Stimulated Optical Emission in Fluorescent Solids. I. Theoretical Considerations

T. H. Maiman*

Hughes Research Laboratories, A Division of Hughes Aircraft Company, Malibu, California

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An analysis of stimulated emission processes in fluorescent solids is presented. The kinetic equations are discussed and expressions for pumping power and effective temperature of the exciting source are given in terms of the material parameters. A comparison of excitation intensity for three- and four-level systems is given. The spectral width of the stimulated radiation is discussed with particular attention to imperfect crystals.

INTRODUCTION

EXPERIMENTS culminating in the achievement of stimulated optical radiation from Cr^{3+} in Al_2O_3 (ruby) were recently reported by the author[1,2]; in addition, stimulated emission from other solids has since been reported.[3] A discussion of stimulated emission processes in the infrared-optical spectral region was presented previously by Schawlow and Townes,[4] with particular emphasis on an alkali vapor system; however, the modifications of both the analytical and experimental problems encountered with solids are considerable. Some of these extensions and revisions are presented here.

The general class of materials to be considered are fluorescent solids whose emission spectra consist of one or more sharp spectral lines. Excitation is normally supplied to these solids by radiation of frequencies which produce absorption into one or more bands. Some of this excitation energy is lost by a combination of spontaneous emission and thermal relaxation to lower-lying states; however, if the solid has a relatively high fluorescent efficiency[1] most of the energy is transferred to the sharp fluorescent levels by means of a non-radiative process. Subsequently, by a combination of spontaneous emission and thermal relaxation, the excited atoms (ions) return either to the ground state, or another low-lying state. The spontaneous emission from these sharp levels is the observed fluorescent radiation. If the exciting radiation is sufficiently intense it is possible to obtain a population density in one of the fluorescent levels greater than that of the lower-lying terminal state. In this situation, spontaneously emitted (fluorescent) photons traveling through the crystal stimulate upper state atoms to radiate, and a net component of induced emission is superimposed on the spontaneous emission.

We distinguish at this point between two useful

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* Now at Quantatron, Inc., 2520 Colorado Avenue, Santa Monica, California.

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

[2] T. H. Maiman, Nature **187**, 493 (1960); Brit. Comm. and Electronics **7**, 674 (1960); Program of the 45th Annual Meeting of the Optical Society of America, October 12–14, 1960 (unpublished), p. 14.

[3] P. P. Sorokin and M. J. Stevenson, Phys. Rev. Letters **5**, 557 (1960); IBM J. Research Develop. **5** (1961).

[4] A. L. Schawlow and C. H. Townes, Phys. Rev. **112**, 1940 (1958).

solid-state systems: a three-level scheme typified by the level diagram shown in Fig. 1, and a four-level arrangement as shown in Fig. 2. Other configurations are, of course, possible, such as one in which all of the levels are relatively sharp and therefore more analogous to the gaseous situation. However, one of the features of the solid system is the possible use of a broad absorption band for the pump transition. This situation allows a relatively high pumping efficiency to be realized since most high-power optical sources have very broad spectral distributions in their radiant energy.

PUMPING POWER CONSIDERATIONS

Three-Level System

In the configuration shown in Fig. 1, the ground state is the terminal level for the spontaneous emission transition $2 \rightarrow 1$. Therefore, to produce a net stimulated emission component it is necessary to have an incident pump radiation intensity large enough to excite at least one-half of the total number of ground-state atoms into level 2. The extreme difficulty of achieving such a radiation intensity led early investigators to ignore the three-level system here described.[5]

A more quantitative description of this scheme can be obtained from a solution of the steady-state rate equations:

$$dN_3/dt = W_{13}N_1 - (W_{31}+A_{31}+S_{32})N_3 = 0, \quad (1)$$

$$dN_2/dt = W_{12}N_1 - (A_{21}+W_{21})N_2 + S_{32}N_3 = 0, \quad (2)$$

$$N_1 + N_2 + N_3 = N_0. \quad (3)$$

These processes are indicated in Fig. 1, where W_{13} is the induced transition probability per unit time for the transition $(1 \rightarrow 3)$ caused by the exciting radiation of frequency ν_{13}, W_{21} is the induced probability $(2 \rightarrow 1)$ due to the presence of radiation of frequency ν_{21}, A_{31} and A_{21} are Einstein A coefficients to account for the spontaneous radiation, and S_{32} is the transition probability for the nonradiative process $(3 \rightarrow 2)$. N_1, N_2, and N_3 are the respective level population densities, and N_0 is the total active ion density in the crystal. It has been assumed that the thermal processes for both $(2 \rightarrow 1)$ and $(3 \rightarrow 1)$ are negligible compared to the correspond-

