P. P. Sorokin
N. Braslau

Some Theoretical Aspects of a Proposed Double Quantum Stimulated Emission Device*

Abstract: A double quantum stimulated emission device is proposed and some operating characteristics and relevant systems of materials are discussed.

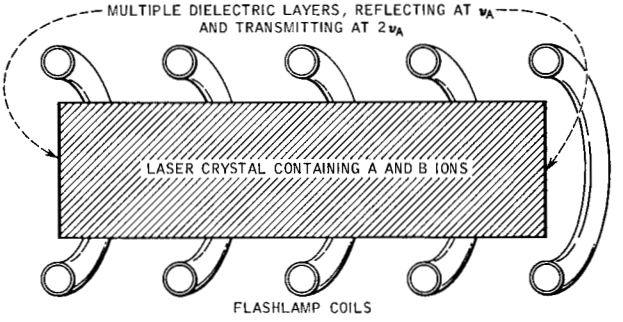
1. Introduction

A number of recent experiments[1-5] have demonstrated that double quantum transitions in the optical region of the electromagnetic spectrum can be induced by laser beams. Kaiser and Garrett have observed,[1] for example, that irradiation of a $CaF_2 : Eu^{+2}$ crystal with intense red light ($\nu = 14\,400$ cm^{-1}) from a ruby optical maser produces a small amount of characteristic Eu^{+2} (blue) fluorescence. It is known that there are no Eu^{+2} energy levels below $\approx 23\,500$ cm^{-1}. Thus, $CaF_2 : Eu^{+2}$ crystals cannot directly absorb ruby light. However, a strong $CaF_2 : Eu^{+2}$ absorption band does occur between 25 000 and 30 000 cm^{-1}, corresponding to $4f - 5d$ transitions of the Eu^{+2} ion. If $CaF_2 : Eu^{+2}$ crystals are allowed to absorb light in this wavelength range, there is produced the brilliant blue fluorescence (band width ≈ 1700 cm^{-1}) which originates at levels around 23 500 cm^{-1} and terminates on the ground state. Kaiser and Garrett proposed, then, that the intensity of a ruby laser beam was great enough to excite Eu^{+2} ions into the $5d$ states via double quantum transitions. In the unit process considered, simultaneous absorption of two ruby photons accompanies the exictation of an Eu^{+2} ion from the ground state to a $5d$ level near 28 800 cm^{-1}. (A subsequent nonradiative decay then quickly drops the Eu^{+2} ion to one of the fluorescing levels at 23 500 cm^{-1}). A theoretical description of double quantum radiative transitions which has existed for some time[6] indicates that the number of Eu^{+2} fluorescent ions produced per unit volume per unit time should be proportional to the square of the incident photon flux (cf. Eq. (3) below). This was found by Kaiser and Garrett to be indeed the case. These authors were able also to determine quantitatively the efficiency of the process in $CaF_2 : Eu^{+2}$. Three other studies[2-4] also examined absorptive double quantum transitions in solids but they, like that for Eu^{+2}, involved *broad band* spectral transitions. One other experiment[5] has given a clear-cut description of an absorptive double quantum transition in a gas.

In the present paper a coherent light generator making use of double quantum stimulated *emission* is considered. The device is shown to be realizable under assumptions compatible with current knowledge and art regarding optically pumped solid state lasers. Expected properties of the device are calculated and presented and some relevant systems of materials are discussed.

A brief examination of Figs. 1 and 2 enables the basic idea of the proposed device to be understood. A crystal contains two species of fluorescent ions, A and B, of which species A can serve as the active ion in a four-level laser. The crystal is fabricated to have a conventional laser geometry in which, for example, the ends are polished

Figure 1 **Schematic diagram of the device.**



* This research was supported jointly by International Business Machines Corporation and the United States Army Research Office, Durham, North Carolina.

