Resonance effects during the interaction of a two-level system with intense polychromatic radiation

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(Submitted 1 June 1983)
Zh. Eksp. Teor. Fiz. **86**, 423-433 (February 1984)

An analysis is given of the interaction between a two-level system and an electromagnetic field with an equidistant spectrum and arbitrary and independent phases of the spectral components (multimode laser radiation). The concept of generalized Rabi frequencies is introduced for a two-level system in a polychromatic field, and the values of these frequencies are calculated. It is shown that the dependence of the harmonics of the population difference and of the absorption coefficients for the individual modes on the intermode interval and field strength exhibits extrema (resonances), the positions of which do not depend on the mode phases. The resonances occur when the combination frequencies of mode beats are equal to the generalized Rabi frequencies.

§1. INTRODUCTION

The classical problem of the interaction of a two-level quantum-mechanical system with a strong resonance electromagnetic field is now the subject of a voluminous literature. Two lines of approach can be identified in the development of the theory.

One of them originates in spectroscopy, and is concerned with the quantum-mechanical system in a given field. The case of a strictly monochromatic field has been treated most thoroughly (see, for example, the review[1]). In particular, it has been established that the fluorescence spectrum of a quantum-mechanical system placed in a strong monochromatic field acquires additional extrema that are shifted relative to the central extremum by the so-called Rabi frequency (this is the dynamic Stark effect).

Papers devoted to interaction with the biharmonic field have investigated in detail the situation where one of the fields (the probing field) is weak and the other strong.[2,3] Apart from the fact that the absorption profile of the probing field exhibits additional extrema (resonances), the susceptibility can change sign at certain frequencies and the medium will not absorb but, on the contrary, will amplify the weak field. The last step in this direction was made in Refs. 4 and 5, in which the two fields acting on the quantum-mechanical system were allowed to be strong. The shape of the absorption profile of the retuned field then becomes very complicated: a whole series of additional extrema appears and was, in fact, seen experimentally prior to these calculations.[6]

The problem arises whether a qualitatively similar picture of resonance effects will arise during the interaction between a two-level atom and a strong polychromatic field. This situation is interesting because powerful lasers usually generate multimode radiation. It would seem, at first sight, that the two-mode situation constitutes a special case because the phase relation between the modes becomes important when the number of modes is greater than two. In addition, it is known that absorption resonances that are observed for a weak probing field when a two-level system is exposed to a strong noise field, become broadened when the spectral width of the noise exceeds the Rabi frequency.[7-9] It must be remembered, however, that the model of structureless noise is more representative of a thermal source than a laser.[7,10] Moreover, the fact that the correlation function of a polychromatic field with equidistant spectrum and arbitrary component phases is periodic may give rise to a number of specific effects.[11] Nevertheless, the question of the absorption resonances has not, in fact, been investigated directly.[1)]

The other line of approach to this problem has been confined to laser theory. Here, the characteristic formulation is that of the self-consistent problem, where neither the state of the medium nor the radiation field is given, and the analysis is directed toward establishing the connection between nonlinear mode interaction and the dynamic nature of the laser. The problem is exceedingly complicated and only certain special cases have so far been examined, for example, the effects of combination tones on the generation spectrum,[14] the stability of stationary generation in the two-mode laser,[15,16] and certain aspects of the theory of mode locking.[17]

The two lines of approach, the spectroscopic and the laser-theoretic, cross one another in intracavity laser spectroscopy. Here, we draw attention to a number of experimental publications devoted to the anomalous increase in the spectral density of radiation near strong absorption lines of gases in the interior of the laser cavity.[18,19] The development of a satisfactory theory of this effect and certain other problems in laser physics and nonlinear spectroscopy require a preliminary analysis of the interaction between the two-level system and a given strong polychromatic field.

This analysis is given in the present paper for the case of an equidistant mode spectrum and homogeneous broadening of an undisturbed spectral line of the medium. Particular attention is devoted to the conditions for the existence of resonances in the mode absorption coefficients and to their position. Generally speaking, one would expect that the positions of the resonances would be random for arbitrary independent mode phases. However, it will be shown below that there is a group of resonances whose positions are not dependent on the random mode phases.

 0038-5646/84/020245-07$04.00

§2. CHARACTERISTIC EQUATION

The well-known equations describing a two-level system in a given electromagnetic field will be written in the truncated form:[17]

$$dy/d\tau=-i\Gamma\Delta_0 y+\Gamma(nx-y), \qquad (2.1a)$$

$$dn/d\tau=\alpha-n-(x^*y+xy^*)/2, \qquad (2.1b)$$

where we have introduced the following dimensionless variables: the time $\tau = t/T_1$, $x = \mathscr{E}/\mathscr{E}_{\rm sat}$, the complex field amplitude divided by the amplitude of the saturating field ($\mathscr{E}_{\rm sat} = \hbar/\mu(T_1,T_2)^{1/2}$, the normalized population difference $n = N/\overline{N}$, the unsaturated population difference $\alpha = N_0/\overline{N}$, the normalized complex polarization amplitude of the medium $y = \mathscr{P}/\overline{\mathscr{P}}$, $\Gamma = T_1/T_2$, and $\Delta_0 = T_2(\omega_{21} - \omega_0)$. It is important to note that the normalizing factors are related by $\overline{N}/\overline{\mathscr{P}} = 2i/\mu\Gamma^{1/2}$ where μ is the matrix element of the transition dipole moment, T_1 and T_2 are the population and polarization relaxation times, ω_{21} is the transition frequency, and ω_0 is the field carrier frequency.