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[5] A. L. Schawlow, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), pp. 553–563.
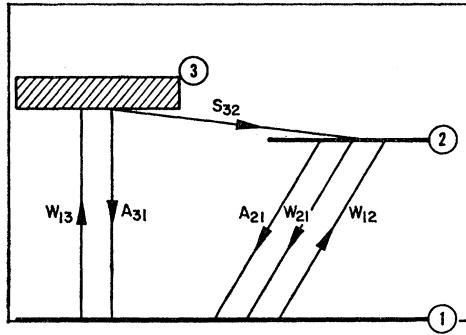
FIG. 1. Optical energy-level diagram
for a three-level fluorescent solid.

ing radiative processes but that the reverse is true for the transition $(3 \rightarrow 2)$.[1] It is also assumed that each of the energy-level separations is large compared to kT so that the Boltzmann factors are negligible.

A solution to the above equations is

$$\frac{N_2}{N_1} = \left[W_{13}\left(\frac{S_{32}}{W_{31}+A_{31}+S_{32}}\right) + W_{12} \right] \Big/ (A_{21}+W_{21}). \quad (4)$$

If the solid has a high fluorescent quantum efficiency, e.g., ruby, $A_{31} \ll S_{32}$. Also, it will be found that even for very high pumping powers $W_{31} \ll S_{32}$. These approximations imply $N_3 \ll N_1$, $N_3 \ll N_2$ and (4) simplifies to

$$N_2/N_1 \cong (W_{13}+W_{12})/(A_{21}+W_{21}), \quad (5)$$

alternatively,

$$(N_2-N_1)/N_0 \cong (W_{13}-A_{21})/(W_{13}+A_{21}+2W_{12}). \quad (6)$$

In order to obtain stimulated emission at the frequency ν_{12}, it is necessary that $N_2 > N_1$ and therefore, from (6) we must have $W_{13} > A_{21}$. This is a minimum condition. In order to achieve a useful stimulated-emission amplifier or oscillator, it is further required that the excess population (N_2-N_1) be sufficient to overcome circuit losses.[4] The degree of population inversion needed can be conveniently represented for the optical case as follows.
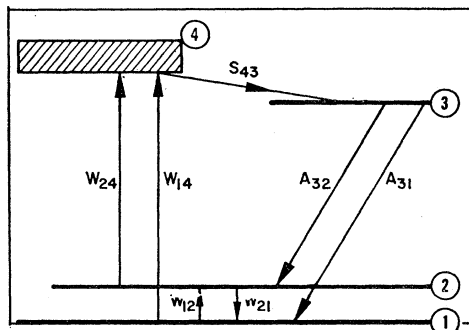

FIG. 2. Energy-level diagram in a fluorescent solid to illustrate a four-level method of producing stimulated optical radiation.

The energy in a wave propagating through a material with an absorption coefficient α is attenuated by the factor $e^{-\alpha l}$, if l is the length of material. The absorption coefficient is related to the population difference by

$$\alpha_{12} = (N_1-N_2)\sigma_{12}, \quad (7)$$

where σ_{12} is the absorption cross section for the pertinent transition. If $N_2 > N_1$, α is negative and $e^{-\alpha l} > 1$, i.e., amplification instead of absorption ensues. However, if $|\alpha|$ is small, a useful degree of amplification will be obtained only with an impractically long crystal. In such cases the interaction of radiation with the material can be increased by placing it between reflecting plates and advantage taken of the resulting regeneration due to waves reflected back and forth many times through the crystal. A wave that has traveled through the crystal once and reflected once has changed in energy content by the factor $e^{-\alpha l}r$, where r is the reflection coefficient of the end plate. In order to achieve a high amplification, or in the limit a coherent oscillation, the requirement is $e^{-\alpha l}r \cong 1$. In most cases $-\alpha l \ll 1$ so that the oscillation condition reduces to

