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ON THE THEORY OF COHERENT SPONTANEOUS EMISSION

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Possible extensions of Dicke's[1] theory of coherent spontaneous emission to multi-level systems and procedures for taking account of emitter correlations are discussed. The experimental data[4] are interpreted as evidence for such correlations between the oscillations in molecular crystals.

A quantum theory of coherent spontaneous emission was developed by Dicke.[1] As the stationary states of a system of n molecules with two levels one may, according to this theory, take the eigenstates of the operator R^2 which commutes with the Hamiltonian $H = H_0 + \epsilon_{12}R_3$ (ϵ_{12} is the spacing between the levels):

$$R^2\psi_{rm} = r(r+1)\psi_{rm}, \qquad R_3\psi_{rm} = m\psi_{rm}, \tag{1}$$

where $m = (N_2 - N_1)/2$, i.e., m is equal to half the difference of the populations of the levels. The intensity of the spontaneous radiation I is determined by the matrix elements of the operator for the interaction between the molecules and the radiation field, and hence

$$I = I_0(r+m)(r-m+1), \tag{2}$$

where I_0 is the intensity of the spontaneous radiation from an isolated molecule. In all radiative transitions of the system the quantum number r does not change (R^2 is an integral of motion).

There is the possibility of the existence of particular bound states of the system where the intensity of the spontaneous radiation is proportional to the square of the number of molecules (superradiant states):

$$r = {}^1\!/_2 n, \qquad m = 0, \qquad I \sim {}^1\!/_4 n^2 I_0. \tag{3}$$

In the general case

$$I = I_0 \sum_{rm} P_{rm}(r+m)(r-m+1), \tag{4}$$

where P_{rm} is the probability for finding the system in the state (rm). A number of questions connected with the formation of the superradiant states, with the width and shape of the lines, and with the analysis of the properties of the coherent radiation, have been considered in various papers.[2] However, in all these papers only a two-level scheme and non-interacting molecules have been discussed. In the present paper we indicate ways of extending Dicke's method to the case of many levels and correlations between the emitters.

If the interaction with the field has resonance character, then, as a rule, the two-level scheme is a good approximation. However, in a number of problems (in particular, in the description of the interaction with very intense radiation fluxes, or in the investigation of the radiation of a system with account of the relaxation of the levels) it is important to take many levels into account. If, according to Dicke, the states of a system of molecules with two levels are described by the representations D(P) of the group SU_2, then systems with three, four, and more levels are described by the representations D(P0), D(P00), D(P000), ... of the corresponding groups SU_3, SU_4, SU_5, ...

The calculational apparatus of Dicke[1] is directly extended to the case of an arbitrary number of levels. Let us consider a three-level system with the energy intervals ϵ_{12}, ϵ_{13}, ϵ_{23} ($\epsilon_{12} + \epsilon_{23} = \epsilon_{13}$) between the levels 1, 2, and 3. The Hamiltonian for the case $\epsilon_{12} = \epsilon_{23}$ has the form

$$H = \varepsilon_{13}({}^3\!/_4 Y + {}^1\!/_2 t). \tag{5}$$

Here Y is the hypercharge; T and t are the isospin and its projection, with $t = (N_2 - N_1)/2$. The wave functions ψ_{PTt} differ by the three quantum numbers P, T, and t. It is easy to see that the number P remains unchanged in any radiative processes in a three-level system. The degree of coherence is given by the numbers T and P (or Y and P), in contrast to the two-level system, where the single cooperation number r was sufficient. The formulas for the intensity of the radiation are determined by these numbers:

$$I_{21} = I_0^{(21)}(T+t)(T-t+1), \qquad I_{31} = I_0^{(31)}(P-2T)(T+t+1),$$
$$I_{32} = I_0^{(32)}(P-2T)(T-t+1).$$

A detailed investigation of the relaxation process with account of relations of the type (6) may be very important for the analysis of the possibilities of using a multi-level system for excitation (cf.[3]) and also for the analysis of methods for making superradiant states.

It is interesting to note that the representations D(P) and D(P0) of the groups SU_2 and SU_3 are also described by vibrational levels with the number p for two- and threefold frequencies. The degrees of degeneracy $p+1$, $(p+1)(p+2)/2$ as well as the level splitting are determined purely by group theory relations. The density matrix formalism may also be translated into group theory language.

The second possibility of extending the Dicke method is connected with the account of the correlations of the radiating molecules owing to their interaction. Let us consider a two-level scheme. The coordination of the radiations of the molecules (in phase and in anti-phase) is taken into account with the help of a single parameter—the cooperation number r. The simplest model which includes correlations is in many respects reminiscent of a ferromagnet. The radiation of the molecules is correlated in groups: k emitters are coordinated (the cooperation number for this group is equal to k/2). Each of these groups radiates independently. The total spin r is the result of coupling n/k spins k/2 (in contrast to the coupling of n spins 1/2 in the Dicke model). Calculating the intensity according to (4) and comparing it with the experimental data, one can determine the number of correlated molecules (in particular cases, when all emitters are coordinated, and when there are two groups, three groups, etc., we obtain r = n/2, r = n/4, r ~ n/6.)

In this connection the experiment of Ryazanov and Sushchinskiĭ[4] is of great interest, who measured the absolute intensity of the combined scattering in the liquid and crystaline states of matter (stillbene, benzene). Here the intensity for the crystalline state was about 20 times greater than for the liquid state. Assuming that the emitters in the liquid are not correlated and using (4), we find the following relation for the intensities:

$$I_{\text{sol}}/I_{\text{liq}} \approx \sum_{rm} P_{rm}(r-m).$$

Estimates with the help of this formula indicate that under the conditions of this experiment (with various model assumptions) some ten molecules are correlated. This interpretation of the experiment brings out the importance of the work[4], since it opens the possibility of investigating a new range of phenomena connected with the interaction of vibrations in solid matter.

The present note is of qualitative nature. In the future it will certainly be necessary to make a detailed analysis of other possible interpretations, to propose an elementary model of the interaction, and also to perform further experiments (to determine the scattering characteristics, the temperature dependencies, and to extend the range of materials investigated).

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