Quantum Mechanical Effects in Stimulated Optical Emission

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An analysis of a three-level system using time-dependent perturbation theory shows that two distinct photon-emission processes take place within such a system when stimulated optical emission occurs: one- and two-photon transitions. The two-photon transition consists of the emission of a stimulated photon followed by the absorption of a coherent pump photon.

In three-level systems which possess a narrow pump level, two-photon transitions are dominant at high pump powers while the maser line is split into two lines due to the modulation of the wave function at an angular frequency determined by the rate of pumping. This might lead to the possibility of frequency-modulating the maser line. These results are extended qualitatively to include optical masers which possess broad pump-bands.

THIS paper is concerned with multiple-quantum transitions in optically pumped masers. A three-level system will be examined where the pump is a continuous coherent light source. The analysis does not apply to those optical masers[1–3] that are operated by pulsed incoherent broad band sources, but the effect of using these sources will be discussed briefly as an extension of the present analysis.

Consider a three-level system, as shown in Fig. 1, where a continuous coherent pump field of amplitude E_0' and frequency $\omega'\sim\omega_{13}$ is applied between levels 1 and 3, another field of amplitude E_0'' and frequency ω'' is applied between 3 and 2 while E_0 is the amplitude and ω the frequency of the stimulated emission that occurs between levels 2 and 1. The population of level 2 is inverted with respect to the population of level 1 because of the action of the pump.

Javan[4] has shown, for microwave frequencies, that when $E_0''=0$ state 2 is split into two lines at saturation pump powers due to the modulation[5] of the wave function between states 1 and 3 at an angular frequency determined by the rate of pumping. In addition to the splitting, two-photon transitions between states 2 and 3 become dominant transitions at highly saturated pump powers. This two-photon transition involves an emission between states 2 and 1, followed by an absorption between states 1 and 3.

This paper, which is primarily concerned with optical transitions, shows that by connecting states 2 and 3 by a fast transition, i.e., $E_0''\neq 0$, the splitting in such a system under certain conditions takes place at threshold, while the two-photon transitions become dominant at easily attainable source temperatures.[6] In addition, an effect can be seen due to the interference between the three transitions which involves the relative phases of the $\boldsymbol{\mu}_{jk}$, the matrix elements of the dipole moments between the states j and k.

If Ψ_k, $k=1, 2, 3$ are the stationary states belonging, respectively, to levels 1, 2 and 3, then the wave function for the atom, after it has gone through either a spin-spin or spin-lattice relaxation process, is[7]:

$$\psi=\sum_{k=1}^{3} a_k(t-t_0)\exp[-iE_k(t-t_0)/\hbar]\psi_k. \tag{1}$$

The state of the atom at any given time t is thus a mixture of all three states. The a_k are the probability amplitudes of the states k.

The transition probabilities $p_{jk}=|a_{jk}|^2$, where the first subscript now indicates the initial condition, i.e., a_{23} indicates the probability amplitude for state 3 with the initial condition that $a_2=1$, $a_1=a_3=0$.

It is convenient to introduce the parameters

$$|x_{12}|=e|\mathbf{E}_0|\cdot|\boldsymbol{\mu}_{12}|/2\hbar,\quad |y_{13}|=e|\mathbf{E}_0'|\cdot|\boldsymbol{\mu}_{13}|/2\hbar,$$
$$|z_{23}|=e|\mathbf{E}_0''|\cdot|\boldsymbol{\mu}_{23}|/2\hbar. \tag{2}$$

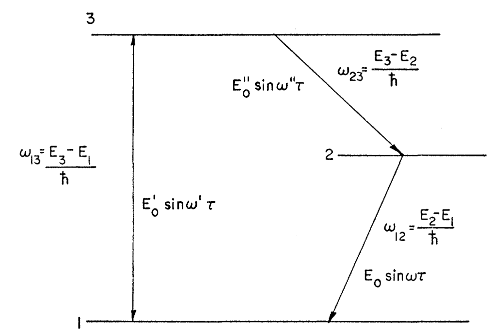

FIG. 1. Energy level diagram of a three-level system showing the amplitudes and frequencies of the fields that are applied between the various levels.

[1] T. H. Maiman, Brit. Commun. and Electronics **7**, 674 (1960).