$$-\alpha l \cong 1-r \quad (8)$$

or

$$(N_2-N_1) = (1-r)/\sigma_{12}l. \quad (9)$$

A different approach is to recognize that the parallel plate material system is an optical cavity, although not quite the same as a Fabry-Perot etalon because of the presence of the dielectric material; this latter point is discussed under "Spectral Width." The "Q" of the axial modes in this cavity can be obtained by solving for the mean decay time τ of photons moving perpendicular to the end plates.[4] When $(1-r) \ll 1$,

$$\tau \cong l\epsilon^{\frac{1}{2}}/[c(1-r)], \quad (10)$$

where l is the distance between the plates and ϵ is the dielectric constant of the material between the plates. Therefore,

$$Q_c = \omega\tau = 2\pi l\epsilon^{\frac{1}{2}}/[\lambda(1-r)]. \quad (11)$$

To account for the absorption (or emission) of energy within the material itself, we can define a material Q and find by similar reasoning to that above

$$Q_M = 2\pi\epsilon^{\frac{1}{2}}/\alpha\lambda. \quad (12)$$

In these terms the oscillation condition is $Q_c \cong -Q_M$ and is seen to be the same as Eq. (8). If we denote as α_0 the normally measured absorption coefficient for the transition $1 \rightarrow 2$ under low-power excitation, then, at the point of oscillation,

$$(N_2-N_1)/N_0 \cong (1-r)/\alpha_0 l \quad (13)$$

for a typical situation $(1-r) \ll \alpha_0 l$; therefore,

$$N_2/N_1 \cong 1+2(1-r)/\alpha_0 l. \quad (14)$$

and only a small excess over equal populations is required.

As the pumping power is increased above the point where $W_{13} = A_{21}$ (equal populations), the crystal begins to emit stimulated as well as spontaneous radiation. The spontaneous emission power is $N_2 h \nu_{12} A_{21}$, and since N_2 normally increases by only a small amount over the equal population value, as indicated above, almost all additional power developed at the frequency ν_{21} appears as stimulated emission. Since only this latter component constitutes useful output it would be desirable to pump at levels where $W_{13} \gg A_{21}$; unfortunately, however, it is difficult in practice even to reach the condition $W_{13} = A_{21}$.

Consider a parallel beam of light incident on the surface of an optically thin sample of the material. The power in the beam is

$$P_0 = nh\nu_p, \tag{15}$$

where n is the number of incident photons per second in the spectral region of the absorption band and ν_p is the center frequency of the pumping light. From the definition of transition probability

$$W_{13} = n\sigma_p/A \tag{16}$$

where σ_p is the absorption cross section for the pumping transition $(1 \rightarrow 3)$ and A is the area of the crystal perpendicular to the incident beam. Therefore, the flux density to produce equal populations $(W_{13} = A_{21})$ is

$$F = P_0/A = A_{21}(h\nu_p/\sigma_p). \tag{17}$$

Thus, in the case of an optically thin sample $(\alpha_p l \ll 1)$ the required flux density is independent of crystal concentration. Of course, in this case, only a small fraction of the incident power is absorbed, but increasing the ion density in the crystal results in nonuniform pumping so that in practice a compromise is made. The problem is alleviated somewhat if the back surface of the crystal is coated with reflecting material or if the entire crystal surface is illuminated uniformly.

The energy density U_p in the crystal is related to the flux density by $U_p = F\epsilon^{\frac{1}{2}}/c$ in the case of a parallel beam; therefore, using (17),

$$U_p = W_{13}h\epsilon^{\frac{1}{2}}/\lambda_p\sigma_p. \tag{18}$$

Although derived for the case of a parallel beam, the energy density is the fundamental quantity that determines transition probabilities, and, therefore, (18) is correct for other pumping geometries as long as the crystal is optically thin. The flux density, on the other hand, will depend on the particular configuration used, [e.g., $F = (c/4\epsilon^{\frac{1}{2}})U_p$ in the case of uniform illumination].

A configuration more likely to be realized in practice is one in which the material is more or less uniformly illuminated over most of its surface. Such an arrangement might be comprised of an appropriate reflector, lens, or reflector-lens combination. An alternative technique would be to use an extended source and place the active material in close proximity to it. For example, the source may be in the form of a hollow cylinder or

helix with the active material placed inside. A disadvantage of the latter arrangement is that unused radiation from the exciting source aggravates heat dissipation problems; however, the over-all structure is much more compact.

