physical meaning of its own, this indicates that $g_c^{(2)}$ may not be considered the correct distribution function and that improvements upon the chain-diagram approximation are necessary.

This is indeed achieved by taking the tick-tack-toe diagrams into consideration as we have done in the previous section. It is not hard to obtain the tick-tack-toe diagram contribution to the ground-state energy. We end up with

$$E_T = \tfrac{1}{2} \sum P_0^4/2p^2. \tag{8.20}$$

This is exactly what we needed to add to E_0 of Eq. (8.1) to remove the divergence. Thus, if we use the correct distribution function we get

$$\begin{aligned} E &= 4\pi aN - \tfrac{1}{2} \sum \{P_0^2 + p^2 - p(p^2+2P_0^2)^{1/2} - P_0^4/2p^2\} \\ &= 4\pi anN\{1+(128/15\pi^{1/2})(a^3n)^{1/2}\}, \end{aligned} \tag{8.21}$$

which is the result first obtained by Lee and Yang by the binary-kernel method.

It is remarked that the divergence in the ground-state energy is due to the appearance of a term proportional to $1/r$ at short distances. Thus, the correct pair distribution function should not contain $1/r$, in conformity with our result.

Summarizing, we may describe the situation as follows: the operator $(\partial/\partial r)r$ in the pseudopotential requires taking diagrams other than chain diagrams into consideration.

The above observation justifies our result at least for both small and large distances. The behavior of $g^{(2)}(r)$ for the intermediate range requires a numerical evaluation. However, it is interesting to observe that $g^{(2)}(r)$ is less than n^2 at large distances. Thus, in a certain intermediate range the $g^{(2)}(r)$ curve might possibly come out above the n^2 line.

Note added in proof. The authors thank Professor Garcia-Colin for informing them of the following important articles: L. Colin and J. Peretti, Compt. Rend. **248**, 1625 (1959); J. Math. Phys. **1**, 97 (1960); L. Colin, *ibid.* **1**, 87 (1960). The discussions of these articles will be given in a later article.

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Quantum-Mechanical Effects in Stimulated Optical Emission. II

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The existence of multiple-quantum transitions in optically pumped lasers, along with splitting of the laser line due to the modulation of the wave function at an angular frequency determined by the rate of pumping, is demonstrated for a certain class of incoherent broad-band sources pumping large pump bands in crystals. The source consists of a large number of stationary elements emitting wave fields continuously at various arbitrary frequencies and arbitrary phases. The distribution of frequencies and phases among the various elements is random. The pump band belongs to the class found in laser crystals of the ionic type. The analysis shows that such sources pumping such bands act like narrow-line sources pumping narrow lines. The effective linewidth is directly related to the pump rate.

THE existence of multiple-quantum transitions in optically pumped lasers and splitting of the laser line due to the modulation of the wave function at an angular frequency determined by the rate of pumping was shown theoretically by the author in "Quantum Mechanical Effects in Stimulated Optical Emission,"[1,2] hereinafter called "QMESOE I."

The theory set forth in that article showed that the splitting would become manifest at threshold when there are a large number of transitions occurring between the pump band and metastable level along with a high pump rate. Since the splitting is dependent upon the pump rate, it increases directly with the magnitude of the electric intensity of the pump field. In addition to this, it was also shown that at high pump powers most of the emitted power would be due to two-photon transitions.

It was not shown in that article if such splittings and multiple photon transitions would occur if both the source and the pump were broad bands instead of being monochromatic lines. Since the source in QMESOE I was chosen to be a coherent monochromatic source, it is not evident that incoherent broad-band sources pumping broad-pump bands will produce the same effect as coherent monochromatic sources pumping narrow-pump bands.

This question will be examined in this paper and it will be shown that indeed certain classes of incoherent broad-band sources pumping broad-pump bands do

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[1] R. C. Williams, Phys. Rev. **126**, 1011 (1962).
[2] R. C. Williams, Appl. Opt. Suppl. **1**, 63 (1962), reprinted from Phys. Rev. **126**, 1011 (1962).

produce splittings and multiple photon transitions.

