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# APPLICATION OF THE STATISTICAL METHOD TO CALCULATION OF THE OPTICAL PROPERTIES OF LASERS*

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THE statistical method of calculation was proposed by Einstein[1] as early as 1917. During the 40's and 50's it was used successfully to calculate the optical properties of matter within the framework of linear optics. The starting equations of this method have a definite limit of applicability and in general do not follow from the more general equations of quantum electrodynamics[2]. Nevertheless the method allows one to obtain the correct expressions for the rates of absorption and emission, for the laws governing the excitation and decay of luminescence, for the quantum yield of luminescence, and for a number of other optical properties[3-5]. However one cannot use the statistical method to solve problems related to the shape and displacement of energy levels, non-resonance interactions, etc.

It was shown[6] in a paper at the Thirteenth Conference on Spectroscopy that the statistical method of calculation can be successfully applied to the calculation of various nonlinear effects which arise when matter is irradiated by light of very great intensity or in systems with metastable energy levels. In such cases Bouger's law no longer holds, and the absorption coefficient depends on the radiation density and may become negative. Nonlinearity manifests itself also in the occurrence of stimulated dichroism, in the depolarization of luminescence, and in many other optical phenomena.

The fruitfulness of the statistical method has recently been reemphasized by the possibility of calculating the optical properties of nonlinear systems of a special type—optical masers. Using the statistical method and resonator theory, the basic properties of lasers have been successfully explained, and, in a series of important cases, quite good agreement with experiment has been obtained.

In using the probability method the energy levels and the transition probabilities between them play the role of initial conditions†. The basic problem is the determination of the individual level populations as a function of the illumination intensity and the subsequent calculation of the number of optical transitions. In lasers it is necessary to consider transitions caused both by the pump radiation and by the action of the laser light itself.

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†They can be obtained only by comparison of the results of the calculation with experiment.

## 1. ENERGY LEVEL POPULATIONS[7]

Consider a set of n particles, each of which has N energy levels. The particles interact with external radiation, with black body radiation, and with the surrounding medium. The probability for transitions between levels i and j is designated $p_{ij}$. Using the Einstein coefficients $A_{ij}$ and $B_{ij}$, which give the probabilities for spontaneous and stimulated optical transitions, the $p_{ij}$ can be written in the form

$$\left.\begin{aligned} p_{ij} &= A_{ij} + B_{ij}u^0_{ij} + d_{ij} + B_{ij}u_{ij} = p^0_{ij} + B_{ij}u_{ij}, \\ p_{ji} &= B_{ji}u^0_{ij} + d_{ji} + B_{ji}u_{ij} = p^0_{ji} + B_{ji}u_{ij}. \end{aligned}\right\} \quad (1)$$

Here $p^0_{ij}$ and $p^0_{ji}$ are the transition probabilities in thermodynamic equilibrium: the $B_{ij}u_{ij}$ are the probabilities for transitions stimulated by external radiation, assuming that there is no angular anisotropy in the excitation of the particles; the $d_{ij}$ and $d_{ji}$ are the probabilities for radiationless transitions, and $u^0_{ij}$ is the density of black body radiation at the frequency $\nu_{ij}$. The quantities (1) have the dimension $\sec^{-1}$ and vary from system to system within wide limits, usually from less than 1 to $10^{10}$ $\sec^{-1}$.

Under stationary illumination and for laser action without pulsations (spiking), the energy level populations do not vary with time. The number of particles leaving the i-th level in a given time interval equals the number of particles making transitions to the level in the same time. Hence the populations of the levels $n_k$ satisfy the following system of kinetic balance equations:

$$n_i \sum_{j=1}^{N} p_{ij} - \sum_{j=1}^{N} n_j p_{ji} = 0. \quad (2)$$

Since the total number of particles per unit volume is constant, we have

$$\sum_{j=1}^{N} n_j = n. \quad (3)$$

Hence only $N-1$ of the N equations of the system (2) are linearly independent.

Putting $n_N$ from (3) in (2) and dropping the N-th equation we have

$$\sum_{j=1}^{N-1} a_{ij} n_j = n p_{Ni}. \quad (4)$$

The following notation has been introduced

$$a_{ij} = p_{Ni} - p_{ji}, \quad a_{ii} = p_{Ni} + \sum_{j=1}^{N} p_{ij}. \quad (5)$$

As is well known, the solution of the system (4) has the form

$$n_j = n\frac{D_j}{D}, \tag{6}$$

where

$$D = \begin{vmatrix} a_{11} & a_{12} & \dots & a_{1,N-1} \\ a_{21} & a_{22} & \dots & a_{2,N-1} \\ \dots & \dots & \dots & \dots \\ a_{N-1,1} & a_{N-1,2} & \dots & a_{N-1,N-1} \end{vmatrix} \tag{7}$$

is the determinant of the system (4). The determinants $D_j$ for all j from 1 to $N-1$ are obtained from D by replacing the j-th column with the column formed from the coefficients $p_{Ni}$. The elements of the determinant $D_n$ are equal to $a_{ij} - p_{Ni}$, where i is the number of the row. According to (3) $\sum_{j=1}^{N} D_j = D$.

With the help of (5)—(7) it is easy to exhibit the explicit form of the particle distribution function for quantum mechanical systems with an arbitrary number of energy levels. In the simplest case of only two levels, $D = a_{11} = p_{21} + p_{12}$, $D_1 = p_{21}$, $D_2 = a_{11} - p_{12} = p_{12}$. For N = 3 we find from (5)—(7)

$$a_{11} = p_{31} + p_{12} + p_{13}, \qquad a_{12} = p_{31} - p_{21},$$
$$a_{21} = p_{32} - p_{12}, \qquad a_{22} = p_{32} + p_{21} + p_{23}$$

and hence

$$n_3 = \frac{n}{D}(p_{13}p_{21} + p_{13}p_{23} + p_{12}p_{23}), \tag{8}$$

$$n_2 = \frac{n}{D}(p_{32}p_{12} + p_{32}p_{13} + p_{31}p_{12}), \tag{9}$$

$$n_1 = \frac{n}{D}(p_{31}p_{21} + p_{31}p_{23} + p_{32}p_{21}), \tag{10}$$

where

$$D = p_{21}(p_{32} + p_{12} + p_{31}) + p_{12}(p_{32} + p_{23} + p_{31}) + p_{13}(p_{23} + p_{32}) + p_{23}p_{31}.$$

Similarly, for N = 4 we have

$$n_4 = \frac{n}{D}\{p_{12}[p_{24}(p_{31} + p_{32}) + p_{34}(p_{24} + p_{23})] + p_{13}[p_{34}(p_{21} + p_{23}) + p_{24}(p_{32} + p_{34})] + p_{14}[(p_{24} + p_{21})(p_{31} + p_{32} + p_{34}) + p_{23}(p_{34} + p_{31})]\}, \tag{11}$$

$$n_3 = \frac{n}{D}\{p_{12}[p_{43}(p_{23} + p_{24}) + p_{42}p_{23}] + p_{13}[p_{43}(p_{21} + p_{23} + p_{24}) + p_{23}p_{42}] + p_{14}[p_{43}(p_{21} + p_{23} + p_{24}) + p_{42}p_{23}] + p_{41}[p_{13}(p_{21} + p_{23} + p_{24}) + p_{12}p_{23}] + p_{21}p_{42}p_{13}\}, \tag{12}$$

$$n_2 = \frac{n}{D}\{p_{12}[p_{42}(p_{31} + p_{32} + p_{34}) + p_{43}p_{32}] + p_{13}[p_{42}(p_{32} + p_{34}) + p_{43}p_{32}] + p_{14}[p_{42}(p_{31} + p_{32} + p_{34}) + p_{43}p_{32}] + p_{41}[p_{13}p_{32} + p_{12}(p_{31} + p_{32} + p_{34})] + p_{31}p_{43}p_{12}\}, \tag{13}$$

$$n_1 = \frac{n}{D}\{p_{21}[p_{42}(p_{31} + p_{32} + p_{34}) + p_{43}p_{32}] + p_{31}[p_{43}(p_{21} + p_{23} + p_{24}) + p_{42}p_{23}] + p_{41}[(p_{24} + p_{21})(p_{31} + p_{32} + p_{34}) + p_{23}(p_{31} + p_{34})]\}, \tag{14}$$

where D is the sum of all terms inside the curly brackets.

A system with N energy levels can have no more than $N-1$ transition probabilities $p_{ij}$. If none of the $p_{ij}$ are equal to zero, the determinant D contains $N^{N-1}$ terms, each of which is a product of $N-1$ probabilities $p_{ij}$. Hence for large N the distribution function has in general a very complicated form. Therefore in treating systems with N greater than four it is necessary from the start to set equal to zero all those transition probabilities which do not play a significant role in the processes being studied.