We shall assume that the field can be represented by a set of equidistant modes

$$x=\sum_{k=0}^{r} x_k \exp(-i\Gamma k\Delta\tau), \qquad (2.2)$$

where $\Delta = T_2\Delta\omega$ is the dimensionless mode spacing. To be specific, the modes in (2.2) are labeled in the order of increasing frequency. The total number of modes is $r+1$ and the total spectrum width $r\Delta$ is small in comparison with the carrier frequency. The mode phases are assumed arbitrary; and independent, which is typical for the radiation emitted by the multimode laser. For this type of field, the polarization excited by it will also contain an equidistant set of components:

$$y=\sum_{m=-\infty}^{\infty} y_m \exp(-i\Gamma m\Delta\tau), \qquad (2.3)$$

but the number of these components is greater than that in the exciting field. This is so because of oscillations in the population difference, which can also be represented in the form of a series:

$$n=\sum_{l=-\infty}^{\infty} n_l \exp(-i\Gamma l\Delta\tau), \qquad n_{-l}=n_l^*. \qquad (2.4)$$

We shall seek the steady-state solution of (2.1) and, with this in mind, we substitute (2.2)–(2.4) in (2.1a), and obtain the following relation from the harmonic balance condition:

$$y_m=\sum_{k=0}^{r}\frac{n_{m-k}x_k}{1+i(\Delta_0-m\Delta)}. \qquad (2.5)$$

Equations (2.2)–(2.5) are now, in turn, substituted in (2.1b), and the result is

$$C_0^l n_l=-\sum_{j=-r,\, j\neq 0}^{r} C_j^l n_{l+j}+\alpha\delta_{l,0}, \qquad l=0,\pm1,\dots \,. \qquad (2.6)$$

The coefficients of this expansion are given by

$$C_0^l=\frac{1}{2}\sum_{k=0}^{r}X_k^2\left[\frac{1}{1+i[\Delta_0-(l+k)\Delta]}+\frac{1}{1-i[\Delta_0+(l-k)\Delta]}\right]-i\Gamma l\Delta+1, \qquad X_k^2=|x_k|^2, \qquad (2.7)$$

$$C_j^l=\frac{1}{2}\left[\sum_{k=\max(j,0)}^{\min(r,r+j)}\frac{x_k^* x_{k-j}}{1+i[\Delta_0-(l+k)\Delta]}+\sum_{k=\max(-j,0)}^{\min(r,r-j)}\frac{x_k x_{k+j}^*}{1-i[\Delta_0+(l-k)\Delta]}\right], \quad j\neq 0, \qquad (2.8)$$

and satisfy the following symmetry relations:

$$C_{-j}^{-l}=(C_j^l)^*, \quad C_0^{-l}=(C_0^l)^*. \qquad (2.9)$$

After solving (2.6), we can use (2.5) to calculate the polarization coefficients and, hence, the mode absorption coefficients:[5,12]

$$K_m=K\alpha^{-1}\mu\,\mathrm{Re}\,(y_m/x_m), \qquad (2.10)$$

where K is the absorption coefficient for the probing field at the line center.

The solution (2.6) was found in Ref. 5 in the form of a continued fraction for the case $r = 1$ (two modes). It can be shown that the exact solution has the form of a matrix continued fraction when the number of modes is arbitrary. It is capable of yielding exhaustive information on the absorption coefficients for the individual modes as functions of intensity and of the parameters of the mode spectrum. However, for the resonances whose position is independent of the modes phases, the situation is much simpler: to determine the positions and the necessary conditions for the existence of such resonance, it is sufficient to investigate the equation $C_0^l = 0$. It is clear from (2.7) that it is precisely in the neighborhood of the value $C_0^l = 0$ that the diagonal coefficient C_0^l in (2.6) can experience a sharp relative change because two large terms in the imaginary part of C_0^l cancel out under these conditions: one of them is $-i\Gamma l\Delta$, and the other is the imaginary part of the sum in (2.7). When the real part of C_0^l is small in comparison with each of the above terms in the imaginary part (this occurs when the mode intensities X_k^2 are sufficiently high and $\Gamma l\Delta \gg 1$), the quantity $|C_0^l|$ will vary rapidly with Δ or X_k^2 and will pass through a deep minimum near $C_0^l = 0$. All the other coefficients C_j^l will not change very much in this region, and the reason for this in the case of the off-diagonal coefficients is that the mode phases are random. According to (2.6), the extrema of the amplitudes of the population-difference harmonics are connected with this result and, by virtue of (2.5) and (2.10), so are the extrema of the mode absorption coefficients.