Section I is concerned with defining the nature of the incoherent broad-band source. Section II constructs the Hamiltonian for the problem, and the analysis is performed using the time-dependent perturbation theory.

The effect of an incoherent broad-band-source pumping a broad pump band is considered in Sec. III.

SECTION I

The approach adopted here is that because of the inherent complex nature of the problem under investigation, the most convenient representation of an incoherent broad-band source will be selected. The first question that arises is whether the photon field of the source should be represented classically or quantum mechanically.

Glauber[3–5] has discussed the coherence and incoherence of radiation fields in a series of articles. He has shown in Ref. 5 that incoherent fields can be represented by superposing the outputs of many stationary sources. This enables him to deduce the distribution of quanta in a particular mode. The use of density operators enables him to provide a detailed discussion on the correlation and coherence properties of the photon field. These points are certainly important, but at present a simple classical description of the source photon field will be adopted because of the complexity of the problem.

Consider the following definition of an incoherent radiation field. Suppose that a source consists of a number of atoms that are rapidly moving, each atom randomly emitting a sequence of photons of various random energies during a time interval which has no correlation with the emitting time interval of any other atom. This would constitute the most general type of incoherent broad-band source, but one that is extremely difficult to describe analytically.

One can arrive at an incoherent broad-band source that can be easily described analytically by first placing all of the atoms, or elements of the source, at rest. One then has an assemblage of stationary sources distributed over an extended volume. The next restriction imposed is on the emission of the various wave fields. Each element of the source is supposed to emit a wave field with a particular frequency and arbitrary phase continuously. The phases of the waves are supposed to be completely uncorrelated with each other, and the distribution of the source elements emitting wave fields at the various frequencies is supposed to be random. If the distribution of frequencies is broad and continuous due to the large number of source elements, one has constructed an incoherent broad-band source, but a source which can be represented easily.

Let the electric-field intensity of a wave of frequency ω' emitted by the rth source element be

$$\boldsymbol{\mathcal{E}}'_{\omega' r} = \mathbf{E}'_{\omega' r} e^{j[\omega' t - \phi_r(\omega')]}\,, \tag{1}$$

where $\mathbf{E}'_{\omega' r}$ is the amplitude and $\phi_r(\omega')$ the phase. The resultant electric intensity of frequency ω' at any point P away from the source is

$$\boldsymbol{\mathcal{E}}'_{\omega'} = \sum_{r=1}^{N} \boldsymbol{\mathcal{E}}'_{\omega' r}, \tag{2}$$

where there are N source elements. Since there are many waves emitted of various frequencies ω', one must sum over all ω' to find the total resultant electric intensity.

$$\boldsymbol{\mathcal{E}}' = \int_0^\infty \boldsymbol{\mathcal{E}}'_{\omega'} d\omega' = \int_0^\infty \sum_{=1}^{N} \mathbf{E}'_{\omega' r} e^{j[\omega' t - \phi_r(\omega')]}, \tag{3}$$

which is of the form

$$\boldsymbol{\mathcal{E}}' = \int_0^\infty \mathbf{A}(\omega') e^{j\omega' t} d\omega', \tag{4}$$

with

$$\mathbf{A}(\omega') = \sum_{r=1}^{N} \mathbf{E}'_{\omega' r} e^{-j\phi_r(\omega')}. \tag{5}$$

Equation (4) is the vector form of the component equation given by Born and Wolf,[6] which is a Fourier-integral representation of a polychromatic wave. However, Eq. (5) generalizes the definition to an extended, incoherent, broad-band source.

This source will be completely incoherent if it is assumed that the light waves from these various source elements are completely independent and that their mean value is zero. If Γ_{rs} is the time average of the product of the intensities from two different source elements r and s, then the requirement for complete incoherence is

$$\Gamma_{rs} = \langle \boldsymbol{\mathcal{E}}_{\omega r}' \cdot \boldsymbol{\mathcal{E}}_{\omega s}' \rangle_{\text{av}} = 0\,. \tag{6}$$

This is the condition that is in force throughout this discussion.