It should be pointed out that the distribution of particles among the energy levels follows certain rules which hold for all systems; these rules can be investigated without writing out the determinants explicitly. In particular, if the external radiation of frequency $\nu_{ij}$ induces transitions between just one pair of levels $i \rightleftarrows j$, then all $D_j$ and D depend linearly on $u_{ij}$ and hence D may be written in the form [7]

$$D = D(u_{ij}=0) + \Delta_{ij}B_{ji}u_{ij} = D(u_{ij}=0)(1 + \alpha_{ij}u_{ij}). \tag{15}$$

Here $D(u_{ij} = 0)$ and $\Delta_{ij}$ do not depend on $u_{ij}$, and $\alpha_{ij}$ is a nonlinear parameter which occurs in all formulas of nonlinear optics. One may also show [7] that the populations of the interesting pair of levels may be expressed for the formulae

$$n_i = \frac{n_i(u_{ij}=0) + l_i u_{ij}}{1 + \alpha_{ij}u_{ij}}, \tag{16}$$

$$n_j = \frac{n_j(u_{ij}=0) + l_j u_{ij}}{1 + \alpha_{ij}u_{ij}}, \tag{17}$$

where $l_i g_j = l_j g_i$ ($g_i$ and $g_j$ are statistical weights). The quantities $l_i$ and $l_j$ do not depend on $u_{ij}$. The quantities $n_i(u_{ij} = 0)$ and $n_j(u_{ij} = 0)$ determine the populations of the levels in the absence of external exci excitation at the frequency $\nu_{ij}$.

It follows (16) and (17) that, for sufficiently large intensities of exciting light of frequency $\nu_{ij}$ and for $g_j = g_i$, the populations of the i-th and j-th levels will become equal and as $\alpha_{ij}u_{ij} \to \infty$ approach a common limit equal to $l_i/\alpha_{ij}$. If $n_i = n_j$ for $u_{ij} = 0$, they are equal for arbitrary $u_{ij}$.

## 2. POWER ABSORBED AND THE ABSORPTION COEFFICIENT

Considering stimulated emission as negative absorption [3-5] and using (16) and (17) we arrive at the following expression for the power absorbed from the external radiation:

$$W_{ji}^{\text{abs}} = (n_j B_{ji} - n_i B_{ij})u_{ij}h\nu_{ij} = \frac{n_j(u_{ij}=0) - n_i(u_{ij}=0)g_j/g_i}{1 + \alpha_{ij}u_{ij}} B_{ji}h\nu_{ij}, \tag{18}$$

where we have used the equality

$$g_i B_{ij} = g_j B_{ji}. \tag{19}$$

As can be seen from (18), the power absorbed can be either positive or negative. The sign of $W_{ji}^{abs}$ depends on the populations of the levels which obtained prior to excitation at frequency $\nu_{ij}$. If $g_i n_j$ $(u_{ij} = 0) > g_j n_i$ $(u_{ij} = 0)$, the power absorbed is positive. If the reverse inequality holds the absorption is negative for arbitary intensities of the incident radiation at frequency $\nu_{ij}$. It follows that excitation of the system at frequency $\nu_{ij}$ can never change the sign of the absorption at this same frequency.

It also follows from (18) that the dependences of the power absorbed on the intensity of the external radiation for media with either negative or positive absorption are given by the same formula.

Recalling that the integrated absorption coefficient is

$$k_{ij} = \frac{W_{ji}^{abs}}{\nu u_{ij}},$$

we find from (18)

$$k_{ji} = \frac{k_{ji}^0}{1 + \alpha_{ij} u_{ij}}. \tag{20}$$

Here

$$k_{ji}^0 = \left[ n_j (u_{ij} = 0) - n_i (u_{ij} = 0) \frac{g_j}{g_i} \right] B_{ji} h \nu_{ij} / v$$

is the absorption coefficient as $u_{ij} \to 0$ (or for the absence of a dependence for $k_{ji}$ on $u_{ij}$).

According to (20) the absorption coefficient can be either positive or negative; its sign however is completely determined by the sign of $k_{ji}^0$ and does not depend on the magnitude of $u_{ij}$. In general $k_{ji} \to 0$ when the energy density becomes infinite, since the quantity $u_{ij}$ occurs only in the denominator of expression (20). Equation (20), which has found wide application in the theory of laser oscillators and amplifiers, is valid for particles with an arbitrary number of levels, if two conditions are fulfilled: first, that there be no anisotropy in the angular distribution of excited molecules, and second, that the incident radiation at frequency $\nu_{ij}$ excite transitions only between levels i and j. In systems which do not satisfy the first condition, the dependence of $k_{ji}$ on $u_{ji}$ is more complicated. However in this case equation (20) gives a good approximation[8].

## 3. THE NONLINEARITY PARAMETERS

It is clear from the above formulae for the level populations and absorption coefficients that the nonlinearity parameters are the most important parameters characterizing the interaction of the particles with the incident radiation. All of the nonlinear effects which have been considered[5,8,9], and which involve excitation at a single frequency $\nu_{ij}$, are simple functions of the product $\alpha_{ij} u_{ij}$. Hence the study of nonlinear effects reduces essentially to a study of the parameters $\alpha_{ij}$. We will now consider the nonlinearity parameters of the simplest quantum-mechanical systems.

For particles with two energy levels $\alpha_{21}$ is given by

$$\alpha_{21} = \frac{B_{12}(1 + g_1/g_2)}{A_{21} + B_{12} + u_{21}^0 (1 + g_1/g_2) + d_{21} + d_{12}}. \tag{21}$$

According to (21) the product $\alpha_{ij} u_{ij} / (1 + g_1/g_2)$ does not exceed the ratio of the probability for stimulated transitions $B_{12} u_{21}$ to the probability $A_{21}$. In visible light and even more so in the ultraviolet, the probabilities for stimulated transitions are small compared to the probability for spontaneous transitions for all practically achievable densities of pump radiation (excluding laser light). Hence it is difficult to observe nonlinear effects in such a system.

When one goes to the infrared or further to the radio region of the spectrum, the ratio $B_{12} u_{21}^0 / A_{21}$ increases rapidly. In the far infrared the ratio is larger than unity even at room temperature. However since the incident radiation density is considerably larger than the thermal emission background, $B_{12} u_{21}^0 / A_{21} \gg 1$ and the laws of linear optics no longer hold. It is characteristic that the value of the nonlinearity parameter for a two-level system is not related simply to the absolute value of the transition probabilities or to the lifetime of the excited state $\tau$. If

$$A_{21} \gg d_{21} + d_{12} + 2B_{12} u_{21}^0,$$

then $\alpha_{21}$ is a simple function of the energy level separation and is completely independent of $\tau$;

$$\alpha_{21}^0 = \frac{c^3}{4\pi h \nu^3}. \tag{21a}$$

The thermal emission background and the radiationless transitions simply lead to a reduction in the nonlinearity parameter, $\alpha_{21} \le \alpha_{21}^0$.

A system of particles with three levels is characterized by three nonlinearity parameters:

$$\alpha_{31} = \frac{B_{13}\,[p_{21}(1 + g_1/g_2) + p_{23}(1 + g_1/g_3) + p_{32} + p_{12}]}{p_{21}(p_{32} + p_{31}^0) + p_{12}(p_{32} + p_{23} + p_{31}^0) + p_{23} p_{31}^0}, \tag{22}$$

$$\alpha_{21} = \frac{B_{12}\,[p_{32}(1 + g_1/g_2) + p_{31}(1 + g_1/g_2) + p_{13} + p_{23}]}{p_{21}^0(p_{32} + p_{13} + p_{31}) + p_{13}(p_{23} + p_{32}) + p_{23} p_{31}}, \tag{23}$$

$$\alpha_{32} = \frac{B_{23}\,[p_{12}(1 + g_2/g_3) + p_{13}(1 + g_2/g_3) + p_{21} + p_{31}]}{p_{21}(p_{32}^0 + p_{13} + p_{31}) + p_{12}(p_{32}^0 + p_{31}) + p_{13} p_{32}^0}. \tag{24}$$

These formulas are valid for simultaneous excitation of the particles by isotropic radiation at three frequencies.

We will apply Eqs. (22)—(24) to a numerical calculation of the nonlinearity parameters of a system of particles whose second level is metastable. Let the excitation occur in turn at frequencies $\nu_{31}$, $\nu_{21}$, and $\nu_{32}$, and let the transition probabilities be $p_{31}^0 = 3 \times 10^5$, $p_{21}^0 = 3 \times 10^2$, $p_{32}^0 = 10^8$ sec$^{-1}$, and $p_{13}^0 = p_{12}^0 = p_{23}^0 = 0$. Neglecting for simplicity the radiationless transitions and using the Einstein relation $A/B = 8\pi h \nu^3 / c^3$, for $\nu_{31} = 20{,}000$ cm$^{-1}$, $\nu_{21} = 14{,}000$ cm$^{-1}$, and $\nu_{32} = 6{,}000$ cm$^{-1}$ we find the following values for the $\alpha_{ij}$ (in cm$^3$/sec$^2$ erg): $\alpha_{31} = 7.8 \times 10^{14}$, $\alpha_{21} = 6.5 \times 10^{12}$, $\alpha_{32} = 4.5 \times 10^{16}$.