Let us write the detuning of the mode with $k = 0$ from the transition frequency in the form $\Delta_0 = (r_0 + \rho)\Delta$, where r_0 is the "coarse" tuning parameter equal to the whole-number part of the ratio Δ_0/Δ and the "fine" tuning parameter is $\rho(|\rho| \leqslant 1/2)$. Rewriting $C_0^l = 0$ in terms of these quantities, we obtain the following characteristic equation:

$$\frac{1}{2}\sum_{k=0}^{r} X_k^2\left[\frac{1}{1+i[r_0+\rho-l-k]\Delta}+\frac{1}{1-i[r_0+\rho+l-k]\Delta}\right]$$

$$-i\Gamma l\Delta+1=0. \quad (2.11)$$

To begin with, we shall assume that the parameters r, r_0, ρ and X_k^2 are fixed[2] and will investigate the values of the mode spacing for which there is a resonance in the population-difference harmonics. For the sake of simplicity, we shall assume that all the mode intensities are equal: $X_k^2 = X^2$.

Each value of l has its own equation (2.11) that determines the position of the resonances. Its roots $\tilde{\Delta}_l$ are complex, but the mode spacing is real. Consequently, the resonance value is the real part of the root $\Delta_l = Re\tilde{\Delta}_l$. For a given resonance to appear, at least two further conditions must be satisfied. The first of them is

$$|\text{Im}\,\tilde{\Delta}_l| \leqslant \Delta_l, \quad (2.12)$$

and restricts the possible "width" of the resonance, and the second is

$$|\text{Im}\,\tilde{\Delta}_{l+1}+\text{Im}\,\tilde{\Delta}_l| \leqslant |\Delta_{l+1}-\Delta_l|, \quad (2.13)$$

and prevents the approach of neighboring resonances to a distance for which they overlap and cease to be resolved. The latter condition corresponds to the inequality

$$\| C_0^{l+1}(\Delta_l)| - |C_0^l(\Delta_l)\| \geqslant |C_0^l(\Delta_l)|. \quad (2.14)$$

§3. RESONANT VALUES OF INTERMODE FREQUENCY

Before we proceed any further, we note that there are no resonances in the case of a small mode spacing, $\Delta \ll 1$, when several modes fall within the undisturbed line. Leaving the explanation of this fact until we come to Sec. 4, we merely note that none of the roots of (2.11) satisfies condition (2.13) for $\Delta \ll 1$. Bearing this in mind, we confine our attention to the case of relatively large Δ and begin by considering the situation where one of the modes coincides with the transition frequency: $0 \leqslant r_0 \leqslant r, \rho = 0$. In this case, the resonance values of the mode spacing satisfying (2.12) are given by

$$\Delta_l = (X^2 S/2\Gamma l)^{1/2}, \quad l=1, 2, \ldots, \quad (3.1)$$

where

$$S=\sum_{\substack{k=0\\ k\neq r_0\pm l}}\left(\frac{1}{l+k-r_0}+\frac{1}{l-k+r_0}\right). \quad (3.2)$$

The imaginary parts of the roots (3.1) will now be found by the perturbation method. It is clear from (2.11) that the essential point is whether there is an equation of the form $k = r_0 \pm l$ for any mode number $k \in [0, r]$. Such equations are not possible for

$$l > \bar{l} = \max\{r_0, r-r_0\}, \quad (3.3)$$

which corresponds to combination frequencies $l\Delta$ exceeding the separation between the line center and the boundaries of the mode spectrum. This ensures that $\text{Im}\,\tilde{\Delta}_l$ is independent of the radiation intensity:

$$\text{Im}\,\tilde{\Delta}_l = -\frac{1}{2}\left(\frac{1}{\Gamma l}+\frac{|dS/dl|}{S}\right), \quad l > \bar{l}. \quad (3.4)$$

It follows from (3.1) that the initial assumption that $\Delta_l \geqslant 1$ is justified when the field intensity is high enough:

$$X^2 > 2\Gamma l/S \ . \quad (3.5)$$

For the group of resonances (3.3), this inequality is more stringent than (2.13) and, consequently, defines the region in which these resonances exist. When $l \leqslant \bar{l}$, the imaginary parts of the roots are proportional to the intensities of modes with indices $k = r_0 \pm l$. Since $\text{Im}\,\tilde{\Delta}_l$ is large in this range of values of l, the inequality given by (2.13) is not satisfied, so that the low-order resonances do not appear when the mode is accurately tuned to the line center.

