THE INFLUENCE OF ABSORPTION FROM THE METASTABLE LEVEL OF A SENSITIZING ON THE CHOICE OF LASER PUMPING MODE

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The influence of pump pulse duration on the self-excitation of a laser is considered, where the laser uses active materials in which the sensitizers possess resonant absorption from a metastable level. It is shown that, given strong absorption of this type, the trailing edge duration \( \tau \) of the pump pulse must be quite short to satisfy the generation conditions. The sensitizers can produce threshold energy reduction only when \( \tau \ll 1/N_a \), where \( 1/N_a \) is the life of the excited state of the activator, if saturation of the resonant absorption is possible, generation takes place as a single giant pulse or as a regularly repeated series of such pulses.

51. One way of improving pumping efficiency in a laser is to sensitize the luminescence. Luminescent organic compounds and uranics are particularly effective sensitizers for rare earth ions. But it is difficult to make workable lasers based on these materials, and sometimes, when the pumping is continuous, it may even be impossible, due to absorption at the operating frequency of the activator as a result of transitions from the sensitizer metastable level.

The energy transmission processes in such systems are discussed below in the context of absorption from the sensitizing metastable level, in an attempt to find the optimal pumping conditions.

Consider an elementary scheme of sensitizer level (s) and activator level (a) (Fig. 1), and the processes occurring in the system when pumped by light. In Fig. 1, 1a and 1s are respectively the lowest working level of the activator and the fundamental level of the sensitizer, and 2a and 2s are the metastable levels. Induced radiation occurs at the frequency of the 2a \( \rightarrow \) 1a transition. The transition 2s \( \rightarrow \) 2s has a frequency close to the operating frequency, and causes absorption from the sensitizer metastable level.

As usual, the 2s metastable level is populated by pumping at the 2s' level, with the subsequent radiationless transition 2s' \( \rightarrow \) 2s. The time of this transition can be assumed to be negligible compared to the characteristic times in the system. The working metastable level (2a) of the activator is excited both by pumping at the 2a' level, with the subsequent transition 2a' \( \rightarrow \) 2a, and as a result of radiationless energy transmission from the excited metastable level 2s, of the sensitizer.

The equations describing the processes in the system before the generation threshold is reached are

\[
\begin{align*}
N_a &= PN_a = D_s A(t), \\
N_s &= W_s N_a = D_s A(t) + SN_a,
\end{align*}
\]

where \( N_a \) is the population of the 2s level; \( N_s \) is the population of the 2a level; \( P = S + W_s \), \( S \) is the probability of the radiationless transition 2s \( \rightarrow \) 2a in unit time; \( W_s \) is the probability of the spontaneous radiative transition 2s \( \rightarrow \) 1s in unit time; \( W_a \) is the probability of spontaneous radiation at the frequency of the operating transition 2a \( \rightarrow \) 1a; \( \int \frac{\omega}{h} A(v) dv = E(t) \) is the optical pump power; \( \int A(v) dv = A(t) \) is the number of photons radiated by the pump source per unit time; and

\[
\begin{align*}
\int D_s(v) A(v) dv &= D_s A(t), \\
\int D_s(v) A(v) dv &= D_s A(t)
\end{align*}
\]

is the number of photons absorbed in unit time by the sensitizer and the activator respectively. The initial conditions of the problem may be written as

\[ N_s = N_a = 0. \]  

The equation for the number of photons \( n \) at the frequency of the 2a \( \rightarrow \) 1a transition is

\[ \frac{dn}{dt} = W_s N_a + n(\sigma_a N_a - \beta - \gamma N_a) / \gamma, \]  

where \( \sigma_a \) is the cross section of the induced radiation at the working transition 2a \( \rightarrow \) 1a; \( \sigma_s \) is the cross section of the absorption at the transition 2s \( \rightarrow \) 3s; \( \beta = \beta_s + (1 - r)/L \) is the loss factor per unit length, including the nonresonant loss in the substance (\( \beta_s \)) and

* The probability of the transition 2s \( \rightarrow \) 3s does not enter into the probability \( P \) because fast radiationless transitions 3s \( \rightarrow \) 2s are present in the substances considered here. The duration of this transition is \( 10^{-6} \) sec for \( \text{UO}_2^{2+} \), while for dyes it is even less.
In the mirrors \((1 - r)/L\) where \(r\) is the reflection coefficient of the mirrors, and \(L\) is the length of the active element; \(c\) is the velocity of light, and \(n\) is the refractive index. Unlike ordinary active materials with low losses, no superluminescence exists when \(N_{o}g_{a} \sim N_{a}g_{a}^{*}\).