If the second level were absent or were not metastable, the value of $\alpha_{31}$ would be three orders of magnitude smaller. However, if the system has a metastable level the nonlinear effects in the transition $3 \rightarrow 1$ occur at excitation energy densities approximately $p_{21}^0/p_{31}^0$ times smaller than in a system of particles with two energy levels. The deviations from linearity at the transition $2 \rightarrow 1$ become significant at higher incident radiation densities. The large value at $\alpha_{32}$ is explained by the fact that the frequency $\nu_{32}$ is in the infrared.

The temperature dependence of the nonlinearity parameters of arbitrary systems is quite complicated. However, beginning at a certain temperature, the values of $\alpha_{ij}$ will decrease with increase in temperature and in the limit $(T \rightarrow \infty)$ go to zero[7]. The high temperature prevents the system from deviating from thermodynamic equilibrium and reduces all nonlinear effects.

In introducing the nonlinearity parameters it was assumed that radiation of a given frequency causes stimulated transitions between a single pair of levels only. Hence the determinants $D_j$ and $D$ were linear functions of the density of incident radiation on $u_{ij}$. It is possible, however, to have systems in which a single spectral line induces transitions between two, three, or more levels. In such cases the distribution functions, the absorption coefficients, and the other optical properties will depend not only on $u_{ij}$ but on $u_{ij}^2$, $u_{ij}^3$, etc. as well. Only the optical properties of one quantum mechanical system—the harmonic oscillator—are independent of the density of the exciting light[10,11].

A treatment of the above systems in this more general case is quite complicated. Hence it is advisable to make the calculation separately for each particular system, making use of simplifying factors.

## 4. THE LASER THRESHOLD

If matter with a negative absorption coefficient is placed in a resonator, e.g., between two plane parallel reflecting plates, oscillation will occur under certain conditions. The statistical method of calculation makes it possible to calculate rather simply such energetic properties as the threshold value of the pump radiation density, the rate of absorption of pump power, and the laser power.

Let the particles be excited by isotropic pump radiation at the frequency $\nu_{kl}$, and let laser action occur at one of the frequencies $\nu_{ij}$. For a stationary oscillation to exist it is necessary to fulfill the condition[12]

$$-k_{ji}(\nu_{las}) = k_{ji}^{loss}, \tag{25}$$

where $k_{ji}(\nu_{las})$ is the negative absorption coefficient, and $k_{ji}^{loss}$ is a coefficient describing the radiation losses in the cavity. Since the laser frequency usually is close to the peak of the line (a luminescence band) the absorption coefficient $k_{ji}(\nu_{las})$ may be calculated from the Kravetz integral by the formula

$$k_{ji}(\nu_{las}) = \frac{k_{ji}}{\Delta\nu_{ij}^{abs}}.$$

Here $\Delta\nu_{ij}^{abs}$ is the absorption line width at the laser transition.

The loss coefficient for a plane parallel cavity is related to the reflection coefficients of the coatings $r_{ij}$ and $r'_{ij}$, and to the thickness of the cavity, by the expression[13]

$$k_{ji}^{loss} = \frac{1}{l} \ln\left(\frac{1}{\sqrt{r_{ij} r'_{ij}}}\right) + \varrho_{ji}, \tag{26}$$

where $\rho_{ji}$ is a parameter giving the energy loss due to scattering and absorption by impurities. The first term in (26) is due to the output of energy at the ends of the cavity.

Using the fact that

$$k_{ji}(\nu) = (n_j B_{ji} - n_i B_{ij}) \frac{h\nu_{ij}}{v\Delta\nu_{ij}^{abs}},$$

the condition for stationary laser action (25) may be put in the form

$$n_i \frac{g_j}{g_i} - n_j = \delta_{ji} n, \tag{27}$$

where

$$\delta_{ji} = \frac{k_{ji}^{loss}(\nu_{las})\, v\Delta\nu_{ij}^{abs}}{nB_{ji} h\nu_{ij}} = \frac{k_{ji}^{loss}(\nu_{las})}{\varkappa_{ji}(\nu_{las})} \tag{28}$$

is a dimensionless quantity equal to the ratio of the loss coefficient to the absorption coefficient for $n_j = n$ $(\kappa_{ji}(\nu_l) = k_{ji}(\nu_l, n_j = n))$.

Equation (27), which expresses conservation of energy inside the cavity, leads to a number of general conclusions. It follows first that in the absence of external excitation, when

$$n_i^0 \frac{g_j}{g_i} - n_j^0 = -n_j^0[1 - \exp(-h\nu_{ij}kT)] < 0,$$

laser action is impossible even in the ideal cavity $(\delta_{ji} \rightarrow 0)$. In order to have laser action one must remove the system from the state of thermodynamic equilibrium. Two qualitatively different cases must be distinguished. In the first case the level j is the ground state, and in the second case it is an excited state.

If j is the ground state and if prior to excitation it contained $10^{17}$ to $10^{19}$ particles, then in order to have laser action at frequency $\nu_{ij}$ it is necessary to transfer a huge number of particles, comparable in order of magnitude to n, to the i-th level. At present it is feasible to do this for those media in which the i-th level is metastable and in which a build-up of excited particles occurs. A simple model of such media is a system of particles with three energy levels, the second of which is metastable.

If the j-th level is an excited state it will contain as small a number of particles as one wishes at low enough temperatures. Hence the preparation of inverted populations in the levels i and j, necessary for laser action, is possible without significant depletion of the number of particles in the ground state. In accord with the above distinction, one speaks of three-level and four-level lasers.

According to (27) the larger $g_j/g_i$ the easier it is to excite laser action. It further follows from (27) and (28) that in order to excite laser action it is necessary that the absorption line at $\nu_{ij}$ be sufficiently narrow ($\Delta\nu_{ij}^{abs}$ small) and sufficiently intense ($B_{ji}$ large). Decreasing the loss coefficient and in particular decreasing the transmission of the coatings, and increasing the concentration of particles facilitate laser action.

If we put for the density of laser emission $u_{ji} = 0$, we can use (27) to determine the threshold value of the pump radiation $u_{kl}^{thr}$. Using (6) and putting the determinants in the form

$$D_i(u_{ij}=0)=D_i^0+\Delta_i B_{kl}u_{kl}, \quad D_j(u_{ij}=0)=D_j^0+\Delta_j B_{kl}u_{kl},$$
$$D(u_{ij}=0)=D^0+\Delta_{kl}B_{kl}u_{kl},$$

we find from (27)

$$u_{kl}^{thr}=\frac{1}{B_{kl}}\frac{D_j^0-D_i g_j/g_i+\delta_{ji}D^0}{\Delta_i g_j/g_i-\Delta_j-\delta_{ji}\Delta_{kl}}. \tag{29}$$

It can be shown that $\Delta_{kl} \geq \Delta_i$ and hence (29) has a finite positive value only if

$$\delta_{ji}<\frac{g_j}{g_i}. \tag{30}$$

Systems which do not satisfy condition (30) will not oscillate even for infinitely large pump power.

Multiplying numerator and denominator in (29) by $n/D^0$ and making use of (6) we find the following simple expression for the threshold:

$$u_{kl}^{thr}=\frac{1}{nB_{kl}}\frac{n_j^0(1-e^{-h\nu_{ij}/kT})+\delta_{ji}n}{\alpha_i g_j/g_i-\alpha_j-\delta_{ji}\alpha_{kl}}, \tag{31}$$

where $\alpha_i$, $\alpha_j$ are positive parameters, and $\alpha_{kl} = \sum_{i=1}^{N}\alpha_i$ is the nonlinearity parameter for the transition $k \rightarrow l$. The number of particles in the j-th level in the absence of external excitation is denoted $n_j^0$.

In three level laser $n_j^0$ is nearly or exactly equal to n, and hence the threshold pump power is not zero even in the ideal resonator with $\delta_{ji} \rightarrow 0$. In four-level lasers the j-th level is an excited state and $n_j^0 \rightarrow 0$ if the temperature of the medium is sufficiently low. Hence for $\delta_{ji} \rightarrow 0$ the threshold $u_{kl}^{thr} \rightarrow 0$. In principle laser action in four-level systems is possible for very small pump powers.

As special cases of (31) one can derive the expressions for three and four level lasers which have been given in [14-17] and which will be considered below.

