nu}$ in a gaseous system is much smaller than in a denser system it is not a disadvantage as a similar reduction also applies to the linear electric polarisation. Obvious advantages like phase-matching over longer distances and favourable $P_{2\nu}$ to P_{linear} ratio (Armstrong et al 1962) give added interest to the study of $P_{2\nu}$ in a gaseous system.

A general expression for the lowest-order second-harmonic electric polarisation is derived in section 2. In section 3 it is averaged over a Maxwellian velocity distribution to obtain $P_{2\nu}$ for a gaseous system. As the assumption of a Doppler limit is not made, it is not necessary to restrict the discussions of the paper entirely to low pressure gases. However, the discussions are predominantly true for gases. Expressions for $E_{2\nu}$ and the threshold condition are derived in section 4 and various aspects of the results are critically analysed.

2. General Expression for Second-Order Electric Polarisation

As in I the presence of the diagonal elements of the matrix representing the electric dipole moment operator μ is taken for granted. The calculations are based on the Lamb's theory of optical masers (Lamb 1964) which adequately explains most observed features of gas laser operation (Riska & Stenholm 1970). A general equation of motion describing the response of TLS interacting with two electromagnetic fields $E(z, t)$ and $E_d(z, t)$ may be given in terms of the density matrix ρ of the TLS, as follows (Mohnanty 1974)

$$\dot{\rho}_{ab} = -(i\omega + \gamma_{ab})\rho_{ab} - iV_d(t)\rho_{ab} + iV(t)(\rho_{aa} - \rho_{bb})$$

$$\rho_{ba} = \rho^*_{ab}$$

$$\dot{\rho}_{aa} = -\gamma_a\rho_{aa} + iV(t)(\rho_{ab} - \rho_{ba})$$

$$\dot{\rho}_{bb} = -\gamma_b\rho_{bb} - iV(t)(\rho_{ab} - \rho_{ba}) \qquad \ldots (1)$$

ω is the (positive) transition frequency $(E_a - E_b)/\hbar$, γ_a and γ_b are the respective decay constants for the upper (a) and the lower (b) levels and γ_{ab} is the transverse decay rate generally unrelated to γ_a or γ_b (Shimoda & Uehara 1971). The interaction terms are $V(t) = 2r\hbar^{-1}E(z, t)$ and $V_d(t) = 2r_d\hbar^{-1}E_d(z, t)$ where r and r_d are the matrix elements ρ_{ab} and $(\rho_{aa} - \rho_{bb})$. Under the initial condition $\rho_{aa}^{(0)} = \exp.(-\gamma_a(t - t_0))$, $\rho_{bb}(t_0) = \rho_{ab}(t_0) = \rho_{ba}(t_0) = 0$ and following *I*, the second order term in ρ_{ab} is defined by the equation

$$\dot{\rho}_{ab}^{(2)} + (i\omega + \gamma_{ab})\rho_{ab}^{(2)} = iV_d(t)\rho_{ab}^{(1)} \qquad \ldots (2)$$

where the first order term for the density matrix, $\rho_{ab}^{(1)}$ is defined in *I*.

The solution of eq. (2) is then

$$\rho_{ab}^{(2)}(a, z, v, t) = \lambda_a/\gamma_a \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' V_d(t')V(t'')\exp[(i\omega + \gamma_{ab}).$$

$$(t'' - t')].\exp.[(i\omega + \gamma_{ab})(t' - t)] \qquad \ldots (3)$$

where λ_i ($i = a, b$) is the excitation rate density to state i.

The optical fields are

$$E_d = E_\zeta(t)U_\zeta[z - v(t - t')]\cos(\nu_\zeta t + \phi_\zeta(t))$$

$$E = E_\xi(t)U_\xi[z - v(t - t')]\cos(\nu_\xi t + \phi_\xi(t)) \qquad \ldots (4)$$

