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Coherent Spontaneous Emission of Multilevel Systems

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Using a group-theoretical approach, we generalize Dicke's theory[1] to include the case of multilevel emitters. The case of three-level particles is considered in detail and is solved completely. It is shown that state are possible for which the emission intensities for all transitions are proportional to the number of particles squared. Methods of producing such states from various initial states of the system are discussed (including a state in thermodynamic equilibrium). Photon-echo effects (intensities, angular distributions) are considered for extended systems.

1. INTRODUCTION

SPONTANEOUS emission of individual atoms or molecules, in the case when the linear dimensions of the systems are small in comparison with the wavelength, do not occur independently. There is always an indirect interaction between particles via the electromagnetic radiation field, which can lead both to enhancement of the radiation (superradiant states) and to its suppression[1,2]. Interest in phenomena connected with coherence of radiating molecules has greatly increased of late, owing to the experimental observation of a number of effects produced when a short electromagnetic pulse acts on a resonant medium (photon echo[3], effects of "self-intransmission" of a pulse[4,5], change of momentum spectrum[6,7]).

The principles of the quantum theory of coherent spontaneous emission were formulated by Dicke[1], who has shown that as a result of interaction via the field two-level molecules behave like a single-quantum-mechanical system whose radiation intensity is determined by its coherence characteristics. Dicke's problem was subsequently investigated, in various aspects, by many workers[2,8,9], but always within the framework of two-level idealization. Yet in many problems (molecules with equidistant spectrum, action of several resonant pulses, interaction with intense radiation fluxes) such an idealization cannot be used. Moreover, the spontaneous emission from multilevel molecules, as will be shown below, exhibits by itself a number of singularities that cannot be predicted solely from results pertaining to two-level molecules. Finally, analysis of the multilevel problem enables us to establish accurately the limits of applicability of the two-level approximation.

The possibility of generalizing Dicke's theory to include multilevel molecules was considered by one of us in[10], where it was noted that an important factor in Dicke's theory is the choice of irreducible representations of the SU_2 group in the energy space of the system as the stationary states of the unperturbed Hamiltonian. A similar role is played for multilevel systems systems by the basis of irreducible representations of the SU_n group (n is the number of levels). Teplitskiĭ obtained[11], by group-theoretical methods, the explicit form of the density matrix of a multilevel system interacting with an electromagnetic field.

The present paper is devoted to coherent emission in multilevel systems on the basis of a group-theoretical approach. We investigated in detail the case $n = 3$ (we present a complete classification of the coherent state, obtain the coherent characteristics, and analyze the properties of the sponteneous emission). The analysis implies experimental possibilities of obtaining superradiance at several frequencies, and also observation of "photon echo" in a multilevel scheme (in this case the angular distributions of the superradiant responses have, as will be shown below, a highly unique character). It must be emphasized that the transition from the two-level to the three-level one is the principal step that makes it possible to investigate a number of subtle effects in nonlinear interactions of light with matter. The analysis of the present paper shows that even in the three-level scheme one can observe the main features of the phenomena that occur in real multilevel systems.

2. THREE-LEVEL SYSTEMS. GROUP APPROACH[1)]

We consider a system of emitters with three nondegenerate energy levels. The ensemble of particles occupies a volume with dimensions that are small in comparison with the wavelengths emitted by the system. We neglect inelastic collisions (low-density gas). Then the variables of the molecule internal energy and the coordinates of their mass centers can be separated.