## 5. LASER POWER

In discussing stimulated transitions caused by external radiation one must keep in mind the fact that the probability Bu is an approximation to the integral $\int B(\nu)u(\nu)d\nu$. If a substance with narrow energy levels is irradiated by light with broad spectral width, $u(\nu)$ may be brought out from the integral sign and one may use the usual expression for the transition probability:

$$\int B(\nu)u(\nu)d\nu=u(\nu)\int B(\nu)d(\nu)=Bu(\nu).$$

The width of the laser line $\Delta\nu_{ij}^{las}$ is usually smaller than the width of the absorption line $\Delta\nu_{ij}^{abs}$, given by the function $B_{ji}(\nu)$. Hence

$$\int B_{ji}(\nu)u_{ij}(\nu)d\nu=B_{ji}(\nu_{las})\int u_{ij}(\nu)d\nu$$
$$=B_{ji}(\nu_{las})u(\nu_{las})\Delta\nu_{ij}^{las}.$$

Если частота генерации совпадает с максимумом линии поглощения, то

$$B_{ji}(\nu_{las})=\frac{B_{ji}}{\Delta\nu_{ij}^{abs}}.$$

Using this last remark and (20) and (25), we have

$$u_{ij}(\nu_{las})\Delta\nu_{ij}^{las}=-\frac{k_{ji}^0(\nu_{las})+k_{ji}^{loss}(\nu_{las})}{\alpha_{ji}(\nu_{las})k_{ji}^{loss}(\nu_{las})} \tag{32}$$

Expression (32) determines the laser radiation density inside the resonator and is a generalization of an analogous formula for the three level laser [12] to the case of a system with an arbitrary number of energy levels.

The laser power, or more precisely the amount of radiant energy emitted per unit volume of the active medium per second is equal to

$$W_{ij}^{las}=vk_{ji}^{loss}u(\nu_{las})\Delta\nu_{las}=\delta_{ji}nB_{ji}h\nu_{ij}u_{ij}(\nu_{las})\Delta\nu_{ij}^{las}/\Delta\nu_{ij}^{abs}$$
$$=v\frac{|k_{ji}^0(\nu_{las})|-k_{ji}^{abs}(\nu_{las})}{\alpha_{ji}(\nu_{las})}. \tag{33}$$

Using the explicit expression for $k_{ji}^0(\nu_{las})$ and making use of (28), we find after a simple transformation:

$$W_{ij}^{las}=\frac{n_i(u_{ij}=0)g_j/g_i-n_j(u_{ij}=0)-\delta_{ji}n}{\alpha_{ji}/B_{ji}}h\nu_{ij}. \tag{34}$$

Here $n_i(u_{ij}=0)$ and $n_j(u_{ij}=0)$ are as before the level populations for a given pump energy density at transition $k \rightarrow l$ and in the absence of laser emission, $u_{ij} = 0$.

Equation (34) allows one to calculate the laser power if one knows the properties of the resonator and the quantities characterizing the interaction of the active laser medium with the pump radiation in the absence of the resonator.

## 6. THREE-LEVEL LASERS[14,18-22]

We now consider in more detail the simplest laser system, having three levels (Fig. 1) with optical pumping of the transition $1 \to 3$. Real materials have additional levels whose influence, as a rule, is to increase the threshold and decrease the laser power. To elucidate the detailed properties of a three level laser we will examine the dependence of the level populations, the luminescence absorption, and the laser power on the pump radiation density both with and without reflecting coatings present. As before, laser action is assumed to be stationary and to occur at the transition $2 \to 1$. The second level is metastable ($p^0_{32} \gg p^0_{21} + p^0_{23}$).

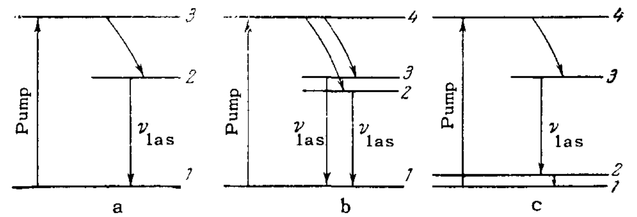


FIG. 1. Energy level diagrams for three level (a & b) and four-level (c) lasers.

If the particles are outside the resonator, they interact only with the pump radiation. Inside the resonator the process of laser action subjects the particles to the strong effects of the laser radiation. In general the level populations are determined by Eqs. (8)—(10), in which one puts $u_{21} = 0$ for the case of no resonator present; when the system is lasing one uses the value of $u_{21}$ calculated from (32).

The dependence of the level populations on pump power is shown graphically in Fig. 2a. The solid lines refer to the medium when outside the resonator; the dashed lines to the medium when inside the resonator. The values of $u_{31}$ are given in units of the threshold, $x = u_{31}/u^{thr}_{31}$. On this scale, $n_3$ is merged with the x axis.

When $u_{31}$ is very small the value of $n_1$ is essentaily constant and $n_2$ increases linearly with $u_{31}$. This is the region of linear optics. As the build-up of particles in the metastable level begins, $n_1$ begins to decrease and the growth of $n_2$ slows down. The solid curve $n_2(u_{31})$, referring to particles outside the resonator tends to an upper limit close to n. For $u_{31} \to \infty$ the populations of the first and third levels approach zero (more precisely, $np^0_{21}/p^0_{32}$).

Now let reflecting coatings be added. The graph of $n_i(u_{31})$ will then be given by the solid curves only up to the laser threshold. For $u_{31} > u^{thr}_{31}$ laser action occurs, and inside the resonator a high radiation density build up at $\nu_{21} = \nu_{las}$; this radiation induces transitions $2 \rightleftarrows 1$. As soon as laser action begins, and for further increases in pump power, the level populations satisfy the relation

$$n_2 \frac{g_1}{g_2} - n_1 = \delta_{12} n = \text{const}.$$

Since $n_3 \approx 0$, and $n_1 + n_2 \approx n$, it follows that the level populations are essentially independent of $u_{31}$ in the lasing regime. The increasing supply of particles to the metastable level with increasing $u_{31}$ is offset by more intense transitions $2 \to 1$ due to the increase in probability of stimulated transitions $B_{12}u_{21}$.

All this remains true as long as $n_2$ is negligibly small. In principle $B_{13}u_{31}$ may become comparable with $A_{31}$. At this point nonlinear behavior begins to appear in the transition $3 \to 1$. In the limit the number of particles in all levels becomes equal and all further changes in the populations cease. This nonlinearity is ordinarily not attained experimentally and hence our further investigations will relate only to the region adjoining the laser threshold, $0 \le x \le p^0_{31}/p^0_{21}$.

According to (29) and (8)—(10) the threshold value of pump power of a three level laser equals

$$u^{thr}_{31} = \frac{1}{B_{13}} \frac{[p^0_{31}(p^0_{21} + p^0_{23}) + p^0_{32}p^0_{21}](1 + \delta_{12})}{p^0_{32}g_1/g_2 - (p^0_{21} + p^0_{23})g_1/g_3 - \delta_{12}[(p^0_{21} + p_{23})(1 + g_1/g_2) + p^0_{32}]}. \quad (35)$$

In deriving (35) it was assumed that for frequencies in the visible region and for temperatures T between 0 and 300°K the probabilities $p^0_{12} = p^0_{13} = 0$.

Using the condition that the second level be meta-

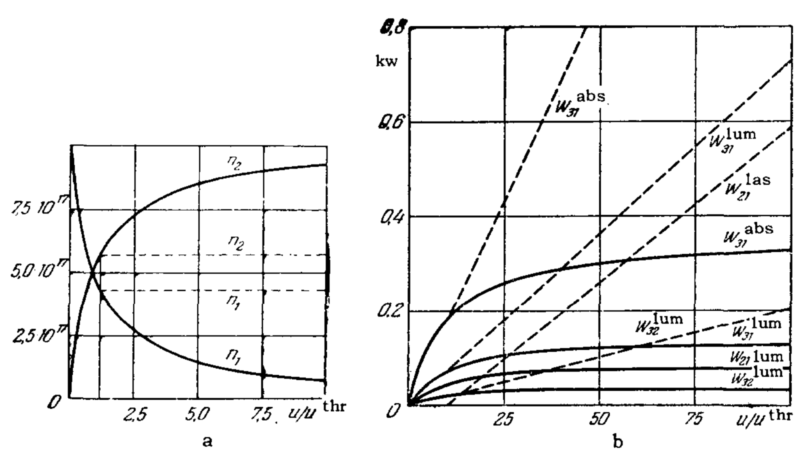


FIG. 2. Dependence of level populations (a), power absorbed, power in luminescence, and laser power (b) on pump radiation density. The dashed lines refer to the lasing regime, the solid lines to the absence of laser action.

stable ($p^0_{21} + p^0_{23} \ll p^0_{32}$) and putting $T = 0$ ($p^0_{23} = 0$), Eq. (35) can be greatly simplified:

$$u^{thr}_{31} = \frac{p^0_{21}}{\eta B_{13}} \frac{1+\delta_{12}}{g_1/g_2-\delta_{12}}, \tag{36}$$

where $\eta = p^0_{32}/(p^0_{32} + p^0_{31})$ is the ratio of the number of excited particles going from the labile to the metastable level to the total number of transitions out of the third level.

