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Abstract

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Properties of a Four-Level Optical Quantum Generator

Let us consider the optical properties of the active substance of a four-level quantum generator. Level 3 is metastable. The transition probabilities are equal to:

$$p_{ij} = A_{ij} + B_{ij}(u^0_{ij} + u_{ij}) + d_{ij} = p^0_{ij} + B_{ij}u_{ij}; \tag{1}$$

$$p_{ji} = B_{ji}(u^0_{ji} + u_{ij}) + d_{ji} = p^0_{ji} + B_{ji}u_{ij} \quad (i > j), \tag{2}$$

where A_{ij} and B_{ij} are the Einstein coefficients integrated over frequency; u^0_{ij} is the density of equilibrium radiation of frequency ν_{ij}; u_{ij} is the density of external radiation (u_{41} is the density of the pumping radiation, u_{32} is the density of the generated radiation). For the system under consideration one may put

$$p^0_{13} = p^0_{14} = p^0_{23} = p^0_{24} = p^0_{34} = 0, \qquad u_{31} = u_{21} = u_{43} = u_{42} = 0, \tag{3}$$

$$p^0_{12} = p^0_{21} \exp(-h\nu_{21}/kT), \qquad B_{14}u_{41} \ll A_{41}. \tag{4}$$

This means that upward thermal transitions are absent in all channels except $2 \rightleftarrows 1$. Inequality (4) can practically never be violated.

The stationary populations of the levels for a system of particles with any number of levels have been calculated in [1]. Applied to a system with four levels and taking (3)–(4) into account, they are expressed by the formulas:

$$n_1 = n\{(p^0_{41} + p^0_{42} + p^0_{43} + B_{14}u_{41})[p^0_{21}(p^0_{31} + p^0_{32}) + B_{23}u_{32}(p^0_{21} + p^0_{31})]\}/D; \tag{5}$$

$$\begin{aligned} n_2 = n\{&(p^0_{41} + p^0_{42} + p^0_{43} + B_{14}u_{41})[p^0_{12}(p^0_{31} + p^0_{32}) + B_{23}u_{32}p^0_{12}] \\ &+ B_{14}u_{41}[(p^0_{42} + p^0_{43})(p^0_{32} + B_{23}u_{32}) + p^0_{42}p^0_{31}]\}/D; \end{aligned} \tag{6}$$

Fig. 1

Figure 1: Fig. 1

$$n_3 = n\{B_{14}u_{41}p^0_{43}p^0_{21} + B_{23}u_{32}[p^0_{12}(p^0_{43} + p^0_{42} + p^0_{41}) + B_{14}u_{41}(p^0_{43} + p^0_{42} + p^0_{12})]\}/D; \quad (7)$$

$$n_4 = n\{B_{14}u_{41}[p^0_{21}(p^0_{31} + p^0_{32}) + B_{23}u_{32}(p^0_{21} + p^0_{31})]\}/D, \quad (8)$$

where n is the number of particles per unit volume, and D is the sum of all terms standing in the braces in (5)–(8).

If in formulas (5)–(8) one sets u_{32} equal to zero, then they will determine the dependence of n_i on the pump density u_{41} in the absence of a resonator and, consequently, in the absence of generation. In the presence of a resonator they are valid in the region from $u_{41} = 0$ to the threshold value u_{41}^{thr}, characterizing the onset of radiation generation.*

The approximate course of the dependences $n_i(u_{41})$ for $u_{32} = 0$ and

$$p^0_{21} > p^0_{32} + (p^0_{32} + p^0_{31})p^0_{42}/p^0_{43} \quad (9)$$

* Generally speaking, in the presence of a resonator u_{32} cannot be considered equal to zero even in the absence of generation. Taking this circumstance into account is complicated; its result depends on the specific properties of the resonator. Thus, for a cylindrical sample of radius r, without taking into account reflection of the radiation from its lateral surface, in the subsequent formulas for the threshold pump power one should replace p^0_{32} by $p^0_{32}(1 + r/K_{32}^{\text{thr}})$ (see (13)). In other cases, for example in the presence, at the frequency ν_{32}, of total internal reflection from the lateral surface, the correction may be larger.

shown in Fig. 1 by solid lines. The values of n_4 are practically equal to zero. If condition (9) is not fulfilled, then $n_2 > n_3$, and generation at the frequency ν_{32} is impossible.

