COHERENT DIVISION OF QUANTA

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An analysis is carried out of the features of amplification in a quantum-mechanical system where the energy spectrum has a band bounded above and composed of a large number of effectively equidistant levels. The conditions for stationary amplification are considered in the limiting cases of low and high intensity of the amplified radiation.

In this paper we consider a typical example of a problem in which the same frequency corresponds to transitions between a large number of different pairs of energy levels of a quantum-mechanical system. Consider, for instance, the principle involved in an efficient frequency divider, i.e., a physical device achieving a considerable reduction in frequency while preserving a large fraction of the power of the incoming radiation. The essential element of such a device—the amplifying medium—is a quantum-mechanical system whose energy spectrum contains a band close to the ground state and bounded on the upper side, in which induced transitions are important only for a frequency close to some fixed value Ω. This band may be either of the "line" type, i.e., consist of effectively equidistant levels, or it may be continuous; in the latter case the frequency Ω is determined by the selection rules for radiative transitions in the medium, and also by the selective conditions for accumulation of photons in the resonator. If now we bring about population inversion in this band by means of intense radiation of frequency Ω_0, this will lead to generation of power at the frequency Ω.

As a qualitative guide, let us consider the simplest possible example—a quantum oscillator with weak anharmonicity which increases with increasing energy (cf. also[1]). To start with we may confine ourselves to the approximation that part of the energy levels form a band of equidistant levels (Fig. 1):

$$E_n - E_{n-1} = h\Omega, \qquad n' \leqslant n \leqslant n'', \qquad n'' \gg n', \qquad (1)$$

while at the same time the spacings of the levels outside this band, $h\Omega_n = E_n - E_{n-1}$, are appreciably different from $h\Omega$:

$$|\Omega - \Omega_n| > 2\Delta\Omega_n, \qquad n < n' \quad \text{or} \quad n > n'', \qquad (2)$$

where $\Delta\Omega_n$ is the spectral width of the spontaneous

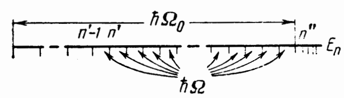

FIG. 1.

transition $E_n \to E_{n-1}$.

The photon produced in a given transition $E_n \to E_{n-1}$ for $n' \leq n \leq n''$ may induce in the band described by (1) new radiative transitions between any pair of neighbouring levels. So if we can create population inversion in the levels of this band, e.g., by means of optical irradiation at a frequency $\Omega_0 = E_{n''} - E_{n'}/\hbar$ and suitable relations between the transition probabilities, then, radiation of frequency Ω will be amplified in the medium described by this oscillator pattern of levels. By placing such a medium in a sufficiently high-Q resonator, we can obtain the required quantum generator[1)], which will effect the frequency transformation $\Omega_0 \to \Omega$. If now the contribution of nonradiative transitions is sufficiently small, the energy of input irradiation is transformed into a coherent signal at frequency Ω with quantum efficiency $\sim n'' - n' \gg 1$ and with comparatively high power efficiency, of order $1 - E'_n/E''_n$.

The system of equations for the populations N_n of the energy levels of the medium has the form

$$\frac{dN_n}{dt} = \sum_{m \neq n} [(a_{m,n}N_m - a_{n,m}N_n) + G_{m,n}(N_m - N_n)], \qquad (3)$$

1)As in the more commonly considered case of a one-quantum laser ($n'' - n' = 1$) the many-quantum generator (amplifier) considered here does not require specifically optical irradiation. In principle amplification in a medium may be effected by other types of energy input; in that case practically all the considerations advanced here remain valid.

where $\alpha_{m,n}dt$ is the probability of a spontaneous transition in time dt from the level E_m to the level E_n; $G_{m,n}dt$ is the probability of an induced transition, which differs appreciably from zero only for neighboring levels ($|m-n| = 1$) of the band (1), and also (in view of the intensive irradiation and the oscillator anharmonicity) for the transition $E_1 \leftrightarrow E''_n$.

Notice that the system (3) satisfies the condition

$$\sum_n N_n = \text{const}$$

or

$$\sum_n \sum_{m \neq n} [(\alpha_{m,n} N_m - \alpha_{n,m} N_n) + G_{m,n}(N_m - N_n)] = 0, \quad (3')$$

i.e., the matrix of the coefficients is degenerate.

