Collective radiative effects in a two-level system

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Quantum field theory methods are used in a consideration of the dynamics of a two-level system which is coupled in thermodynamic equilibrium with the radiation field of a resonator. The collective radiative effects depend on the ratios of the following quantities: the resonator's length a, the size L of the atomic specimen, the temperature T of the system, the wavelength k^{-1} of the radiation, the frequency ω_0 of the atomic transitions, and the density ρ_S of atoms in the system. In the uniform-field case $k^{-1} \gg L$ the result of Faĭn [Sov. Phys. JETP 5, 501 (1957] are valid for the natural line width if $\omega_0 \gg 2\pi c/a$. Under such conditions equations are given for the collective radiation shift and for the change of the g factor. All three effects are proportional to the number of atoms in the resonator. The interaction of the atoms with the longitudinal component of the field gives an additional radiative shift. If, however, $\omega_0 \sim 2\pi c/a$, all collective radiative effects disappear, and the natural line width is determined only by the figure of merit Q of the resonator. For $k^{-1} \lesssim L$ a new radiative effect appears; this is a smearing out of the resonance frequencies into a band of width. For $\omega_0 < \mu^2 \rho_S$, where μ is the matrix element for the dipole transition, there exists a Bose-Einstein transition temperature for the condensation of photons of all modes of the radiation field of the resonator, independent of the wave vector and polarization of the mode, provided that the position of the atomic specimen does not destroy the symmetry of the resonator. Otherwise the interaction of the atoms with the field leads to an induced symmetry breaking and the phase transition does not occur.

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1. INTRODUCTION

An important achievement of quantum electrodynamics is the remarkable agreement with experiment of the theory of radiative effects for a single atom. For a set of atoms, however, radiation effects can be decidedly changed owing to exchange symmetry.[1] In particular, Faĭn,[2] in work based on results of Dicke,[1] on the spontaneous decay of a system of two-level atoms, showed that the natural width of the line from a gas of N atoms in thermodynamic equilibrium is

$$\gamma = N\hbar\omega_0\gamma_0/2kT. \tag{1}$$

Here ω_0 is the frequency of the atomic transition, γ_0 is the natural width of the line for a single atom, and T is the temperature of the gas. This result is derived on the assumption that the system of atoms interacts with only one mode of the radiation field of a resonance cavity and that its wavelength is much larger than the size of the two-level system ($k^{-1} \gg L$).

Equation (1) is a phenomenological formula, since γ_0 is itself the result of the interaction with the entire infinite set of modes of the radiation field. Doubt is aroused by the two-stage approach to the calculation of the natural line width, first for one atom, and then for the entire system. Besides this, some authors believe that in the case of thermodynamic equilibrium in a system of atoms there are no cooperative effects in spontaneous decay.[3]

We believe that a natural way to avoid these difficulties is to use the methods of quantum field theory in statistical physics.[4] In this paper we consider the dynamics of a two-level system which is in thermal equilibrium with all the radiation modes of a cavity resonator, for an arbitrary ratio of the sizes of the resonator and the system of atoms. The purpose of this work is to investigate the effects of collective action and of the temperature of the system on such well known radiative phenomena as the natural width of the line, the radiative shift, and the change of the g factor, which occur when the atomic system is affected uniformly by the field, and also to consider new radiative effects that arise in the case when the wavelength of the radiation field is less than the size of the system of atoms.

Particular attention is given to the dynamics of a system close to the temperature of the phase transition to the superradiative state,[5,6] which can occur when a threshold condition on the density of atoms in the resonator is satisfied. This phase transition is the Bose-Einstein condensation of the photons of the radiation field,[7] and therefore the thermodynamic phase of low symmetry is characterized by spontaneous coherence of the field.[8,9] We shall call this phase transition the transition to the spontaneous coherent state (SCS). The connection between the Bose-Einstein condensation and the spontaneous appearance of a coherent state in a Bose system has also been considered in Refs. 10–12.

In studying radiative and thermodynamic effects in a two-level system the Hamiltonian usually used is that of Dicke, in which the interaction is of the following form:

$$N^{-1/2}\sum_{j=1}^{N}\sum_{\mathbf{k}\sigma}\lambda_{\mathbf{k}\sigma}(S_j^+ b_{\mathbf{k}\sigma}e^{i\mathbf{k}\mathbf{r}_j} + S_j^+ b_{\mathbf{k}\sigma}^+ e^{-i\mathbf{k}\mathbf{r}_j}) + \text{H.c.},$$

where $S_j^\pm$ are atomic operators, and $b_{\mathbf{k}\sigma}$ and $b_{\mathbf{k}\sigma}^+$ are field operators. Since in the resonator there is already present a longitudinal component of the electromagnetic field, it is necessary to take into account also the longi-

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tudinal interaction of the atoms with the radiation field, $S_j^z(b_{\mathbf{k}\sigma}+b_{\mathbf{k}\sigma}^+)$. The longitudinal interaction is of no particular interest from the point of view of radiative effects, but it is essential in treating the phase transition to the SCS. For $T \leqslant T_c$, owing to the Bose condensation of the photons, a spontaneous macroscopic field arises, which breaks the high-temperature symmetry of the system. However, if the location of the atoms in the resonator is such that this symmetry is already removed, according to the theory of phase transitions of the second kind as a theory of spontaneous symmetry breaking,[14] the phase transition in the system atoms +field must disappear. Owing to the longitudinal interaction, this is the situation in the present case.

When the atoms and the field are in thermal equilibrium with each other, the singling out of a single mode or a finite set of modes of the field is improper. Obviously the singling out of "one mode" of the atomic subsystem, i.e., the approximation of an actual many-level atom with a two-level system is equally improper. Nevertheless there are actual systems to which the present treatment can be applied, namely paramagnetic systems in a resonator. Therefore a number of the formulas and diagrams in the present paper refer to these systems.