As the temperature is increased, $2 \to 3$ transitions begin to occur. The probability $p^0_{23} = p^0_{32} \exp(-h\nu_{32}/kT)$. According to (35) the value of $p_{23}$ begins to influence the threshold at temperatures for which $p^0_{23}$ becomes comparable with $p^0_{21}p^0_{32}/p^0_{31}$, that is

$$e^{-h\nu_{32}/kT} \sim \frac{p^0_{21}}{p^0_{31}}.$$

For $\nu_{32} = 6{,}000\ \mathrm{cm}^{-1}$, $p^0_{21} = 3 \times 10^2\ \mathrm{sec}^{-1}$, and $p^0_{31} = 3 \times 10^5\ \mathrm{sec}^{-1}$ the threshold increases by a factor of two as T increases from 0 to 2,000 K.

The experimentally observed temperature dependence of the threshold is related to the growth of $\Delta\nu^{abs}_{21}$ with increasing T[23] and, as will be shown below, to the presence of a splitting of the metastable level. The quantity $\Delta\nu^{abs}_{21}$ occurs in the expression for threshold via $\delta_{21} \sim \Delta\nu^{abs}_{21}$. If $\delta_{12} = 0$, the threshold is a minimum, $B_{13}u^{thr}_{31} = p^0_{21}g_2/g_1$. Laser action occurs when the probability for stimulated transitions $1 \to 3$ becomes equal to the probability for the outflow of particles from the metastable level multiplied by $g_2/g_1 \sim 1$. The magnitude of threshold is related only to the internal losses in the active medium (luminescence and non-radiative transitions $2 \to 1$ and $3 \to 1$). In high quality resonators, for which $\delta_{12} \ll (1 + g_1/g_2)$, the dependence of threshold on $\delta_{12}$ and hence on the line width $\Delta\nu^{abs}_{21}$ and on temperature disappears. Such dependence is observed only in resonators with low Q's where $\delta_{12}$ is comparable with $g_1/g_2$.

It also follows from (36) that the laser threshold is small for those media for which $B_{13}$ is large, i.e., for which the integrated absorption coefficient for pump radiation is large, for which $g_1/g_2 > 1$, and for which moreover the value $\eta = p^0_{32}/(p^0_{32} + p^0_{31})$ is close to one.

The dependence of threshold on $p^0_{21}$ or, equivalently, on $\tau_2 = 1/p^0_{21}$ requires special attention since $p^0_{21}$ enters the expression for $u^{thr}_{31}$ not only in the numerator but also via $\delta_{12}$ in the denominator. Putting the values $\delta_{12} = k^{abs}_{21}/\varkappa_{12}(\nu)$ and $p^0_{21} = A_{21} + d_{21}$ in (36) we obtain

$$u^{thr}_{31} = \frac{1}{B_{13}\eta} \frac{(A_{21}+d_{21})(1+k^{abs}_{12}/\varkappa_{12}(\nu_{las}))}{g_1/g_2 - k^{abs}_{12}/\varkappa_{12}(\nu_{las})}. \tag{37}$$

We assume further that the absorption band (line), whose maximum corresponds to the laser frequency, has no structure and hence that

$$\varkappa_{12}(\nu_{las}) = \frac{\varkappa_{12}}{\Delta\nu^{abs}_{21}} = \frac{nB_{12}h\nu_{21}}{v\Delta\nu^{abs}_{21}}.$$

Expressing $B_{12}$ in terms of $A_{21}$ we find from (37)

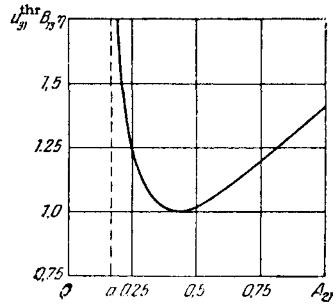


FIG. 3. Dependence of threshold on $A_{21}$.

$$u^{thr}_{31} = \frac{1}{B_{13}\eta} \frac{(A_{21}+d_{21})(A_{21}+ag_1/g_2)}{g_1/g_2\,(A_{21}-a)}, \tag{38}$$

where

$$a = 8\pi\nu^2\,\Delta\nu_{21}\frac{k^{abs}_{12}\,v}{n}, \tag{39}$$

and where $\nu$ and $\Delta\nu_{21}$ are expressed in $\mathrm{cm}^{-1}$.

Figure 3 shows a graph of the dependence of $u^{thr}_{31}B_{13}\eta$ on $A_{21}$ for $\nu = 20{,}000\ \mathrm{cm}^{-1}$, $n = 10^{19}\ \mathrm{cm}^{-3}$, $k^{loss}_{12} = 0.01\ \mathrm{cm}^{-1}$, $d = 0$, and $v = c/1.76$. It is clear from the figure that the left-hand portion of the graph is very steep, whereas to the right of the minimum the curve varies much more gradually and becomes a straight line. For $A_{21} = a$, the threshold goes to infinity and laser action is impossible. The value of $u^{thr}_{31}$ is a minimum when

$$A^{min}_{21} = a - \frac{d}{2} + \sqrt{\left(a - \frac{d}{2}\right)^2 + a^2\frac{g_1}{g_2} + ad} \tag{40}$$

or, if $d = 0$,

$$A^{min}_{21} = a\left(1 + \sqrt{1 + \frac{g_1}{g_2}}\right). \tag{40a}$$

The threshold is equal to

$$u^{thr}_{31\,min} = \frac{1}{B_{13}\eta}\frac{a}{g_1/g_2}\left(1 + \sqrt{1 + \frac{g_1}{g_2}}\right)^2. \tag{41}$$

For the previous values of the parameters and for $g_1 = g_2$, and for $\Delta\nu^{abs}_{21} = 1\ \mathrm{cm}^{-1}$ we find from (39)–(41) $A^{min}_{21} \approx 0.4\ \mathrm{sec}^{-1}$, $(B_{13}u^{thr}_{31})_{min} \sim 1\ \mathrm{sec}^{-1}$.

The expression for laser power (34) applicable to a three level laser takes the form

$$\begin{aligned} W^{las}_{21} = nh\nu_{21}\Big\{&p^0_{32}\frac{g_1}{g_2} - (p^0_{21} + p^0_{23})\frac{g_1}{g_3} \\ &- \delta_{21}\left[p^0_{32} + (p^0_{21} + p^0_{23})\left(1 + \frac{g_1}{g_3}\right)\right]\Big\} \\ &\times B_{13}(u_{31} - u^{abs}_{31})\left[(p^0_{31} + p^0_{32})\left(1 + \frac{g_1}{g_2}\right)\right. \\ &\left.+ p^0_{23} + B_{13}u_{31}\left(\frac{g_1}{g_3} + \frac{g_1}{g_2} + \frac{g_1^2}{g_1g_3}\right)\right]^{-1}. \end{aligned} \tag{42}$$

For ordinary pump powers the probability for stimulated transitions $B_{13}u_{31}$ is several orders of magnitude smaller than $p^0_{31}$ and may be neglected in comparison with the latter.

Furthermore, making use of the condition for metastability we have

$$W_{21}^{\mathrm{las}} = \eta n h\nu_{21} \frac{g_1/g_2 - \delta_{12}}{1 + g_1/g_2} B_{13}\left(u_{31} - u_{31}^{\mathrm{thr}}\right). \tag{42a}$$

The laser power is proportional to the Einstein coefficient $B_{13}$, determining the area of the absorption band at the pump frequency, and increases linearly with $u_{31}$. The maximum laser power occurs for $\eta = 1$, i.e., in the absence of luminescence and radiationless transitions $3 \rightarrow 1$. The smaller $\delta$ (i.e., the smaller the value of both $k_{12}^{\mathrm{loss}}$ and the absorption line width $\Delta\nu_{21}^{\mathrm{abs}}$ and the larger $n$ and the Einstein coefficient $B_{12}$) the larger the laser power.

It is clear from (42a) that the threshold alone is insufficient to characterize the tendency of a material to intense laser action. Along with the threshold one must know the tangent of the straight line $W_{12}^{\mathrm{las}}(u_{31})$, which is given by

$$\tan\varphi = \frac{W_{21}^{\mathrm{las}}}{u_{31} - u_{31}^{\mathrm{thr}}} = \eta n B_{13} h\nu_{21} \frac{g_1/g_2 - \delta_{21}}{1 + g_1/g_2}. \tag{43}$$

The parameters $u_{31}^{\mathrm{thr}}$ and $\tan\varphi$ determine the lasing behavior of a medium for a given pump power. One may find systems having a small threshold accompanied by a small value of $\tan\varphi$.

