AMPLIFICATION OF LIGHT BY FOUR-LEVEL QUANTUM SYSTEMS

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Stationary and transient regimes of light amplification in active media operating according to a four level scheme are considered. Amplification of light in $CaF_2:Sm^{2+}$ is investigated. The decrease in the lifetime of an excited state in the case of appreciable inversion is calculated. Two types of losses in an active medium are discussed.

IN this paper we investigate the amplification of light in an active medium operating according to a four level scheme and traversed by monochromatic radiation of wavelength corresponding to the maximum in the coefficient of negative absorption in the medium. Such a case occurs, for example, when stimulated radiation characteristic of the active medium under consideration [1,2] is being amplified.

The theoretical investigation is based, as before [3], on the method of probabilities.

STATIONARY REGIME

We consider the stationary regime of light amplification in a sample of length l. The ends of the sample are flat with a coefficient of reflection R equal to zero. Thus, it is assumed that there is no feedback in the amplifier. It can be easily seen that this simplification is valid when the condition $(MR)^2 \ll 1$ is satisfied where M is the coefficient of light amplification in the sample.

The differential equation for the density of radiation U in the amplifier can be written in the form

$$dU / U = (K - \rho)\, dx, \qquad (1)$$

where K and ρ are respectively the coefficients of amplification and of losses per cm length.

The relation between the coefficient of amplification K and the density of radiation U can be obtained by means of the equation for the stationary population N_3 of the upper working level:

$$N_3 / \tau + N_3 B_{32} U - N_0 B_{14} U_{14} = 0, \qquad (2)$$

where $\tau = 1/(A_{31} + A_{32})$ is the lifetime of the excited state; A_{31}, A_{32}, B_{32}, B_{14} are the Einstein coefficients, N_0 is the density of the activator atoms; U_{14} is the density of the pumping radiation (it is assumed that the population of the lower working level and, moreover, $N_3 \ll N_0$; both conditions are as a rule satisfied [3] in the case of real four level systems with actually attainable pumping rates).

It follows from (2) that

$$N_3 = N_3^0 / (1 + \alpha U),$$

where $N_3^0 = B_{14} U_{14} \tau N_0$ is the population of the upper level for $U = 0$; $\alpha = B_{32}\tau$.

Since the coefficient of amplification $K \sim N_3$, we can write

$$K = K_0 / (1 + \alpha U); \qquad (3)$$

K_0 corresponds to the coefficient of amplification K for $U = 0$, i.e., in the absence of a signal being amplified.[1)]

Substituting relation (3) into (1) and carrying out the integration we obtain the following relation for the general coefficient of amplification M in the sample:

$$\ln M = \ln \frac{U(l)}{U(0)} = (K_0 - \rho) l + \frac{K_0}{\rho} \ln \left[1 - \frac{M - 1}{(K_0/\rho - 1)/\alpha U(0) - 1} \right], \qquad (4)$$

where $U(l)$ and $U(0)$ are the values at the output and the input of the amplifier respectively of the density of the radiation being amplified.

TRANSIENT REGIME

We now estimate the time required to establish the stationary regime of amplification restricting

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1)In accordance with [4] expression (3) is valid also in those cases when relation (2) is not applicable. However, further analysis and comparison of results of theoretical estimates with experimental results are limited to the case when (2) is applicable.

our consideration to the case when at the instant of time $t = 0$ the radiation to be amplified arrives at the input of the amplifier with an intensity of n quanta per second. Neglecting the losses in the medium we can write

$$\frac{dN_3}{dt} = \frac{N_3^0}{\tau} - \frac{N_3}{\tau} - \frac{n}{V}(M-1).$$

Here N_3^0 is the population of the upper level before the arrival of the signal to be amplified, V is the volume of the sample.

From this relation one can easily go over to the expression describing the variation with time of the amplification coefficient M, if we take into account that $\ln M \sim N_3$:

$$\frac{dM}{dt} = M\left[\frac{\ln M_0}{\tau} - \frac{\ln M}{\tau} - \frac{n}{VN_3^0}(M-1)\ln M_0\right]. \quad (5)$$

If the radiation being amplified comes from a source formed in a similar manner and placed in a resonator, then for equal pumping intensities for the generator and the amplifier it can be easily shown that expression (5) assumes the form

$$\frac{dM}{dt} \approx -\frac{M}{\tau}[\ln M + (M-2)\ln M_0]. \quad (5')$$

From relation (5′) it follows that for large values of M_0 the rate of establishment of a stationary amplification coefficient (in the present case its value does not exceed 2) is quite large. Numerical integration of (5′) for the cases $M_0 = 10$ and $M_0 = 100$ gives for the instants of time at which M has been reduced to a value M_0/e the values $10^{-1}\tau$ and $4 \times 10^{-3}\tau$.