Equations (3.1), (3.2), and (3.4) are valid for an arbitrary number of modes, including the special case of the biharmonic field ($r = 1$) analyzed in Refs. 4–6. We note that, when the number of modes is large ($r \ll 1$), the sum in (3.2) can be replaced by an integral, and this yields[3]

$$S=\ln|(l+r_0)(l+r-r_0)/(l-r_0)(l-r+r_0)|. \quad (3.6)$$

In the limiting case where $l \gg \bar{l}$, the last formula becomes much simpler:

$$S=2r/l \quad (3.7)$$

and the resonance mode spacing is

$$\Delta_l=(rX^2\Gamma^{-1})^{1/2}/l \quad (3.8)$$

and is determined by the resultant intensity of all the modes, rX^2, which is inversely proportional to the number of the resonance.

Thus, in the above situation ($\rho = 0$), the only resonances that are possible are those of sufficiently high ($l > \bar{l}$) order, but this order must not exceed $l \sim (rX^2/\Gamma)^{1/2}$, when $\Delta_l \sim 1$. The largest number of resonances should be observed for the smallest value of $\bar{l}$, i.e., for the symmetric disposition of the mode spectrum relative to the line ($r_0 = r/2$). The minimum number is $\bar{l} + 1 \simeq r/2$ and corresponds to the largest mode spacing $\Delta \approx (2X^2 \ln r/\Gamma r)^{1/2}$. As the mode spectrum shifts, the lowest-number resonances are successively lost until the largest resonant value of the mode spacing reaches the linewidth for $r_0^2 = rX^2/\Gamma$ and none of the resonances remains. The inequality given by (3.5) imposes a relatively stringent condition on the field intensity. Since the function $S(l)$ decreases monotonically in the region defined by (3.3), the resonances will be present if the intensity of one mode exceeds the coherent value Γ by a factor of at least $r/\ln r$. This situation is connected with the fact that the line is strongly saturated by one of the modes.

Let us now suppose that none of the modes falls into the line, i.e., $\rho \neq 0$ and, moreover, $|\rho\Delta| \gg 1$. The roots of (2.11) which satisfy (2.12) are now given by expressions that differ from (3.1) and (3.4) by the fact that S must be replaced with

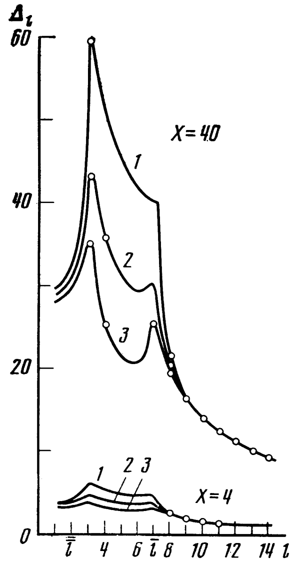

FIG. 1. Resonance value of the normalized mode spacing Δ_l (3.1) as a function of the number l of the resonance. The graphs were plotted numerically for the following values of the parameters: $r = 9$, $r_0 = 2$, $\rho = 0.1; 0.25; 0.5$ (curves 1, 2, and 3, respectively). $X = 4; 40$ (lower and upper curves, respectively). Circles represent resonances satisfying condition (3.11).

$$S_\rho=\sum_{k=0}\left(\frac{1}{l+k-r_0+\rho}+\frac{1}{l-k+r_0+\rho}\right). \tag{3.9}$$

Comparison of (3.2) with (3.9) will show that, for resonance of a high order (3.3), the accuracy with which the tuning to the transition frequency is performed is unimportant, and all the results obtained for $\rho = 0$ remain in force (cf. the descending branch on the right of Fig. 1). However, this group of resonances is joined by a group of resonances of a lower order, $l \ll \bar{l}$. For these, we have $|\mathrm{Im}\tilde{\Delta}_l| \sim |\rho^2 S_\rho|^{-1}$. When $|\rho| \ll 1$, for which

$$S_\rho=\begin{Bmatrix} S & (l\ll\bar{\bar{l}}) \\ S+\rho^{-1} & (\bar{\bar{l}}<l\ll\bar{l};\ \ r_0<r/2) \\ S-\rho^{-1} & (\bar{\bar{l}}<l\ll\bar{l};\ \ r_0>r/2) \end{Bmatrix};\qquad \bar{\bar{l}}=\min\{r_0, r-r_0\}, \tag{3.10}$$

the resolution condition (2.13) will be satisfied when the intensity of one mode satisfies the inequality

$$X^2>\frac{\Gamma l^3}{\rho^4 S_\rho[l(dS/dl)^2-S_\rho]^2}. \tag{3.11}$$

For the lowest-order resonances $l<\bar{\bar{l}}$, the last inequality is very stringent because $S_\rho \approx S \lesssim \ln r$. However, for $\bar{\bar{l}} \ll l \ll \bar{l}$, when $S_\rho = \pm\rho^{-1} \gg S$ and