SECTION II

Consider any three-level system with a ground state, labeled (1), a metastable level (2), and a broad pump band (3). Level (1) and band (3) are connected by an incoherent radiation field from an incoherent broad-band source, in particular of the type discussed in Sec. I. The pump band and the metastable level can be considered to be connected by another incoherent radiation field, or by nonradiationless transitions such as the phonon transitions that exist in ionic crystals doped

[3] Roy J. Glauber, Phys. Rev. Letters **10**, 3, 84 (1963).
[4] Roy J. Glauber, Phys. Rev. **130**, 2529 (1963).
[5] Roy J. Glauber, Phys. Rev. **131**, 2766 (1963).

[6] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Ltd., London, 1959), Chap. 10, Sec. 10.2.

by transition-metal elements or by rare earths. We shall represent these transitions phenomenologically, i.e., assign a matrix element to these nonradiationless transitions and measure it by measuring the splitting, since the splitting depends upon it directly. This was shown in QMESOE I. The metastable level is connected to the ground state by stimulated emission. The total Hamiltonian for the system H is then

$$H=H_0+H_{ep}+H'. \tag{7}$$

H_0 is the unperturbed Hamiltonian, H_{ep} is the electron-phonon interaction that gives rise to the nonradiationless transitions between the pump band and metastable level, H' is the perturbation due to the two wave fields, the incoherent field from the source and the coherent field from the laser transitions.

If ψ_k, $k=1, 2, 3$ are the stationary states belonging, respectively, to levels (1), (2) and band (3), then the wave function for the atom, after it has gone through some relaxation process, is

$$\psi=\sum_{k=1}^{3} a_k(t-t_0) \exp\left[-iE_k\frac{(t-t_0)}{\hbar}\right]\psi_k. \tag{8}$$

The time-dependent equations for the state amplitudes are

$$j\hbar\dot{a}_1=-\frac{e}{2j}\mathbf{E}_0\cdot\boldsymbol{\mu}_{12}a_2e^{j(\omega-\omega_{21})t} -\frac{e}{2j}\left(\sum_{r=1}^{N}\mathbf{E}'_{\omega' r}\cdot\boldsymbol{\mu}_{13}e^{-j\phi_r(\omega')}\right)a_3e^{j(\omega'-\omega_{31})t}, \quad 9(\text{a})$$

$$j\hbar\dot{a}_2=\frac{e}{2j}\mathbf{E}_0{}^*\cdot\boldsymbol{\mu}_{12}{}^*a_1e^{-j(\omega-\omega_{21})t} +\langle 2|H_{ep}|3\rangle a_3e^{j(\omega''-\omega_{32})t}, \quad 9(\text{b})$$

$$j\hbar\dot{a}_3=\frac{e}{2j}\left(\sum_{r=1}^{N}\mathbf{E}'_{\omega' r}{}^*\cdot\boldsymbol{\mu}_{13}e^{j\phi_r(\omega')}\right)a_1e^{-j(\omega'-\omega_{31})t} +\langle 2|H_{ep}|3\rangle^*a_2e^{-j(\omega''-\omega_{32})t}. \quad 9(\text{c})$$

These equations have been written out to show that the laser wave field is coherent and monochromatic, while the incoherence of the source is clearly displayed. It can be seen now that if the incoherent source was allowed to have its various source elements emit during various uncorrelated time intervals, the problem would be almost intractable. The fact that each stationary element of the source is continuously emitting a wave of a particular frequency and phase renders the problem tractable. One can now solve these equations in exactly the same manner that it was in QMESOE I.