REDUCTION IN THE LIFETIME OF AN EXCITED STATE IN THE CASE OF APPRECIABLE INVERSION

In the case of an appreciable population inversion it is necessary to take into account the reduction in the lifetime of the excited state due to the increase in the spontaneous radiation in the medium as the result of induced transitions which leads to a reduction of N_3 and consequently also of the amplification coefficient K. The existence of these induced transitions can be taken into account by replacing in (2) τ by τ_{eff}; it can be easily shown that

$$\gamma = \frac{\tau}{\tau_{eff}} = 1 + \eta\frac{N_i}{N_s},$$

where η is the quantum yield of luminescence of frequency ν_{32}; N_s and N_i are the number of events of spontaneous and induced (by the quanta of spontaneous transitions) radiation of frequency ν_{32} taking place per second in the sample. The value of $N_i/N_s = \xi$ depends in an essential manner on the geometry of the sample. An estimate has been made of ξ for the case of a circular cylindrical sample with a coefficient of amplification K which is constant over its volume. The end surfaces, just as before, were regarded as flat and translucent, while the side surface is either translucent or completely dull, i.e., scattering isotropically the energy incident on it.

For the power of the radiation emerging from the sample we can write the relation

$$h\nu_{32}\left[N_s + N_i\left(1 - \frac{\rho}{K}\right)\right] = \frac{1}{2}\int_{S_d} E\,dS + \int_{S_t} E\,dS,$$

where E is the illumination (from the inside) of the surface of the sample by radiation of frequency ν_{32}; S_d and S_t are the areas of the dull and of the translucent parts of the surface of the sample. From this it follows that

$$\xi = \frac{K}{K-\rho}\left[\left({}^1/_2\int_{S_d} E\,dS + \int_{S_t} E\,dS\right)\Big/ h\nu_{32}N_c - 1\right].$$

Thus, in order to find ξ and, consequently, γ, it is sufficient to evaluate the illumination of the surface of the sample. Without repeating the fairly awkward calculations, we note only that the calculation reduces to the numerical solution of the integral equation for E(s).

Figure 1 shows the calculated values of ξ and γ for $\rho = 0$ and $l/D = 4$ (D is the diameter of the sample). In the evaluation of the value of γ it was assumed that $\eta = 0.2$ (the case of $CaF_2:Sm^{2+}$ for 20°K [5]). It can be seen from Fig. 1 that the reduction in the lifetime for large values of Kl can be appreciable. In particular, in the case of samples with a dull side surface the effect of the reduction in the lifetime becomes noticeable already for $Kl \geq 1.6 - 2$, which corresponds to the value of $M \geq 5 - 7$.

We note that in order to estimate the value of γ one can utilize the relation

$$\gamma = 1 + \eta DK \exp[l_e(K-\rho)],$$

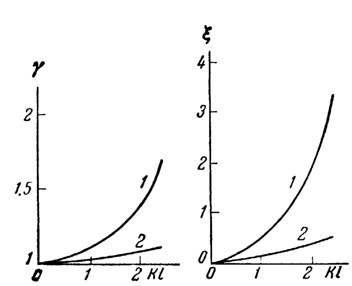

FIG. 1. Calculated values of γ and ξ: 1—dull side surface, 2—translucent side surface.

obtained on the assumption that each spontaneous quantum of frequency ν_{32} before leaving the sample traverses a length l_e (for $l/D \geq 10$ for a dull sample $l_e \approx 5D/3$, while for a translucent sample $l_e \approx 2D/3$).

RESULTS OF INVESTIGATIONS

We now discuss the results of our investigations. Crystals of $CaF_2:Sm^{2+}$ at a temperature of 20°K were used as the active medium. The absence of "spikes" in the stimulated radiation of this material renders it a very convenient object for the study of amplification of light in an active medium.

The measurements of the coefficient of light amplification which were carried out refer to the stationary regime of amplification; transient regimes whose duration for the case of $CaF_2:Sm^{2+}$ was less than 1 μsec have not been studied.