According to (6) and (7), at a certain value of u_{41} the population of level 3 begins to exceed the population of level 2, and the absorption coefficient k^0_{23} becomes negative.

Fig. 1. Dependence of the absorption and generation powers (in kW) and of the level populations (in relative units) on the pump-radiation density at $T = 100°$K.
$p^0_{41} = 4 \cdot 10^7,\ p^0_{42} = 0,\ p^0_{43} = 10^8,\ p^0_{31} = 10^4,\ p^0_{32} = 3 \cdot 10^4,\ p^0_{21} = 10^5,\ p^0_{24} = p^0_{34} = p^0_{13} = p^0_{23} = 0,\ p_{14} = B_{14}u_{41}$ sec^{-1}; $\delta_{23} = 0.1,\ p^0_{12} = 2.27 \cdot 10^3,\ u_{41}^{\text{thr}} =$

$0.67 \cdot 10^{-13}$ erg/cm^3 sec^{-1}. The solid curves are calculated for $u_{21} = 0$; the dashed curves correspond to the generation regime.

Formulas (5)–(8) make it possible to determine the values of the threshold and of the generation density at the frequency ν_{32}. The average radiation density inside the resonator is determined by the expression[2]

$$u_{32}(\nu)\Delta\nu_{32}^{\mathrm{g}} = \frac{\left|k_{23}^0(\nu)\right| - k_{23}^{\mathrm{loss}}}{\alpha_{32}k_{23}^{\mathrm{loss}}}, \tag{10}$$

where $u_{32}(\nu)$ is the spectral density of the generated radiation; $\Delta\nu_{32}^{\mathrm{g}}$ is the width of the generated line; $k_{23}^0 = B_{23}(n_2^0 - n_3^0)h\nu_{32}/\nu\Delta\nu_{32}^{\mathrm{a}}$ is the value of the absorption coefficient at the maximum of the absorption band for a given pump and in the absence of a resonator (i.e., for $u_{32} = 0$); $\Delta\nu_{32}^{\mathrm{a}}$ is the width of the absorption line for the transition $2 \to 3$; α_{32} is a nonlinearity parameter of the system, determined with the aid of (6) and (7) from the relation

$$k_{23} = B_{23}(n_2 - n_3)h\nu_{32}/\nu\Delta\nu_{32}^{\mathrm{a}} = k_{23}^0/(1 + \alpha_{32}u_{32}(\nu)\Delta\nu_{32}^{\mathrm{g}}); \tag{11}$$

k_{23}^{loss} is the coefficient characterizing losses of the generated radiation when the radiation emerges beyond the layer, losses in the coatings, and losses due to scattering and absorption by impurities. The coefficient k_{23}^{loss} is calculated per unit length of the resonator and is equal to[3]

$$k_{23}^{\mathrm{loss}} = \frac{1}{l}\ln\frac{1}{\sqrt{r_{23}r_{23}'}} + \rho_{23}. \tag{12}$$

The threshold pump power is determined by equating expression (10) to zero. Using (10), (12), and formulas (6) and (7) at $u_{32} = 0$, we obtain:

$$u_{41}^{\mathrm{thr}} = \frac{1}{B_{14}}\frac{(p_{31}^0 + p_{32}^0)[p_{12}^0 + \delta_{23}(p_{21}^0 + p_{12}^0)]}{\eta_{43}(M - \delta_{23}N)}, \tag{13}$$

where

$$M = p_{21}^0 - p_{32}^0 - (p_{31}^0 + p_{32}^0)(p_{42}^0 + p_{12}^0)/p_{43}^0; \tag{14}$$

$$N = (p_{21}^0 + p_{32}^0) + (p_{31}^0 + p_{32}^0)(2p_{21}^0 + p_{42}^0 + p_{12}^0)/p_{43}^0; \tag{15}$$

$$\delta_{23} = k_{23}^{\mathrm{loss}}\nu\Delta\nu_{32}^{\mathrm{a}}/nB_{23}h\nu_{32} = k_{23}^{\mathrm{loss}}/\chi_{23}, \qquad \eta = \frac{p_{43}^0}{p_{43}^0 + p_{42}^0 + p_{41}^0}. \tag{16}$$