For the various n these equations take the following form:

$$\frac{dN_n}{dt} = \sum_{m \neq n} (\alpha_{m,n} N_m - \alpha_{n,m} N_n) + G_{n+1,n}(N_{n+1} - N_n) + G_{n,n-1}(N_{n-1} - N_n), \quad n' \leqslant n < n'',$$

$$\frac{dN_n}{dt} = \sum_{m \neq n} (\alpha_{m,n} N_m - \alpha_{n,m} N_n), \quad n < n' - 1 \text{ or } n > n'',$$

$$\frac{dN_1}{dt} = \sum_{m > 1} (\alpha_{m,1} N_m - \alpha_{1,m} N_1) + G_{n'',1}(N_{n''} - N_1),$$

$$\frac{dN_{n'-1}}{dt} = \sum_{m \neq n'-1} (\alpha_{m,n'-1} N_m - \alpha_{n'-1,m} N_{n'-1}) + G_{n',n'-1}(N_{n'} - N_{n'-1}),$$

$$\frac{dN_{n''}}{dt} = \sum_{m \neq n''} (\alpha_{m,n''} N_m - \alpha_{n'',m} N_{n''}) + G_{n'',n''-1}(N_{n''-1} - N_{n''}) + G_{n'',1}(N_1 - N_{n''}). \quad (3'')$$

In lowest order we have for the electronic dipole transitions

$$G_{n,n-1} = n\Gamma J, \quad \Gamma = 2\pi^2 e^2 / 3\hbar mc\Omega\Delta\Omega, \quad (4)$$

where m and e are respectively the mass and charge of the electron; J is the intensity of radiation of frequency Ω; $\Delta\Omega$ is the spectral line width of the transition $E_n \rightarrow E_{n-1}$, which we take to be identical for all the levels of the band (1). The analogous formula for $G''_{n,1}$ has the form $G''_{n,1} = \gamma I$, where I is the intensity of irradiation at frequency Ω_0.

The amplification coefficient per centimeter path length of the photons in the medium, $\kappa = \kappa(\Omega)$, is defined by the formulae:

$$\frac{dJ}{ds} = \varkappa J, \quad \varkappa = \hbar\Omega P\Gamma,$$

$$P = (n''+1)N_{n''} - N' - n'N_{n'-1}, \quad N' = \sum_{m=n'}^{n''} N_m. \quad (5)$$

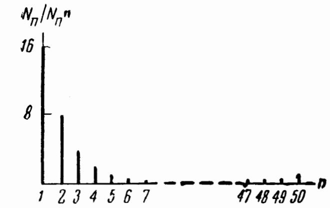

FIG. 2.

We shall say that the band (1) is "actively populated" if the inequality $P > 0$ is satisfied; for $P < 0$ the population is "passive." If conditions (1) and (2) are met (with $\Delta\Omega_n = \Delta\Omega$, $n' \leq n \leq n''$) a medium with an actively populated equally-spaced band will amplify radiation of frequency Ω.

Two of the simplest ways of obtaining active population may be noted. First, the inequality $P > 0$ is valid if within the band (1) the level population is a non-decreasing function of the energy of the level (inverted population of the band):

$$N_n \geqslant N_{n-1} \ (n' \leqslant n \leqslant n''), \qquad N_{n''} > N_{n-1'}. \quad (6)$$

An alternative condition, considerably easier to satisfy, is the following one connecting the average population of the levels of the band (1)

$$\bar{N} = N' / (n'' - n' + 1)$$

with the populations of the levels at its edge:

$$N_{n''} = \bar{N} + p, \quad N_{n'} \leqslant \bar{N} + n''p / n', \ p > 0. \quad (6')$$

In the case considered, where the spectral line width is the same for all lines of the band (1), the parameter P is determined primarily by the upper levels, owing to the increase with n of the probability of radiative transitions [Eq. (4)]. An example will clarify this point; suppose

$$N_n = \mathcal{N}(q^{n-n'-3} + q^{n''-n}), \quad 0 < q < 1, \quad q^{(n''-n')/2} \ll 1.$$

Then

$$N_{n''} \approx \mathcal{N}, \quad N_{n'-1} \approx N_{n''} q^{-4}, \quad N' \approx \frac{1 + q^{-3}}{1 - q},$$

$$P \approx N_{n''}\left(n'' + 1 - \frac{1 + q^{-3}}{1 - q} - n'q^{-4}\right).$$

If we put, for example, $n' = 2$, $n'' = 50$, $q = 1/2$, we get $P \approx N''_n > 0$, so that the population scheme of the equally-spaced band illustrated in Fig. 2 has an "active" character in spite of the normal population of the low levels.

When the temperature of the medium is low, sufficiently strong pulsed irradiation with monochromatic light will always lead to pulsed activity in the population of the band (1). However, whether or not stationary negative absorption at a given frequency under conditions of constant irradiation is possible, is determined by the relations between the probabilities for induced and spontaneous transitions (radiative or non-radiative) within the band (1), the distribution of spontaneous transition probabilities throughout the band, and also the rate of outflow of energy from its lower edge. A general analysis of the stationary solution ($dN_n/dt = 0$, $n = 1, 2, 3, \ldots$) of the system (3) is very difficult, and the resultant expressions for N_n are complex and hard to visualize. We shall consider only the simplest limiting cases, and assume that the mean energy of the particles of the medium is low and the probability of non-radiative electron transitions to higher energy levels negligibly small; we shall also assume that the only spontaneous transitions of importance in the band (1) are those between neighboring levels ($E_n \to E_{n-1}$) and those to levels below the lower edge of the band ($E_n \to E_m$, $m < n' - 1$).