E_ζ, E_ξ and ϕ's are slowly varying functions of time and only one mode is taken into account. The expression for the second order electric polarisation is simplified by neglecting the small z dependence of E_ζ and E_ξ (Mohanty 1974). Then,

$$P_2^{(\nu_\zeta+\nu_\xi)}(z, t) = r\int_{-\infty}^{+\infty} dV[\rho_{ab}^{(2)}(a, z, v, t) + \rho_{ab}^{(2)}(b, z, v, t)] + \text{complex conj.}$$

$$= -\tfrac{1}{2}rAN(z)\cos(k_\zeta + k_\xi)z\exp[-i\{(\nu_\zeta + \nu_\xi)t + \phi_\zeta + \phi_\xi\}].$$

$$\int_{-\infty}^{+\infty} dvW(v)\int_0^\infty dT\int_0^\infty dT'\cos[(k_\zeta + k_\xi)vT + k_\xi vT']\exp.$$

$$[-(\gamma_{ab} + i\Delta)T]\exp.[-(\gamma_{ab} + i\Omega)T'] + \text{c.c.} \qquad \ldots (5)$$

where $A = 4E_\zeta E_\xi \gamma\gamma_a \hbar^{-2}$, $\Delta = \omega-\nu_\xi-\nu_\zeta$, $\Omega = \omega-\nu_\zeta$, $T = (t-t')$, $T' = (t'-t'')$ and $N(z) = (\Lambda_a/\gamma_a-\Lambda_b/\gamma_b)$ with $\Lambda_i = \Lambda_i W(v)$, $W(v)$ being the atomic velocity distribution.

3. $P_2^{(\nu_\xi+\nu_\zeta)}(z, t)$ in the Case of Fluids

In the case of fluids a Maxwell distribution of velocities is assumed for $W(v)$

$$W(v) = (\pi^{\frac{1}{2}}u)^{-1}\exp.(-v^2/u^2).$$

Unlike *I* the integrations over T and T' are carried out first, giving for P_2

$$P_2^{(\nu_\zeta+\nu_\xi)}(z,t) = -\tfrac{1}{2}rAN(z)\cos(k_\zeta+k_\xi)z\exp[-i\{(\nu_\zeta+\nu_\xi)t+\phi_\zeta+\phi_\xi\}]\int_{-\infty}^{+\infty}dvW(v)$$

$$\frac{(\gamma_{ab}+i\Omega)(\gamma_{ab}+i\Delta)-k_\xi(k_\zeta+k_\xi)v^2}{\{(\gamma_{ab}+i\Omega)^2+k_\zeta^2v^2\}\{(\gamma_{ab}+i\Delta)^2+(k_\zeta+k_\xi)^2v^2\}}\quad \text{c.c.} \tag{6}$$

The integral may be rewritten to give :

$$P_2^{(\nu_\zeta+\nu_\xi)}(z,t) = -\frac{rAN(z)}{2D\pi^{\frac{1}{2}}}\cos(k_\zeta+k_\xi)z\exp[-i\{(\nu_\zeta+\nu_\xi)t+\phi_\zeta+\phi_\xi\}]$$

$$\left[\frac{2(\gamma_{ab}+i\Omega)}{k_\zeta u}\int_0^\infty \frac{\exp(-v^2/u^2)}{\left(\frac{\gamma_{ab}+i\Omega}{k_\zeta}\right)^2+v^2}dv\right.$$

$$\left.-\frac{2(\gamma_{ab}+i\Delta)}{(k_\zeta+k_\xi)u}\int_0^\infty \frac{\exp(-v^2/u^2)}{\left(\frac{\gamma_{ab}+i\Delta}{k_\zeta+k_\xi}\right)^2+v^2}dv\right]+\text{c.c.} \tag{7}$$

where $D = k_\xi(\gamma_{ab}+i\Delta)-(k_\zeta+k_\xi)(\gamma_{ab}+i\Omega)$.

Substituting $\alpha = (i\gamma_{ab}-\Omega)/k_\zeta u$ and $\beta = (i\gamma_{ab}-\Delta)/(k_\zeta+k_\xi)u$ and $v/u = x$, the two integrals within the square brackets of eqn. (7) are seen to be

$$\mathcal{J}_p = -i\alpha\frac{2}{\pi^{\frac{1}{2}}u}\int_0^\infty\frac{\exp(-x^2)dx}{(-i\alpha)^2+x^2}+i\beta\frac{2}{\pi^{\frac{1}{2}}u}\int_0^\infty\frac{\exp(-x^2)dx}{(-i\beta)^2+x^2}.$$

Although these integrals cannot be evaluated in a closed form, they can be rearranged so that $\mathcal{J}_p$ reduces to

$$\mathcal{J}_p = [(\pi^{\frac{1}{2}}u^{-1})\theta(\alpha)-(\pi^{\frac{1}{2}}u^{-1})\theta(\beta)]$$

where $\theta(\alpha)$ and $\theta(\beta)$ are the probability integrals (gaussian) of the complex function, which is an extensively tqbulated function (Faddeyeva & Terent'ev 1961).