Let

$$x_{12}=e\mathbf{E}_0\cdot\boldsymbol{\mu}_{12}/2\hbar, \tag{10}$$

$$y_{13}=\frac{e}{2\hbar}\sum_{r=1}^{N}\mathbf{E}'_{\omega' r}\cdot\boldsymbol{\mu}_{13}e^{-j\phi_r(\omega')}, \tag{11}$$

$$z_{23}=\langle 2|H_{ep}|3\rangle. \tag{12}$$

These equations then go over to those obtained in QMESO I, except that since the solutions of (9) yield the transition probabilities $|a_{jk}|^2$, these transition probabilities contain the squares of the transition rates $|x_{12}|^2$, $|y_{13}|^2$, and $|z_{23}|^2$. The item that makes the broad-band source analysis different from a monochromatic source is the evaluation of

$$|y_{13}|^2{}_{\omega'}=\frac{e^2}{4\hbar^2}\left|\sum_{r=1}^{N}\mathbf{E}'_{\omega' r}\cdot\boldsymbol{\mu}_{13}e^{-j\phi_r(\omega')}\right|^2. \tag{13}$$

Since the $\mathbf{E}'_{\omega' r}$ are complex terms all unequal, this problem can be solved using the theory of random walk. The essential results were first obtained by Lord Rayleigh,[7] while Chandrasekhar[8] and Kennard[9] both provide extremely thorough treatments.

Their results show that, if N is large enough to provide a good statistical sample, then

$$\left|\sum_{r=1}^{N}\mathbf{E}'_{\omega' r}\cdot\boldsymbol{\mu}_{13}e^{-j\phi_r(\omega')}\right|^2=N\langle(\mathbf{E}'_{\omega'}\cdot\boldsymbol{\mu}_{13})^2\rangle_{\text{av}}.$$

Therefore

$$|y_{13}|^2{}_{\omega'}=(e^2/4\hbar^2)N\langle(\mathbf{E}'_{\omega'}\cdot\boldsymbol{\mu}_{13})^2\rangle_{\text{av}}. \tag{14}$$

The pump rate at a particular frequency ω' is given by

$$|y_{13}|_{\omega'}=\frac{e}{2\hbar}(N)^{1/2}[\langle(\mathbf{E}'_{\omega'}\cdot\boldsymbol{\mu}_{13})^2\rangle_{\text{av}}]^{1/2}, \tag{15}$$

and a high pump rate at a given frequency ω' can be obtained by providing a source with a *large number N of individual elements that are emitting wave fields at that frequency.*

This is accomplished in practice by using extended sources and providing a suitable optical system to focus their radiation.

It was shown in QMESOE I that at high pump rates the two-photon process was several orders of magnitude greater than the one-photon process. Therefore, the one-photon process will not be considered. The one-photon process was the emission of a photon by an electron undergoing a transition between level (2) and the ground state, level (1). The two-photon process in that article, also called multiple photon transitions, consisted of photon emission by an electron undergoing a transition between level (2) and (1), followed by photon absorption from the coherent radiation field of the source, forcing the electron to make a transition between levels (1) and (3).

The two-photon process in this article consists of photon emission by an electron undergoing a transition between levels (2) and (1), followed by photon absorption from the incoherent radiation field of the incoherent broad-band source, forcing the electron to make a transition between the ground state [level (1)] and the pump band.

[7] Lord Rayleigh, *Scientific Papers* (Cambridge University Press, London, 1899, 1903), Vol. I, p. 491 and Vol. IV, p. 370.

[8] S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943). Reprinted in *Selected Papers on Noise and Stochastic Processes* (Dover Publications, Inc., New York, 1954).

[9] E. H. Kennard, *Kinetic Theory of Gases* (McGraw-Hill Book Company, Inc., New York, 1938).