It is convenient to express the Hamiltonian of the system with the aid of generators of the SU_3 group (we use the notation introduced in[13]). The internal-energy operator of the j-th molecule has eigenvalues $\epsilon_1 = -\epsilon/2$, $\epsilon_2 = \epsilon/2$ and ϵ_3, and can be represented in the form

$$H_1^{(j)} = \varepsilon I_3^{(j)} + \varepsilon_3(1/3 - Y)^{(j)}. \tag{1}$$

The eigenfunctions of this operator $\chi_i^{(j)}$ ($i = 1, 2, 3$) correspond to the eigenvalues $(-1/2, 1/3)$, $(1/2, 1/3)$, $(0, -2/3)$ of the isospin and hypercharge operators $I_3^{(j)}$ and $Y^{(j)}$, and correspond to the molecule occupying the first, second, and third-level, respectively. The function $\chi_i^{(j)}$ transforms in accordance with the representation $D(1, 0)$ of the group SU_3. The unperturbed Hamiltonian of a system of N molecules is

$$H = H_0(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) + \varepsilon I_3 + \varepsilon_3(1/3 - Y), \tag{2}$$

where H_0 is the energy of translational motion and of

[1)] A detailed derivation of all the mathematical statements made here can be found in[12].

the intermolecular interaction of the molecules.

$$I_3=\sum_{j=1}^{N} I_3^{(j)},\quad {}^1/_3-Y=\sum_{j=1}^{N} ({}^1/_3-Y)^{(j)}.$$

H_0 acts only on the coordinates $r_1, r_2, \ldots r_N$ of the molecule mass centers. The eigenfunctions of the unperturbed Hamiltonian are

$$\Psi_g(N_1,N_2,N_3)=U_g(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)\chi_{p_1}^{(1)}\chi_{p_2}^{(2)}\ldots\chi_{p_N}^{(N)}. \tag{3}$$

Here $U_g(r_1, \ldots, r_N)$ is the coordinate part of the wave function ($H_0U_g = \epsilon_gU_g$); the functions $\chi^{(j)}$ indicate the level occupied by each of the molecules; N_1, N_2, and N_3 are the level populations. The interaction of the molecules with a classical radiation field is described in the dipole approximation by the operator

$$H_{int}=-\mathbf{A}[(\mathbf{e}_1-i\mathbf{e}_2)I_+ +(\mathbf{e}_3+i\mathbf{e}_4)K_- +(\mathbf{e}_5+i\mathbf{e}_6)L_- + \text{h.c.}],$$

where **A** is the vector potential of the field at the location of the system; the vectors $\mathbf{e}_i$ are connected with the dipole-moment matrix elements by the relations

$$\mathbf{e}_1-i\mathbf{e}_2=-\frac{i\varepsilon}{\hbar c}\mathbf{d}_{12},\quad \mathbf{e}_3-i\mathbf{e}_4=\frac{i}{\hbar c}\left(\varepsilon_3-\frac{\varepsilon}{2}\right)\mathbf{d}_{32},$$
$$\mathbf{e}_5-i\mathbf{e}_6=\frac{i}{\hbar c}\left(\varepsilon_3+\frac{\varepsilon}{2}\right)\mathbf{d}_{31}.$$

Thus, the transition probabilities are determined by the matrix elements of the nondiagonal generators of the SU_3 group (see the figure).

In analogy with the two-level case[1], it is necessary to use as the initial states not those in (3) but their linear combinations, such that H_{int} couples one stationary state with a minimum number of other states. Such a set of states is the basis of irreducible representations $|P, Q, I, I_3, Y\rangle$ of the SU_3 group. The electromagnetic transitions conserve the quantum numbers P and Q (the Hamiltonian is made up of group generators with which the Casimir operators commute). It can be shown[12] that when N_1, N_2, N_3 and I are specified, the numbers s ($s = {}^1/_3(N - P + Q)$ and a ($a = {}^1/_3(N - P - 2Q)$) run through values satisfying the system of inequalities

$$\begin{gathered} I_M-I\leqslant s\leqslant I_M+I,\\ a\leqslant I_M-I,\qquad s+a\leqslant I_M-I+N_3, \end{gathered} \tag{4}$$

where $2I_M = N_1 + N_2$, $|N_2 - N_1| \le 2I \le N_1 + N_2$. Using the matrix elements of the operators $L_\pm$, $K_\pm$, and $L_\pm$[12] we obtain for the intensities of the spontaneous emission from the state $|s, a, I, I_3, Y\rangle$