$$\Delta_l=(X^2/2\Gamma\rho l)^{1/2}, \tag{3.12}$$

the conditions imposed on the intensity are much softer because S_ρ increases.[4)] According to (3.11), when the detuning is small enough, $|\rho| \leqslant 1/l$, the resonances given by (3.12) exist if $X^2 \geqslant 8\Gamma l^3/\rho$ (we note that this quantity is independent of the number of modes). The last inequality leads us to the conclusion that resonances in this group are already possible for intensity X^2 that exceeds the coherent value Γ by one or two orders of magnitude. The most favorable conditions are established for $\rho \sim 1/2$ and $\bar{\bar{l}} = 0$, when the line lies between the outermost and the nearest modes. The first resonance to appear as X^2 increases should then be the resonance with $\bar{\bar{l}} = 1$. For given intensity, the resonance appear for $\bar{\bar{l}} < l \lesssim l_* = (X^2|\rho|/8\Gamma)^{1/3}$, i.e., in the interval $(2l_*/\rho) \lesssim \Delta_l < (l_*/\bar{\bar{l}})^{1/2} \cdot 2l_*/\rho$, and their number increases with increasing fine-tuning parameter ρ. Figure 1 illustrates the overall picture.

In this section, we have concentrated our attention on the situation where the transition frequency lies within the mode spectrum ($0 \leqslant r_0 \leqslant r$). No complications are found to arise when this conditionis relaxed. All that we have said above remains in force even for the external position of the line, say, for[5)] $r_0 < 0$. All that needs to be done is to substitute $\bar{\bar{l}} = |r_0|$, $\bar{l} = r + |r_0|$ and remember that S is negative for $l < |r_0(r - \bar{\bar{r}}_0)|^{1/2}$. Hence, in particular, the lowest-order resonance, $l \leqslant \bar{\bar{l}}$, are absent because, according to (3.10), for such resonances.

§4. GENERALIZATION OF RABI FREQUENCY

We must now consider the physical interpretation of the above resonances. We begin with the most obvious limiting case presented by (3.8), which is conveniently rewritten in the form

$$l\Delta=\Omega. \tag{4.1}$$

The quantity $\Omega = (rX_2/\Gamma)^{1/2}$ on the right of this equation can be interpreted as the frequency of Rabi oscillations produced by a monochromatic field of intensity rX^2, tuned to the transition frequency. The effect of this field on the two-level system is equivalent to the effect of the above polyharmonic radiation if the Rabi frequency Ω is much greater than the width $r\Delta$ of the mode spectrum and the detuning[9,11] $|r_0\Delta|$. It is precisely these assumptions that lie at the basis of (3.8). (The interpretation of the resonances in this case is also discussed in Ref. 20, where use is made of the idea of quasienergy states found in the adiabatic approximation.) We shall show that, in general, the resonance condition is still that the mode beat frequencies, or the beat harmonics, are equal to certain natural frequencies of the system, which may be referred to as the generalized Rabi frequencies. To show this, we shall seek the solution of (2.1) for $\alpha = 0$ (for the sake of simplicity, without relaxation) in the form

$$n=\bar{n}\exp(-i\Gamma\Omega\tau)+\text{c.c.}; \tag{4.2}$$

$$y=\sum_{k=0}^{r}[y_k'\exp(-i\Gamma(k\Delta-\Omega)\tau)+y_k''\exp(-i\Gamma(k\Delta+\Omega)\tau)]. \tag{4.3}$$

When we substitute the amplitudes of the polarization harmonics, given by (2.1a) in terms of $\bar{n}$ and x_k, into the right-hand side of (2.1b), we shall omit terms that depend on the mode phases, i.e., we shall average over the random phases. Eliminating in this way the phase effects in which we do not interested, we find that the population difference oscillates in the polyharmonic field with specific natural frequencies satisfying the condition

$$\frac{X^2}{\Gamma}\sum_{k=0}^{r}\frac{1}{\Omega^2-(\Delta_0-k\Delta)^2}=1. \qquad (4.4)$$

The number of generalized Rabi frequencies is thus seen to be equal to the number $r+1$ of field modes.

In the special case of one mode ($r=0$), we obtain the well-known expression for the Rabi frequency:[1] $\Omega_0^2=(X^2/\Gamma)+\Delta_0^2$. When there are two modes ($r=1$), Eq. (4.4) turns out to be a biquadratic equation and there are two generalized Rabi frequencies:

$$\Omega_{0,1}^2=\frac{\Delta_0^2+(\Delta_0-\Delta)^2}{2}+\frac{X^2}{\Gamma}\mp\left[\frac{\Delta^2(2\Delta_0-\Delta)^2}{4}+\frac{X^4}{\Gamma^2}\right]^{1/2}. \qquad (4.5)$$