The quantity χ_{23} determines the value of the absorption coefficient k_{23}^0 that would be obtained if all particles were on the second level.

If $p_{12}^0 = 0$ (large $h\nu_{21}/kT$) and $\delta_{23} = 0$ (no external losses), then the generation threshold is equal to zero. This is the principal advantage of a four-level quantum generator.

Formula (13) takes the simple form:

$$u_{41}^{\text{thr}} = \frac{1}{B_{14}\eta_{43}} \frac{(p_{32}^0 + p_{31}^0)\{\exp(-h\nu_{21}/kT) + \delta_{23}[1 + \exp(-h\nu_{21}/kT)]\}}{(1 - p_{32}^0/p_{21}^0) - \delta_{23}(1 + p_{32}^0/p_{21}^0)}. \quad (17)$$

provided that the condition

$$p_{43}^0 \gg (p_{32}^0 + p_{31}^0)(1 + p_{42}^0/p_{21}^0) \quad (18)$$

is satisfied.

If, in addition, it is also true that

$$p_{21}^0 \gg p_{31}^0 + p_{32}^0, \quad (19)$$

then (9) is certainly fulfilled, and the threshold can be high only for large δ_{23}, i.e., for large external losses or a small value of χ_{23}. It follows from (17) that the most advantageous substances are those with large p_{21}^0 and small $p_{32}^0 + p_{31}^0$ (large τ_{32}). For p_{32}^0 close to p_{21}^0, the four-level scheme loses its advantages.

The amount of energy generated per unit volume of the resonator can be determined from the formula $W_{32}^{\text{r}} = \nu k_{23} u_{32}(\nu)\Delta\nu_{32}$ with the aid of (10), calculating $k_{23}^0(\nu)$ and α_{32} from (11), (6), and (7), and taking into account the condition of stationary generation ($k_{23} = k_{2\infty}^{\text{tot}}$).

The calculation gives

$$W_{32}^{\text{gen}} = \eta_{43} \frac{nh\nu_{32}(M - \delta_{23}N)\, B_{14}(u_{41} - u_{41}^{\text{thr}})}{p_{21}^0 + 2p_{12}^0 + p_{31}^0 + 2\pi_{43}B_{14}u_{41}\left[1 + (p_{42}^0 + p_{21}^0 + p_{12}^0 + p_{31}^0)/p_{43}^0\right]}. \quad (20)$$

If the pumping is small, the generation power is proportional to the difference $u_{41} - u_{41}^{\text{thr}}$. For large u_{41} (when $B_{14}u_{41} \sim p_{31}^0 + p_{21}^0$), the tangent of the angle of inclination on the curve W_{32}^{gen} begins to decrease (see Fig. 1). Taking into account conditions (18), (19), and (4), formula (20) assumes a simpler form:

$$W_{32}^{\text{gen}} = n\eta_{43}h\nu_{32}\frac{[1-p_{32}^0/p_{21}^0-\delta_{23}(1+p_{32}^0/p_{21}^0)]\,B_{14}(u_{41}-u_{41}^{\text{thr}})}{1+p_{31}^0/p_{21}^0+2\exp(-h\nu_{21}/kT)+2\left(1-\frac{p_{41}^0}{p_{43}^0+p_{42}^0+p_{41}^0}\right)B_{14}u_{41}p_{21}^0}. \quad (21)$$