2. THE GENERAL APPROACH

Applying the procedure of quantizing the electromagnetic field in a resonator in terms of standing waves,[15] we write the magnetic field in the form ($\hbar=1$)

$$\mathbf{H}(\mathbf{r})=-N^{-1/2}\sum_{\mathbf{k}\sigma}\left(\omega_{\mathbf{k}\sigma}\frac{\rho}{2}\right)^{1/2}\varphi_{\mathbf{k}\sigma}H_{\mathbf{k}\sigma}(\mathbf{r}), \tag{2}$$

where $\omega_{\mathbf{k}\sigma}$ are the eigenfrequencies of the resonator with volume V, $\rho=N/V$ is the density of atoms in the resonator, and

$$\varphi_{\mathbf{k}\sigma}=b_{\mathbf{k}\sigma}+b_{\mathbf{k}\sigma}^+, \tag{3}$$

where $b_{\mathbf{k}\sigma}, b_{\mathbf{k}\sigma}^+$ are photon operators for the field mode with propagation vector $\mathbf{k}$ and polarization σ. The functions $\mathbf{H}_{\mathbf{k}\sigma}(\mathbf{r})$ satisfy the normalization condition

$$\int_V d^3\mathbf{r}\,\mathbf{H}_{\mathbf{k}\sigma}(\mathbf{r})\mathbf{H}_{\mathbf{k}'\sigma'}(\mathbf{r})=4\pi\delta_{\mathbf{k}\mathbf{k}'}\delta_{\sigma\sigma'}. \tag{4}$$

The electric field $\mathbf{E}(\mathbf{r})$ is quantized analogously.

When direct spin-spin interactions are neglected, the Hamiltonian of the system of spins plus radiation field is

$$\mathscr{H}=\sum_{\mathbf{k}\sigma}\omega_{\mathbf{k}\sigma}b_{\mathbf{k}\sigma}^+b_{\mathbf{k}\sigma}-\omega_0\sum_{j=1}^{N}S_j^z-g\mu_B\sum_{j=1}^{N}\mathbf{S}_j\mathbf{H}(\mathbf{r}_j). \tag{5}$$

Going over to the fermion representation for spin $\frac{1}{2}$, we write

$$\mathscr{H}=\sum_{\mathbf{k}\sigma}\omega_{\mathbf{k}\sigma}b_{\mathbf{k}\sigma}^+b_{\mathbf{k}\sigma}-\frac{\omega_0}{2}\sum_{j=1}^{N}(a_{j2}^+a_{j2}-a_{j1}^+a_{j1})$$
$$+N^{-1/2}\sum_{\mathbf{k}\sigma}\sum_{j=1}^{N}\sum_{\alpha,\beta=1,2}\Lambda_{k\sigma}^{\alpha\beta}(\mathbf{r}_j)a_{j\alpha}^+a_{j\beta}\varphi_{k\sigma}, \tag{6}$$

where a_{j1}, a_{j1}^+ are operators for the lower energy level, and a_{j2}, a_{j2}^+ are those for the upper level.

$$\Lambda_{k\sigma}^{\alpha\beta}(\mathbf{r})=\begin{pmatrix}-\xi_{\mathbf{k}\sigma}(\mathbf{r}) & \lambda_{\mathbf{k}\sigma}(\mathbf{r})\\ \lambda_{\mathbf{k}\sigma}^*(\mathbf{r}) & \xi_{\mathbf{k}\sigma}(\mathbf{r})\end{pmatrix},$$
$$\lambda_{\mathbf{k}\sigma}(\mathbf{r})=-{}^1/_2\mu(\omega_{\mathbf{k}\sigma}\rho/2)^{1/2}(H_{\mathbf{k}\sigma}^x(\mathbf{r})-iH_{\mathbf{k}\sigma}^y(\mathbf{r})),$$
$$\xi_{\mathbf{k}\sigma}(\mathbf{r})=-{}^1/_2\mu(\omega_{\mathbf{k}\sigma}\rho/2)^{1/2}H_{\mathbf{k}\sigma}^z, \tag{7}$$

with $\mu=g\mu_B$. This is precisely the same as the form of the fermion representation of the Hamiltonian of a system of two-level atoms plus the field, the only exception being that for the latter the interaction is written in the form

$$iN^{-1/2}\sum_{\mathbf{k}\sigma}\sum_{j=1}^{N}\sum_{\alpha,\beta}\left(\omega_{\mathbf{k}\sigma}\frac{\rho}{2}\right)^{1/2}\mathbf{d}_{\alpha\beta}E_{\mathbf{k}\sigma}(\mathbf{r}_j)a_{j\alpha}^+a_{j\beta}(b_{\mathbf{k}\sigma}-b_{\mathbf{k}\sigma}^+), \tag{8}$$

where $\mathbf{d}_{\alpha\beta}$ are the matrix elements of the dipole moment of the atom.

Let us introduce the temperature-dependent photon Green's functions

$$D(\mathbf{k}\sigma,\mathbf{k}'\sigma';\omega_m)=\frac{1}{\beta}\int_0^\beta e^{i\omega_m\tau}\langle T\{\varphi_{\mathbf{k}\sigma}(\tau)\varphi_{\mathbf{k}'\sigma'}(0)\}\rangle\,d\tau \tag{9}$$

and the fermion functions

$$G_{\alpha\beta}(\mathbf{r}_j,\omega_n)=\frac{1}{\beta}\int_0^\beta e^{i\omega_n\tau}\langle T\{a_{j\beta}(\tau)a_{j\alpha}^+(0)\}\rangle\,d\tau, \tag{10}$$