For $\nu_\xi = \nu_\zeta$ and $k_\xi = k_\zeta$, eq (7) can be rewritten as :

$$P_2^{(2\nu)}(z,t) = \frac{rAN(z)\pi^{\frac{1}{2}}\cos 2kz}{ku(\gamma_{ab}^2+\omega^2)}[\Theta_{re}\cos(2\nu t+\phi_\zeta+\phi_\xi)+\Theta_{im}\sin(2\nu t+\phi_\zeta+\phi_\xi)] \quad \ldots (8)$$

where the subscripts with k and ν are lifted and

$$\Theta_{re} = \omega[\theta_{im}(\alpha)-\theta_{im}(\beta)]+\gamma_{ab}[\theta_{re}(\alpha)-\theta_{re}(\beta)]$$

$$\Theta_{im} = \gamma_{ab}[\theta_{rm}(\alpha)-\theta_{im}(\beta)]-\omega[\theta_{re}(\alpha)-\theta_{re}(\beta)].$$

The subscripts *re* and *im* denote the real and the imaginary parts of the quantities respectively.

Some aspects of the behaviour of Θ_{re} and Θ_{im} are displayed in figures 1 and 2. As usual there are two resonances at optical frequencies, with the resonanc frequencies slightly shifted from the values predicted for the case where there is no atomic motion (Bloembergen & Shen 1964). As expected, these shifts are marginal and increase when relaxation increases. It may be noted that the results here and those in *I* show a difference of a negative sign which is due to the difference in the shape of the $W(v)$ function; the sign in *I* had to be arbitrarily chosen The expected broadening for increasing γ_{ab} for the same value of ω/ku is displayed in figure 2. It is seen to be rather small, the halfwidth being $0{\cdot}033\omega$ for $\gamma_{ab} = 4\times10^{-4}\omega$ and $0{\cdot}038\omega$ for $\gamma_{ab} = 4\times10^{-3}\omega$. However, the influence of a change in ku on Θ is seen to be much more striking. This situation is shown in figure 1. The resonance values are much more flattened for a change in $|ku/\omega|^{-1}$ ratio comparable to the γ_{ab}/ω ratio. The insensitivity of Θ_{im} and Θ_{re} values to a change in frequency for large ku values (Doppler limit, $k\,u \gg \gamma_{ab}$) is reported elsewhere for comparable situations (Icsevgi & Lamb 1969). In the literature quoted the spectral quantities are shown to be independent of frequency, a result which would have been obtained if the delta-function approximation used by Lamb (1964) were applied to eq. (6).