First, suppose the relation between induced and spontaneous transitions is disadvantageous from the point fo view considered here, i.e., from the point of view of the efficiency of the frequency divider; then the intensity of radiation J in the medium at the resonance frequency is sufficiently small so that

$$n''\Gamma J \ll r_n, \quad n' \leqslant n \leqslant n'';$$

$$r_n = a_n + s_n, \quad a_n = a_{n,n-1}, \quad s_n = \sum_{m=1}^{n'-2} a_{n,m}.$$

In this limiting case we may neglect in zeroth approximation the effect of the induced transitions on the level population; then from (3″) we have

$$a_n N_n = r_{n-1} N_{n-1}, \quad n' \leqslant n \leqslant n''. \tag{7}$$

We can write the solution of (7) in the form

$$N_n = N' D^{-1} D_n, \quad D = \sum_{n=n'}^{n''} D_n, \quad D_n = \prod_{m=n+1}^{n''} \frac{a_m}{r_{m-1}},$$

$$D_{n''} = 1, \quad n' - 1 \leqslant n \leqslant n'' - 1. \tag{7'}$$

From the equation

$$\Delta N_n = N_n - N_{n-1} = N_n b_n / r_{n-1}, \; b_n = r_{n-1} - a_n \tag{7''}$$

it follows that if the parameter b_n in the band (1) is positive, the criterion (6) for active population of an equally-spaced band is fulfilled, and a medium of this kind will amplify radiation of frequency Ω. If on the other hand $b_n < 0 (n' \leq n \leq n'')$ we have passive population, and radiation of the resonance frequency is absorbed. Finally, if the parameter b_n changes sign within the band (1), analysis of the amplifying properties for weak intensities of the resonance radiation requires a more detailed theory.

Let us now consider the case when the resonance radiation passing through the medium is so intense that the probability of spontaneous transitions in the band (1) is small compared to the probability of induced transitions. We introduce a small parameter $\lambda (0 < \lambda \ll 1)$ and use (3′), which expresses the fact that the matrix of the system (3) is degenerate; in the case considered this takes the form

$$\sum_{m=2}^{n''} a_{m1} N_m = \sum_{m=n'-1}^{n''} s_n N_n.$$

Then we can rewrite (3) in the new notation in the form

$$(n+1) N_{n+1} + n N_{n-1} - (2n+1) N_n = \lambda [R_n N_n - A_{n+1} N_{n+1}],$$

$$n' \leqslant n < n'', \tag{8}$$

$$n' (N_{n'} - N_{n'-1}) = \lambda [s_{n'-1} N_{n'-1} - A_{n'} N_{n'}], \tag{8'}$$

$$n'' (N_{n''} - N_{n''-1}) = \lambda \Big[\sum_{m=n'-1}^{n''-1} s_m N_m - A_{n''} N_{n''} \Big]; \tag{8''}$$

$$\frac{s_n}{\Gamma J} = \lambda S_n, \quad \frac{a_n}{\Gamma J} = \lambda A_n, \quad \frac{b_n}{\Gamma J} = \lambda B_n, \quad \frac{r_n}{\Gamma J} = \lambda R_n.$$

The solution of the system of equations (8), (8′), and (8″) to zeroth order in λ,

$$N_n^{(0)} \approx N_{n''}^{(0)}, \quad n' - 1 \leqslant n \leqslant n'' - 1$$

implies that

$$P^{(0)} = (n'' + 1) N_{n''}^{(0)}$$

$$- (n'' - n' + 1) N_{n''}^{(0)} - n' N_{n''}^{(0)} = 0.$$

Thus in order to analyze the amplifying properties of the medium at high intensities of the resonance field it is not sufficient to restrict oneself to the zeroth approximation in λ.

Putting

$$N_n = N_{n''} (1 + \lambda H_n), \quad H_{n''} = 0,$$

we can write down the equations for the first approximation in λ:[2)]

$$(n+1) H_{n+1} + n H_{n-1} - (2n+1) H_n = B_{n+1},$$

$$n' \leqslant n \leqslant n'' - 1, \tag{9}$$

$$H_{n''-1} = \frac{1}{n''} \Big(A_{n''} - \sum_{m=n'-1}^{n''-1} s_m \Big). \tag{9'}$$

The solution of the system of equations (9) and (9′) can be put in the compact form

$$H_n = \sum_{m=n+1}^{n''} \frac{1}{m} \Big[\sum_{p=m+1}^{n''} B_p + n'' H_{n''-1} \Big] = - \sum_{m=n+1}^{n''} \frac{1}{m} \sum_{p=n'}^{m} B_p.$$

2)The conditions (8′) and (8″) are equivalent in view of the degeneracy of the system of equations (8), (8′) and (8″).