The generation condition is that the term describing the induction processes on the right side of (3) must become positive at some instant:

\[
\sigma_{a}N_{a} > \beta + \sigma_{a}N_{a}. \tag{4}
\]

The special feature of our case, therefore, is that high-power pumping leads on the one hand to inversion at the \(2a \rightarrow 1a\) transition, but on the other hand produces resonant absorption at the frequency of the \(2s \rightarrow 3s\) transition.

\$\S 2.\$ We consider three cases, taking into account Eq. (1) and the generation condition (4).

a) Continuous (stationary) pumping \((A(0) = A_{0} > 0)\). Putting \(N_{o} = N_{a} = 0\) in (1), we obtain from (1) and (4) the self-excitation condition

\[
A_{0} > P \frac{P}{\sigma_{a}(D_{s} + D_{p} + S)} D_{s}W_{a} = \frac{1}{(c/\omega_{a})}. \tag{5}
\]

It is easily seen that generation is possible only when

\[
\sigma_{a}D_{s}S + D_{p}P > 1. \tag{6}
\]

For illustration, take the case of sensitization of \(Nd^{3+}\) and \(Eu^{3+}\) by uranyl. In the case of phosphoric glass coactivated by \(1\% UO_{2}^{2+}\) and \(3\% Nd\), we have \(\sigma_{Nd} = 3 \times 10^{-23} \text{ cm}^{-3} \text{ sec}^{-1}, W_{Nd} = 2 \times 10^{4} \text{ sec}^{-1}, S = 3 \times 10^{4} \text{ sec}^{-1}, \gamma_{UO_{2}} = 10^{-48} \text{ cm}^{2}, \gamma_{Eu} = 10^{-30} \text{ cm}^{2}\). Then, in the same glass, we have \(\sigma_{Eu}(7020 \AA) \sim 10^{-23} \text{ cm}^{2} \text{ sec}^{-1}, W_{Eu} = 5 \times 10^{4} \text{ sec}^{-1}, S = 3 \times 10^{4} \text{ sec}^{-1}\). Obviously, in both cases, when an optical pump source with a brightness temperature of \(7000^\circ\) K is used, \(D_{p} = 0.2, D_{s} = 0.10\), and \(D_{s}W_{a} = 0.01\), condition (6) is not satisfied.

b) Pumping by a very short pulse. We shall show that, with this method of energy transfer, the restrictions indicated in the previous example no longer apply. Let pumping be by a pulse of duration \(\tau < 1/P\). We shall assume that the pulse is switched off at the instant \(t = 0\); we then find, for the \(N_{o}\) and \(N_{a}\) in (1) (as is usually the case), that

\[
N_{a} = \frac{\varepsilon_{o}}{D_{s}e^{-PT}},
\]

\[
N_{a} \approx \varepsilon_{o}D_{s}S(1 - e^{-\tau}) + \varepsilon_{o}D_{s}. \tag{7}
\]

\[
\varepsilon = \int_{t}^{\infty} A(t) dt. \tag{8}
\]

The quantity \(\varepsilon\) is proportional to the total pump energy.

Substituting (7) in (4), the condition to be satisfied by the pump energy \(P\) for generation to result becomes

\[
\varepsilon > \varepsilon_{o} = \frac{\beta P}{\sigma_{a}(SD_{s} + PD_{s})}. \tag{9}
\]

Given sufficient pump energy, this condition can be satisfied for any \(\sigma_{a}\). In fact, after the pump pulse has terminated, along with the increase in the inverse population at the \(2a\) level, the population at the \(2a\) level falls and hence the absorption in the \(2s \rightarrow 3s\) transition is reduced. It is clear from (9) that the presence of an effective sensitizer \((P \gg W_{a}\text{ and }P \gg S)\) considerably reduces the threshold energy, which in the absence of sensitization is \(\varepsilon = \beta D_{s}P_{a}\). Hence, in the case of a pump source with a temperature of 7000$^\circ$ K, sensitization of neodymium by uranyl should roughly halve the threshold energy, while sensitization of europium by uranyl should reduce the threshold by more than an order of magnitude.

c) Pumping by a long pulse. If the duration of the pulse is \(\tau > 1/P\), the generation condition becomes more difficult. The most important pulse characteristic here is the leading-edge duration \(\tau\). As \(\tau\) increases, i.e., as we approach the continuous pumping mode, the pump threshold energy must also increase and become infinite at some \(\tau > \tau_{cr}\) (when condition (6) is not satisfied).