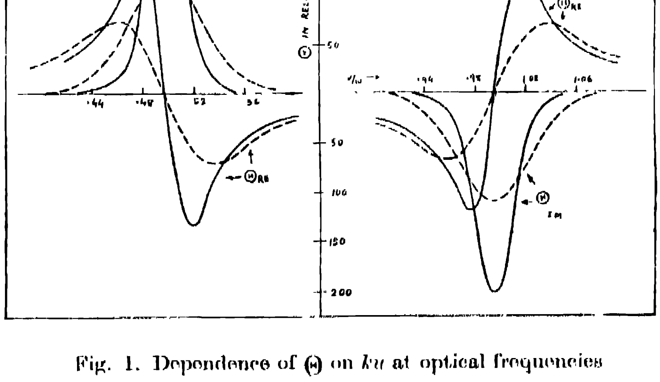

Fig. 1. Dependence of Θ on ku at optical frequencies

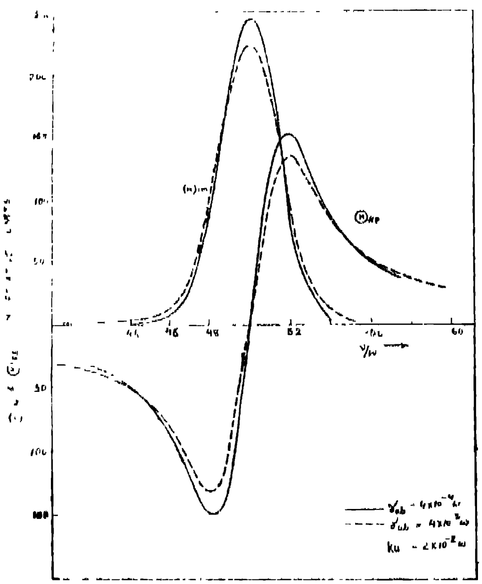

Fig. 2 Dependence of Θ on γ_{ab} at optical frequencies

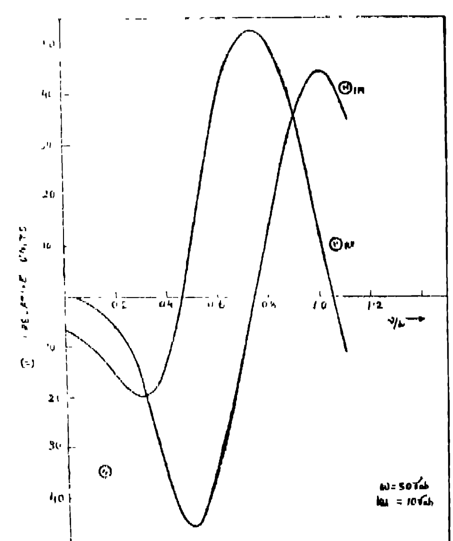

Fig. 3 Frequency characteristics of Θ at low frequencies $\omega = 50\gamma_{ab}$ and $ku = 10\gamma_{ab}$.

Figure 3 describes a situation which is probably true for microwave rather than optical frequencies The merger of the two peaks at $\omega = 2\nu$ and $\omega = \nu$, resulting in a single peak somewhat in the mid-frequency range is qualitatively similar to the one referred to in *I*.

4. Discussion

As in *I*, the expression for $P_2^{2\nu}$, given by eq. (8) is introduced into the one dimensional form of Maxwell's equations and the expressions for $E_{2\nu}$ and Q are calculated. However, unlike *I*, the wave number $k_{2\nu}$ associated with 2ν propagation can be straightaway put equal to $2k$ for gases. The reason for this is the same as given by Armstrong *et al* (1962) and experimentally this situation can be achieved by admixture of gases and change of pressure. As the results of the present work are free from Doppler limit restrictions, any resonable ratio of ku and γ_{ab} can be utilised. However, as this result is derived by an iteration procedure, it should not be trusted for $ku \to 0$. This can be regarded as a definite asset of this treatment.

Under the above assumption, and following *I*, the second harmonic field $E^{2\nu}$ is

$$E^{2\nu} = E_0^{2\nu}(z) \cos(2\nu t + \phi_{2\nu}) \sin k_{2\nu} z$$

The amplitude $E_0^{2\nu}$ is

$$E_0^{2\nu} = -\frac{8\nu^2}{c^2 \epsilon_0 k_2^{\nu}} \cdot x \cdot \frac{\sqrt{\pi}}{ku(\omega^2 + \gamma_{ab}^2)} \cdot \frac{r^2 r_d E_z E_y}{\hbar^2}$$

$$[\Theta_{re} \cos \Delta\phi + \Theta \sin \Delta\phi] \int N(z) dz \qquad \ldots \quad (9)$$

where $\Delta\phi = \phi_{2\nu} - \phi_z - \phi_y$, and the limit of integration extends over the length of the resonance cavity. The value of the quality factor Q is given by

$$Q \geqslant -\frac{2\nu^2 z \tan k_{2\nu} z}{c^2 k_{2\nu}} \cdot f(\Theta) \qquad \ldots \quad (10)$$

with

$$f(\Theta) = \frac{[\Theta_{re} \cos \Delta\phi + \Theta_{im} \sin \Delta\phi]}{[\Theta_{im} \cos \Delta\phi - \Theta_{re} \sin \Delta\phi]}.$$

The usual features of parametric frequency upconversion are apparent in eqs. (9) and (10), which have similar dependence on $\Delta\phi$ and z as described in *I*. It is interesting to note the ku dependence of Q when operating at threshold. At the two resonances ($2\nu = \omega$ or $\nu = \omega$), Q is independent of ku. It is to be expected from the form of eq. (10). But away from resonances, for an operating value above resonance ($2\nu > \omega$ or $\nu > \omega$) Q decreases as ku increases. Near the Doppler limit it approaches the resonance value of Q, being frequency independent beyond Doppler limit. This is a direct result of the frequency independence of Θ_{im} and Θ_{re} very near resonance and at the Doppler limit. The conclusion that the integrals similar to the ones denoting Θ_{im} and Θ_{re} are frequency independent has been discussed by Jesevgi & Lamb (1969). It is to be stressed that the method used in the present paper is not restricted to the Doppler limit and so it is more general. The actual dependence, near resonance and for a small γ_{ab}/ku ratio is of the form Q is proportional to $[1-(\pi^{\frac{1}{2}}ku)^{-1}x(-g)]$ where $x(-g)$ is a positive function of $g(=\Delta$ or $\Omega)$ and depends on $\Delta\phi$. It is true for negative Δ and Ω. For a change of sign in g the function $x(-g)$ changes sign. But the exception is $\Delta\phi = \pi/2$, where Q changes sign. This condition is discussed in I as the unstable condition. The value of Q for negative Δ is given in figure 4.