[2] R. J. Collins, D. F. Nelson, A. L. Schawlow, W. Bond, C. G. B. Garrett, and W. Kaiser, Phys. Rev. Letters **5**, 303 (1960).

[3] P. P. Sorokin and M. J. Stevenson, Phys. Rev. Letters **5**, 557 (1960).

[4] A. Javan, Phys. Rev. **107**, 1579 (1957). Javan has examined this problem for E_0 and E_0' finite and nonzero while $E_0''=0$.

[5] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).

[6] The pump power is given in terms of an effective source temperature, the source being a blackbody that irradiates the crystal isotropically.

[7] H. A. Weakleim states that in the application of this analysis to ruby it should be realized each level is made up of many eigenstates of the system. We have assumed the states have random phase, which yields an average phase of zero for a given level. This maximizes the effect of the interference terms. There may be systems where the phase is not zero and estimable, which would give results different from these.

If one then solves the time-dependent Schrödinger equation[4] for the state amplitudes subject to the restriction $|y_{13}| \gg |x_{12}|$ and $|z_{23}| > |x_{12}|$, one obtains the transition probabilities $p_{jk} = |a_{jk}|^2$. The p_{jk} are then multiplied by $f(t-t_0) = \tau^{-1} dt_0 \exp[-(t-t_0)/\tau]$ and integrated over time t_0 in order to obtain the net power emitted under the conditions that a population inversion exists between levels 2 and 1. $f(t-t_0)$ is the fraction of atoms which have experienced a collision at time t_0 and exist for a time interval $(t-t_0)$ before suffering a second collision in a time dt_0. The parameter τ is the mean collision time and equals T_2 for a solid, the spin-spin relaxation time.

The power emitted by one-photon transitions between states 2 and 1 is denoted by P_{21} and the emitted power due to two-photon transitions between states 2 and 3 is denoted by P_{23}.

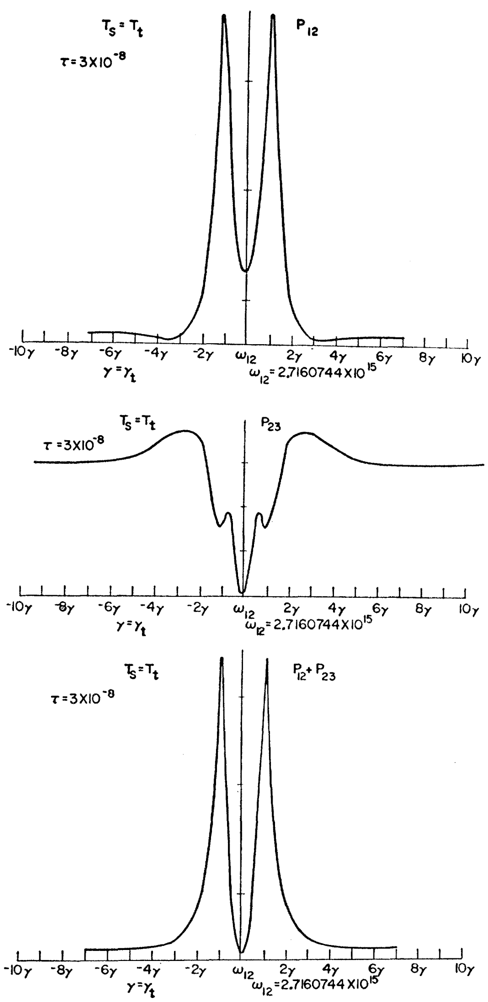

FIG. 2. Plot of power output vs frequency for (a) P_{12}, a one-photon process; (b) P_{23}, a two-photon process; and (c) $P_{12}+P_{23}$ for $T_s = T_t$ and $\tau = 3\times10^{-8}$ sec. The power is normalized by the factor $Nh\nu/\tau$. The abscissa is marked in units of $\gamma = \gamma_t$. Curve in 2(b) is magnified by a factor of 3.