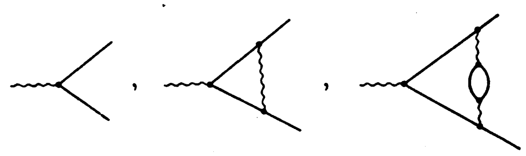
with $\omega_m=2\pi m/\beta$, $\omega_n=\pi(2n+1)/\beta$, $m,n=0,\pm1,\ldots.$ In diagrams the photon Green's functions will be represented by wavy lines and the fermion functions, by solid lines. An ordinary arrow on a fermion line will indicate the lower atomic state $|1\rangle$, and an open arrow, the upper state $|2\rangle$. According to Eq. (6) the zeroth-order fermion Green's functions are

$$G_{11}^0(\omega_n)=-(i\omega_n+\omega_0/2)^{-1},\quad G_{22}^0(\omega_n)=-(i\omega_n-\omega_0/2)^{-1}. \tag{11}$$

The zeroth-order Green's functions of the field are

$$D_0(\mathbf{k}\sigma,\omega_m)=-2\omega_{\mathbf{k}\sigma}/[(i\omega_m)^2-\omega_{\mathbf{k}\sigma}^2]. \tag{12}$$

Formally the number of modes of the radiation field of the resonance cavity in interaction with the system of atoms is infinite. Actually the number of modes is bounded, since the boundary conditions with which the quantization of the field in the resonator is carried out are violated for frequencies larger than the plasma frequency $\omega_p\sim10^{16}$. This makes it possible to retain only the bare part of the vertex function. For example, the condition that the second graph in the expression



(13)

be small compared with the first is as follows:

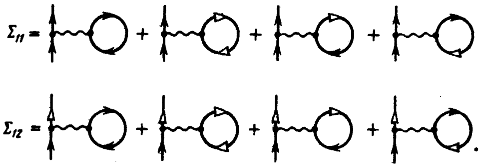
$$\mu^2\rho kTn/N\omega_0^2\ll1, \tag{14}$$

where $n\sim V\omega_p^3/\pi^3c^3$ is the number of modes of the radiation field in the resonator. For the region interesting for our problem, temperatures $kT\sim\omega_0$ and atomic frequencies $\omega_0\sim\rho\mu^2$, the inequality (14) becomes $n\ll N$ or $\rho\gg10^{15}$. Comparison of the second and third graphs gives a similar inequality.

Accordingly the Dyson equations take the form

$$G_{11}=(G_{22}^{0^{-1}}-\Sigma_{22})/R,$$
$$G_{12}=\Sigma_{12}/R, \tag{15}$$
$$R=(G_{11}^{0^{-1}}-\Sigma_{11})(G_{22}^{0^{-1}}-\Sigma_{22})-|\Sigma_{12}|^2,$$

where



(16)

The existence of nondiagonal (anomalous) Green's functions G_{12} and mass operators (16) is possible only because of the longitudinal interactions in the Hamiltonian (6).

Using the relations between the fermion Green's functions and the spin polarizations[9]

$$\langle S_j^+\rangle=-\frac{1}{\beta}\sum_n G_{21}(\mathbf{r}_j,\omega_n)\exp[-i\omega_n(\tau\to-0)],$$
$$2\langle S_j^z\rangle=-\frac{1}{\beta}\sum_n [G_{22}(\mathbf{r}_j,\omega_n)-G_{11}(\mathbf{r}_j,\omega_n)]\exp[-i\omega_n(\tau\to-0)], \tag{17}$$

we can readily write out the mass operators (16) in the following form:

$$\Sigma_{11}(\mathbf{r})=-\sum_{\mathbf{k}\sigma}\xi_{\mathbf{k}\sigma}(\mathbf{r})c_{\mathbf{k}\sigma}, \tag{18}$$
$$\Sigma_{12}(\mathbf{r})=\Delta=-\sum_{\mathbf{k}\sigma}\lambda_{\mathbf{k}\sigma}(\mathbf{r})c_{\mathbf{k}\sigma},$$

where

$$c_{\mathbf{k}\sigma}=-\frac{2}{\omega_{\mathbf{k}\sigma}N}\sum_j[\lambda_{\mathbf{k}\sigma}(\mathbf{r}_j)\langle S_j^+\rangle+\lambda_{\mathbf{k}\sigma}^*(\mathbf{r}_j)\langle S_j^-\rangle+2\xi_{\mathbf{k}\sigma}(\mathbf{r}_j)\langle S_j^z\rangle]. \tag{19}$$

From the equations of motion for the field variables it follows that

$$c_{\mathbf{k}\sigma}=\langle\varphi_{\mathbf{k}\sigma}N^{-1/2}\rangle.$$

Substituting Eqs. (11) and (18) in Eq. (15), we get

$$G_{11}(\mathbf{r},\omega_n)=-\frac{i\omega_n-E(\mathbf{r})}{(i\omega_n)^2-E^2(\mathbf{r})-|\Delta(\mathbf{r})|^2},$$
$$G_{12}(\mathbf{r},\omega_n)=\frac{\Delta(\mathbf{r})}{(i\omega_n)^2-E^2(\mathbf{r})-|\Delta(\mathbf{r})|^2}. \tag{20}$$

The quantity

$$E(\mathbf{r})=\frac{\omega_0}{2}-\sum_{\mathbf{k}\sigma}\xi_{\mathbf{k}\sigma}(\mathbf{r})\left\langle\frac{\varphi_{\mathbf{k}\sigma}}{N^{1/2}}\right\rangle, \tag{21}$$

has the meaning of an effective longitudinal field, as can be seen from the Hamiltonian (6) or (8), and the quantity

$$(E^2(\mathbf{r})+|\Delta(\mathbf{r})|^2)^{1/2}=\omega_0\eta(\mathbf{r})/2, \tag{22}$$

is obviously the total effective magnetic field acting on a spin located at the point r (or the total electric field acting on the atom).