$$J_{21}=J_{21}^{(0)}(I+I_3)(I-I_3+1),\ J_{32}=J_{32}^{(0)}[\gamma_1^2(I+I_3+1)+\gamma_2^2(I-I_3)],$$
$$J_{31}=J_{31}^{(0)}[\gamma_1^2(I-I_3+1)+\gamma_2^2(I+I_3)], \tag{5}$$

where

$$\gamma_1^2=(I_M+I-s+1)(I_M+I-a+2)(I_M+I-s-a+N_3)\times[(2I+1)(2I+2)]^{-1},$$
$$\gamma_2^2=(I-I_M+s)(I_M-I-a+1)(I_M+I-s-a+N_3+1)\times[2I(2I+1)]^{-1};$$

$J_{21}^{(0)}$, $J_{32}^{(0)}$, and $J_{31}^{(0)}$ are the intensities of the spontaneous emission of an isolated molecule. Thus, the intensities depend on the three quantum numbers s, a, and I, one of which (I) changes in the transitions 3 → 1 and 3 → 2. These numbers are the cooperative characteristics of the system of molecules and determine the rate of its radiative decay.

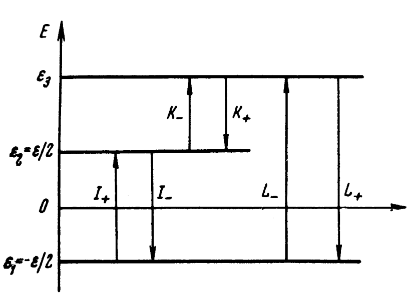

Diagram of transitions between the energy levels. The transition between each pair of levels is described by the non-diagonal I−, K−, or L−spin components.

In particular cases when the number of particles at one of the levels is equal to zero, the correlation between the two other levels is described, just as in the case of the two-level scheme, by the corresponding isospin (I, K, or L). The emission via the other channels is proportional to the number of particles at the level from which the transition is considered. For example, if $N_2 = 0$, then we have from (4) $I = -I_3 = N_1/2$, $a = 0$, $0 \le s \le \min(N_1, N_3)$. With the aid of (5) we obtain

$$J_{31}=J_{31}^{(0)}(L+L_3)(L-L_3+1),\quad J_{32}=J_{32}^{(0)}N_3,\ J_{21}=0, \tag{6}$$

where

$$L={}^1/_2(N_1+N_3)-s,\quad |L_3|\leqslant L\leqslant {}^1/_2(N_1+N_3),\quad L_3={}^1/_2(N_3-N_1).$$

At $s = 0$ the intensities of emission between levels coupled by the corresponding isospin are proportional to N^2 (Dicke's superradiant states).

On the whole, one encounters in the analysis of formulas (4) and (5) a great variety of possibilities. Let, for example, $I = I_{max}$. Then $a = 0$ and $s = \min(N_1 + N_2, N_3)$. If $s = 0$, we have

$$J_{21}=J_{21}^{(0)}N_2(N_1+1),\ J_{32}=J_{32}^{(0)}N_3(N_2+1),\ J_{31}=J_{31}^{(0)}N_3(N_1+1). \tag{7}$$