Then we have:

$$P = -\lambda N_{n''}\left(\sum_{n=n'}^{n''-1} H_n + n'H_{n'-1}\right)$$

$$= \frac{N_{n''}}{\Gamma J}\sum_{n=n'}^{n''}(n''-n+1)\,b_n. \qquad (9'')$$

These results show that for sufficiently large values of the intensity J the modulus of the linear amplification coefficient κ decreases as 1/J. This result, which is also well-known for single-quantum media (the "saturation effect") means that the dependence of the radiation intensity in the medium on the coordinate is no longer exponential (as for small J where we have $J_S = J_0 e^{\kappa S}$) but linear:

$$J_S = J_p(1+ks),\ k = \varkappa J = \text{const.}$$

At high intensities the most important factor is the relation between the spontaneous transitions into the low levels of the equally-spaced band.

According to (7″), for small J the degree of population inversion ΔN_m of the m-th level is determined by the value of the parameter b_n for n = m alone; for large J it is determined by the mean value of this parameter for all levels from n′ to m:

$$\Delta N_m = N_m - N_{m-1} = \frac{N_{n''}}{m\Gamma J}\sum_{p=n'}^{m} b_p.$$

If the sign of b_n is the same throughout the band (1) the active or passive character of the population is the same for small and for large values of J. If however b_n changes sign, the situation is quite different; by a suitable choice of the functions α_n and s_n we may ensure either amplification of the resonance radiation for small and absorption for large J, or absorption for small and amplification for large J. The latter possibility is of some interest; it should be possible in principle to construct a many-quantum medium to work under "selective conditions of amplification" i.e., such that resonance radiation is absorbed for intensities less than some threshold value and amplified to any appreciable extent only in some definite region of values of J (cf. also[2]).

The above analysis of amplification in a many-quantum system is of course based on a very simplified model. One of our assumptions—that the spontaneous radiation width $\Delta\Omega_n$ is independent of the position of the level in the band (1)—could certainly be removed with advantage when we come to make a more detailed consideration of concrete physical models. Another important assumption made above is that the edges of the equally-spaced band are sharply defined; since in general the anharmonicity increases with energy, this assumption is less justified for the lower edge than for the upper one. The "blurring" of the lower edge leads to a gradually decreasing participation by the lower levels in the induced transitions; in many cases this may make it easier to construct an amplifying medium.

We shall just indicate a few examples of physical media which possess the properties of interest to us in this work. One example is a gas composed of diatomic molecules excited in a non-equilibrium manner; the equally-spaced band in the spectrum is formed by the vibrational levels. Or consider the current carriers of a semiconductor in a magnetic field; in this case the anharmonicity with respect to the cyclotron transitions is provided by the energy-dependence of the carrier effective mass in the conduction band.[3] As a final example, the energy spectrum of the electrons in a solid in a strong electric field will also possess the required band of equally spaced levels.[4]

Each of the above examples only partially conforms to the model which we have used in this paper to simplify the discussion of the properties of a many-quantum maser. In each of the media considered, the formation of active population in the equally-spaced band is beset by characteristic difficulties. In particular, the probability of spontaneous transitions within the band increases, as a rule, with increasing energy of the level. Thus, for example, if we wish to analyze the possibility of effecting steady amplification by means of the vibrational transitions of a molecule of type A, it is convenient to consider a gaseous mixture of molecules of types A and B. The molecule B is chosen so that the transition frequencies between its low vibrational levels are close to the vibrational frequency corresponding to transitions near the lower edge of the equally-spaced band in the spectrum of the molecule A used for the amplification.

If now the concentration of molecules of type B is very much larger than that of type A, the lifetime against spontaneous transitions of the low levels of the equally-spaced band of molecule A may be considerably reduced compared to the lifetime of higher levels. On the other hand we must remember that too large a relative concentration of molecules of type B will lead to an increase of absorption at the frequency intended for amplification. It is desirable to choose the molecule A so that the anharmonicity of its vibrational levels is as small as possible; this will increase the number of levels we can use for a given value of the linewidth. In this case there is a difference from the model used above, in that the population of the upper levels of the equally-spaced band must be produced by excitation of the appropriate electronic levels or by effecting a high degree of non-equilibrium dissociation in the molecules of type A. A complete investigation of this problem is impossible at present because of the inadequacy of the experimental data.

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