To obtain the equations for self-consistency, we substitute the Green's functions (20) in Eq. (17). Then from Eqs. (19) we find

$$c_{\mathbf{k}\sigma}=-\frac{2}{N}\sum_j\frac{1}{\omega_{\mathbf{k}\sigma}\eta(\mathbf{r}_j)}\operatorname{th}\frac{\beta\omega_0\eta(\mathbf{r}_j)}{2}\Big\{\xi_{\mathbf{k}\sigma}(\mathbf{r}_j)$$
$$-\sum_{\mathbf{k}'\sigma'}[\lambda_{\mathbf{k}\sigma}(\mathbf{r}_j)\lambda_{\mathbf{k}'\sigma'}^*(\mathbf{r}_j)+\xi_{\mathbf{k}\sigma}(\mathbf{r}_j)\xi_{\mathbf{k}'\sigma'}(\mathbf{r}_j)+\text{c.c.}]\Big\}. \tag{23}$$

Then from the usual rules of the diagram technique we write the diagram equation for the photon Green's function:[9]

(24)

If we introduce the matrices

$$\mathbf{D}(\omega_m)=D(\mathbf{k}\sigma,\mathbf{k}'\sigma';\omega_m),$$
$$\mathbf{D}_0(\omega_m)=D_0(\mathbf{k}\sigma,\omega_m)\delta_{\mathbf{k}\mathbf{k}'}\delta_{\sigma\sigma'},$$
$$\Xi(\omega_m)=\frac{1}{N}\sum_j\sum_{\alpha\beta\gamma\delta}\Lambda_{\mathbf{k}\sigma}^{\alpha\beta}(\mathbf{r}_j)\Pi_{\alpha\beta\gamma\delta}(\mathbf{r}_j,\omega_m)\Lambda_{\mathbf{k}'\sigma'}(\mathbf{r}_j), \tag{25}$$
$$\Pi_{\alpha\beta\gamma\delta}(\mathbf{r}_j,\omega_m)=\frac{-1}{\beta}\sum_n G_{\delta\alpha}(\mathbf{r}_j,\omega_n)G_{\beta\gamma}(\mathbf{r}_j,\omega_n+\omega_m), \tag{26}$$

Eq. (24) takes the simple form

$$\mathbf{D}=\mathbf{D}_0+\mathbf{D}_0\Xi\mathbf{D}. \tag{27}$$

In the one-mode case the temperature at which the photon Green's function with $\omega_m=0$ has a singularity gives the temperature of the Bose-Einstein condensation.[7] Generalizing this condition to the many-mode case, we find from Eq. (27) the following equation for the temperature of the Bose-Einstein condensation T_c:

$$\operatorname{Det}[\Xi(0)-\mathbf{D}_0^{-1}(0)]=0. \tag{28}$$

The transverse susceptibility of the system is determined by analytic continuation of the polarization Green's function[4,9]

$$\chi_{+-}(\omega_m)=\sum_{ij}\chi_{ij}(\omega_m),$$
$$\chi_{ij}(\omega_m)=\frac{1}{\beta}\int_0^\beta e^{i\omega_m\tau}\langle T\{a_{i2}^+(\tau)a_{i1}(\tau)a_{j1}^+(0)a_{j2}(0)\}\rangle d\tau.$$

In analogy with the photon Green's function (24) we can write out the equation for the polarization function

$$\chi_{ij}^{\alpha\beta}(\omega_m)=\Pi_{\alpha\beta21}(\mathbf{r}_i,\omega_m)\delta_{ij}+\frac{1}{N}\sum_l\sum_{\mathbf{k}\sigma}\sum_{\gamma\delta\mu\nu}\Pi_{\alpha\beta\gamma\delta}(\mathbf{r}_i,\omega_m)$$
$$\times\Lambda_{\mathbf{k}\sigma}^{\gamma\delta}(\mathbf{r}_i)D_0(\mathbf{k}\sigma,\omega_m)\Lambda_{\mathbf{k}\sigma}^{\mu\nu}(\mathbf{r}_l)\chi_{lj}^{\mu\nu}(\omega_m), \tag{29}$$
$$\chi^{12}=\chi_{+-}.$$

Let us introduce the notation

$$X_{\mathbf{k}\sigma}(\mathbf{r}_j,\omega_m)=\frac{1}{N}\sum_l\sum_{\mu\nu}\Lambda_{\mathbf{k}\sigma}^{\mu\nu}(\mathbf{r}_l)\chi_{lj}^{\mu\nu}.$$

Then Eq. (29) takes the form of a system of linear algebraic equations

$$\sum_{\mathbf{k}'\sigma'}[D_0(\mathbf{k}\sigma,\omega_m)\Xi_{\mathbf{k}\sigma,\mathbf{k}'\sigma'}(\omega_m)-\delta_{\mathbf{k}\mathbf{k}'}\delta_{\sigma\sigma'}]X_{\mathbf{k}'\sigma'}(\mathbf{r},\omega_m)$$
$$=\sum_{\alpha\beta}\Pi_{\alpha\beta21}(\mathbf{r},\omega_m)\Lambda_{\mathbf{k}\sigma}^{\alpha\beta}(\mathbf{r}). \tag{30}$$

Accordingly the resonance frequencies can be found as the roots of the equation

$$\operatorname{Det}[\Xi(\Omega)-\mathbf{D}_0^{-1}(\Omega)]=0, \tag{31}$$

where $\Xi(\Omega)$ is defined in Eq. (25). Comparison of Eqs. (28) and (31) shows that the spectrum of the collective excitations must include a soft mode.

Because of the Bose condensation of the photons a nonvanishing average field $\langle\varphi N^{-1/2}\rangle$ appears for T

$\leq T_c$.[16] However, if the first sum on the right side of Eq. (23) is not equal to zero, a condensate of the field exists at arbitrary temperature. Accordingly, the longitudinal interaction can make the phase transition to a SCS forbidden, because at any temperature the system is in a state with induced symmetry breaking.