For large p_{32}^0/p_{21}^0, large δ_{23}, and small η_{43}, the generation power is very small. The maximum possible power for an ideal four-level generator ($p_{32}^0/p_{21}^0 \to 0,\ B_{14}u_{41}/p_{21}^0 \to 0,\ \eta_{43}=1$) is

$$W_{32}^{\text{gen}} = \frac{nh\nu_{32}(1-\delta_{23})}{1+2\exp(-h\nu_{21}/kT)}\times$$

$$\times\left[B_{14}u_{41}-\frac{(p_{31}^0+p_{32}^0)\{\delta_{23}+(1+\delta_{23})\exp(-h\nu_{21}/kT)\}}{1-\delta_{23}}\right]. \quad (22)$$

In the case of small external losses and very low temperatures, the efficiency of an ideal four-level generator is very high. At $T=0$ and $\delta_{23}=0$, the generation power is equal to $nB_{24}u_{41}h\nu_{32}$. Under these conditions all particles are on the lower level, and the generation power, to within Stokes losses in the channels $4\to 3$ and $2\to 1$, is equal to the absorption power for $n_1=n$.

Substituting the value $M-\delta_{23}N$ from (13) into (20), it is not difficult to obtain a convenient formula relating the generated power to the excess over threshold $x=u_{41}/u_{41}^{\text{th}}$:

$$W_{32}^{\text{gen}} = (x-1)\,\frac{(p_{31}^0+p_{32}^0)[\delta_{23}+(1+\delta_{23})\exp(-h\nu_{21}/kT)]}{1+p_{31}^0/p_{21}^0+2\exp(-h\nu_{21}/kT)}\,nh\nu_{32} \quad (23)$$

(the term with u_{41} in the denominator of (20) may be neglected).

By measuring W_{32}^{gen} at different pump powers and temperatures, with the aid of (23) it is not difficult to determine the three principal characteristics of the generator, $p_{31}^0+p_{32}^0$, δ_{23}, and p_{31}^0/p_{21}^0.

After the generation threshold is reached, very large radiation densities of frequency ν_{32} arise inside the resonator, which strongly changes the population of the levels and hence the absorption power and other optical properties of the working substance. All the calculations are readily carried out by substituting in (5)–(8) the values of u_{32} from (10). The form of the functions $n_i(u_{41})$ in the generation regime is shown in Fig. 1 by the dashed lines. The dependence of the absorption power on u_{11} for a substance with a resonator (the dashed curve in Fig. 1) also differs sharply from the analogous dependence without a resonator (the solid curve).

Dividing (20) by the absorption power, calculated from the formula $W_{14}^{\text{abs}} = B_{14}u_{41}(n_1-n_4)h\nu_{41}$ with account taken of (5), (8), and (10), we find the energy efficiency of generation

$$\gamma_{\mathrm{g}} = \frac{\nu_{32}}{\nu_{41}} \left(1 - \frac{u_{41}^{\mathrm{th}}}{u_{41}}\right) \times$$

$$\times \frac{p_{43}^0 p_{21}^0 (1-\delta_{23}) - [(p_{31}^0 + p_{32}^0)(p_{12}^0 + p_{42}^0) + p_{43}^0 p_{32}^0](1+\delta_{23}) - 2p_{21}^0 (p_{31}^0 + p_{32}^0)\delta_{23}}{(p_{41}^0 + p_{42}^0 + p_{43}^0)[p_{21}^0 (1-\delta_{23}) + p_{31}^0 (1+\delta_{23})]} \simeq$$

$$\simeq \frac{\nu_{32}}{\nu_{41}} \eta_{43} \left(1 - \frac{1}{x}\right). \tag{24}$$

It is remarkable that γ_{g} depends only on η_{43}, and not on the total quantum yield of luminescence of the transition $3 \to 2$.

The simplified expression for γ_{g} is valid if the third level is metastable ($p_{21}^0 \gg p_{31}^0 + p_{32}^0$). The energy efficiency of a three-level quantum generator is expressed by an analogous formula [4].

The formulas given make it possible to investigate the influence of the parameters of the generator substance and of the resonator on the processes of absorption and generation.

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CITED LITERATURE

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Note: Figure translations are in progress. See original paper for figures.