Lowering $g_1/g_2$ increases the laser threshold and simultaneously decreases $\tan\varphi$. The smaller $g_1/g_2$ the greater the requirements on the resonator in which laser action will be possible for a given material. In the general case the pump power absorption is given by

$$\begin{aligned} W_{13}^{\mathrm{abs}} &= (n_1 B_{13} - n_3 B_{31}) u_{31} h\nu_{31} = \left(n_1 - n_3 \frac{g_1}{g_3}\right) B_{13} u_{31} h\nu_{31} \\ &= \frac{n B_{13} u_{31} h\nu_{31}}{D} \left[ p_{31}^0 p_{21}^0 + p_{31}^0 p_{23}^0 + p_{32}^0 p_{21}^0 \right. \\ &\quad \left. + B_{12} u_{21} \left( p_{31}^0 \frac{g_1}{g_2} + p_{32}^0 \frac{g_1}{g_2} - p_{23}^0 \frac{g_1}{g_3} \right) \right]. \end{aligned} \tag{44}$$

If we put $u_{21} = 0$, then (44) will give the radiation absorbed by the medium in the absence of laser action. Inserting the value of $u_{21}$ from (32) in (44) we obtain an expression for the absorption power in the lasing regime as a function of the pump power and all other parameters describing the system of particles and the resonator. Taking account of the metastability of the second level and putting $T = 0$ this expression takes the form

$$W_{13}^{\mathrm{abs}} = \frac{n}{1 + g_1/g_2} B_{13} u_{31} h\nu_{31} \left( \frac{g_1}{g_2} - \delta_{21} \right). \tag{45}$$

Dividing (42a) by (45) we obtain the laser efficiency (energy yield)

$$\gamma_{\mathrm{las}} = \eta \frac{\nu_{21}}{\nu_{31}} \left( 1 - \frac{u_{31}^{\mathrm{thr}}}{u_{31}} \right) = \eta \frac{\nu_{21}}{\nu_{31}} \left( 1 - \frac{1}{x} \right), \tag{46}$$

where $x$ is the pump power in units of the threshold. The value of $\gamma_{\mathrm{las}}$ increases with increasing pump power. In the limiting case $\eta \rightarrow 1$, $x \gg 1$, all of the pump energy (with the exception of Stokes' losses at transition $3 \rightarrow 2$) goes into the laser output.

Graphs of the dependence of the absorption power, the laser output and the luminescence ($W_{ij}^{\mathrm{lum}} = nA_{ij}h\nu_{ij}$) on the density of pump radiation are shown in Fig. 2b. The solid curves refer to the case of no resonator. These curves exhibit a tendency to saturation which manifests itself in the breakdown of the linear absorption and in a nonlinear dependence of $W_{ij}^{\mathrm{lum}}$ on $u_{31}$. The introduction of coatings leads to a build-up of radiation at frequency $\nu$ inside the cavity and to the preponderance of stimulated transitions $2 \rightarrow 1$ over spontaneous transitions; this strong radiation at $\nu$ causes a sharp change in all optical properties of the medium (cf. the dashed lines).

The appearance of significant probabilities of the stimulated transitions $2 \rightleftarrows 1$ is equivalent to the removal of the metastability of the second level. An intense exchange of particles takes place between the first and second levels and may proceed much more rapidly than any other process in the system. Since at this point the numbers of particles $n_1$ and $n_2$ cease to depend on the pump power (cf. Fig. 2a), the system reverts to having a linear dependence of all its optical properties on pump power. The power absorbed increases linearly with $u_{31}$. The luminescence power $W_{31}^{\mathrm{lum}}$ is also proportional to $u_{31}$, which is due to the linear dependence of $n_3$ on pump power.* The quantity $W_{21}^{\mathrm{lum}}$ remains constant with increasing $u_{31}$ since $n_2 \approx$ const.

An analysis of the curves in Fig. 2b shows how the distribution of energy among the various transitions changes with the introduction of reflecting coatings and the consequent occurrence of laser action.

We assume that the particles outside the resonator are subject to the effects of a powerful pump $u_{31} \gg u_{31}^{\mathrm{thr}}$. In this case the values of all of the properties under study are given by a point on the corresponding solid curves. We then assume that reflecting coatings are introduced and that after a certain interval a stationary state is attained. The transition from the first case to the second is accompanied by a decrease in the luminescence power in the transition $2 \rightarrow 1$. However the onset of laser action is connected not with a decrease in the luminescence and other losses but rather with a sharp step-like increase in the amount of energy absorbed. Part of this additional absorbed energy is inevitably lost to the growth of luminescence and thermal losses in transitions $3 \rightarrow 1$ and $3 \rightarrow 2$, and the remaining part goes into the laser radiation.

The laser action which occurs with the introduction of reflecting coating is related not to an increase in the number of active molecules $n_2 - n_1$, as is often stated in popular articles, but rather to a decrease in $n_2$ and an increase in $n_1$. It is precisely this increase

*The value of $n_3$ is several orders of magnitude smaller than $n_1$ and $n_2$ and hence the linear increase of $n_4$ with increasing $u_{31}$ has practically no effect on the magnitude of $n_1$ and $n_2$.

in $n_1$ which leads to the increase in the power absorbed and to the occurrence of laser action. An increase in the pump power under lasing conditions does not increase the number of active particles, which remains constant, but rather leads only to an increase in the power absorbed.

## 7. THE EFFECT OF A SPLITTING OF THE METASTABLE LEVEL[17]

It is well known that in a real system such as a ruby the metastable level is split and a more rigorous solution of the problem must take account of the presence of two metastable levels (Fig. 1b). In the stationary state the populations of the levels of such a system are determined by Eqs. (11)—(14). If we put in the expressions for $n_i$ $u_{31} = u_{21} = u_{32} = 0$, they will give the energy level populations as functions of the pump radiation density $u_{41}$ in the absence of laser action. Since the second and third levels are metastable, that is since

$$p^0_{31} + p^0_{21} \ll p^0_{42},\ p^0_{43}, \tag{47}$$

it is essentially only the populations of these levels which increase with increasing $u_{41}$. For sufficiently large pump powers the sum $n_2 + n_3$ is nearly equal to the total number of particles $n$.

It follows from (12) and (13) that $n_2 > n_3$ if the following inequality is satisfied:

$$p^0_{32}(p^0_{42} + p^0_{43}) + p^0_{42}p^0_{31} > p^0_{13}(p^0_{42} + p^0_{43}) + p^0_{43}p^0_{21}, \tag{48}$$

If the reverse inequality holds, we have $n_2 < n_3$ for arbitrary values of the pump power. In cavities of equal Q at the $R_1$ and $R_2$ lines (frequencies $\nu_{21}$ and $\nu_{31}$ respectively), the laser will operate on the $R_1$ line in the first case and on the $R_2$ line in the second.

In ruby lasers as a rule one observes stimulated emission at the $R_1$ line. We will treat this case first. Restricting the temperature interval to be from 0 to 300°K for the present system we can put

$$p^0_{14} = p^0_{13} = p^0_{12} = p^0_{24} = p^0_{34} = 0, \qquad p^0_{23} = p^0_{32}\exp(-h\nu_{32}/kT), \tag{49}$$

$$u_{42} = u_{43} = 0, \qquad B_{14}u_{41} \ll A_{41}. \tag{50}$$

This means that thermal excitation is absent in all transitions except $2 \rightarrow 3$. The inequality (50) will be violated only for pump powers $10^3$—$10^5$ times larger than $u^{\text{thr}}_{41}$.

The threshold value of the pump power $u^{\text{thr}}_{41}$ ($\nu_{\text{las}} = \nu_{21}$) leading to laser action at frequency $\nu_{21}$ can be found easily from (29) and (11)—(14) with the help of (49) and (50). It has the form

$$u^{\text{thr}}_{41}(\nu_{\text{las}} = \nu_{21}) = \frac{1}{B_{14}}\frac{D^0(1+\delta_{12})}{\Delta_2 g_1/g_2 - \Delta_1 - \delta_{12}\Delta_{14}}, \tag{51}$$

where

$$D^0 = (p^0_{41} + p^0_{42} + p^0_{43})(p^0_{32}p^0_{21} + p^0_{31}p^0_{23} + p^0_{21}p^0_{31}),$$
$$\Delta_2 = p^0_{32}(p^0_{42} + p^0_{43}) + p^0_{42}p^0_{31}, \quad \Delta_1 = p^0_{21}p^0_{31} + p^0_{21}p^0_{32} + p^0_{23}p^0_{31},$$
$$\Delta_{14} = (p^0_{43} + p^0_{42})(p^0_{23} + p^0_{32}) + p^0_{43}p^0_{21} + p^0_{42}p^0_{31} + 2(p^0_{21}p^0_{31} + p^0_{21}p^0_{32} + p^0_{31}p^0_{23}).$$

Making use of the metastability of the second level and putting $p^0_{32} \gg p^0_{21} + p^0_{21}$ it is not difficult to obtain

$$u^{\text{thr}}_{41}(\nu_{\text{las}} = \nu_{21}) = \frac{1}{\eta' B_{14}}\frac{p^0_{21}(1+\delta_{12})}{g_1/g'_2 - \delta_{12}}, \quad kT \ll h\nu_{32}, \tag{52}$$

$$u^{\text{thr}}_{41}(\nu_{\text{las}} = \nu_{21}) = \frac{1}{\eta' B_{14}}\frac{(p^0_{21} + p^0_{31})(1-\delta_{12})}{g_1/g'_2 - 2\delta_{12}}, \quad kT \gg h\nu_{32}, \tag{52a}$$

where $\eta' = (p^0_{42} + p^0_{43})/(p^0_{41} + p^0_{42} + p^0_{43})$. Expression (52) formally coincides with (36), which was obtained without taking account of the splitting of the metastable level. There is numerical agreement in the magnitudes of the thresholds if $\eta = \eta'$ and if in (36) we set $g_2$ equal to the statistical weight of the lower sublevel of the metastable state $g_2 = g'_2$. If in real systems the metastable level can either be split or not, $g_2$ gives the degeneracy of the unsplit level where $g'_2$ gives that of its lower component. In this case for $kT \ll h\nu_{32}$ the splitting of the metastable level causes a decrease in the laser threshold. Laser action becomes possible in resonators with lower Q, i.e., for $g_1/g_2 < \delta_{12} < g_1/g'_2$.