3. THE UNIFORM-FIELD CASE $k^{-1} \gg L$

Let us suppose that the size of the atomic specimen is much smaller than the wavelength of any mode of the radiation field in the resonator. Clearly this is an unrealistic condition, since it would require an enormous density of two-level atoms in a specimen of microscopic dimensions. Nevertheless, this limiting case is interesting for two reasons. First, it can be compared with Faĭn's result, Eq. (1), and second, it models the experimental situation when the dimensions of an atomic or paramagnetic specimen are much smaller than those of the resonator.

Let us find the coupling constants between the radiation field of a resonator and an atomic system. Here we take the rectangular resonator with sides a, b, and d. The structure of the field in such a resonator has been shown, for example, in Poole's book.[17] Normalizing the field according to Eq. (4), we have from (7) for TE waves

$$\lambda_{k1}(\mathbf{r}) = 2\mu(\pi\omega_k\rho)^{1/2}\frac{k_z/k}{(k_x^2+k_y^2)^{1/2}}(k_x \sin k_x x \cos k_y y - ik_y \cos k_x x \sin k_y y)\cos k_z z,$$
$$\xi_{k1}(\mathbf{r}) = -2\mu(\pi\omega_k\rho)^{1/2}(k_x^2+k_y^2)^{1/2}k^{-1}\cos k_x x \cos k_y y \sin k_z z; \tag{32}$$

and for TM waves

$$\lambda_{k2}(\mathbf{r}) = -2\mu(\pi\omega_k\rho)^{1/2}(k_x^2+k_y^2)^{-1/2}(k_x \cos k_x x \sin k_y y - ik_y \sin k_x x \cos k_y y)\cos k_z z,$$
$$\xi_{k2}(\mathbf{r}) = 0, \tag{33}$$

where $k_x = \pi m/a$, $k_y = \pi n/b$, $k_z = \pi p/d$; m, n, and p are positive integers.

We give the solutions of the compatibility equation (23) for three cases: 1. A paramagnetic system is placed in such a way that $\xi_{k1}(\mathbf{r}) = 0$. One can see easily from Eq. (32) how this can be done. As for a system of two-level atoms, this condition is fulfilled if the diagonal matrix element of the dipole moment is equal to zero. 2. The weak-coupling approximation, with

$$\sum_{k\sigma} |\lambda_{k\sigma}(\mathbf{r})|^2/\omega_0\omega_{k\sigma} \ll 1. \tag{34}$$

3. The one-mode approach.

1. In the case in which the longitudinal interactions are equal to zero, the mass operators (16) vanish, and the polarization operators can be calculated by means of the Green's functions (11). The result contains only the following polarization diagrams:

$$\Pi(\omega_m) = \Pi_{1221}(\omega_m) = \Pi_{2112}(-\omega_m) = -\text{th}(\beta\omega_0/2)(i\omega_m-\omega_0)^{-1}, \tag{35}$$
$$\pi(\omega_m) = \Pi_{1111}(\omega_m) = \Pi_{2222}(\omega_m) = \begin{cases} 0, & m \neq 0 \\ {}^1/_2\beta\,\text{ch}^{-2}(\beta\omega_0/2), & m = 0 \end{cases}.$$

For $m \neq 0$ the matrix (25) takes the simple form

$$\Xi_{k\sigma,k'\sigma'}(\omega_m) = \lambda_{k\sigma}(\mathbf{r})\lambda^*_{k'\sigma'}(\mathbf{r})\Pi(\omega_m) + \lambda^*_{k\sigma}(\mathbf{r})\lambda_{k'\sigma'}(\mathbf{r})\Pi(-\omega_m).$$

Substituting this expression in Eq. (31), we find

$$\begin{vmatrix} 1+a_1 & 1 & 1 & \dots \\ 1 & 1+a_2 & 1 & \dots \\ 1 & 1 & 1+a_3 & \dots \\ \dots & \dots & \dots & \dots \end{vmatrix} = 0, \tag{36}$$

where according to Eqs. (12) and (35)

$$a_n = a_{k\sigma}(\Omega) = -(\Omega^2-\omega_0^2)(\Omega^2-\omega_{k\sigma}^2)/4\omega_0\omega_{k\sigma}|\lambda_{k\sigma}(\mathbf{r})|^2\text{th}(\beta\omega_0/2).$$

Expanding the determinant (36) into a sum, we get the equation for the resonance frequencies of the system atoms plus radiation field:

$$\Omega^2-\omega_0^2 = 4\,\text{th}\frac{\beta\omega_0}{2}\sum_{k\sigma}\frac{|\lambda_{k\sigma}|^2\omega_0\omega_{k\sigma}}{\Omega^2-\omega_{k\sigma}^2}. \tag{37}$$

If the characteristic dimension a of the resonator is such that the frequencies Ω and ω_0 are comparable with $2\pi c/a$, then, as can be seen from the graphical solution of Eq. (37) shown in Fig. 1,

$$\chi_{+-}(\Omega) = \sum_{k\sigma}\alpha_{k\sigma}\delta(\Omega-\Omega_{k\sigma}),$$

where $\Omega_{k\sigma}$ are the points of intersection of the curves in Fig. 1. Accordingly, in this case the natural broadening is determined only by the figure of merit of the resonator.