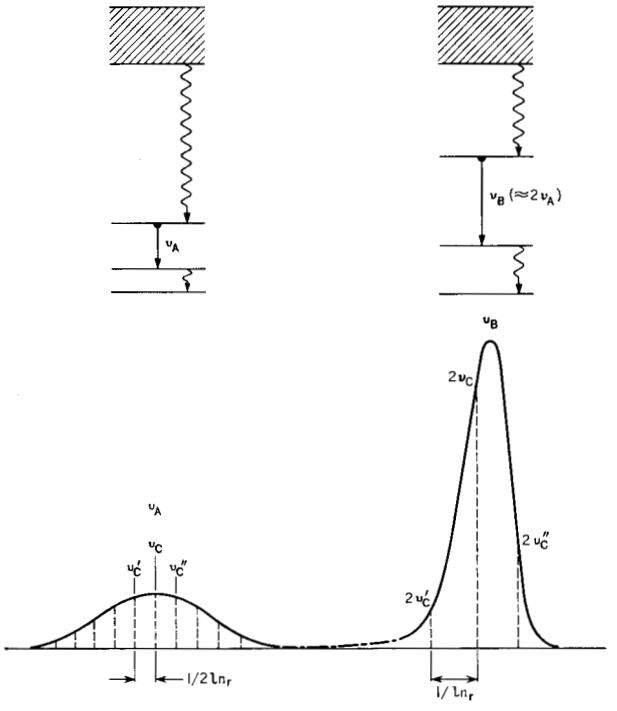


Figure 2 **Relationship between A and B ions.**

into plane or hemispherical surfaces. The ions A and B are required to bear a special relationship to each other: when multiplied by two, the frequency of the laser light generated by the A ions must fall within the spontaneous emission line width of a B ion fluorescent transition which can be inverted by the action of the same surrounding flashlamp that activates the A ions. Deposited on the ends of the crystal are reflecting films which must be highly reflective at frequency ν_A and highly transmissive at $2\nu_A$. (Both of these needs can be achieved through the use of multiple dielectric layer films.)

The action of the device is as follows. The flashlamp pumps both A and B ions, inverting the population of each in the aforementioned transitions. Since the end plate reflectivity is high at ν_A, the crystal will begin to oscillate at this frequency, emitting "spikes" of light in the manner characteristic of most optically pumped four-level solid state lasers. Large photon populations will thus be sporadically produced in certain modes having frequencies ν_c, ν_c', ν_c'', $\cdots$ which lie near the peak ν_A of the A ion fluorescence. Although no stimulated emission involving the inverted B ion population can occur, because of the low end-plate reflectivity at $2\nu_A$, there remains the possibility that if the photon density becomes large enough in one of the modes coherently excited by the A ions (ν_c, for example), the cross-section for a double quantum transition in which a B ion is de-excited and two photons are simultaneously added to the ν_c mode will then become appreciably large. Since the cross section for this process continues to increase with the addition of more photons to the ν_c mode, an avalanche effect producing ν_c photons can occur. This would result in a "giant" pulse at the ν_c frequency. As is shown below, this pulse would continue until the population excess of B ions is largely reduced.

Requirements for triggering

A fundamental question to be answered is whether normal "spiking" of the A ions can produce enough photons in the cavity mode ν_c to "trigger" the double quantum avalanche which involves and is energized by the B ions. Recently Kleinman[7] reconsidered the original theory of Göppert-Mayer[6] in the light of the successful experiment performed by Kaiser and Garrett. In Kleinman's article, the following expression is derived for the coefficient expressing the strength of two-photon absorptive processes

$$\sigma_1 = \frac{r_0^2 c^2 f^2}{n_r^2 \nu_A^2 \Delta\nu}, \tag{1}$$

where n_r is the refractive index of the medium, r_0 is the classical electron radius, and $\Delta\nu$ is the width of the real, excited state at $2h\nu_A$. In the derivation of Eq. (1) it is assumed that absorption occurs through a single, intermediate, virtual state coupled to both initial and final states by an oscillator strength f. Although Eq. (1) must, *a priori*, be regarded as only approximate, it nonetheless accounts surprisingly well for the data for CaF_2:Eu^{+2} presented in Ref. 1. To obtain the double quantum *cross section* σ, the coefficient σ_1 appearing in Eq. (1) must be multiplied by the incident flux F; thus we have

$$\sigma = \sigma_1 F \tag{2}$$

and the decrease in power of a propagating plane wave inducing double quantum absorptive transitions is

$$\frac{dP}{dx} = -\left(\frac{N}{V}\right)\sigma_1 \frac{P^2}{h\nu_A A}, \tag{3}$$

where A is the beam area and (N/V) is the density of ions. Additionally, as will be important subsequently, Eqs. (1), (2), and (3) should also apply in the case of stimulated emission, with the exception that the *minus* in Eq. (3) should be replaced by a *plus*. For the double quantum stimulated emission process to become dominant, resulting in the formation of a giant pulse, the cavity energy gained by this process must exceed the energy lost by transmission through the end walls. If $\bar{u}_T$ is the average total energy density in the cavity, (i.e., the total for both electric and magnetic energy), the "trigger" condition may be written

$$\int_{x=0}^{x=l} \left(\frac{d\bar{u}_T}{dt}\right)_{DQ} dx = \frac{1}{t_c}\int_{x=0}^{x=l} \bar{u}_T \, dx. \tag{4}$$