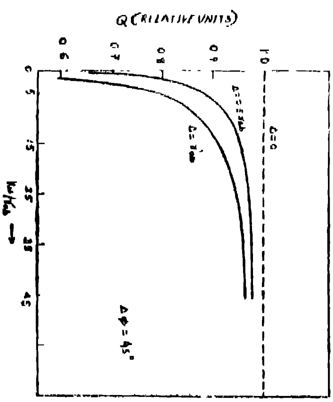

Fig. 4 Value of Q for changing Doppler parameter, ku/γ_{ab}, at and near resonance

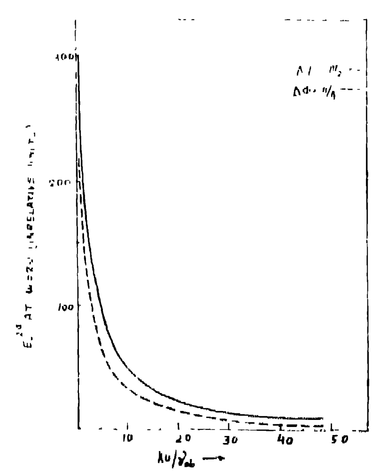

Fig. 5. Influence of ku/γ_{ab} on field amplitude $E_0^{2\nu}$. The value is at resonance.

From eq. (9) it is clear that $E_0^{2\nu}$ depends on $\int N(x)dx$, which is directly related to population difference between the levels. It is seen that there is negligible second harmonic conversion at $\Delta\phi = 0$ for near resonance operation where Θ_{rr} is extremely small. For $\Delta\phi = \pi/2$ and $\pi/4$, as shown in figure 5, $E_0^{2\nu}$ decreases for increasing ku/γ_{ab}. Though the diagram is shown for only one of the resonances, it is true for both of them. This decrease can be shown to be steeper when ω is near 2ν. For high ku/γ_{ab} values the decrease in $E_0^{2\nu}$ is of the form $(ku)^{-1}$. This is a direct result of the fact that near the Doppler limit Θ_{lm} behaves as exp. $-g^2/ku$ approximately. This approximate form of the integral, as well as the $(ku)^{-n}$exp. $-g^2/ku$ form for similar physical quantities (e.g., photon parameters) has been discussed by other other authors also (Kiska & Stenholm 1970). Figure 5 predicts an infinite value of $E_0^{2\nu}$ when ku tends to 0. This is not a serious defect, as the results of this paper, though valid for a much higher value of γ_{ab} then predicted by either Lamb ($\gamma_{ab} \leqslant 1/2ku$) (Lamb 1964) or Shimoda ($\gamma_{ab} \leqslant 0{\cdot}1ku$ Shimoda & Uehera 1965) should not be used in the limit of extremely large ratio (of γ_{ab}/ku for the reason given in the first paragraph of this section.

References

Armstrong J. A., Bloembergen N., Ducing J. & Pershan P. S. 1962 *Pyhs. Rev.* **127**, 1918.

Bloembergen N. & Shen Y. R. 1964 *Phys. Rev.* **133**, A37.

Faddeyeva V. N. & Terent'ev N. M. 1961 *Tables of Probability Integral for Complex Argument*, Pergamon Press, (New York).

Icsevgi A. & Lamb Jr W. E. 1969 *Phys. Rev.* **185**, 517.

Lamb Jr. W. E. 1964 *Phys. Rev.* **134**, A1429.

Mohanty B. K. 1974 *Physica* **77**, 192.

Riska D. O. & Stenholm S. 1970 *J. Phys.* **A3**, 189.

Shimoda K. & Uehdra K. 1965 *Japan J. Appl. Phys.* **4**, 921.

Shimoda K. & Uehara K. 1971 *Japan J. Appl. Phys.* **10**, 460.