If, however, $\Omega \sim \omega_0 \gg 2\pi c/a$, we can replace the sum over $k\sigma$ with an integral[18]

$$\sum_{k\sigma} = \frac{V}{\pi^3}\int k^2\,dk\,d\cos\theta\,d\varphi.$$

The result then is that the numerator in Eq. (37) takes the form

$$\Omega^2-\omega_0^2(1+2\delta/\omega_0-2i\gamma/\omega_0),$$

where

$$\gamma = N\frac{128}{3}\frac{\mu^2}{c^3}\Omega^3\,\text{th}\frac{\beta\omega_0}{2}, \tag{38}$$

$$\delta = N\frac{64}{3\pi}\frac{\mu^2}{c^3}\text{th}\frac{\beta\omega_0}{2}\,\text{P}\int\frac{\omega_k^3\,d\omega_k}{\Omega-\omega_k}. \tag{39}$$

Equation (38) is the expression for the collective radiative broadening of the line, and Eq. (39) is the collective radiative line shift. The change of the g factor is obviously given by

$$\Delta g = g\delta/\omega_0, \tag{40}$$

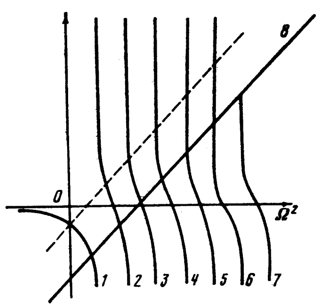


FIG. 1. Graphical solution of Eq. (33). Curves 1–7 give the function $4\sum_{k\sigma}|\lambda_{k\sigma}|^2\omega_0\omega_{k\sigma}/[\Omega^2-\omega_{k\sigma}^2]$; curve 8 is the function $(\Omega^2-\omega_0^2)\text{th}(\beta\omega_0/2)$. The dashed straight line corresponds to the temperature T_c of the Bose condensation.

where g is the Landé factor for an isolated paramagnetic atom.

For the line width (38) to have meaning, it is necessary that

$$\gamma \ll \omega_0.$$

In the radiofrequency range $\omega_0 \sim 10^{10}$ this condition is satisfied for $N \ll 10^{24}$, i.e., practically always. In the optical region, on the other hand, this condition leads to the inequality $N \ll 10^7$, which violates the condition for the applicability of the theory in question, Eq. (14).

Since $\xi_{\mathbf{k}\sigma}=0$, the thermodynamic averages of the field quantities vanish, $\langle \varphi_{\mathbf{k}\sigma} N^{-1/2}\rangle = 0$. They can appear spontaneously, however, for $T \leqslant T_c$. We find the temperature of the Bose condensation from Eq. (37) by setting $\Omega = 0$:

$$4\,\mathrm{th}\frac{\omega_0}{2kT_c}=\omega_0 \Big/ \sum_{\mathbf{k}\sigma} \frac{|\lambda_{\mathbf{k}\sigma}(\mathbf{r})|^2}{\omega_{\mathbf{k}\sigma}}.$$

This result was first derived by Wang and Hioe.[6]

For $kT_c > \omega_0$ we have from Eq. (29), to good accuracy,

$$kT_c \approx 4\pi\mu^2\rho \sum_{\mathbf{k}} \left(\frac{k_z^2}{k^2}+1\right).$$

Since the argument of this section is based on the uniform-field assumption $k^{-1} \gg L$, the sum over k actually has $(a/L)^3$ as an effective upper limit. Consequently,

$$kT_c \sim 4\pi\mu^2\rho_s, \tag{41}$$

where ρ_S is the density of atoms in the specimen. In a previous paper[19] it was shown by simple examples that modes with $k^{-1} \lesssim L$ give no contribution to T_c.

2. *Weak coupling approximation.* It follows from Eqs. (32) and (33) that an inequality analogous to (34) also holds for the longitudinal coupling constants. Then from Eq. (22) we get

$$c_{\mathbf{k}\sigma} \approx -\frac{2\xi_{\mathbf{k}\sigma}(\mathbf{r})}{\omega_0}\,\mathrm{th}\frac{\beta\omega_0}{2},$$

and Eqs. (21) and (22) take the following form:

$$\frac{\omega_0}{2}\eta(\mathbf{r}) = E(\mathbf{r}) \approx \frac{\omega_0}{2}\left(1+4\,\mathrm{th}\frac{\beta\omega_0}{2}\sum_{\mathbf{k}\sigma}\frac{\xi_{\mathbf{k}\sigma}^2}{\omega_0\omega_{\mathbf{k}\sigma}}\right). \tag{42}$$

If we substitute Eq. (42) in the Green's function (20) and use the fact that $\Delta \approx 0$, it is not hard to guess that all the results of this approach can be obtained from Eq. (37) by replacing $\omega_0/2$ with $E(\mathbf{r})$. Accordingly, as before the natural width will be of the form shown in Eq. (38), and to the radiative shift (39) and the g-factor correction (40) we must add the quantity

$$2\,\mathrm{th}\frac{\beta\omega_0}{2}\sum_{\mathbf{k}\sigma}\frac{\xi_{\mathbf{k}\sigma}^2(\mathbf{r})}{\omega_{\mathbf{k}\sigma}}. \tag{43}$$

Thus, unlike the natural line width, the radiative line shift depends on the position of the two-level system in the resonator.

3. *The one-mode approach.* This approach is only a model, but it enables us to elucidate some features of the influence of the longitudinal interaction for the case of an arbitrary coupling between the two-level system and the radiation field. The compatibility condition (23) takes the following form:

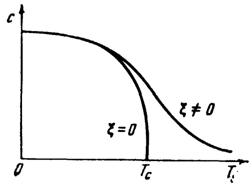


FIG. 2. Temperature dependence of the mean field amplitude $\langle \varphi N^{-1/2}\rangle$.

$$c\left(1-\frac{2}{\eta}\frac{|\lambda|^2+\xi^2}{\omega_0\omega}\,\mathrm{th}\frac{\beta\omega_0\eta}{2}\right) = -\frac{\xi}{\omega\eta}\,\mathrm{th}\frac{\beta\omega_0}{2}\eta, \tag{44}$$

where

$$\eta = \left[\left(1-\frac{2\xi c}{\omega_0}\right)^2 + 4\frac{|\lambda|^2c^2}{\omega_0^2}\right]^{1/2}.$$

For simplicity the subscripts for the mode and the radius vector have not been written explicitly.