In Eq. (4) t_c is the cavity decay time given by

$$t_c = \frac{n_r l}{c(1 - R)} \tag{5}$$

provided, as is assumed, the cavity energy decays via an equal transmission loss $(1 - R)$ through each end wall. The expression $(d\bar{u}_T/dt)_{DQ}$ represents the rate of increase of average total energy density inside the cavity as a result of the double quantum process. For the case of a propagating plane wave, the following holds true

$$P = \frac{A\bar{u}_T c}{n_r}. \tag{6}$$

Thus, from Eq. (3) one obtains

$$\left(\frac{d\bar{u}_T}{dt}\right)_{DQ} = \left(\frac{N^b}{V}\right)\frac{\sigma_1 c^2 \bar{u}_T^2}{h\nu_A n_r^2} \tag{7}$$

where N^b/V is the inverted population density of B ions. Noting that only the electric field is active in a two-photon process, since the "intermediate" states are of opposite parity, one may rewrite Eq. (7) as

$$\left(\frac{d\bar{u}_T}{dt}\right)_{DQ} = 4\left(\frac{N^b}{V}\right)\frac{\sigma_1 c^2 \bar{u}_E^2}{h\nu_A n_r^2} \tag{8}$$

and assume that this last equation holds true generally. Inside the cavity the following expressions obtain

$$\bar{u}_E = \tfrac{1}{2} K \cos^2 kx \tag{9}$$

$$\bar{u}_H = \tfrac{1}{2} K \sin^2 kx, \tag{10}$$

where K is a constant of proportionality. By making combined use of Eqs. (4), (5), (8), (9), and (10), one obtains the following expression for the peak power emitted from either end of the crystal in the smallest primary spike capable of triggering the double quantum avalanche:

$$P_{\text{spike}} = \frac{A\int \bar{u}_T\, dx}{2t_c} = \frac{(1 - R)^2 h\nu_A A^2}{8\sigma_1 N^b \langle\cos^4\rangle}. \tag{11}$$

Since $\langle\cos^4\rangle = \frac{3}{8}$,

$$P_{\text{spike}} = \frac{1}{3}\frac{(1 - R)^2 h\nu_A A^2}{N^b \sigma_1}. \tag{12}$$

Using Eq. (1) with $f \approx 1$, $n_r = 1.5$, $\nu_A = 1.5 \times 10^{14}$ cps, and $\Delta\nu \approx 9.6$ cm^{-1}, one finds $\sigma_1 \approx 4.8 \times 10^{-45}$ cm^4 sec. Using this value of σ_1 in Eq. (12), together with the values $N^b/V \approx 10^{18}$/cm^3, $l = 4$ cm, $A = 0.5$ cm^2, and $(1 - R) = 0.05$, one computes a required minimum output power $P_{\text{spike}} \approx 2.18$ kw. This peak output power should be attainable with most existing four-level optically pumped lasers. The number of photons S_0 in the cavity corresponding to this power is determined by

$$\frac{S_0 h\nu_A}{2t_c} = P_{\text{spike}}. \tag{13}$$

Thus, for the conditions assumed here, $t_c \approx 4 \times 10^{-9}$ sec, and $S_0 \approx 1.74 \times 10^{+14}$ photons. From the above the triggering requirements are seen to be reasonable. Although there is obvious necessity for fairly heavy pumping, sufficient pumping intensity should be available with the crystal placed within the coils of a cylindrical flashlamp such as the FT-524.