At equal populations, in particular, the emission intensities in all channels are proportional to the square of the total number of molecules (superradiance at all frequencies). If $s = N_3$, the emission at the frequencies ω_{13} and ω_{32} is suppressed; at the same time, the system emits coherently at the frequency ω_{12}: $J_{21} = J_{21}^{(0)}N_2(N_2 + 1)$. Let $I = -I_3$. Then $N_2 \le s \le N_1$, $a \le N_2$, and $s + a \le N_2 + N_3$. At $s = N_2$ and $a = N_3$, the system does not emit through any of the channels. It is easy to show (cf.[1,2]) that whereas the intensities of the spontaneous emission of the system depend on the states of the system (on the cooperation numbers s, a, and I), the intensity of the total emission (or absorption) in the absence of external resonant fields is always proportional to the difference between the numbers of molecules at the corresponding levels ($J_{32} - J_{23} \propto N_3 - N_2$, etc.). The superradiant states can be excited by starting from the ground state ($kT = 0$): $N_1 = N$, $N_2 = N_3 = 0$. Then $I = -I_3 = N/2$, $s = a = 0$. If a strong resonant pulse of frequency ω_{12} acts on the system, then it goes over into a state with $I_3 \sim 0$ (I remains maximal). The pulse action is followed by an intense response at the frequency ω_{12}: $J_{21} = J_{21}^{(0)}(N/2 + 1)N/2$. When a pulse of frequency ω_{13} acts on the initial state, we arrive at the case (6) with $s = 0$ (coherent emission at the frequency

ω_{13}). Finally, by using excitation pulses with frequencies ω_{12} and ω_{13} we can transfer the gas into a state with $s = a = 0$, $N_1 = N_2 = N_3$, and $I = I_M = N/3$ (when a pulse of frequency ω_{12} is absorbed, the number I does not change; I remains maximal when the second pulse is absorbed[12]). The system in this state emits with an intensity proportional to N^2 at all frequencies.

Superradiant states can be obtained also by molecule sorting. Unlike in the two-level case, equal sorting of the molecules can lead to essentially different results, depending on the concrete properties of the multilevel system. If one sorts the molecules at the upper level, a state with $s = a = 0$ is produced. During its decay at $J_{32}^{(0)} \gg J_{31}^{(0)}$ (which is usually satisfied for vibrational transitions), two successive intense signals are observed, first at the frequency ω_{32} and then at ω_{21}. If $J_{31}^{(0)} \gg J_{32}^{(0)}$, one signal at the frequency ω_{31} is observed. At $J_{32}^{(0)} \approx J_{31}^{(0)}$, the picture of the evolution is more complicated. The system goes through a state that is intermediate between the superradiant and the ordinary one (the degree of proximity of this state to (7) calls for further study). Formulas (4) and (5) make it also possible to obtain the time dependences of the intensities in the case of systems that are not fully sorted out. As to the sorting methods, one can propose, in addition to the known methods used in masers[2], a chemical method in which the reaction of hydrogen with molecular fluorine is used[14]. The rate of this reaction decreases rapidly with increasing number of excitation of the hydrogen levels. One can observe superradiance on the hydrogen atoms. In general, by virtue of the difference between the properties of the atoms or molecules located at relatively remote levels, the sorting in multilevel systems becomes much easier in comparison with the two-level case.

Let us stop and discuss the case of a system in thermodynamic equilibrium. We assume that

$$kT \gg \varepsilon_3 + \varepsilon/2. \tag{8}$$

Obtaining the mean values of the corresponding operators with the aid of the equilibrium density matrix we can show that if the conditions

$$\bar{I}_3^2 \gg \bar{N}_1 + \bar{N}_2 \gg 1, \quad \bar{L}_3^2 \gg \bar{N}_1 + \bar{N}_3 \gg 1, \quad \bar{K}_3^2 \gg \bar{N}_2 + \bar{N}_3 \gg 1$$

are satisfied ($\bar{N}_i$ are the equilibrium populations), the values of I, s, and a deviate slightly from the respective values

$$\bar{I} = N\frac{\varepsilon}{6kT}, \quad \bar{s} = N\left(\frac{1}{3} + \frac{\varepsilon_3}{9kT} - \frac{\varepsilon}{6kT}\right), \quad \bar{a} = N\left(\frac{1}{3} - \frac{2\varepsilon_3}{9kT}\right). \tag{9}$$