If we set $\xi=0$ in Eq. (44), we get an equation of state which has been found in a number of papers.[5,6,9] The solution of Eq. (44) is shown in Fig. 2. The smaller the value of ξ, the closer the behavior of the mean field $\langle \varphi N^{-1/2}\rangle$ as a function of temperature comes to spontaneous appearance. We call attention to the fact that for $\omega_0=0$ the equation of state (44) goes over into an equation of the Curie-Weiss type:

$$c = \frac{(|\lambda|^2+\xi^2)^{1/2}}{\omega}\,\mathrm{th}\frac{(|\lambda|^2+\xi^2)^{1/2}}{2kT}c$$

with the temperature of the phase transition given by

$$kT_c = (\xi^2+|\lambda|^2)/2\omega.$$

4. COMPLETE FILLING OF THE RESONATOR, $L \gtrsim k^{-1}$

Let us consider another limiting case, in which the resonator is filled completely with atoms with macroscopically uniform density. If we take spins as the two-level objects, a paramagnetic-crystal resonator is a practical realization of this case. In the normal unordered thermodynamic phase $T \leqslant T_c$ a system of two-level atoms coupled with the radiation field of a rectangular resonator must be invariant under a number of discrete transformations, for example, rotation through 180° around the z axis, or an inversion of coordinates, $x \to -x$ and $y \to -y$. We recall that the external field along which the spin is quantized is in the z direction. In the case of a cylindrical resonator the Hamiltonian (6) must obviously be invariant under rotations around the z axis. The thermodynamic averages $\langle S_x\rangle$ and $\langle S_y\rangle$ must be equal to zero, because there is no preferred direction in a plane perpendicular to the z axis. In fact, according to the solution of the Dyson equation (15) we find that $\Sigma_{11}=\Sigma_{12}=0$. Then it follows at once from Eq. (17) that there is no polarization in the normal phase and no transverse field, these being effects that can arise only owing to spontaneous symmetry breaking in the system during a phase transition.[8,9]

If, however, the atoms are located in the resonator

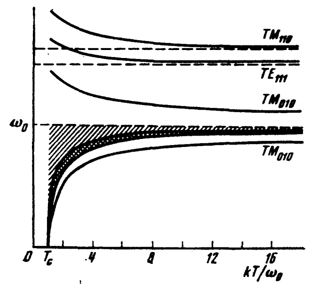


FIG. 3. Temperature spectrum of resonance frequencies of a two-level system placed in a cylindrical resonator. Natural frequencies of the resonator are shown as dashed straight lines. The length of the resonator is equal to its radius, and $\varkappa = \frac{1}{2}$, $\omega_0 = \omega^{TM010}$.

in such a way that there are no such symmetries as those mentioned above, the diagrams (16) are different from zero. There is no phase transition, since, as can be seen from Eq. (23), an order parameter exists at any temperature; it is the constant field given by the linear superposition of the $\langle \varphi_{\mathbf{k}\sigma} N^{-1/2} \rangle$. All of this is in agreement with Landau's theory of phase transitions of the second kind, considered as a theory of spontaneous symmetry breaking.[14]

Substituting Eq. (35) in the matrix (25), after summing over the atoms for $\Omega \neq 0$ we find

$$\Xi_{\mathbf{k}1,\mathbf{k}1}(\Omega) = {}^1/_2 \pi\mu^2\omega_{\mathbf{k}}\rho (k_z^2/k^2)[\Pi(\Omega)+\Pi(-\Omega)],$$
$$\Xi_{\mathbf{k}2,\mathbf{k}2}(\Omega) = {}^1/_2 \pi\mu^2\omega_{\mathbf{k}}\rho [\Pi(\Omega)+\Pi(-\Omega)], \tag{45}$$

with all of the other elements equal to zero.[1)] For $\Omega = 0$,

$$\Xi_{\mathbf{k}1,\mathbf{k}1}(0) = \pi\mu^2\omega_{\mathbf{k}}\rho(k_z^2/k^2)\Pi(0) + \pi\mu^2\omega_{\mathbf{k}}\rho(1-k_z^2/k^2)\pi(0),$$
$$\Xi_{\mathbf{k}2,\mathbf{k}2}(0) = \pi\mu^2\omega_{\mathbf{k}}\rho\Pi(0). \tag{46}$$

Substituting (45) into Eq. (31), we find the resonance frequencies

$$\Omega_{\mathbf{k}1}^2 = \frac{\omega_0^2+\omega_{\mathbf{k}1}^2}{2} \pm \left[\left(\frac{\omega_0^2-\omega_{\mathbf{k}1}^2}{2}\right)^2 + \omega_0\omega_{\mathbf{k}1}^2 2\pi\mu^2\rho \frac{k_z^2}{k^2} \operatorname{th}\frac{\beta\omega_0}{2}\right]^{1/2},$$
$$\Omega_{\mathbf{k}2}^2 = \frac{\omega_0^2+\omega_{\mathbf{k}2}^2}{2} \pm \left[\left(\frac{\omega_0^2-\omega_{\mathbf{k}2}^2}{2}\right)^2 + \omega_0\omega_{\mathbf{k}2}^2 2\pi\mu^2\rho \operatorname{th}\frac{\beta\omega_0}{2}\right]^{1/2}. \tag{47}$$

By sorting all of the states of the radiation field according to $\mathbf{k}\sigma$, we cannot construct the temperature spectrum of resonance frequencies of the system for $T \geq T_c$, shown in Fig. 3. This spectrum has the following features.

1) The spectrum of the frequencies $\Omega_{\mathbf{k}2}$ given by the interaction of the two-level system with the *TM* radiation field contains a bunch of closely spaced soft modes, whose frequencies decrease as the temperature is lowered according to a T^{-1} law, and near T_c go to zero according to a law $(T-T_c)^{1/2}$. The thickness of the bunch goes to zero as $(T-T_c)^{1/2}$. In Fig. 3 this bunch of soft modes is shown by double shading.

