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# Thermodynamics and Electrodynamics of Superradiant Phase

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The thermodynamics and electrodynamics of the superradiant phase are analyzed on the basis of the Emeljanov-Klimontovich model by means of a mean field approach and random phase approximation. The model considers infinite modes of radiation field. The superradiant phase is characterized by a static and homogeneous Bose condensation of the transverse collective mode with zero wave vector. The thermodynamic quantities and the dispersion relations for the collective mode are obtained in closed forms. While the thermodynamic properties of the present model are the same as those of the Dicke model, the electrodynamics differs in form from the latter. A softening of the lower branch of the collective mode behaves as  $(T-T_c)^{1/2}$  for  $T > T_c$ , whereas for  $T < T_c$  it obeys a law  $T_c - T$  or  $(T_c - T)^{1/2}$  according to the different regions of temperature and the polarizations. A light velocity is renormalized with an anisotropic constant.

# § 1. Introduction

Properties of a system of n identical atoms with two levels interacting with a radiation field are now widely investigated. In the most of the works a simplification that takes account of only a single rotating field has been made. Within this simplified model it is shown that a mean field treatment is sufficient to obtain the equilibrium properties, which is shown to be exact in the limit of large  $n.^{1)\sim6}$  It was predicted that if the coupling between the two subsystems—the atoms and the radiation field—is sufficiently strong, the system exhibits a phase transition to an ordered state called a superradiant phase.

In spite of this success, however, up to the present time a natural problem of infinite number of modes of the radiation field and without trancation to a single rotating field has been remained less clear. Emeljanov and Klimontovich<sup>8)</sup> (referred to as E. K. hereafter) studied briefly this problem on the basis of a model which is a generalization of the Dicke model. They derived a set of operator equations for the field and the atomic polarization. By linearizing them with respect to fluctuations they obtained a critical behaviour of collective modes which suggests a phase transition to a superradiant phase at a critical temperature analogous to that of the Dicke model. Recently, the superradiant ground state of the E. K. model is discussed by the author within a framework of a mean field theory.<sup>10)</sup>

The purpose of the present paper is to work out the analysis about thermodynamic and electromagnetic properties of the E. K. model.<sup>8)</sup> Following the idea that the superradiant state is characterized by the presence of the static and uniform Bose condensation of a collective mode,<sup>5),6)</sup> we introduce anomalous quasi-averages of an electric field and off-diagonal atomic polarizations, which play the role of the order parameter of the new phase.

In § 2 the Hamiltonian of the E. K. model is introduced. In order to make easy the analysis in subsequent sections Fermi operators are used to represent the atomic system. In such a representation the present model can be thought of as a member of a family of the Lee models,<sup>11)</sup> as well.

In § 3 a collective mode—transverse polariton—in the normal phase is examined in a random phase approximation (RPA). It is shown that the lower branch of the mode becomes soft as  $\omega_- \propto (T-T_c)^{1/2}$  as a temperature is lowered. This form of temperature behaviour of the softening differs from that of the single rotating wave where the softening law as  $\omega_- \propto T-T_c$  is obtained. This softening brings out the instability of the normal phase below a certain critical temperature and predicts a phase transition accompanied with the condensation of the unstable collective mode. It is pointed out that the mode with k=0 is most unstable. This is important to take the property of the low temperature phase to be characterized by a homogeneous condensation as a natural consequence. This was assumed by E. K.<sup>8)</sup> without proof.

In § 4 the thermodynamic properties are discussed on the basis of an effective Hamiltonian which is constructed by a mean field treatment suitable for the superradiant phase. The presence of the condensation field gives rise to the Stark shift of the atomic energy levels, which brings out a net free energy gain of the superradiant phase at an expence of energy of the static electric field as compared with the normal phase. The transition is of the second kind as in the case of the Dicke model.<sup>1)</sup>

As is well known,  $^{12)}$  the time development of a Bose condensation field is governed by its chemical potential  $\mu$  as  $e^{i\mu t}$ . In considering that the chemical potential of a photon is zero the condensation field in the present case is anticipated to be static in time. In § 5 it is shown that this is indeed the case by examining equations of motion of the condensation fields.  $^{10)}$ 

In § 6 the collective mode in the superradiant phase is obtained. They are anisotropic because of the broken rotational symmetry. The lower branch of them is gapless and linear in k for small k with renormalized anisotropic light velocity. This is the Goldstone mode accompanying in the new phase and becomes soft as  $(T_c - T)^{1/2}$  near  $T_c$ .