The type of source most likely to be used is a high-pressure gaseous or vapor arc lamp. The spectral lines characteristic of the particular gas or vapor are practically nonexistent in such a lamp because of a combination of pressure and Stark broadening. The resultant output has some vague peaks, but is otherwise a more or less continuous spectrum with an effective radiation temperature typically between 4000°K and 10 000°K. Therefore, it is convenient and not too unrealistic, to consider that the active material is immersed in isotropic blackbody radiation.

We again assume an optically thin crystal so that the radiation density is uniform throughout its volume. Equation (18) is applicable to monochromatic radiation; in the case of broad-band radiation,

$$W_{13} = \int \frac{c\sigma(\nu)\rho(\nu)d\nu}{h\nu_p\epsilon^{\frac{1}{2}}}, \tag{19}$$

where $\rho(\nu)$ is the energy density per unit volume per unit frequency interval. Taking $\sigma(\nu)$ as Lorentzian, and using

$$\rho(\nu) = \frac{8\pi h\nu^3\epsilon^{\frac{3}{2}}}{c^3(e^{h\nu/kT} - 1)}, \tag{20}$$

we get

$$W_{13} \cong \frac{4\pi^2\epsilon\Delta\nu_p\sigma_p\nu_p^2}{c^2(e^{h\nu/kT} - 1)}, \tag{21}$$

where $\Delta\nu_p$ is the half-width of the absorption band. From radiation theory,

$$A_{31} = 4\pi^2\epsilon\Delta\nu_p\sigma_p\nu_p^2/c^2 \tag{22}$$

for a Lorentzian line; therefore,

$$W_{31} = A_{31}/(e^{h\nu_p/kT_s} - 1). \tag{23}$$

The source temperature required for equal populations $(N_2 = N_1, W_{13} = A_{21})$ is therefore

$$T_s = \frac{h\nu_p}{k\ln[1 + (A_{31}/A_{21})]}. \tag{24}$$

If the lifetime of the fluorescent level is not wholly radiative, $1/A_{21}$ should be replaced by τ_2, the actual lifetime of this state.

The important material parameters connected with pumping power requirements for the three-level system are indicated in (24). The effective source temperature is a measure of the power per unit area per unit frequency interval and is, therefore, germane to the pumping problem. Since the necessary temperature is proportional to the pump frequency ν_p, operation at shorter wavelengths becomes increasingly difficult. The main

consideration at a given wavelength is to maximize the ratio $A_{31}:A_{21}$.

One of the main problems in obtaining stimulated emission from the three-level solid is the very high power density required for the pump source with its associated heat-dissipation problem. A possible solution is to use a pulsed rather than a continuous source, i.e., a high-power electronic flash lamp. If the pulse is short compared to the life of the fluorescent level $(\tau_s \ll \tau_2)$ the atoms excited to the fluorescent level do not appreciably decay during such an exciting pulse. The total energy absorbed when equal populations are attained is $E = \frac{1}{2}N_0 h\nu_p V$. If the crystal is isotropically irradiated, the absorbed energy is also (from the definition of Q_M at the pump frequency)

$$E = (2\pi\nu_p U_p V/Q_M)\tau_s. \quad (25)$$

Combining with previously derived relations, we find that the required energy per unit area radiated in one pulse from the flash tube is

$$J = F\tau_s \cong h\nu_p/4\sigma_p. \quad (26)$$

If, on the other hand, the pulse is long compared to the fluorescent decay time then the necessary *peak power* developed by the source is the same as that derived previously for a continuous source. The *average* power, however, can be made arbitrarily small by keeping the flash rate low. If large average powers are desired, larger flash tubes and consequently larger crystals can be employed. Although it requires about the same average input power to obtain a given average output power, the power dissipation problem is much less severe with pulsed operation than with continuous operation because of the greater volume of material involved.

Four-Level System

The level scheme depicted in Fig. 2 requires, in principle, lower exciting intensity than the three-level system discussed above.[5] If the low-lying level 2 is high enough above the ground state so that $e^{-h\nu_{12}/kT} \ll 1$, then level 2 will be relatively unpopulated. It will be as difficult as before to obtain an excess population with respect to the ground state, but negative temperatures for the transition $3 \rightarrow 2$ will be obtained at much lower intensities. The rate equations for this system are:

$$dN_3/dt = W_{14}N_1 + W_{24}N_2 - (A_{32} + A_{31})N_3 = 0, \quad (27)$$

$$dN_2/dt = w_{12}N_1 - (W_{24} + w_{21})N_2 + A_{32}N_3 = 0, \quad (28)$$

$$N_1 + N_2 + N_3 = N_0. \quad (29)$$

We have neglected the induced rates $W_{32} = W_{23}$ which do not become appreciable until inversion is produced and have assumed as previously that the nonradiative process S_{43} is much greater than either A_{42}, A_{41}, W_{14},

or W_{24}. The solution is then

$$\frac{N_3}{N_2} \cong \frac{W_{14}(W_{24} + w_{21}) + (W_{24} + w_{12})W_{24}}{W_{14}A_{32} + w_{12}(A_{32} + A_{31})}. \quad (30)$$