Time development of the giant pulse

For the parameters considered above, it is possible to show that a giant pulse would indeed occur if the photon population in the primary cavity exceeds S_0. A simple set of rate equations describing the kinetics of the process (neglecting spontaneous emission of B ions and stimulated emission of A ions) is

$$\frac{dS}{dt} = 2B_1 S^2 N^b - \frac{S}{t_c} \tag{14}$$

$$\frac{dN^b}{dt} = -B_1 S^2 N^b, \tag{15}$$

where S is the number of photons at any time in the ν_c mode and N^b is the total inversion of B ions in the crystal at any time. The coefficient B_1 appearing in Eqs. (14) and (15) is related to the quantity σ_1 appearing in Eq. (1) by

$$B_1 = \frac{3}{4}\frac{c^2\sigma_1}{V^2 n_r^2}, \tag{16}$$

Let S_i be the initial number of photons placed in the mode at time $t = 0$ as a result of stimulated emission from the A ions. In terms of Eqs. (14) and (15) the threshold number of photons S_0 is that number which makes $(dS/dt)_{t=0} = 0$. This is found from Eq. (14) to be

$$S_0 = \frac{1}{2B_1 N_i^b t_c}, \tag{17}$$

where N_i^b is the initial B ion population inversion.

For a choice of parameters satisfying $N_i^b \gg S_i$, one expects the number of inverted B ions N_i^b to remain essentially constant until the number of photons S exceeds the threshold value by at least an order of magnitude. Initially then, Eq. (14) can be integrated directly with N^b constant and equal to N_i^b. If the initial number of photons is given as a fraction η of the threshold number S_0,

$$S = S_0[1 - (1 - 1/\eta)\exp(t/t_c)]^{-1}. \tag{18}$$

For $\eta > 1$ this expression remains finite until the time

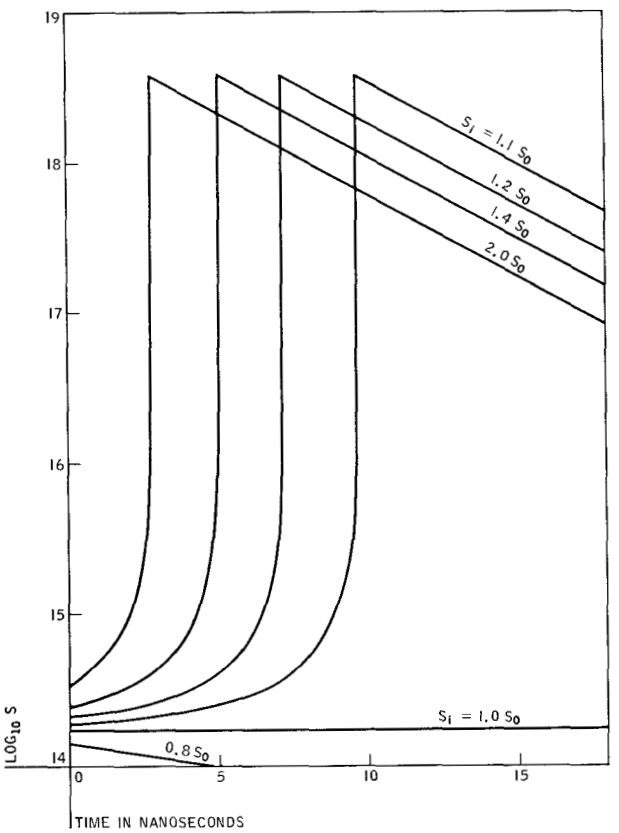
$$t_G = t_c \ln[\eta/(\eta - 1)], \tag{19}$$

when it rapidly diverges. One thus expects the number

of photons in the cavity to be given by Eq. (18) up to about the time t_G. At this time, the number of photons will be large enough to affect the B ion population inversion. The latter will suddenly be driven rapidly to zero with the number of photons increasing to a value approximately equal to $2N_i^b$. Thereafter, the number of photons will decay with the cavity time constant t_c.

Solution of the coupled set of equations (14) and (15) was implemented by a program written for the IBM 7094 employing a method of successive iterations. Examples of the results obtained are shown in Fig. 3. These agree very closely with predictions made using Eqs. (18) and (19). The following values of parameters were used in these machine calculations: $N_i^b = 2 \times 10^{18}$, $B_1 = 3.6 \times 10^{-25}$ sec^{-1}, and $t_c = 4 \times 10^{-9}$ sec. The threshold number of photons S_0 has the value 1.74×10^{14}. (These numbers are consistent with those of Sec. 2.) As shown in Fig. 3, for $\eta \leq 1$ the number of photons will simply decay to zero, but for $\eta > 1$ the number rapidly builds up to the maximum value $2N_i^b$ at a time very close to t_G and then decays with the cavity time constant t_c. For the parameters chosen, the spike rises in a time of the order of one nanosecond and decays with a half life of 4 nanoseconds. Thus a giant pulse occurs.