The experimental arrangement is schematically shown in Fig. 2. In the cryostat 1 there are placed two cylindrical samples 2 and 3 each of 30 mm length and 8 mm diameter with a dull side surface and flat ends. Special experiments were conducted to check the complete identity of the material of samples 2 and 3 (cut out from the same initial batch) and, in particular, the equality of the values of the threshold pumping power (when the samples are placed in the resonator) was verified. Suitable coatings ($R_1 = 1$ and $R_2 = 0.66$) were applied to the ends of sample 2. This sample is the source of the radiation being amplified. Sample 3 is the amplifier. Between it and sample 2 there was placed diaphragm 4 of 2.5 mm diameter which served to eliminate edge effects. In order to suppress feedback in amplifier 3 due to the reflecting coating on the end of sample 2 the axis of sample 3 was inclined with respect to the axis of sample 2 by an angle of ~3°. The pumping of samples 2 and 3 was carried out by pulsed lamps 5 and 6 respectively; the mirrors 7 and 8 served to focus the radiation from the lamps onto the samples. The duration of a pumping pulse in the case of sample 2 was 25-30 μsec, and in the case of sample 3 it was 150 μsec (the shape of the pumping pulse is rectangular). Lamp 5 was turned on ~70 μsec after lamp 6 had been turned on. The intensity of pumping in samples 2 and 3 was recorded by means of two photomultipliers before the entrance windows of which filters were placed which selected the portion of the spectrum 0.6–0.65 μ corresponding to the absorption band of $CaF^2:Sm^{2+}$.

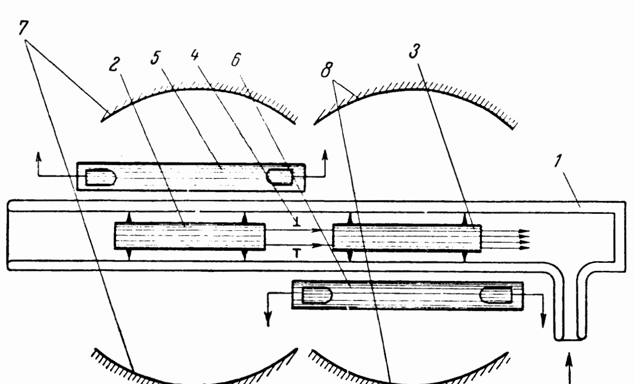

FIG. 2. Schematic diagram of the experimental arrangement.

The radiation from sample 2 after passing through sample 3 was recorded by means of a third photomultiplier. By comparing the values of the intensity of this radiation when sample 3 was "pumped" and when it was not "pumped," it was possible to determine the value of $M_1 = Me^{\rho l}$ (since in the absence of pumping in sample 3 $U(l) = U(0)\,e^{-\rho l}$). Measurements were carried out for different pumping intensities of samples 2 and 3; a change in the pumping intensity of 2 brought about a corresponding change in the density of radiation $U(0)$ falling on the input of the amplifier. The results of the measurements are shown in Fig. 3.

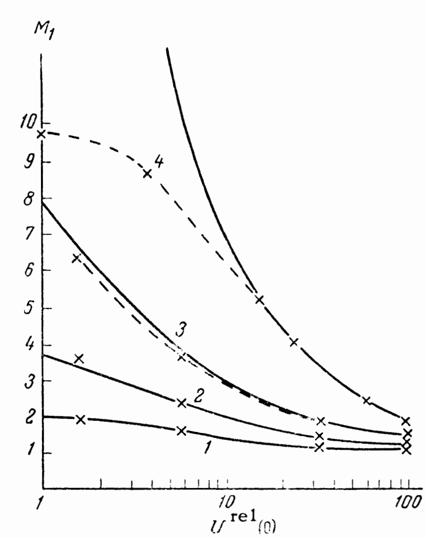

FIG. 3. Dependence of the coefficient of amplification on the pumping. Solid curves—calculation according to (6) for $\sigma = 0.11$ and $\rho = 0.03$, dotted curves and points—experimental results; 1—$U^{rel}(0) = 1.4$; 2—$U^{rel}(0) = 2.8$; 3—$U^{rel}(0) = 5.2$; 4—$U^{rel}(0) \approx 12$.

Along the vertical axis we have plotted the value of M_1, and along the horizontal axis the value of the density of the signal being amplified $U^{rel}(0)$ in relative units, with the value unity being assigned to the value of $U(0)$ when the pumping intensity of sample 2 exceeds the threshold intensity by a factor two. Curves 1-4 correspond to the pumping intensity of sample 3 P_{14}^{rel} in relative units being equal respectively to 1.4, 2.8, 5.2, and 12, with the value unity being assigned to the pumping intensity equal to the threshold pumping intensity of sample 2.

As can be seen from the results shown the coefficient of amplification M_1 is reduced as the intensity of the radiation being amplified is increased; this dependence is most strongly manifested for large values of the amplification coefficient.