The solution of the system of Eqs. (9) subject to the restriction $|y_{13}|\gg|x_{12}|$ and $|z_{23}|\gg|x_{12}|$ enables one to obtain the state amplitudes a_{jk}. a_{23} indicates the probability amplitude for state (3) with the initial condition $a_2=1$, $a_1=a_3=0$. The transition probabilities are

$$p_{jk}=|a_{jk}|^2.$$

The net power emitted under the conditions that a population inversion exists between levels (2) and (1) is obtained by multiplying the p_{jk} by

$$f(t-t_0)=(1/\tau)dt_0\exp[-(t-t_0)/\tau]$$

and integrating over time t_0. The function $f(t-t_0)$ 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 is defined as follows:

$$\tau=T_2=1/\pi\Delta\nu_2,$$

where $\Delta\nu_2$ is the natural linewidth of metastable level (2) for spontaneous emission, or, the linewidth of one of the cavity Fabry-Perot modes, whichever is the smaller.

Most optical masers, both solid (ionic crystals) and gaseous, contain many Fabry-Perot modes within the natural linewidth of the metastable level,[10–13] which permits the observation of beats between the various modes. Natural linewidth here refers to the resultant linewidth due to all the various physical effects, such as Doppler broadening in gases and phonon broadening in solids.

The integration over t_0 yields the following result for the power emitted due to two-photon transitions between states (2) and (3) as a function of ω', ω'', and ω:

$$\begin{aligned}
P_{23}(\omega',\omega'',\omega)={}&(N_2-N_3)\frac{h\nu}{\tau^2}\bigg\{\frac{\frac{1}{2}\tau^3|x_{12}|^2|y_{13}|^2}{\gamma^2_{\omega'}[1+(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'})^2\tau^2]}+\frac{\frac{1}{2}\tau^3|x_{12}|^2|y_{13}|^2}{\gamma^2_{\omega'}[1+(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'})^2\tau^2]}\\
&+\frac{\frac{1}{2}\tau^3|z_{23}|^2[1+(\omega'-\omega_{21})/2\gamma_{\omega'}]^2}{[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})^2\tau^2]}+\frac{\frac{1}{2}\tau^3|z_{23}|^2[1-(\omega'-\omega_{31})/2\gamma_{\omega'}]^2}{[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'})^2\tau^2}\\
&-\frac{|x_{12}|^2|y_{13}|^2\tau}{\gamma^2_{\omega'}[(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21}))^2-\gamma^2_{\omega'}]}\bigg[\frac{1+2\gamma_{\omega'}{}^2\tau^2}{1+4\gamma^2_{\omega'}\tau^2}-\frac{1}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'})^2\tau^2]}\\
&-\frac{1}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})^2\tau^2]}\bigg]+\frac{\tau|z_{23}|^2[1-(\omega'-\omega_{31})^2/4\gamma^2_{\omega'}]}{[(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31}))^2-\gamma^2_{\omega'}]}\\
&\times\bigg[\frac{1+2\gamma^2_{\omega'}\tau^2}{1+4\gamma^2_{\omega'}\tau^2}-\frac{1}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'})^2\tau^2]}-\frac{1}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})^2\tau^2]}\bigg]\\
&+\frac{\tau|x_{12}||y_{13}||z_{23}|[1+(\omega'-\omega_{31})/2\gamma_{\omega'}]}{\gamma_{\omega'}[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'}][\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'}]}\bigg[\frac{(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21})\tau}{2[1+(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21})^2\tau^2]}\\
&+\frac{[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega}']\tau}{2[1+(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'})^2\tau^2]}-\frac{(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})\tau}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})^2\tau^2]}\bigg]\\
&+\frac{\tau|x_{12}||y_{13}||z_{23}|[1-(\omega'-\omega_{31})/2\gamma_{\omega'}]}{\gamma_{\omega'}[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'}][\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'}]}\bigg[\frac{(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21}-2\gamma_{\omega'})\tau}{2[1+(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21}-2\gamma_{\omega'})^2\tau^2]}\\
&+\frac{[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'}]\tau}{2[1+(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})+\gamma_{\omega'})^2\tau^2]}-\frac{[\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'}]\tau}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'})^2\tau^2]}\bigg]\\
&-\frac{\tau|x_{12}||y_{13}||z_{23}|[1+(\omega'-\omega_{31})/2\gamma_{\omega'}]}{\gamma_{\omega'}[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'}][\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'}]}\bigg[\frac{(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21}+2\gamma_{\omega'})\tau}{2[1+(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21}+2\gamma_{\omega'})^2\tau^2]}\\
&+\frac{[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'}]\tau}{2[1+(\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'})^2\tau^2]}-\frac{(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})\tau}{2[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'})^2\tau^2]}\bigg]\\
&+\frac{\tau|x_{12}||y_{13}||z_{23}|[1-(\omega'-\omega_{31})/2\gamma_{\omega'}]}{-2\gamma_{\omega'}[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'}][\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'}]}\bigg[\frac{[\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21}]\tau}{[1+(\omega''-\omega_{32}-(\omega'-\omega_{31})+\omega-\omega_{21})^2\tau^2]}\\
&+\frac{[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'}]\tau}{[1+[\frac{1}{2}(\omega'-\omega_{31})-(\omega-\omega_{21})-\gamma_{\omega'}]^2\tau^2]}-\frac{[\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega}']\tau}{[1+(\omega''-\omega_{32}-\frac{1}{2}(\omega'-\omega_{31})-\gamma_{\omega'})^2\tau^2]}\bigg]\bigg\}
\end{aligned}\tag{16}$$