Figure 3 **Time development of the giant pulse.**



Discussion

The number of near-infrared radiative transitions existing in the case of the trivalent rare earths is large enough to suggest that the attainment of the required 2 : 1 frequency match is not out of the question. For example, the ions Nd^{+3} and Ho^{+3} both work as four-level laser ions in CaF_2, with trivalent neodymium oscillating very near to 1.046μ in CaF_2 and trivalent holmium oscillating very near to 2.092μ in the same lattice.[8] This particular system is one which would afford design opportunities were it not for the fact that selection rules contained in a recently developed theory[9] appear to raise serious objections for this specific system. Other possibilities remain for success, however. One promising approach involves the use of Yb^{+3} as the B ion. Trivalent ytterbium has a single, very intense luminescent group ${}^2F_{5/2} \rightarrow {}^2F_{7/2}$ located near one micron. In many lattices such as CaF_2 the fluorescence of Yb^{+3} is primarily concentrated in a pair of very sharp lines ($\Delta\nu \approx 1$ cm^{-1} at 300°K). Although these are resonance lines, and hence correspond to three-level laser transitions, population inversion can be achieved by pumping into the wide intense Yb^{+3} band covering the region 33 000 to 45 000 cm^{-1}. Selection rules favor a large double quantum cross-section for ${}^2F_{5/2} \rightarrow {}^2F_{7/2}$ transitions. The fluorescent decay time of Yb^{+3} is long ($\tau \approx 10^{-2}$ sec), and the luminescence is notoriously unsusceptible to concentration quenching. Since operation at room temperature should be feasible, the crystal containing the Yb^{+3} ions can be fabricated with Brewster-angle ends so that it can be physically isolated from the triggering crystal but yet remain in the primary cavity. Triggering possibilities are afforded by silicate glass rods doped with either Ho^{+3} or Tm^{+3}. These ions lase around 2μ; the exact frequency will be a function of the selective reflectance of multiple dielectric layers.

To find all systems that could lead to successful two-photon lasers will require careful study and thorough cataloguing of the fluorescent spectra (principally in the i-r range) of the rare earth ions Yb^{+3}, Tm^{+3}, Er^{+3}, Ho^{+3}, Dy^{+3}, Nd^{+3}, and Pr^{+3} in such lattices as CaF_2, CdF_2, and LaF_3. A better understanding of $4f \rightarrow 4f$ double quantum cross sections should soon be available since they are related to electronic Raman cross sections and the necessary understanding can be inferred from studies of the latter. It seems most likely that the results of future studies will enable one to judge whether Eq. (1) is in fact adequate for estimating the order of magnitude of σ_1 in the case of $4f \rightarrow 4f$ transitions.

Acknowledgments

We are indebted to our colleagues, Drs. G. J. Lasher, W. A. Hardy, and J. D. Axe, Jr., for most helpful dis

cussions. Also stimulating were two discussions with Dr. H. R. Robl of the U. S. Army Research Office—Durham.

References

1. W. Kaiser and C. G. B. Garrett, "Two-Photon Excitation in CaF_2:Eu^{2+}," *Phys. Rev. Letters* **7**, 229 (1961).
2. W. L. Peticolas, J. P. Goldsborough, and K. E. Rieckoff, "Double Photon Excitation in Organic Crystals," *Phys. Rev. Letters* **10**, 43 (1963).
3. R. G. Kepler, J. C. Caris, P. Avakian, and E. Abramson, "Triplet Excitons and Delayed Fluorescence in Anthracene Crystals," *Phys. Rev. Letters* **10**, 400 (1963).
4. J. L. Hall, D. A. Jennings, and R. M. McClintock, "Study of Anthracene Fluorescence Excited by the Ruby Giant-Pulse Laser," *Phys. Rev. Letters* **11**, 364 (1963).
5. I. B. Abella, "Optical Double-Photon Absorption in Cesium Vapor," *Phys. Rev. Letters* **9**, 453 (1962).
6. M. Göppert-Mayer, "Über Elementarakte mit zwei Quantensprungen," *Ann. Physik* **9**, 273 (1931).
7. D. A. Kleinman, "Laser and Two-Photon Processes," *Phys. Rev.* **125**, 87 (1962).
8. L. F. Johnson, "Optical Maser Characteristics of Rare Earth Ions in Crystals," *J. Appl. Phys.* **34**, 897 (1963).
9. J. D. Axe, Jr., to be published.

Received January 16, 1964.