DISCUSSION OF THE RESULTS

In order to compare the results of these investigations with theoretical results it is necessary to express the quantities K_0 and $\alpha U(0)$ appearing in relation (4) in terms of the experimentally obtainable quantities P_{14}^{rel} and $U^{rel}(0)$. Taking into account the assumptions utilized in deriving relation (2) and the adopted unit for measuring the pumping intensity P_{14}^{rel} we can write

$$K_0 = P_{14}{}^{rel} K_{th},$$

where K_{th} is the threshold value of the coefficient of amplification in sample 2. With respect to the quantity K_{th} the following remark should be made: in accordance with elementary theory $K_{th} = \sigma + \ln(1/R_2)/2l$, where σ is the value of the losses in the resonator expressed per cm length. Measurements have shown that the value of σ is significantly larger than the value of ρ which determines the decrease in the flux in its passage through the active medium. In order to determine σ and ρ a study was made of the dependence of the threshold pumping intensity P_{th} and of the generator power P_g on the value of the transmission of the mirror $\beta = 1 - R_2$ (cf., for example,[3]). Taking into account the fact that $P_{th} \sim K_{th}$ and, as can be easily shown, $P_g \sim [\beta/(\beta + 2\rho l)]\,(P_{14} - P_{th})$, values of σ and ρ were obtained which were respectively equal to 0.1 and 0.033 cm^{-1}. The difference in the values of ρ and σ is apparently due to the fact that the value of ρ is determined, primarily, by the inactive absorption and large angle scattering (> 30-60′), while the value of σ is determined, in addition to the factors indicated above, also by diffraction, by small angle scattering, by imperfections of the resonator (wedge, spherical aberration, waviness, etc.), i.e., purely "resonator" losses make contributions to σ.[2)]

[2)]Apparently, this circumstance was partially responsible for the disagreement observed in a number of papers (cf., for example, [5]) between the calculated and the observed values of the threshold pumping intensity.

We note that a direct determination of the quantity K_{th} which reduces to a measurement of M_1 for $P_{14}^{rel} = 1$ and a very small value of $U(0)$ gave for the quantity σ a value which agreed within experimental accuracy with the value given above. Thus, the quantity K_0 can be expressed in terms of P_{14}^{rel} in the following manner:

$$K_0 = P_{14}{}^{rel}\left(\sigma + \frac{1}{2l}\ln\frac{1}{R_2}\right).$$

Utilizing the results of reference[3] and the boundary conditions at the semitransparent mirror on the end of sample 2 it can be easily shown that the quantity $\alpha U(0)$ can be represented in terms of the units of measurement chosen above in the form

$$\alpha U(0) = \frac{1-R_2}{1+R_2} U^{rel}(0).$$

Substituting the expressions obtained for K_0 and $\alpha U(0)$ into (4) we obtain for the quantity M_1 the following relation:

$$\ln M_1 = P_{14}\left(\sigma + \frac{1}{2l}\ln\frac{1}{R_2}\right)\left\{l + \frac{1}{\rho} - \frac{1}{\rho}(M_1 e^{-\rho l} - 1)\right.$$
$$\times\left[\left(P_{14}{}^{rel}\left(\sigma + \frac{1}{2l}\ln\frac{1}{R_2}\right) - \rho\right)\right.$$
$$\left.\left.\times\left(\rho\frac{1-R_2}{1+R_2}U^{rel}(0) - \rho\right)^{-1} - 1\right]^{-1}\right\}.$$

The calculated values of M_1 are shown in Fig. 3. As can be seen, the agreement between the experimental and the theoretical results is quite satisfactory with the exception of the case when the amplification coefficient $M_1 \geq 7$ (curve 4). The disagreement in this case is apparently due to the reduction in the lifetime of the excited state which leads to a reduction in the amplification coefficient.

[1] J. Heysick and D. Scovil, In the collection of articles on "Lasers" edited by M. E. Zhabotinskiĭ and T. A. Shmaonov, IIL, 1963, p. 135.

[2] P. P. Kisliuk and W. S. Boyle, Proc. Inst. Radio Engrs. 49, 1635 (1961).

[3] Anan'ev, Egorova, Mak, Prilezhaev and Sedov, JETP 44, 1884 (1963), Soviet Phys. JETP 17, 1268 (1963).

[4] B. I. Stepanov and V. P. Gribkovskiĭ, UFN 82, 201 (1964), Soviet Phys. Uspekhi 7, 68 (1964).

[5] Kaiser, Garret, and Wood, In the collection of articles on "Lasers" edited by M. E. Zhabotinskiĭ and T. A. Shmaonov, IIL, 1963, p. 75.

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