[10] D. R. Herriot, J. Opt. Soc. Am. **52**, 31 (1962).
[11] A. Javan, W. R. Bennett, Jr., and D. R. Herriott, Phys. Rev. Letters **6**, 106 (1961).
[12] B. J. McMurty and A. E. Siegman, Appl. Opt. **1**, 51 (1962).
[13] W. Heinlein and D. Roess, Proc. IEEE **51**, 1667 (1963).

$$\gamma_{\omega'}=\tfrac{1}{2}\{(\omega'-\omega_{31})^2+4[|y_{13}|^2+|z_{23}|^2]\}^{1/2} \tag{17}$$

and when $\omega'=\omega_{31}$,

$$\gamma_{\omega'}=(|y_{13}|^2+|z_{23}|^2)^{1/2}=\gamma, \tag{18}$$

the splitting factor of QMESOE I. As an example, if $|y_{13}|\lesssim|z_{23}|$ in ruby,[1] $\gamma_r\sim|z_{23}|=2\times10^7$/sec.

SECTION III

Throughout this section it is assumed that transitions between the pump band and the metastable level occur via single-phonon emission of frequency ω_{23}, hence $\omega''=\omega_{23}$; and P_{23} now becomes a function of ω' and ω.

Let the width of the pump band between the extremes of the band be $\Delta\omega_B$, the extremes being the point where the absorption band drops to 10% of its maximum. Define a relaxation time

$$T_3=2\pi/\Delta\omega_B\,,$$

then the structure factor for the pump band can be represented as $S_p([\omega'-\omega_{13}]T_3)$, where S_p is the function that analytically describes the shape of the pump band.

If the pump band is replaced by a large number of equally spaced monochromatic lines with a separation ζ, then there will be $\Delta\omega_B/\zeta$ lines in place of the band with the same profile as the band. A typical pump band in a laser crystal has $\Delta\omega_B\sim10^{14}$ cps. If $\zeta\sim10^{-6}$, say, then the number of lines replacing the band is $\Delta\omega_B/\zeta\sim10^{20}$. Later on in the analysis, the limit will be taken when $\zeta\rightarrow\epsilon$, ϵ being an infinitesimal different from zero. In that limit all the lines merge into a band. The structure factor for the band can thus be written as

$$S_p(\omega')=\sum_{n=-M/2}^{M/2}S_p[(\omega'-\omega_{13})T_3]\delta(\omega'-\omega_{13}-n\zeta)\,, \tag{19}$$

where $M=\Delta\omega_B/\zeta$ and $n=0,\pm1,\pm2,\cdots\pm M/2$. Consider next the power emitted by a two-photon transition $P_{23}(\omega',\omega)$ due to an exciting pump frequency ω'. Let the shape factor for the broad-band source be $S_s(\omega')$.