The quantities w_{12} and w_{21} are thermal transition probabilities, and the other quantities have the same meaning as previously stated. Since at room temperature thermal relaxation between levels 1 and 2 is apt to be very fast ($\sim 10^{-6}$ sec), $w_{12} \gg W_{14}$ and $w_{21} \gg W_{24}$; also the thermal probabilities are related by $w_{12} = w_{21} \exp[-h\nu_{12}/kT]$. Therefore,

$$\left(\frac{N_3}{N_2}\right) \cong \frac{W_{14}e^{h\nu_{12}/kT} + W_{24}}{A_{32} + A_{31}}. \quad (31)$$

and

$$N_2/N_1 \cong e^{-h\nu_{12}/kT}.$$

If we make the further assumption $W_{14} \approx W_{24}$, and define $A_3 = A_{31} + A_{32}$, we find that

$$\frac{N_3 - N_2}{N_0} \cong \frac{W_{14} - A_3/(1 + e^{h\nu_{12}/kT})}{W_{14} + A_3}. \quad (32)$$

In order to obtain high gain or oscillation the excess population must be

$$N_3 - N_2 \cong (1 - r)/\sigma_{32}l. \quad (33)$$

Combining (32) and (33) and assuming $N_0\sigma_{32}l \gg 1 - r$, we find

$$W_{14}\tau \cong \frac{1 - r}{N_0\sigma_{32}l} + \frac{1}{1 + e^{h\nu_{12}/kT}}, \quad (34)$$

where $\tau = 1/A_3$ is the lifetime of the fluorescent level. This result is to be compared with the equivalent requirement $W\tau \approx 1$ in the three-level system.

SPECTRAL WIDTH

The spectral width of the radiation emitted from an optical maser is discussed by Schawlow and Townes in the ideal limiting case in which the oscillations excite only a single mode in a Fabry-Perot type of resonator. This situation is possible with extremely precise fabrication in a gaseous or vapor system but has not been realized in solid state devices.[2,3,6] The effect of the solid dielectric is twofold. The first is the introduction of "light pipe" modes of propagation, due to internal reflection; the second is distortion of the mode patterns because of strains, inhomogeneities, and deviations from single crystallinity. The first effect is not so serious since it can, in principle, be circumvented, for example, by using a roughened cylindrical surface or immersing the maser material in a liquid (or solid) having nearly the same or higher refractive index; however, this is an additional problem to deal with.

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

Temperature effects in a solid are apt to be more serious than in a gaseous system. Generally speaking solids require a much higher power density because of the much larger density of atoms. For this reason, the magnitude of temperature shifts will be larger, and in addition the same temperature shifts will usually be of more consequence. The chamber for a gaseous system can be made of quartz or other special low temperature material whereas the plate separation in the solid situation is determined by the maser material itself which in turn may have a relatively high expansion coefficient. Finally, the energy levels are temperature dependent in a solid [about 3 (kMc/sec)/°C near room temperature for the ruby R_1 line].

Crystal imperfections, light-pipe modes, and end-plate misalignments generally introduce ambiguity in the mode selection process; i.e., there may be a very large number of modes which are of equal status in contrast to the perfect situation where the axial (or nearly axial) cavity mode closest to the atomic resonance peak is clearly the preferred one. Temperature shifts give rise to frequency sweeping and mode hopping.

As discussed by Schawlow and Townes,[4] multimoding introduces additional noise into the system due to spontaneous emission considerations; in addition, as shown in any text on oscillator theory, this same noise input reduces the monochromaticity of the output radiation from the system when it is oscillating.