The presence of the second component of the metastable level causes an increase in laser threshold only for temperatures $kT \geq h\nu_{32}$, where the deactivation of excited particles is determined not only by the probability $p^0_{21}$ but also by $p^0_{31}$. Equations (12) and (12a) are valid only for $p^0_{32} \gg p^0_{21} + p^0_{31}$. In the opposite case the exchange of particles between the second and third levels is hindered and an increase in temperature does not change the laser threshold. Calculations also show[17] that a splitting of the metastable level causes a decrease in laser power only when $\exp(-h\nu_{32}/kT)$ is comparable with unity.

The presence of laser radiation inside the cavity causes a drastic chante in the dependence of the level populations on the pump radiation density. In (11)—(14) we must take account of the transition probability $B_{12}u_{21}$ due to the laser radiation ($p_{21} = p^0_{21} + B_{12}u_{21}$ and $p_{12} = B_{12}u_{21}$). An analysis of the distribution function appropriate during laser action at frequency $\nu_{21}$ shows that an increase in the pump power causes a gradual growth of $n_3$ and a decrease in $n_2$ and $n_1$. The changes in $n_i$ are most significant when the probabilities $p^0_{23}$ and $p^0_{32}$ are small. If however $p^0_{32}$ and $p^0_{23}$ considerably exceed $p^0_{43}$ and $p^0_{42}$, then according to (11)—(14), the values of $n_i$ remain essentially constant for wide variations of $u_{41}$.

Increasing the pump power when laser action is occurring at frequency $\nu_{21}$ may cause a population inversion sufficient to satisfy the condition for laser action $n_3 - n_1 = \delta_{13}n$ for the transition $3 \rightarrow 1$ without perturbing the conditions for oscillation at frequency $\nu_{21}$.

Putting first for simplicity $\delta_{12} = 0$ and $g_2 = g_1$, we obtain from this equation with the aid of (12) and (14), with $u_{32} = u_{31} = 0$ and $u_{21} \neq 0$, the second laser threshold, i.e., the value of the pump radiation density at which the frequency $\nu_{32}$ is generated simultaneously with the frequency $\nu_{21}$:

$$u_{41}^{*\,\mathrm{thr}}(\nu) = \frac{1}{B_{14}} \frac{(p_{41}^0+p_{42}^0+p_{43}^0)\,[p_{32}^0+p_{31}^0-p_{23}^0+\delta_{13}(2p_{31}^0+2p_{32}^0+p_{23}^0)]}{p_{43}^0+p_{23}^0-p_{31}^0-p_{32}^0-\delta_{13}(3p_{31}^0+3p_{32}^0+p_{43}^0+p_{23}^0)}. \quad (53)$$

According to (51) and (53) the values of the first and second laser thresholds are of the same order of magnitude in systems in which $p_{32}^0 + p_{31}^0 \approx p_{21}^0$, $p_{43}^0 \geq p_{42}^0 \geq p_{41}^0$ if $\delta_{13}$ is sufficiently small. Conversely if $p_{32}^0/p_{21}^0 \gg 1$, the second threshold is increased with respect to the first by the same factor. In the latter case it is simple to take account of the dependence of the second threshold on $\delta_{12}$;

$$u_{41}^{*\,\mathrm{thr}}(\nu) = \frac{1}{B_{14}} \times \frac{(p_{42}^0+p_{43}^0)\,[(1+2\delta_{13}+\delta_{12})-(1-\delta_{13}+\delta_{12})\exp(-h\nu_{32}/kT)]}{[p_{43}^0(1-\delta_{13}-\delta_{12})/p_{32}^0-(1+3\delta_{13}-\delta_{12})+(1-\delta_{13}+3\delta_{12})\exp(-h\nu_{32}/kT)]}. \quad (54)$$

In deriving (54) use was made of the metastability of the second and third levels and of the inequality $p_{32}^0 - p_{23}^0 \gg p_{21}^0 + p_{31}^0$.

It follows from (54) that the second threshold decreases with increasing temperature and with decreasing $\delta_{13}$. For low temperatures the denominator of (53) becomes negative if $p_{32}^0 > p_{43}^0$. This means that once laser action has begun at frequency $\nu_{21}$ it is impossible to excite laser action at the transition $3 \to 1$ in systems in which the probability for the redistribution of particles between the components of the metastable level $p_{23}^0$ is larger than $p_{43}^0$.

We assume now that laser action occurs only at the frequency $\nu_{31}$. In this case the value of the threshold is

$$u_{14}^{\mathrm{thr}}(\nu_{\mathrm{las}} = \nu_{31}) = \frac{1}{B_{14}} \frac{D^0(1-\delta_{12})}{\Delta_3 g_1/g_3 - \Delta_1 - \delta_{13}\Delta_{14}}, \quad (55)$$
$$\Delta_3 = p_{23}^0(p_{42}^0+p_{43}^0)+p_{43}^0 p_{21}^0.$$

The expressions for $D^0$, $\Delta_1$ and $\Delta_{14}$ were introduced previously.

Depending on the actual values of the transition probabilities $\delta_{12}$ and $\delta_{13}$, $u_{41}^{\mathrm{thr}}$ $(\nu_{\mathrm{las}} = \nu_{31})$ can be either larger or smaller than the threshold for laser action at frequency $\nu_{21}$ determined by (51). In the special case that $\delta_{12} = \delta_{13}$, laser action occurs at the transition $2 \to 1$ if the inequality (48) holds, and at the transitions $3 \to 1$ if the opposite inequality holds.

In systems for which (48) is valid laser action can be produced at frequency $\nu_{31}$ only by increasing $\delta_{12}$ and decreasing $\delta_{13}$, which means in particular increasing the transmission of the coatings at frequency $\nu_{21}$ and at the same time increasing the reflectivity at frequency $\nu_{31}$. Laser action at the $R_2$ line in ruby was obtained in this way [24,25]

Thus in lasers with a splitting of the metastable level we can have laser action at either of the frequencies $\nu_{21}$ or $\nu_{31}$. If the probability for redistribution of particles between the components of the metastable level is small compared with the probabilities for populating the metastable state, it is possible to have simultaneous laser action at both frequencies $\nu_{21}$ and $\nu_{31}$ at pump powers comparable with the initial threshold.

## 8. FOUR-LEVEL LASERS [15]

The simplest level scheme for a laser of this type is shown in Fig. 1c. The pump raises particles to the fourth level, from which they make transitions to the third, metastable, level. Population inversion occurs initially at the transition $3 \to 2$, and for larger $u_{41}$ at transition $3 \to 1$. For the scheme shown we can put

$$p_{13}^0 = p_{14}^0 = p_{23}^0 = p_{24}^0 = p_{34}^0 = 0, \qquad u_{31} = u_{21} = u_{42} = 0, \quad (56)$$
$$p_{12}^0 = p_{21}^0 \exp(-h\nu_{21}/kT), \qquad B_{14}u_{41} \ll A_{41}. \quad (57)$$

This means that all thermal transitions upwards except for $1 \to 2$ are negligibly small. The inequality (57) is valid for all practically obtainable values of $u_{41}$.

The equations for the level populations (11)–(14) may be simplified with the help of (56) and (57):

$$n_4 = \frac{n}{D}\{B_{14}u_{41}[p_{21}^0(p_{31}^0+p_{32}^0)+B_{32}u_{32}p_{21}^0+B_{23}u_{32}p_{31}]\}, \quad (58)$$

$$n_3 = \frac{n}{D}\{B_{14}u_{41}p_{43}^0p_{21}^0 + B_{23}u_{32}[p_{12}^0(p_{43}^0+p_{42}^0+p_{41}^0) + B_{14}u_{41}(p_{41}^0+p_{43}^0+p_{12}^0)]\}, \quad (59)$$

$$n_2 = \frac{n}{D}\{(p_{41}^0+p_{42}^0+p_{43}^0+B_{14}u_{41})[p_{12}^0(p_{31}^0+p_{32}^0)+B_{32}u_{32}p_{12}^0] + B_{14}u_{41}[(p_{42}^0+p_{43}^0)(p_{32}^0+B_{32}u_{32})+p_{42}^0p_{31}^0]\}, \quad (60)$$

$$n_1 = \frac{n}{D}\{(p_{41}^0+p_{42}^0+p_{43}^0+B_{14}u_{41})[p_{21}^0(p_{31}^0+p_{32}^0) + B_{32}u_{32}p_{21}^0+B_{23}u_{32}p_{31}]\}, \quad (61)$$

where D is the sum of all terms in the curly brackets in equations (58)–(61).