$$P_{21} = (N_2-N_1)\bigg\{ \frac{\tau^2|x_{12}|^2}{2[1+(\omega-\omega_{21}-\gamma)^2\tau^2]} + \frac{\tau^2|x_{12}|^2}{2[1+(\omega-\omega_{21}+\gamma)^2\tau^2]} + \frac{6\gamma^4\tau^4|z_{23}|^2}{(1+\gamma^2\tau^2)(1+4\gamma^2\tau^2)|y_{13}|^2} + \gamma^2|x_{12}|^2 L_1 - \gamma^2 \frac{|x_{12}||z_{23}|}{|y_{13}|}[L_2+L_3-L_4-L_5] \bigg\}, \quad (3)$$

$$P_{23} = (N_2-N_3)\bigg\{ \frac{\tau^2|x_{12}|^2|y_{13}|^2}{\gamma^2[1+(\omega-\omega_{21}-\gamma)^2\tau^2]} + \frac{\tau^2|x_{12}|^2|y_{13}|^2}{\gamma^2[1+(\omega-\omega_{21}+\gamma)^2\tau^2]} + \frac{2\tau^2|z_{23}|^2}{1+4\gamma^2\tau^2} - |x_{12}|^2|y_{13}|^2 L_1 - |x_{12}||y_{13}||z_{23}|[L_2-L_3+L_4-L_5] \bigg\}, \quad (4)$$

where

$$L_2 = \frac{F_1 - F_2^- - F_4}{2\gamma^2(\delta-\gamma)}, \qquad L_3 = \frac{F_3^- - F_2^- + F_4}{2\gamma^2(\delta-\gamma)},$$

$$L_4 = \frac{-F_3^+ + F_2^+ + F_4}{2\gamma^2(\delta+\gamma)}, \quad L_5 = \frac{-F_1 + F_2^+ - F_4}{2\gamma^2(\delta+\gamma)}, \quad (5)$$

$$L_1 = \frac{1}{2\gamma^2(\delta^2-\gamma^2)}\left[1 + \frac{F_4}{\gamma\tau} - \frac{F_2^+}{(\delta+\gamma)\tau} - \frac{F_2^-}{(\delta-\gamma)\tau}\right],$$

where F functions are given by

$$F_1 = \frac{\delta\tau}{1+\delta^2\tau^2}, \quad F_2^{\pm} = \frac{(\delta\pm\gamma)\tau}{[1+(\delta\pm\gamma)^2\tau^2]},$$

$$F_3^{\pm} = \frac{(\delta\pm2\gamma)}{[1+(\delta\pm2\gamma)^2\tau^2]},$$

$$F_4 = \frac{\gamma\tau}{1+\gamma^2\tau^2}, \qquad \delta = \omega-\omega_{21}, \quad \gamma = (|y_{13}|^2 + |z_{23}|^2)^{\frac{1}{2}}.$$

These formulas are the result of the fields E_0' and E_0'' being on resonance.

In order to apply these results to a three level system a few comments are in order about linewidths. Levels 2 and 3 are supposed to be lines that are sharp enough to show a resonant character, i.e., the separation between levels 3 and 1, $E_3 - E_1$, is distinct enough so that a monochromatic coherent light source could be found so that $\omega' = \omega_{13} = (E_3-E_1)/\hbar$. The same situation applies to level 2, of course.

A well-known three-level system like ruby has this condition satisfied for level 2, the so-called $\bar{E}$ state, but level 3, the ${}^4F_2$ band, has a width of 1000 Å[8] and clearly does not meet the specifications. This does not mean that an optical maser cannot be found whose pump level is sufficiently narrow to meet these requirements be-

[8] T. H. Maiman, Phys. Rev. Letters 4, 564 (1960).

cause one such system does exist, namely Nd^{3+} in $CaWO_4$,[9] a four-level system where the pump levels are less than 50 Å at nitrogen temperatures and should be much sharper at helium temperatures.

Since the analysis applies to a three-level system, we cannot apply these results to Nd^{3+} in $CaWO_4$, but the analysis can be readily extended to include four-level systems.

It will be assumed that a three-level system exists that has a narrow pump level that satisfies the resonant condition; however, to fix ideas, we shall use all the other material constants characteristic of ruby, keeping in mind the fact that this system is artificial, since it has been designed to illustrate an effect. In this system the constant $|z_{23}|$, the number of transitions per second the ions undergo in moving from $3 \rightarrow 2$ due to the action of the electromagnetic field $E_0'' \sin\omega'' t$, will be set equal to 2×10^7/sec,[8] the decay rate S_{32} of the radiationless transition ${}^4F_2 \rightarrow \bar{E}$ in ruby.