The shape factors are defined here to mean the probabilities of emitting or absorbing a photon for source or pump band, respectively. Another way of viewing them is to regard them as the statistical weights attached to photon emission or absorption.

These weights are real functions of ω'. The shape factor for the pump band is given by its absorption spectrum. Similarly, the shape factor for the source is given by its emission spectrum. Since the spectra are obtained by measuring averaged transition rates, the case considered here is identical with that considered in Sec. II, i.e., these functions are dependent only on ω', not on the phases of the source. The average transition rates for the source are evaluated in the same manner as Eq. (13), i.e., by random walk, hence the phases average out. The case where they do not average out is extremely difficult, and would apply to extremely low-intensity sources of infinitesimal spatial extension. This case is concerned with high-intensity sources of large spatial extension.

An integration of $P_{23}(\omega',\omega)$ over both bands yields

$$P_{23}(\omega)=\int_0^\infty P_{23}(\omega',\omega)S_p(\omega')S_s(\omega')d\omega'\,, \tag{20}$$

choosing the broadest source possible, i.e.,

$$S_s(\omega')=1\,,\quad 0<\omega'<\infty\,; \tag{21}$$

and substituting (19) and (21) into (20) yields

$$P_{23}(\omega)=\sum_{n=-M/2}^{+M/2}P_{23}(\omega_{13}+n\zeta,\omega)S_p(n\zeta T_3)\,. \tag{22}$$

The integral converts to a sum because of the delta function $\delta(\omega'-\omega_{13}-n\zeta)$.

Instead of examining the sum (22) for all of the terms in (16), it shall be done for the two leading terms. The analysis can be readily extended to the remaining terms with the same results (see Appendix).

We will express these terms in $P_{23}(\omega',\omega)$ as a function of ω' and ω, all other matrix elements being lumped into one constant. Thus, the leading terms can be written as

$$P_{23}{}^{\pm}(\omega',\omega)=\frac{A_{23}{}^{\pm}}{[(\omega'-\omega_{13})^2+4\gamma^2][1+(\frac{1}{2}[\omega'-\omega_{13}]-\{\omega-\omega_{12}\pm\frac{1}{2}[(\omega'-\omega_{13})^2+4\gamma^2]\}^{1/2})^2\tau^2]}\,. \tag{23}$$

Substituting (23) into (22) yields

$$P_{23}{}^{\pm}(\omega)=\sum_{n=-M/2}^{+M/2}\frac{A_{23}{}^{\pm}S_p(n\zeta T_3)}{[n^2\zeta^2+4\gamma^2]\{1+[\omega-\omega_{12}\pm\frac{1}{2}(n^2\zeta^2+4\gamma^2)^{1/2}-\frac{1}{2}n\zeta]^2\tau^2\}}\,. \tag{24}$$

In the limit when $\zeta\rightarrow\epsilon$, some infinitesimal different from zero, Eq. (24) holds for a band, i.e., a large number of densely packed lines.

If $\gamma\tau>1$, then when $n=0$ the result is the split two-photon power emission spectrum obtained in QMESOE I. For small ζ, the $n=0$ term provides an adequate description of all terms up to $n\zeta \sim \gamma/10$. Thus, those pump frequencies which fall in the range

$$\omega_{13}-(\gamma/10)<\omega'<\omega_{13}+(\gamma/10)$$

act like a monochromatic source of frequency ω_{13}.

When $n\zeta \gg \gamma$, i.e., $n\zeta \sim 10\gamma$, the term $[n^2\zeta^2+4\gamma^2]$ in the denominator attenuates these large n terms by factors of 10^2 and greater. Thus, one needs only to consider the range

$$\omega_{13}-10\gamma<\omega'<\omega_{13}+10\gamma\,,$$

since this is the only region of the pump band that contributes. The effective band width of the pump band is even narrower since we can select only that range which contributes to the main part of the observed power. The power will be down by a factor of 2 when $n\zeta=2\gamma$. Hence, the effective band width of the pump band $\Delta\omega_{BE}=4\gamma$. Since γ contains (14), the larger the spatial extension of the source, i.e., the higher the N, the larger the γ.