2) If a *TM* mode of the field is resonant to the transition frequency ω_0, the spectrum contains an individual soft mode with width determined only by the figure of merit Q of the resonator. With decreasing frequency the frequency of this mode behaves like $T^{-1/2}$, and for $T \to T_c$ it behaves like $(T-T_c)^{1/2}$.

3) Since $k_z^2 < k^2$, the interaction of the atoms with the *TE* modes gives another bunch of closely spaced modes, which are shown in Fig. 3 by shading. Unlike the resonance frequencies $\Omega_{\mathbf{k}2}$, this bunch of frequencies does not contract to zero for $T \to T_c$. In the neighborhood of T_c the width of the *TE* band of resonance frequencies $\Omega_{\mathbf{k}1}$ becomes comparable with the atomic transition frequency ω_0.

4) Independently of the ratio of ω_0 and the frequency of the radiation field, the temperature at which the frequencies of the soft modes go to zero can be determined from the equation

$$\operatorname{th}(\omega_0/2kT_c) = \omega_0/2\pi\mu^2\rho = \varkappa \tag{48}$$

or $kT_c \sim \pi\mu^2\rho$.

If we substitute (46) in Eq. (22), we can verify that at the critical point (48) a Bose-Einstein condensation occurs for both the *TE* and *TM* radiation fields. Substituting the matrix elements (45) in Eq. (30), we find

$$X_{\mathbf{k}\sigma}(\mathbf{r},\Omega) = -\frac{\Pi(\Omega)\lambda_{\mathbf{k}\sigma}^*(\mathbf{r})}{D_0(\mathbf{k}\sigma,\Omega)\Xi_{\mathbf{k}\sigma,\mathbf{k}\sigma}(\Omega)-1}$$

and from this and Eq. (29) we get the transverse dynamic susceptibility of the system atoms plus radiation field

$$\chi_{+-}(\Omega) = N\Pi(\Omega) - 4\omega_0\Pi(\Omega)\sum_j\sum_{\mathbf{k}\sigma}\frac{\omega_{\mathbf{k}\sigma}|\lambda_{\mathbf{k}\sigma}(\mathbf{r}_j)|^2}{(\Omega^2-\Omega_{\mathbf{k}\sigma}^{-2})(\Omega^2-\Omega_{\mathbf{k}\sigma}^{+2})}. \tag{49}$$

The density of states of the collective excitations in the band can be written in a form which is used in band theory[20]

$$\rho(\Omega_{\mathbf{k}\sigma}^-) = \frac{V}{\pi^3}\int\frac{dS}{|\nabla_{\mathbf{k}}\Omega_{\mathbf{k}\sigma}^-|}.$$

According to Eq. (47) we find that Van Hove singularities occur at $T = T_c$ and $T = \infty$.

5. DISCUSSION

The two limiting cases examined in Secs. 3 and 4 show that the ratio of the dimensions of the resonator and of the atomic system plays an important part in the dynamics of a two-level system coupled to a radiation field. A rather important distinction between different types of temperature behavior of this dynamics can be made.

1. If the atoms are placed in a resonator which is so large that the dimensions of the system of atoms are much smaller than the wavelengths of the radiation field of the resonator and the frequency of the atomic transitions is much larger than the fundamental proper frequency of the resonator, there is a collective radiative line width (38) and a collective radiative shift (39). Depending on the position of the atomic specimen in the resonator, there may also be an additional radiative line shift owing to a longitudinal coupling between the radiation field and the atoms. For paramagnetic atoms there is a corresponding radiative change of the g factor, Eq. (40). All of these effects are proportional to the number of two-level atoms in the resonator.

If, on the other hand, the atoms are uniformly distributed in the resonator and the atomic transition fre-

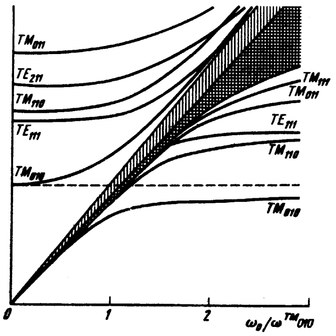


FIG. 4. Field spectrum of resonance frequencies for $T = 4.2$ K, $\omega^{TM_{010}} = 10^{11}$. The shaded region represents the set of frequencies $\Omega_{k1}(TE)$, and the doubly shaded region, the frequencies Ω_{k2} (TM).

quency is comparable with the fundamental frequency of the resonator, as is the case in ordinary EPR research, the spectrum of resonance frequencies consists of a set of discrete lines, as shown in Fig. 1.

2. In the other extreme case considered here, the cavity of the resonator is completely and uniformly filled with atoms. In this case a new radiative effect appears: a band spectrum of resonance frequencies. According to Eq. (43) the width of the band is

$$r \sim \pi\mu^2\rho_s\omega_0/kT, \tag{50}$$

if $kT > \omega_0 > \mu^2\rho_S$. On the other hand, if $T \gg T_c$ and $\omega_0 < \mu^2\rho_S$,

$$r \sim \omega_0 T_c/T. \tag{51}$$

To obtain a more exact expression for the band width r, a specific resonator must be considered. Figure 3 shows the temperature dependence of the resonance frequencies of a two-level system placed in a cylindrical resonator.

Resonance studies of paramagnetic systems usually use scanning over values of the external magnetic field.[17] The dependence of the resonant frequencies for paramagnetic system plus radiation field on the external field H_0 is shown in Fig. 4. To make the radiative effects clearly visible in Fig. 4, we have taken the extreme density $g^2\rho_S \approx 0.9 \cdot 10^{23}$. Physical situations are often encountered in which the atomic specimen is much smaller than the resonator and at the same time much larger than the wavelengths of the leading radiation modes of the resonator. Unfortunately, the calculations are very difficult in this case.

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[1)]If we take into account the microscopic nonuniformity of the distribution of the atoms in the resonator, Eqs. (43) will hold for $k \ll \rho s^{1/3}$.

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