The information on N_2-N_1 and N_2-N_3 is obtained from the steady-state rate equations[10] for the populations, which allows one to solve for the population difference in terms of the W_{jk}'s and the A_{jk}'s, the induced and spontaneous-transition probabilities. As mentioned previously, empirical values for the following constants were used: the W_{jk} and A_{jk},[8] the $\boldsymbol{\mu}_{jk}$,[11] T_2,[12] and temperatures of the source and stimulated emission. $\boldsymbol{\mu}_{13}=3\times 10^{-10}$ cm, $\boldsymbol{\mu}_{12}=8.86\times 10^{-12}$ cm, and $T_2=3\times 10^{-8}$ sec,[12] the spin-spin relaxation time of the $\bar{E}$ state in ruby at helium temperatures.

T_s indicates the source temperature while T_t is that source temperature which corresponds to threshold operation. Using these values, the line shape for 6940 Å stimulated emission from this system is shown in Figures 2, 3, and 4 for $T_s=T_t$, $T_s=2.73T_t$, and $T_s=3.62T_t$. Figure 2 shows that at $T_s\sim T_t$ i.e., just above threshold, the two-photon component P_{23} is reduced relative to the off-resonance radiation. This is due to the interference term (the last term in Eq. 4) which dominates at these low source temperatures. This effect is absent when $|z_{23}|=0$ since the interference term vanishes. The resonances that occur at $\omega=\omega_{21}\pm 2\gamma$ and $\omega=\omega_{21}$ arise due to the interference and are shown quite clearly. The one-photon component is emissive and is split into two lines by the pump field,[4,5] but the interesting fact is that it occurs just above threshold. The total output $P=P_{12}+P_{23}$ has two lines separated by $\sim 2\gamma$ with a linewidth of $\sim\frac{1}{2}\gamma$. Let this threshold γ be denoted by γ_t. The γ's that result at higher pump temperatures will be given in terms of γ_t.

[9] L. F. Johnson and K. Nassau, Proc. Inst. Radio Engrs. **49**, 1704 (1961).
[10] T. H. Maiman, Phys. Rev. **123**, 1145 (1961).
[11] D. S. McClure and H. A. Weakleim (private communication).
[12] A. L. Schawlow reported at the 1961 International Conference on Chemical Physics of Nonmetallic Crystals (unpublished) that he has measured linewidths of ruby crystals at helium temperatures that give a $T_2\sim 10^{-8}$.

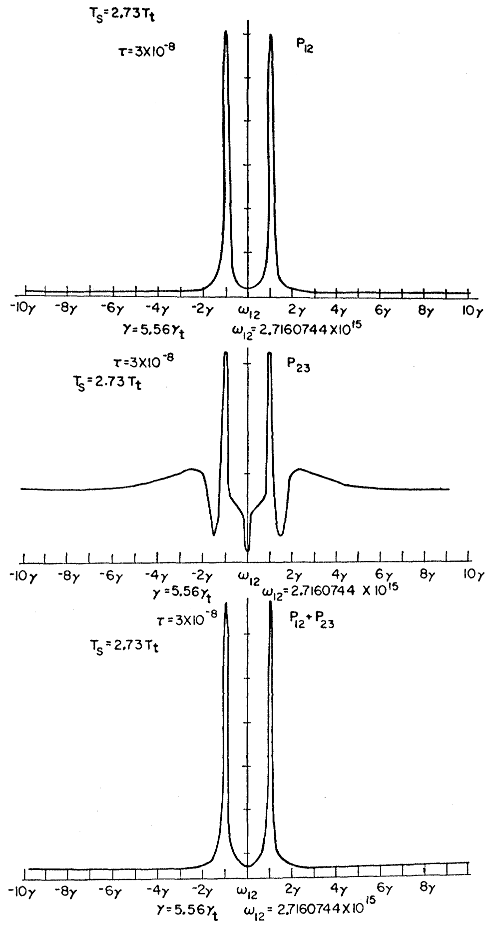

FIG. 3. Plot of normalized power output vs frequency for (a) P_{12}, a one-photon process; (b) P_{23}, a two-photon process; and (c) $P_{12}+P_{23}$ for $T_s=2.73T_t$ and $\tau=3\times 10^{-8}$ sec. The abscissa is marked in units of $\gamma=5.56\gamma_t$. Peak emission of P_{12} is 3 times larger than peak emission of P_{23}.