The intensities of the responses of the system to pulses are determined in the main by the states with the s, a, and I indicated in (9). The action of a strong pulse of frequency ω_{21} will produce an intense response

$$J_{21} = J_{21}^{(0)}\bar{I}(\bar{I}+1) \approx J_{21}^{(0)} N^2(\varepsilon/6kT)^2, \quad J_{31} = J_{32} \approx 0. \tag{10}$$

After excitation by a strong pulse of frequency ω_{23}, the system goes over into a state with $I_3 = \frac{1}{4}(\bar{N}_2 + \bar{N}_3) - \frac{1}{2}\bar{N}_1$, $I = -I_3$. We obtain for the intensities

$$J_{32} \approx J_{32}^{(0)}\frac{N^2}{4}\left(\frac{2\varepsilon_3 - \varepsilon}{6kT}\right)^2, \quad J_{31} = J_{21} \approx 0. \tag{11}$$

The calculation method employed enables us to separate in the intensities only the terms proportional to N^2. Using the density matrix formalism (see Sec. 3, formulas (18) and (19), below), we can obtain exact formulas (without confining ourselves, furthermore, to the condition (8)): for the case (10) we must put in (18) the values $\theta = 0$, $\varphi = 90°$, and $\Gamma(\mathbf{k}_{12}, \mathbf{k}') = 1$, and integrate over the angles; for case (11) we must put $\chi = 90°$ and $\Gamma(\mathbf{k}_{23}, \mathbf{k}'') = 1$ in (19). The result of the action of a pulse of frequency ω_{13} cannot be obtained on the basis of the simple considerations that lead to (10) and (11), for absorption of the pulse results in a superposition of states with different I. We must therefore use the results of Sec. (3) see formula (18) below, with $\varphi = 0$, $\theta = 90°$, and $\Gamma(\mathbf{k}_{13}, \mathbf{k}''') = 1$). A superradiant response at the frequency of the applied pulse is observed in the cases under consideration. By applying in succession two intense pulses (with frequencies ω_{12} and ω_{13}) we can bring the system to a state from which superradiance is observed at all frequencies. The intensities of the responses are determined in this case by formulas (18) with $\theta = \varphi = 90°$ and $\Gamma(\mathbf{k}_{12}, \mathbf{k}') = \Gamma(\mathbf{k}_{13}, \mathbf{k}''') = \Gamma(\mathbf{k}'', \mathbf{k}_{13} - \mathbf{k}_{12}) = 1$. The emission intensities following the action of several pulses depend, generally speaking, on their sequence[12].

3. SYSTEMS WITH LARGE DIMENSIONS. PHOTON ECHO

In the optical band, as a rule, the linear dimensions of the system exceed the wavelength of the spontaneous emission. In this case the molecules can be correlated in such a way that coherent emission is observed at different frequencies in directions that are connected by definite geometrical relations. The Hamiltonian of the interaction between the molecules and the field is

$$H_{int} = -\sum_{\mathbf{k}\lambda}\left(\frac{\hbar}{2\omega V}\right)^{1/2}\{a_{\mathbf{k}\lambda}\mathbf{e}_{\mathbf{k}\lambda}[(\mathbf{e}_1 - i\mathbf{e}_2)I_+(\mathbf{k}) + (\mathbf{e}_3 + i\mathbf{e}_4)K_-(\mathbf{k}) + (\mathbf{e}_5 + i\mathbf{e}_6)L_-(\mathbf{k})] + \text{h.c.}\}, \tag{12}$$

where $a_{\mathbf{k}\lambda}^+$ and $a_{\mathbf{k}\lambda}$ are the photon creation and annihilation operators. The operators in (12) are defined by the formulas

$$I_\pm(\mathbf{k}') = \sum_j I_\pm^{(j)} \exp(\pm i\mathbf{k}'\mathbf{r}_j), \quad K_\pm(\mathbf{k}'') = \sum_j K_\pm^{(j)} \exp(\mp i\mathbf{k}''\mathbf{r}_j),$$

$$L_\pm(\mathbf{k}''') = \sum_j L_\pm^{(j)} \exp(\mp i\mathbf{k}'''\mathbf{r}_j)$$