We consider a system such as described above in which a large number of cavity modes enter into the oscillation because of poor mode selection. For purposes of illustration, we consider a cylindrical rod of three-level fluorescent material (such as ruby) with reflecting end plates to form an optical resonator. The power dissipated in this cavity is the sum of spontaneous and stimulated emission components. The stimulated emission power results primarily from the energy density built up in the cavity by standing waves and so essentially all of the stimulated emission power radiated by the material is coupled to the cavity. On the other hand, only a fraction (denoted by f) of the spontaneous emission power is dissipated in and excites the cavity. When regeneration ensues due to excess upper-state population, only the highest Q (axial and near axial) modes are excited appreciably. Although in practice only a small fraction of the total number of modes may be excited (limited principally by crystal perfection), the absolute number might be very large. (In a ruby of one-cm dimensions, the total number of modes is $\sim 2 \times 10^{11}$.)

The power per unit volume coupled to the cavity is

$$p_c/V = 2\pi\nu_{12}U/Q_c$$
$$= fA_{21}N_2h\nu_{12} + W_{21}(N_2-N_1)h\nu_{12}. \quad (35)$$

Here U is the energy density in the cavity, and Q_c is the loaded cavity Q (not including the material but including output coupling). Because we are dealing with a multimode cavity with dimensions very large compared to a wavelength, the spectral distribution of the spontaneous radiation is essentially the same as the fluorescence radiating into free space. If we assume a Lorentzian line, the frequency distribution of spontaneous power is $N_2A_{21}h\nu g(\nu)$, where

$$g(\nu) = \frac{\Delta\nu}{2\pi} \frac{1}{(\nu-\nu_0)^2 + (\Delta\nu/2)^2} \quad (36)$$

and $\Delta\nu$ is the half-power width of the atomic transition $(2 \to 1)$. Analogous to Eq. (19), we have

$$W_{21} = \int \frac{c\sigma(\nu)\rho(\nu)d\nu}{h\nu\epsilon^{\frac{1}{2}}}, \quad (37)$$

where $\sigma(\nu) = \frac{1}{2}\pi\Delta\nu\sigma_0 g(\nu)$. The power per unit volume per unit frequency interval is then

$$\frac{2\pi\nu\rho(\nu)}{Q_c} = fN_2A_{21}h\nu g(\nu) + \frac{c}{\epsilon^{\frac{1}{2}}}\sigma(\nu)\rho(\nu)(N_2-N_1), \quad (38)$$

therefore, using (12)

$$\rho(\nu) = \frac{1}{4\pi^2} \frac{fN_2A_{21}h\Delta\nu Q_c}{(\nu-\nu_0)^2 + (\Delta\nu/2)^2(1+Q_c/Q_M)}. \quad (39)$$

Thus, the energy density is Lorentzian with a half-amplitude width

$$B = \Delta\nu(1+Q_c/Q_M)^{\frac{1}{2}}. \quad (40)$$

The total power delivered to the cavity is

$$P_c = \frac{2\pi\nu V}{Q_c} \int \rho(\nu)d\nu,$$

therefore, using (36) and (37),

$$B = fN_2A_{21}h\nu\Delta\nu V/P_c. \quad (41)$$

The fraction f in terms of cavity modes is well known to be

$$f = \frac{\bar{n}}{4\pi^2\nu^2\Delta\nu V\epsilon^{\frac{3}{2}}/c^3}, \quad (42)$$

where the denominator is the total number of modes within the integrated width $(\frac{1}{2}\pi\Delta\nu)$ of the atomic transition and $\bar{n}$ is the actual number of modes participating in the oscillation; also since

$$A_{21} = 4\pi^2\epsilon\Delta\nu\sigma\nu^2/c^2,$$

we find

$$B = \bar{n}cN_2\sigma h\nu\Delta\nu/\epsilon^{\frac{1}{2}}P_c. \quad (43)$$

Then, using $N_2/N_1 = e^{-h\nu/kT_e}$ (where T_e is the effective temperature of the two-level system composed of the fluorescent level and the ground state), $|Q_M| = 2\pi\epsilon^{\frac{1}{2}}/|\alpha|\lambda \cong Q_c$ (when the system is oscillating), and $Q_c = \nu/\delta\nu$ (where $\delta\nu$ is the width of a single cavity mode)
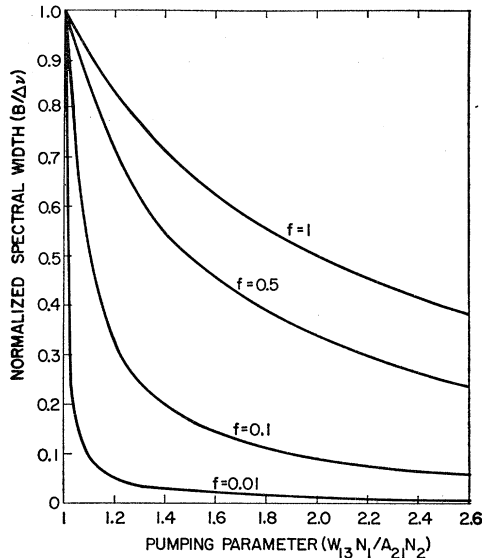
FIG. 3. Spectral bandwidth of the stimulated emission as a function of pumping power and the fractional number of cavity modes entering into the oscillation.