Figure 2 shows the distribution of generalized Rabi frequencies for an arbitrary number of modes, where the frequencies normalized to the mode spacing Δ are shown as functions of the universal parameter $X/\Gamma^{1/2}\Delta$ for different positions of the mode spectrum relative to the quantum transition. Resonances correspond to the points of intersection of the curves $\Delta^{-1}\Omega_k(X/\Gamma^{1/2}\Delta)$ and the straight lines $\Omega/\Delta=l$ ($k=0, 1,\dots, r$; $l=1,2,\dots$). When the intensities are low enough, the Rabi frequencies are formed by each of the modes independently: $\Omega_k^2\approx X^2/\Gamma+(k\Delta-\Delta_0)^2$. As the mode intensity increases, all the Rabi frequencies with the exception of one, the highest, tend to certain fixed values Ω_k lying within the mode spectrum in intervals bounded by the points $|\Delta_0-k\Delta|$. According to (4.4), the highest frequency increases without limit with increasing field intensity and, for $X^2\gg\Gamma(\bar{l}\Delta)^2/r$ is given by $\Omega_r=(rX^2/\Gamma)^{1/2}$. As we have seen, this determines the high-order resonances in (4.1) with $l\gg r$, $|r_0|$. Similarly, in the general case, a resonance with arbitrary l appears when intermode beat resonance frequency $l\Delta$ is equal to one of the $r+1$ generalized Rabi frequencies $\Omega_0, \Omega_1,\dots,\Omega_r$. In fact, replacing Ω in (4.4) with $l\Delta_l$, we immediately obtain the characteristic equation (2.11) that was used in the above analysis [relaxation is taken into account in (4.6)]. Since the first r Rabi frequencies lie within mode intervals with bounding points $|\Delta_0-k\Delta|$, and the highest rth Rabi frequency lies outside the most distant of them relative to the transition frequency, it is clear that of the lowest order resonances $l<\bar{l}$ occur when the intermode beat frequencies coincide with one of the r first Rabi frequencies, whereas the high-order resonances with $l>\bar{l}$ occur when there is a coincidence with the highest Rabi frequency (see Sec. 3 and Fig. 2). The latter are the subharmonics of Rabi frequency Ω_r but the former are not: each of the low-order resonances is coupled to its own Rabi eigenfrequency Ω_k. For example, in the case of the two modes given by (4.5) (for $X^2\gg\Gamma\Delta|\Delta_0-\Delta/2|$), the higher Rabi frequency $\Omega_1\approx(2X^2/\Gamma)^{1/2}$ may coincide for a particular intensity with any of the intermode beat frequencies $l\Delta$, where $l>\bar{l}$, and the lower frequency, which is given by $\Omega_0=([\Delta_0^2+(\Delta-\Delta_0)^2]/2)^{1/2}$ and lies in the intermode interval $[|\Delta_0|, |\Delta-\Delta_0|]$, may coincide with one of the intermode beats, provided both modes lie on one side of the transition frequency.

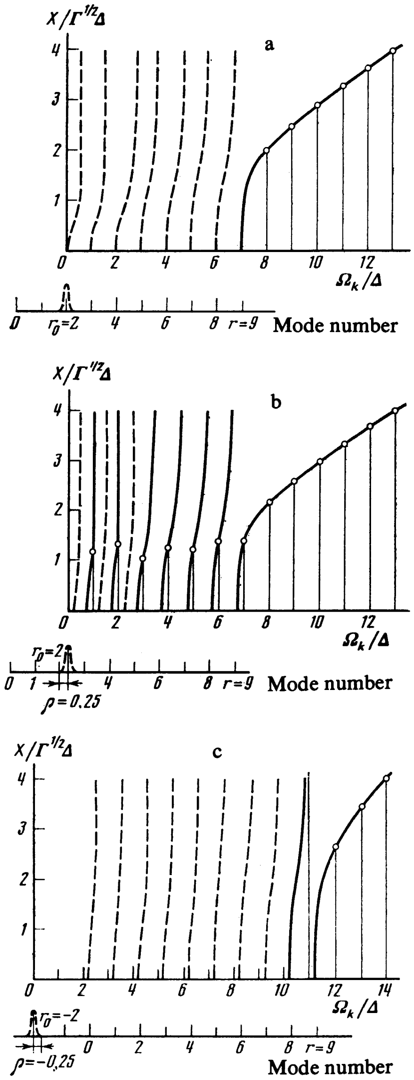

FIG. 2. Generalized Rabi frequencies Ω_k ($k=0, 1,\dots r$), normalized to the mode spacing Δ as functions of the parameter $X/\sqrt{\Gamma\Delta}$ for three different positions of the spectrum, indicated on the graphs. Solid curves show the Rabi frequencies generating at least one resonance. Resonances are indicated by open circles. The graphs were plotted numerically for the case of ten modes.