and if the condition

$$\mathbf{k}' + \mathbf{k}'' = \mathbf{k}''' \tag{14}$$

is satisfied, these operators, together with the diagonal operators I_3 and Y, satisfy the commutation relations of SU_3 algebra. The wave functions of a system of molecules can be chosen to be simultaneously the eigenfunctions of the unperturbed Hamiltonian (2) and the Casimir operators $F^2(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')$ and $G^3(\mathbf{k}', \mathbf{k}'', \mathbf{k}''')$ made up from (13) with condition (14) satisfied. The Casimir operators do not commute with operators of type (13) having wave-vector indices different from those already chosen. In analogy with the reasoning that led to (5), we find that the constructed wave functions $|s, a, I, I_3, Y\rangle$ with momenta $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$ are the system states for which the emission in the directions of $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$ ($\mathbf{k}' + \mathbf{k}'' = \mathbf{k}'''$) at the respective frequencies ω_{12}, ω_{13}, and ω_{23} is coherent. The intensity of the spontaneous emission in these directions is calculated from formulas (5), where J_{21}, $J_{21}^{(0)}$: $J_{32}^{(0)}$; J_{31}, $J_{31}^{(0)}$ should be interpreted as the rates of photon emission in a unit of solid angle in the directions $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$, respectively. Emission in the directions $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$ does not change the quantum numbers s and a. The emission in other direction does change these numbers and destroys the coherence with respect to $\mathbf{k}'$, $\mathbf{k}''$, and $\mathbf{k}'''$.

In analogy with the analysis given in Sec. 2 above, we can show that when two plane waves of high intensity are incident on a system in the ground state, superradiant pulses will be observed at all three frequencies, and the wave vectors of the emitted photons are given by the wave vectors of the incident waves. If, for example, two pulses $\mathbf{k}_{12}$ and $\mathbf{k}_{13}$ (at frequencies ω_{12} and ω_{13}) act on the system, then the photons emitted by the system have wave vectors $\mathbf{k}_{12}$ (frequency ω_{12}), $\mathbf{k}_{13} - \mathbf{k}_{12}$ (frequency ω_{23}), and $\mathbf{k}_{13}$ (frequency ω_{13}). The action of one pulse produces an intense response in the same direction (at the other frequencies the radiation is incoherent and isotropic). Let us calculate the intensities of the photon echo after application of a pulse to an equilibrium system. We use the formulas (see[1])

$$\begin{aligned} J_{21}(\mathbf{k}') &= J_{21}^{(0)}(\mathbf{k}')\,\mathrm{Sp}\,I_-(\mathbf{k}')\,\rho I_+(\mathbf{k}'), \\ J_{32}(\mathbf{k}'') &= J_{32}^{(0)}(\mathbf{k}'')\,\mathrm{Sp}\,K_+(\mathbf{k}'')\,\rho K_-(\mathbf{k}''), \\ J_{31}(\mathbf{k}''') &= J_{31}^{(0)}(\mathbf{k}''')\,\mathrm{Sp}\,L_+(\mathbf{k}''')\,\rho L_-(\mathbf{k}'''), \end{aligned} \tag{15}$$

where ρ is the density matrix of the molecule system after the action of the pulses. The action of the pulse on the molecule is described by a unitary transformation in its energy space. The only difference between the action of the pulse on different molecules is that the time of arrival of the wave at the location of the molecule is different (the molecule locations are assumed fixed). After the action of a pulse of frequency ω_{13} (wave vector $\mathbf{k}_{13}$), for example, we have

$$\rho(t) = \exp(-iH_1t/\hbar)\,T_{13}\rho_0T_{13}^{-1}\exp(iH_1t/\hbar), \tag{16}$$

where ρ_0 is the equilibrium density matrix:

$$\rho_0 = \frac{\exp(-H_1/kT)}{\mathrm{Sp}\exp(-H_1/kT)} = \prod_{j=1}^{N}[\sigma_1P_1^{(j)} + \sigma_2P_2^{(j)} + \sigma_3P_3^{(j)}];$$

the quantities $\sigma_1 = \overline{N}_1/\overline{N}$, $\sigma_2 = \overline{N}_2/\overline{N}$, $\sigma_3 = \overline{N}_3/\overline{N}$ determine the level populations;

$$P_3^{(j)} = (1/3 - Y)^{(j)}, \quad P_1^{(j)} = 2L_3^{(j)} + P_3^{(j)}, \quad P_2^{(j)} = 2K_3^{(j)} + P_3^{(j)}$$

Rotation in L-spin space is described by the operator

$$T_{13} = \exp\Big[\frac{i}{\hbar}\sum_{j=1}^{N}H_1^{(j)}t_j\Big]\cdot$$

$$\prod_{s=1}^{N}\exp\Big[i\frac{\theta}{2}(L_-^{(j)}\beta + L_+^{(j)}\beta^*)\Big]\exp\Big[-\frac{i}{\hbar}\sum_{j=1}^{N}H_1^{(j)}t_j\Big],$$

which yields, after the substitution $t_j = \mathbf{k}_{13}\cdot\mathbf{r}_j/\omega_{13}$,

$$T_{13} = \exp\{{}^1\!/_2 i\theta[L_-(\mathbf{k}_{13})\beta + L_+(\mathbf{k}_{13})\beta^*]\}.$$

Here θ is the angle of rotation around the axis whose direction in the L-spin subspace is determined by a parameter β ($|\beta| = 1$) and which is perpendicular to the quantization axis (all other transformations in energy space have no meaning in this case). The parameter θ is proportional to the area of the pulse (the product of the amplitude by the duration); $\sin^2(\theta/2)$ is the probability of molecule excitation from the ground state to the third level.

Let us calculate the intensities of the echo signal following a succession of two pulses with frequencies ω_{12} (wave vector $\mathbf{k}_{12}$) and $\omega_{13}(\mathbf{k}_{13})$. The distance between pulses is neglected in comparison with the radiative decay time of the system. The density matrix after the action of the pulses is similar to (16), with a transformation matrix

$$T = T_{13}T_{12}, \quad T_{12} = \exp\{{}^1\!/_2 i\varphi[\alpha I_-(\mathbf{k}_{12}) + \alpha^*I_+(\mathbf{k}_{12})]\}.$$

It is convenient to represent the operator $T\rho_0T^{-1}$ in the form of a product

$$T\rho_0T^{-1} = \prod_{j=1}^{N}A_j \tag{17}$$

(the complicated expressions for A_j are given in[12]). Substitution of (17) in (15) yields (the details of the calculation are given in[12]):

$$\begin{aligned} J_{21}(\mathbf{k}') &= J_{21}^{(0)}(\mathbf{k}')N\{\sigma_1 + (\sigma_2 - \sigma_1)\cos^2(\varphi/2) \\ &\quad + {}^1\!/_4\sin^2\varphi(\sigma_2 - \sigma_1)^2\cos^2(\theta/2)\,[N\Gamma(\mathbf{k}_{12}, \mathbf{k}') - 1]\}, \\ J_{32}(\mathbf{k}'') &= J_{32}^{(0)}(\mathbf{k}'')N\{\sigma_3 + \sin^2(\theta/2)\,[\sigma_1 + (\sigma_2 - \sigma_1)\sin^2(\varphi/2) - \sigma_3] \\ &\quad + {}^1\!/_4\sin^2\varphi\sin^2(\theta/2)\,(\sigma_2 - \sigma_1)^2[N\Gamma(\mathbf{k}_{13} - \mathbf{k}_{12}, \mathbf{k}'') - 1]\}, \\ J_{31}(\mathbf{k}''') &= J_{31}^{(0)}(\mathbf{k}''')N\{\sigma_3 + \sin^2(\theta/2)\,[\sigma_1 - \sigma_3 + (\sigma_2 - \sigma_1)\sin^2(\varphi/2)] \\ &\quad + {}^1\!/_4\sin^2\theta[\sigma_3 - \sigma_1\cos^2(\varphi/2) - \sigma_2\sin^2(\varphi/2)]^2[N\Gamma(\mathbf{k}_{13}, \mathbf{k}''') - 1]\}. \end{aligned} \tag{18}$$