we obtain from (43)

$$B = 2\pi \bar{n} h \nu \Delta \nu \delta \nu / P_c (1 - e^{h\nu/kT_e}) \qquad (44)$$

(T_e is a negative quantity).

An alternative expression for the spectral bandwidth is obtained by considering the total power developed at the frequency ν_{12},

$$P_t = N_1 W_{13} h \nu_{12} V = P_c + (1-f) N_2 A_{21} h \nu V, \qquad (45)$$

then using (41), we find

$$B = \frac{f \Delta \nu}{(W_{13}/A_{21})(N_1/N_2) - (1-f)}. \qquad (46)$$

When the pumping power is increased to the point where $W_{13} = A_{21}(N_1 = N_2)$, from (46) $B = \Delta \nu$ independent of f. Any further increase in pumping power results in the production of stimulated emission and narrowing of the spectral width. Equation (46) is plotted in this region for various values of the parameter f and shown in Fig. 3. Thus, with a fairly good crystal, where good mode selection is feasible ($f \ll 1$), an abrupt change in the spectral width occurs with a fairly well-defined threshold pump power, but with a badly strained crystal ($f \sim 1$), the energy is scattered into many modes and no well-defined threshold can be found. Both types of behavior have been observed (see following paper).

The validity of the foregoing analysis breaks down when the spectral width of the "mode packet" is comparable to, or less than, that of the atomic resonance. In this case account must be taken of the modified spectral distribution of the spontaneous

radiation as discussed by Purcell[7] and Pound.[8] In the extreme case of a single mode oscillation one can use a wire circuit analysis[8,9] and arrive at an equation analogous to (44), i.e.,

$$B = \frac{2\pi h \nu (\delta \nu)^2}{P_c (1 - e^{h\nu/kT_e})}. \qquad (47)$$

This expression is similar to the one presented by Schawlow and Townes,[4] but more general in that account is taken of a finite lower state population (their result implies $T_e \rightarrow -0$). Also the width of the cavity mode appears here, as it should, since usually (for both solids and gases) $\delta \nu < \Delta \nu$ and the narrower of these two is the dominant element in determining the spectral width.

CONCLUSIONS

Optical excitation into a broad absorption band is possible with many fluorescent solids; thus, stimulated emission may be produced with relatively efficient utilization of typical broad band pumping sources. A three-level solid system as described here (e.g., ruby), uses the ground state as the terminal level for the stimulated emission transition. The required pumping power for this scheme is quite high; however, it has been shown that the important pumping parameter is the effective temperature of the source. A four-level fluorescent solid may also utilize a broad absorption band; however, in some cases, depending on the material parameters, much less pumping power is required than for the three-level scheme.

Solid-state systems pose additional problems in mode selection not present in a gaseous system. A dielectric rod constitutes a waveguide (light pipe) and thus additional modes of propagation (which may be low loss) are introduced. Even more important are strains, inhomogeneities, and deviations from single crystallinity which tend to scatter energy into undesired modes. Under the assumption that the mode selection is poor, i.e., many modes participate in the oscillation, an analysis was presented for the case of badly strained crystals. The general features of this formulation have been observed in the laboratory as discussed in the following paper.

No attempt has been made here to treat the transient behavior or multiple spectrum observed in pulsed excitation of ruby and other solids. A quantitative description would require an analytical expression for the variation of Q of the various mode possibilities. This in turn is a function of the particular crystal and its history of fabrication (see following paper).

[7] E. M. Purcell, Phys. Rev. **89**, 681(A) (1946).
[8] R. V. Pound, Ann. Phys. (N. Y.) **1**, 24 (1957).
[9] T. H. Maiman, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), pp. 324–332.