Since the pump bandwidth is of the order of 10^{14} cycles, and the splitting factor γ is of the order 10^7, the shape factor $S_p(n\zeta\tau)$ is a constant for $\omega_{13}-10\gamma<\omega'<\omega_{13}+10\gamma$ given by the value it has at its maximum, $S_p(\omega_{13})$. It should be noted that if

$$\int_{-\infty}^{+\infty} S_p(\omega')d\omega'=1\,,\quad \text{then} \quad \sum_{n=-M/2}^{+M/2} S_p[n\zeta\tau]=1\,.$$

Thus, one can state the following very general conclusions about pumping conditions in laser crystals:

Incoherent broad-band sources of the type discussed in Sec. I, pumping broad pump bands act like narrow-line sources pumping narrow lines. The effective linewidth of both source and band is $\Delta\omega_{BE}=4\gamma$. This result is a consequence of the quantum-mechanical splitting factor $\gamma_{\omega'}$, which appears in the denominators of $P_{23}(\omega',\omega)$.

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APPENDIX

Let the various terms of $P_{23}(\omega',\omega'',\omega)$ in (16) be denoted by T_i, $i=1, 2, 3, \cdots$. Consider the third and fourth terms. Setting $\omega''=\omega_{32}$, it is seen that $T_{3,4}$ is independent of ω and does not affect the line shape, only the power level. Term 6 is also independent of ω and does not affect the line shape.

Term 5 has three terms inside the square brackets that are independent of ω, while the factors multiplying the bracket have the form given by (23), hence, term 5 exhibits the same type of behavior as the leading terms.

The remaining four terms have the following behavior. At a particular set of resonant frequencies, $\omega'=\omega_{13}$, $\omega''=\omega_{32}$, $\omega=\omega_{12}+\gamma$, the sum of these four terms is of the order of magnitude

$$\sum_{i=7}^{10} T_i=O\left[\frac{|x_{12}||y_{13}||z_{23}|\,\tau^4}{(1+m\gamma^2\tau^2)(1+\gamma^2\tau^2)}\right],$$

where m is a positive integer greater than unity. To facilitate the evaluation, let $|y_{13}|=|z_{23}|$, then $\gamma^2=2|z_{23}|^2$. If we assume a τ so that $\gamma\tau\sim 10$, say, then

$$\sum_{i=7}^{10} T_i=O\left[\frac{|x_{12}||y_{13}||z_{23}|\,\tau^4}{m\gamma^4\tau^4}\right]=O\left[\frac{|x_{12}|}{4m|z_{23}|^2}\right].$$

The leading terms, at the same set of resonances, are of the order of

$$L=O\left[\tfrac{1}{2}\tau^3\frac{|x_{12}|^2|y_{13}|^2}{\gamma^2}\right]=O[\tfrac{1}{4}\tau^3|x_{12}|^2].$$

Hence,

$$L/\sum_{i=7}^{10} T_i=O[m(\tau^2|z_{23}|^2)(\tau|x_{12}|)]\,,$$

if τ is chosen so that

$$\tau|x_{12}|\gtrsim|\,.$$

Then

$$L/\sum_{i=7}^{10} T_i=O[m10^2]\,,$$

or the last four terms are negligible in comparison to the leading terms and do not affect the line shape or power level. Even so, if one considers the last term T_{10}, and in particular looks at the factors multiplying the square brackets, these have factors in the denominator that lead to rapid convergence when expressed in the form (22), in particular the factors $\gamma_{\omega'}$ and $[\frac{1}{2}(\omega'-\omega_{31})+\gamma_{\omega'}]$; recall that $\omega''=\omega_{32}$. This is also true of $T_7 \rightarrow T_9$.