Figure 3 shows that at $T_s=2.73T_t$, P_{23} now becomes emissive at $\omega=\omega_{21}\pm\gamma$ and displays relative absorption at $\omega=\omega_{21}\pm 1.5\gamma$, and $\omega=\omega_{21}$. The relative absorption in these resonances is again due to the interference term. The emission due to P_{12} is now only $3\frac{1}{2}$ times greater than that due to P_{23}. The splitting of the doublet in P at $2.73T_t$ is approximately 5.6 times greater than the splitting at T_t, i.e. it is $\sim 11.2\gamma_t$. The linewidth is less than one-half of the linewidth at T_t, i.e., it is $\sim\frac{1}{4}\gamma_t$.

At lamp temperatures of $3.62T_t$, the two-photon component P_{23} is now two orders of magnitude greater than P_{12}, the one-photon component (Fig. 4). The splitting is roughly $7\frac{1}{2}$ times greater than that for $T_s=T_t$, i.e., it is $\sim 15\gamma_t$ while the linewidths are about half that at threshold, i.e., $\frac{1}{4}\gamma_t$.

One can conclude from these results that in this system for the above enumerated conditions, a considerable portion of the stimulated emission is due to the two-photon transition. The results also show that because of the high-pump powers required to obtain stimulated

emission, splittings and line widths result which are fairly large. If one is interested in spectral purity, a three-level system such as the one examined could never yield values similar to the gas maser[13] which has a linewidth of about one cps.[14] The state mixing effect which has been shown to exist in this particular three level system does not exist in the gas maser[13] because of the absence of a large dipole moment connecting the He 2^3S metastable with the ground state and the use of low-pump powers to excite the gaseous discharge.

The question of the kind of linewidths one should expect in ruby where the pump band is broad can now be answered. The results of the above analysis show that the splitting and linewidth factor,

$$\gamma=\tfrac{1}{2}\{(\omega'-\omega_{31})^2+4[|y_{13}|^2+|z_{23}|^2]\}^{\frac{1}{2}},$$

is a minimum when the pump level is narrow enough so that $\omega'=\omega_{31}$. The γ defined earlier is the result of ω' being on resonance, i.e., $\omega'=\omega_{31}$. However, when the level goes over into a band, this factor becomes large, because of $(\omega'-\omega_{31})^2$ which is now finite and large, hence larger linewidths will result. An integration over all frequencies in the pump band might wash out the effect of the splitting, but there is no reason why, at high pump powers, two photon transitions could not exist in crystals with broad pump bands.

If high spectral purity is desired from a solid-state optical maser, a four-level system would be a better choice since it requires lower pump powers (and hence a smaller γ), or a solid-state system where one could inject electrons or excite them by collision.

The advantages of the existence of this time-dependent state mixing effect in the three-level system that has been examined is that one can frequency modulate the emission by varying the pump power because the splitting factor γ varies directly as the amplitude of the pump field.

These results also apply to the microwave region, since the analysis is applicable to any part of the electromagnetic spectrum.

The author is indebted to A. Javan, H. Weaklein, D. McClure, H. R. Lewis, and A. Schawlow for extremely useful and enlightening discussions, and to S. W. Kahng for programming the above calculations.

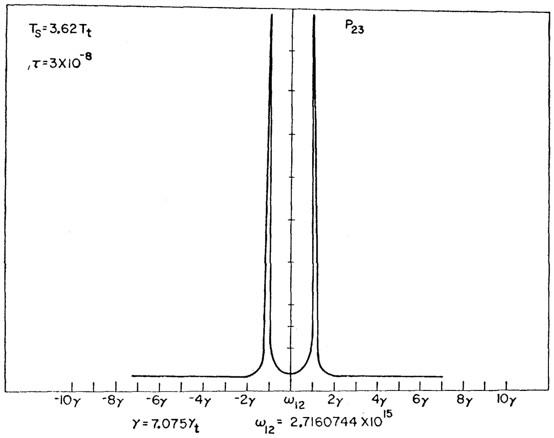

FIG. 4. Plot of normalized power output vs frequency for (a) P_{23}, a two-photon process for $T_s=3.62T_t$. P_{12} is two orders of magnitude less than P_{23}. The abscissa is marked in units of $\gamma=7.075\gamma_t$.

[13] A. Javan, W. R. Bennett, and D. R. Herriott, Phys. Rev. Letters **6**, 106 (1961).

[14] A. Javan (private communication).