By analyzing Figs. 2a, b, and c, we can establish which particular Rabi frequency coincides with the combination frequency $l\Delta$ at the lth resonance. It is clear from Fig. 2a that, even if we neglect relaxation, the lower-order resonances with $l<\bar{l}$ should be absent when one of the modes is accurately tuned to the transition frequency ($\rho=0$). Coincidences between the combination frequencies $l\Delta$ and the first Rabi frequencies are then impossible for finite mode intensities. Coincidences are possible only with the highest Rabi frequency Ω_r and, to reach the resonances, we must satisfy the condition $\Omega_r\geqslant(\bar{l}+1)\Delta$ by increasing the Rabi frequency, for example, by increasing the mode intensity. This means that, when the number of modes is very large, resonances will now appear for higher intensity than the lower-order resonances with $l\ll\bar{l}$ in the case of detuning $\rho\neq 0$ (cf. Fig. 2a and b). Inspection of Figs. 2b and c will show that the existence of resonances with $l<l<\bar{l}$ depends on the sign of the detuning (see footnote 4). If the mode spectrum does not include the transition frequency (Fig. 2c), resonances of very low order $l < e$ are impossible because the lowest Rabi frequency, Ω_0, now exceeds $l\Delta$. It is thus clear that the properties of the resonances, including those established earlier in Sec. 3, are determined by the distribution of the generalized Rabi frequencies, so that the resonances investigated in this paper can be referred to as Rabi resonances.

Relaxation has so far been neglected for the sake of simplicity. However, when it is taken into account, the equation for the Rabi frequencies assumes the form

$$\frac{X^2}{\Gamma}\sum_{k=0}^{r}\frac{\Omega+i}{(\Omega+i)^2-(\Delta_0-k\Delta)^2}=\frac{i}{\Gamma}+\Omega. \tag{4.6}$$

The generalized Rabi frequencies thus become complex:

$$\tilde{\Omega}_k=\Omega_k+i\,\mathrm{Im}\,\tilde{\Omega}_k,$$

where $\mathrm{Im}\,\tilde{\Omega}_k < 0$, which corresponds to the damping of the natural oscillations in the population difference. It is clear from the physical significance of the Rabi frequencies that the following conditions must be satisfied for resonances to occur:

$$\Omega_k \gg |\mathrm{Im}\,\tilde{\Omega}_k|,\ |\Omega_{k+1}-\Omega_k| \gtrsim |\mathrm{Im}\,\tilde{\Omega}_k|+|\mathrm{Im}\,\tilde{\Omega}_{k+1}|$$

[cf. (2.12) and (2.13)]. When $\Gamma = 1$, i.e., $T_1 = T_2$, Eq. (4.6) yields $\mathrm{Im}\,\tilde{\Omega}_k = -1$. Consequently, resonances are possible only with Rabi frequencies that exceed the linewidth: $\Omega_k \gg |\mathrm{Im}\,\tilde{\Omega}_k| = 1$. Moreover, it is now clear why there are no resonances in the case of a dense spectrum, when several modes fall into the medium line. In fact, resonances with $l \leqslant \bar{l}$ are impossible because the first r Rabi frequencies Ω_k are separated by the mode spacing and overlap considerably for $\Delta \leqslant 1 = |\mathrm{Im}\,\tilde{\Omega}_k|$. On the other hand, there are no resonances with $l > \bar{l}$ because broadening of the highest Rabi frequency ($\mathrm{Im}\,\tilde{\Omega}_r = 1$) ensures that its subharmonics $\Delta_l = \Omega_r/l$ overlap because the difference $\Delta_l - \Delta_{l+1} = \Delta_l/l$ is small in comparison with $|\mathrm{Im}\,\tilde{\Delta}_l| = |\mathrm{Im}\,\tilde{\Omega}_r|/l$ for $\Delta_i \ll 1$. We note that even in the general case ($\Gamma > 1$), Eq. (2.14) shows that the condition $|\mathrm{Im}\,\tilde{\Omega}_k| \lesssim \Delta$ must be satisfied on resonance since, otherwise, the broadening of the Rabi frequencies will ensure that the resonance relation $\Omega = l\Delta$ will be satisfied for given Ω and Δ for different neighboring values of l, and the resonances will become unresolvable.

§5. RESONANCE EFFECTS ACCOMPANYING INTENSITY VARIATIONS AND SHIFT OF MODE SPECTRUM

The resonance values of the mode spacing were determined above for fixed other parameters characterizing the field. On the other hand, there are other acceptable formulations of the problem in which the mode spacing is fixed and some other parameter is varied. The analysis is then reduced to the solution of the characteristic equation given by (2.11) for the varied quantity, and to an analysis of the necessary resonance conditions such as (2.14). Thus, when the *intensity* is varied, its resonance values are given by $X_l^2 = 2\Gamma l\Delta/S_\rho$, which corresponds to (3.1), and the necessary condition given by (2.14) reduces to the inequality given by (3.11).