If we put $u_{32} = 0$ in (58)–(61), these equations give the dependence of $n_i$ on the pump radiation density $u_{41}$ in the absence of a resonator and hence in the absence of laser action. They also describe the level populations of particles placed in a resonator for values of $u_{41}$ between 0 and $u_{41}^{\mathrm{thr}}$, which characterize the beginning of laser action.

In the absence of excitation, only the first and second levels are populated:

$$n_1^0 = n\frac{p_{21}^0}{p_{21}^0+p_{12}}, \qquad n_2^0 = n\frac{p_{12}^0}{p_{21}^0+p_{12}}, \qquad n_3^0 = n_4^0 = 0. \quad (62)$$

When one turns on the excitation and gradually increases $u_{41}$, the population of the first level begins to decrease and the populations of all excited levels begin to increase. The value of $n_4$, as in the three-level scheme, is many orders of magnitude smaller than n, since the majority of the excited particles end up in the third level. The curve of $n_3(u_{41})$ intersects the graph $n_2(u_{41})$ if for small T one has

$$p_{21}^0 > p_{32}^0 + (p_{32}^0+p_{31}^0)\frac{p_{42}^0}{p_{43}^0}. \quad (63)$$

In this case one may easily satisfy the condition $n_3 - n_2 = n\delta_{23}$ and obtain laser action. If condition (63) is not satisfied, $n_2 > n_3$ and laser action at frequency $\nu_{32}$ is impossible.

Using (29) and (58)—(60) we obtain the following expression for the laser threshold:

$$u_{41}^{\text{thr}} = \frac{1}{\eta_1 B_{14}} \frac{(p_{31}^0 + p_{32}^0)\left[(1+\delta_{23})\exp(-h\nu_{32}/kT) + \delta_{23}\right]}{g_2/g_3 - \delta_{23} - p_{32}^0(1+\delta)/p_{21}^0 - c_1(1+\delta_{23}) - c_2\delta_{23}}, \quad (64)$$

where

$$\eta_1 = p_{43}^0/(p_{41}^0 + p_{42}^0 + p_{43}^0), \qquad c_1 = (p_{31}^0 + p_{32}^0)(p_{12}^0 + p_{42}^0)/p_{21}^0 p_{43}^0,$$
$$c_2 = 2(p_{31}^0 + p_{32}^0)/p_{43}^0.$$

The coefficients $c_1$ and $c_2$ are $\ll 1$, and may be omitted in (64) if the following conditions hold

$$p_{43}^0 \gg p_{42}^0, \qquad p_{21}^0 \gg p_{31}^0 + p_{32}^0, \quad (65)$$
$$p_{43}^0 \gg p_{31}^0 + p_{32}^0 \quad \text{or} \quad p_{43}^0 \gg p_{21}^0. \quad (66)$$

In the contrary case the threshold may be large. Violation of condition (66) causes a significant increase in $u_{41}^{\text{thr}}$ only for large $\delta_{23}$, i.e., for large external losses or for a small value of $\kappa_{23}(\nu_{\text{las}})$. The first of conditions (66) can be called the condition of metastability. It follows from the above that the metastability of the third level is by no means necessary in the four level scheme.

At low temperatures, where $\exp(-h\nu_{32}/kT) \to 0$ and $\delta_{23} \to 0$, the laser threshold is zero. This of course is the fundamental advantage of a four-level laser.

The level scheme shown in Fig. 1c (with the third level being metastable) may function as an ordinary three level laser at the transition $3 \to 1$. The only special feature of such a system is that the laser threshold at frequency $\nu_{31}$ will be a sensitive function of temperature because of the thermal interaction of the neighboring levels 1 and 2. Neglecting small terms of order $(p_{31}^0 + p_{32}^0)/p_{43}^0$ we find

$$u_{41}^{\text{thr}}(\nu_{\text{las}} = \nu_{31}) = \frac{1}{\eta_1 B_{14}} \frac{(p_{31}^0 + p_{32}^0)\left[(1+\delta_{13}) + \delta_{13}\exp(-h\nu_{21}/kT)\right]}{g_1/g_3 - \delta_{13}}. \quad (67)$$

In the same approximation the ratio of the thresholds (64) and (67) is equal to

$$\frac{u_{41}^{\text{thr}}(\nu_{\text{las}} = \nu_{32})}{u_{41}^{\text{thr}}(\nu_{\text{las}} = \nu_{31})} = \frac{g_1/g_3 - \delta_{13}}{g_2/g_3 - \delta_{23}} \frac{(1+\delta_{23})\exp(-h\nu_{21}/kT) + \delta_{23}}{(1+\delta_{23}) + \exp(-h\nu_{21}/kT)\delta_{23}}. \quad (68)$$

If one has a laser with $g_1 = g_2$ and $\delta_{23} = \delta_{13}$, the magnitude of (68) varies from $\delta_{23}/(1+\delta_{23})$ to 1 as the temperature is increased from 0 to $kT \gg h\nu_{21}$. In this case laser action occurs first at frequency $\nu_{32}$. If however $g_1 > g_2$ or $\delta_{23} > \delta_{13}$, then at low temperatures

$$u_{41}^{\text{thr}}(\nu_{\text{las}} = \nu_{23}) < u_{41}^{\text{thr}}(\nu_{\text{las}} = \nu_{31}),$$

Moreover, the inequality reverses as T increases. Hence in the process of operating the laser at frequency $\nu_{32}$, heating of the active medium may interrupt laser action at frequency $\nu_{32}$ and stimulated emission may occur at frequency $\nu_{31}$. The interruption of laser action may also be due to the fact that in the course of laser operation at frequency $\nu_{32}$ the number of particles in the second level may become larger than at the first.

The exact formula for the laser power may be obtained easily from (34), (58)—(61), but is not very convenient. After the usual transformations it may be simplified:

$$W_{32}^{\text{las}} = n\frac{\left[(1 - p_{32}^0/p_{21}^0) - \delta_{23}(1 + p_{32}^0/p_{21}^0)\right] B_{14}(u_{41} - u_{41}^{\text{thr}})h\nu_{32}}{\left[1 + p_{31}^0/p_{21}^0 + 2\exp(-h\nu_{21}/kT)\right]/\eta_1 + 2B_{14}u_{41}/p_{21}^0}, \quad (69)$$

where we have put $g_2 = g_3$ for simplicity. If $B_{14}u_{41} \ll p_{21}^0$, as is usually the case, the laser power $W_{32}^{\text{las}}$ of the four-level laser depends linearly on the pump power (just as the three-level laser does). Saturation is reached only for large $B_{14}u_{41} \sim p_{21}^0$.

In the limiting case of an ideal four-level laser one may put $p_{32}^0 \ll p_{21}^0$, $B_{14}u_{41} \ll p_{21}^0$, $\eta_1 \to 1$. One then has the maximum possible power

$$W_{32}^{\text{las}} = \frac{nh\nu_{32}(1-\delta_{23})}{1 + 2\exp(-h\nu_{21}/kT)} \times \left\{B_{14}u_{41} - \frac{(p_{31}^0 + p_{32}^0)\left[\delta_{23} + (1+\delta_{23})\exp(-h\nu_{21}/kT)\right]}{1-\delta_{23}}\right\}. \quad (70)$$

According to (70), when $kT \ll h\nu_{32}$, $\delta_{23} \to 0$, the efficiency of the laser is very high $W_{32}^{\text{las}} \to nB_{14}u_{41}h\nu_{32}$. Except for Stokes' losses at transitions $4 \to 3$ and $2 \to 1$, all absorbed energy reappears as stimulated emission.

## 9. CONCLUSION

The optical properties of three and four-level lasers obtained above apply to the case of unit volume of the working substance, stationary pump power, and laser action without spiking. Experimentally however one observes laser action in finite volumes with non-uniform distribution of pump radiation, and a part of the active medium by be occupied by trapped modes which are not coupled out of the resonator. The phenomenon of laser action is greatly complicated by the pulsations which are observed in all solid and liquid lasers. If one adds to this the fact that the transition probabilities and frequently the pump power also are unknown, the difficulty of making quantitative comparisons of the results of calculations with experiment becomes understandable.

Despite this there is already good qualitative agreement between experimental results and the conclusions obtained within the framework of the statistical method of calculation. Experiment confirms the linear dependence of the absorption and laser power on the pump radiation density [26,27], the increase of threshold and the cessation of laser action with increase in temperature [28], and a number of other results obtained by this method.

It is clear that the statistical method has great usefulness in the construction of a complete theory of optical masers.

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