In (18) we have introduced the notation

$$\Gamma(\mathbf{p}_1, \mathbf{p}_2) = |\langle\exp\{i(\mathbf{p}_1 - \mathbf{p}_2)\mathbf{r}\}\rangle|^2,$$

where the symbol $\langle\ldots\rangle$ denotes averaging over the coordinates of all the particles; for systems of sufficiently large dimensions we have

$$\langle\exp\{i(\mathbf{p}_1 - \mathbf{p}_2)\mathbf{r}\}\rangle = \delta(\mathbf{p}_1 - \mathbf{p}_2).$$

It follows therefore that intensive coherent radiation is observed at the frequencies ω_{12}, ω_{23}, and ω_{13} in the directions $\mathbf{k}_{12}$, $\mathbf{k}_{13} - \mathbf{k}_{12}$, and $\mathbf{k}_{13}$, respectively. To obtain the time evolution of the echo signals, it is necessary to take the inhomogeneous broadening into account in the calculations (see[3]).

We present also formulas for the intensities following the action of a pulse of frequency ω_{23} (wave vector $\mathbf{k}_{23}$):

$$\begin{aligned} J_{32}(\mathbf{k}'') &= J_{32}^{(0)}(\mathbf{k}'')N\{\sigma_3 + (\sigma_2 - \sigma_3)\sin^2(\chi/2) \\ &\quad + ({}^1\!/_4)\sin^2\chi(\sigma_3 - \sigma_2)^2[N\Gamma(\mathbf{k}_{23}, \mathbf{k}'') - 1]\}, \\ J_{21}(\mathbf{k}') &= J_{21}^{(0)}(\mathbf{k}')N[\sigma_2 + (\sigma_3 - \sigma_2)\sin^2(\chi/2)], \\ J_{31}(\mathbf{k}''') &= J_{31}^{(0)}(\mathbf{k}''')N[\sigma_3 + (\sigma_2 - \sigma_3)\sin^2(\chi/2)]. \end{aligned} \tag{19}$$

Other cases of pulsed excitation of coherent radiation are considered in[12].

4. CONCLUSION

An examination of the case n = 3 reveals the main features of the multilevel problem. The classification of the states of a molecule system with n levels is in accord with the irreducible representations $D(p_1, p_2, \ldots, p_{n-1})$ of the group SU_n. The cooperative characteristics of the system are the indices of the irreducible representations. If the system is in a superradiant state, then the radiation intensities at all frequencies are proportional to N^2 (the total number of such transitions is $n(n - 1)/2$). The superradiant states can be excited by applying pulses to the system. If the wavelengths are shorter than the system dimensions, then the intensities of the responses can be easily obtained by generalizing the formalism used in Sec. 3. By reducing the n-level problem on the SU_3 subgroups contained in the SU_n group, it is easy to find the angular distributions of the photon-echo signals from the purely geometrical relations between the wave vectors.

It should be noted that a number of problems not touched upon here (equidistant molecules, quasiclassical states of the Glauber type for the multilevel Dicke problem, equations of motion for macroscopic polarization vectors, comparison with paramagnetic systems) are considered in[12]. In the future, besides the realization of the experimental possibilities ensuing from the present paper (simultaneous observation of superradiant states on several transitions, specific photon-echo effects), great interest attaches to the use of the formalism developed here for the study of cooperation effects in processes involving the interaction of light with matter and described by higher orders of perturbation theory.

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