Let us find the function \(\varepsilon(t)\). Since our main interest is in the trailing edge of the pump pulse, we shall assume that the pulse shape is as shown in Fig. 2 (ininitely steep leading edge and time-exponential trailing edge):

\[
A(t) = \begin{cases} 0 & (t < 0) \\ \frac{\varepsilon_{o}e^{-\tau t}}{\tau} & (t > 0) \end{cases} \tag{10}
\]

We shall assume that

\[
P \gg W_{a}. \tag{11}
\]

This is also the condition for effective sensitization and must therefore always be satisfied by materials for lasers.

With the pump shape (10), Eq. (1) gives for the populations \(N_{o}(t)\) and \(N_{a}(t)\), when \(\tau \gg 1/P\) and \(P \gg 1/P\),

\[
N_{o}(t) \approx \frac{\varepsilon_{o}}{P_{a}} e^{-\tau t},
\]

\[
N_{a}(t) \approx \frac{\varepsilon_{o}SD_{s} + PD_{s}}{P_{a}} \left( e^{-\varepsilon_{o}t} - e^{-\varepsilon_{o}t} \right). \tag{12}
\]

The absorption coefficient \(\beta + \sigma_{a}N_{a}\) and gain \(\sigma_{a}N_{a}\) are

\[
\text{Absorption} \quad \beta + \sigma_{a}N_{a}
\]

\[
\text{Amplification} \quad \sigma_{a}N_{a}
\]

Fig. 2
shown for this case as functions of time in Fig. 2; Eqs. (12) give their values in the region of interest, i.e., close to the point where the curves touch one another in the case of threshold pumping. Using these values of $N_b$ and $N_a$ and the generation condition (4), for the case when (6) is not satisfied we get

$$I(t) = \frac{E_{th}(a)}{E_{th}(0)} = a^{\alpha_1} \left( \frac{\alpha_1 - 1}{\alpha_1 - \alpha} \right)^{\alpha_1 (a-1)}, \quad (13)$$

where

$$\alpha = \tau W_a, \quad (14)$$

$$\frac{s_{cr}}{1 - s_{cr}(SD_2 + PD_2)/\alpha S_{cr} D_2} = \frac{\beta P}{\epsilon_0 (SD_2 + PD_2)}, \quad (15)$$

$$E_{th}(0) = \frac{\beta P}{\epsilon_0 (SD_2 + PD_2)}. \quad (16)$$

It is easily seen that $E_{th}(a)$ is defined only when $0 < \alpha < \alpha_{cr}$, while $E_{th}(\alpha) \rightarrow \infty$ as $\alpha \rightarrow \alpha_{cr}$. Hence, if condition (6) is violated, trailing edge duration of the pump pulse must be less than the critical value $\tau_{cr}$ for generation to result:

$$\tau < \tau_{cr} = \frac{s_{cr}}{1/\alpha S_{cr}}, \quad (17)$$

In all cases, a sufficient time condition is

$$\tau < 1/\alpha S_{cr}, \quad (18)$$

(since $\alpha_{cr} > 1$ when (6) is violated). However, given the active material parameters of $\alpha_2 a$, threshold reduction due to sensitization will be observed when $\alpha \leq 0.2$.

Bursts with a trailing edge several orders of magnitude shorter than $1/\alpha S_{cr}$ may easily be obtained, since in the case of Nd$^{3+}$ in crystals, glasses and liquids, $\tau_{Nd}$ amounts to $10^{-2}$ to $10^{-4}$ sec.

Note that we assumed when deriving (13), that $\tau > 0.1/\alpha S_{cr}$, which corresponds to the maximum possible $\tau$. But consideration of $\alpha_2 b$ for the case $\tau < 1/P$ shows that, under condition (11), the threshold energy $E_{th}$ of (9) is the same for this case as the $E_{th}$ of (16). In connection, we can assume that (13) is applicable for any $\tau$ when condition (11) is satisfied.