If we now suppose that the intensity and mode spectrum are fixed (X^2, $r,\Delta = \mathrm{const} > 1$), we can determine the values of the detuning of the center of the mode spectrum $\Delta_0 - \Delta r/2$ from the transition frequency for which resonance is possible. To be specific, we shall consider high-order resonances with $l > \bar{l} \geqslant r/2$. If we substitute $r_0 \to r_0 + \rho$ in (3.1) and (3.6), we obtain

$$\Delta_{0l}-\frac{\Delta r}{2}=\pm\Delta\left\{\left(l-\frac{r}{2}\right)^2-rl\left[\mathrm{cth}\left(\frac{l\Gamma\Delta^2}{X^2}\right)-1\right]\right\}^{1/2}. \tag{5.1}$$

Hence, it follows, in particular, that, when $X^2 \gtrsim \Gamma\Delta^2 r/2$, the required detunings differ from a whole number of mode spacing.[6] Figure 2 presents a clear picture of the resonances if we recall that a shift of the mode spectrum as a whole to the right is accompanied by a shift of the generalized Rabi frequencies in the same direction. Resonances with $l \geqslant \bar{l}$ then correspond to the seccessive crossing of the vertical lines $\Omega/\Delta = l$ by the point of intersection of the curve Ω_r/Δ (representing the highest Rabi frequency) with the horizontal line $X/\Gamma^{1/2}\Delta$, specified by fixed values of X and Δ.

§6. CONCLUDING REMARKS

It is clear from the foregoing that resonance effects will occur when a two-level system interacts with multimode radiation with an equidistant spectrum and arbitrary independent mode phases. Resonances in the mode absorption coefficients correspond to coincidences between the harmonics of intermode beats and generalized Rabi frequencies that are functions of the mode spacing the position of the mode spectrum, and the mode intensities. These Rabi resonances are independent of the phase relationships, and are described by

$$(\Delta\omega)_l=\mu\mathscr{E}\hbar^{-1}(S_\rho/2l)^{1/2},\quad l=1,2,\ldots, \tag{6.1}$$

where S_ρ of (3.9) is determined by the disposition of the mode spectrum.

Resonances occur when the amplitude of each mode exceeds by at least a substantial factor the coherent value $\hbar/\mu T_2$ and the mode spacing $\Delta\omega$ is greater than the width T_2^{-1} of the spectral line of the medium. In the special case of the biharmonic field, examined earlier in Refs. 4–6, the resonance intermode frequencies given by (6.1) are the subharmonics of a single Rabi frequency. In the case of the polychromatic field, there is, in addition to the high-order resonances with $l > \bar{l} = \max\{r_0, r - r_0\}$, which are also the subharmonics of the same (highest) Rabi frequency, a new group of low-order resonances with $l < \bar{l}$, each of which is coupled to its own Rabi frequency. The number of resonances and the maximum value of the resonance mode spacing will, in general, increase with increasing intensity and number of modes. From the practical point of view, the most interesting are the resonances with low numbers $l < r$ that correspond to the main components of the intermode beats. They have a greater effect on the mode absorption coefficients than the higher-order resonances, and occur at relatively low radiation intensities that are completely attainable in multimode lasers (for example, for $T_1 \sim T_2 \sim 10^{-8}$ s, $\mu \sim 1D$, and optimum disposition of the spectrum, the power density of multimode radiation should be of the order of 10, 1000, and 10 000 W/cm^2 for $r \sim 10$, 100, and 1000, respectively).

We note that the conditions for which the above Rabi resonances will occur are very typical for experiments in intracavity laser spectroscopy. However, the given-field approximation has to be abandoned when such phenomena are examined, and resonance effects in the self-consistent formulation must be investigated.

[1)] The exact solution of the equations describing the dynamics of a two-level system in a polychromatic field was obtained in Refs. 12, 13 for certain special cases. However, the analysis of the solution was difficult because of its complexity and did not lead to a clear picture of the mode absorption resonances.

[2)] In particular, when the frequency of the quantum transition lies inside the mode spectrum, the fixing of these parameters means that we have fixed the ratio in which the transition frequency divides the mode spectrum. Other possible formulations of the problem are considered below, in Sec. 5.

[3)] When one of the factors in the numerator or the denominator in the logarithm is equal to zero (i.e., when $l = |r_0|$ or $l = |r - r_0|$) we must replace this particular factor with unity.

[4)] Since $\Delta_l \sim S_\rho^{1/2}$, the necessary condition for resonance is that S_ρ be positive. This sets the sign of ρ, depending on which side of the line contains more modes: $\rho > 0$ for $r_0 < r/2$ and $\rho < 0$ for $r_0 > r/2$.

[5)] We note that, without loss of generality, we may suppose that $r_0 < r/2$ because the opposite situation corresponds to the symmetric position of the mode spectrum relative to the line center.

[6)] It is clear from the foregoing that we are concerned with resonance effects that differ from resonances in the saturation of the quantum transition, which occur when the successive modes cross the line of the medium as the spectrum is shifted.

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Translated by S. Chomet