$\S 3$. The considerations above refer primarily to the conditions for self-excitation in various pumping modes; an examination of the generation dynamics in such systems is of interest in itself, but we can only point to some general features here.

When absorption by the parasitic transition is large compared to nonresonant absorption, and the radiationless relaxation $3s \rightarrow 2s$ is not of particularly short duration ($\sim 10^{-8}$ sec for uranyl), it seems probable that generation will consist of single radiation pulses of the "giant pulse" type, such as occur in systems with Q-switching. In our case, the Q-switching occurs as a result of "clarification" of the $2s \rightarrow 3s$ transition or a rapid decrease in the population $N_b$.

If, after a giant radiation pulse, the population of the $2s$ level is sufficient for a new accumulation of atoms at the $2a$ level and to satisfy generation condition (4), a second giant pulse will follow the first, and so on.

Consider the conditions under which a pulse series can occur. We shall start from the fact that, after the first pulse, a population $N_b \approx \beta P \beta_{2s} S D_3$, representing the threshold for the next pulse, remains in the level. We assume here that the $2s$ transitions are the only ones of importance in destroying the $3s$ level. In this case, the population of the $3s$ level must be the same before the first peak and shortly after it.

Assuming that the pump pulse duration is short ($\tau \ll 1/P$), we find from condition (4) that the starting instant of the first generated pulse is

$$t_1 = \frac{1}{P} \ln \frac{\beta P}{\epsilon_0 E_{th}}, \quad (19)$$

where

$$\rho = 1 + \frac{2 \alpha S_{cr} P D_2}{\epsilon_0 (SD_2 + PD_2)}. \quad (20)$$

At this instant, the population of the $2s$ level is

$$N_s(t_1) = \frac{E_{th} - E_{th}}{\rho}. \quad (21)$$

For generation of a second pulse, this population must be greater than the threshold:

$$N_s(t_1) > E_{th} = E_{th}(D_a = 0) = \frac{\beta P}{\epsilon_0 S D_2} \quad (21.1)$$

Hence the condition for more than one generated pulse to occur during one pump pulse is

$$\frac{E_{th}}{E_{th}} > 1 + \frac{E_{th}}{\rho}. \quad (22)$$

Similarly, the condition for generation of a sequence of n pulses per pump pulse is

$$\frac{E_{th}'}{E_{th}} > \frac{E_{th}'}{E_{th}} + 1, \quad (23)$$

where

$$\frac{E_{th}'}{E_{th}} = \frac{E_{th}(D_a = 0) = 1 + \frac{s_{cr}}{\alpha} S_{cr} D_2}{S_{cr} D_2}. \quad (24)$$

The spacing between the first pulses is

$$\Delta t = \frac{1}{P} \ln \frac{1}{\rho'}. \quad (25)$$

When the probability of a radiationless $3s \rightarrow 2s$ transition is high, a sensitizer molecule will return rapidly to the $2s$ level after absorbing a light quantum and jumping to the $3s$ level, and clarification of the $2s \rightarrow 3s$ transition becomes very difficult. This can make generation of giant pulses impossible. In this case, ordinary beam generation will be observed when the condition $E > E_{th}$ is satisfied.

The present results agree with the experimental data of [1, 2], where generation in glass activated by UO$_2^{3+}$ and Nd$^{3+}$ is described. The oscillograms reproduced in these papers show periodically repeated generation pulses with a power of up to 20 MW on the skirt of the pump pulse. Measurements performed in the laboratory show that, given a short pump ($\tau_p \approx 30 \mu$ sec),
sensitization reduces the threshold by as much as a factor of 2.

To sum up, when we have a strong parasitic resonant absorption from the metastable level of an excited sensitizer, generation is impossible under continuous pumping conditions. But generation becomes possible when the pump pulse power is sufficiently high. The duration $\tau$ of the pump pulse trailing edge should here be less than some definite critical value. The pump threshold energy increases with $\tau$.

If the absorbing transition is clarified, a giant pulse is generated. When the pump energy is well above the threshold, a sequence of giant pulses may occur.

REFERENCES


11 May 1967 Institute of Radio Engineering and Electronics AS USSR