#### § 2. Model Hamiltonian

Now, the Hamiltonian of the E. K. model<sup>8)~10)</sup> is introduced as follows. Let  $a_{k\lambda}$  and  $a_{k\lambda}^+$  be the operators for the electromagnetic field with a wave vector k and a polarization  $\lambda$ ,  $b_{is}$  and  $b_{is}^+$  for an i-th atom at a level s=1 or 2 located at  $R_i$ . The splitting of the levels 1 and 2 is denoted by  $\varepsilon_0$ . The transverse electric induction  $D_{k\lambda}$  is written as

$$D_{\mathbf{k}\lambda} = -i\sqrt{2\pi c} k (a_{\mathbf{k}\lambda}^{+} - a_{\mathbf{k}\lambda}) \tag{2.1}$$

and the transverse component of the electric dipole  $P_i$  of an i-th atom is as

$$P_i = d_{\lambda} (b_{i1}^+ b_{i2} + b_{i2}^+ b_{i1}), \tag{2.2}$$

where  $d_{\lambda} = e \int \psi_1^* (e_{\lambda} \cdot r) \psi_2 dr$  is a matrix element of an atomic dipole in the direction  $e_{\lambda}$ . The Hamiltonian is decomposed into three parts,  $e^{(\lambda)}$ 

$$H = H_0 + H' + H''$$
, (2.3)

where

$$H^{0} = \sum_{\mathbf{k}} ck \ a_{\mathbf{k}\lambda}^{+} a_{\mathbf{k}\lambda} + \frac{\varepsilon_{0}}{2} \sum_{i=1}^{n} (b_{i2}^{+} b_{i2} - b_{i1}^{+} b_{i1}), \tag{2.4}$$

$$H' = -\sum_{k \lambda i} D_{k \lambda} P_i e^{ik\mathbf{R}_i} , \qquad (2.5)$$

$$H'' = 2\pi (1 - \beta) \sum_{k \neq ij} P_i P_j e^{ik(R_i - R_j)}.$$
 (2.6)

H' is a part of an interaction with transverse electric induction. H'' embodies the remaining part of the interaction with the transverse field and the interaction with longitudinal field. The latter comes from the direct dipole-dipole interaction, which is written at present in the form proportional to a local field constant  $\beta$ . The detailed discussion for this simplification is given in the text. As was shown there, the parameter  $\beta$  depends on  $\mathbf{k}$ , but we take it as a constant (of order 1) because we are interested in the behaviour of the system at the uniform equilibrium state and the fluctuations about it with long wave length.

Now, we define operators

$$b_{k} = \frac{1}{\sqrt{n}} \sum_{i} b_{i} \exp(-i\boldsymbol{k}\boldsymbol{R}_{i}). \tag{2.7}$$

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By the same reason mentioned just above, we can replace the summation over i in (2.7) by integral as  $\sum_{i} \cdots \simeq \int d\mathbf{R} \cdots$ . Then, the inverse of (2.7) becomes

$$b_i = \frac{1}{\sqrt{n}} \sum_{k} b_k \exp(ikR_i)$$
 (2.8)

and the commutation relations

$$\{b_{k}^{+}, b_{k'}\} = \delta_{k,k'}, \quad \{b_{k}, b_{k'}\} = 0.$$
 (2.9)

The  $(\mathbf{k}, \lambda)$ -component of the total transverse atomic dipole is defined as

$$P_{k\lambda} = \sum_{i} d_{\lambda} P_{i} \exp(-ik\mathbf{R}_{i})$$

$$= d_{\lambda} \sum_{p} (b_{2,p}^{+} b_{1,p+k}^{+} + b_{1,p-k}^{+} b_{2,p}^{-}).$$
(2.10)

By using definitions  $(2\cdot7)\sim(2\cdot10)$  the Hamiltonians  $(2\cdot2)\sim(2\cdot4)$  are rewritten as

$$H_{0} = \sum_{\mathbf{k}\lambda} c_{\mathbf{k}} a_{\mathbf{k}\lambda}^{+} + \frac{\varepsilon_{0}}{2} \sum_{\mathbf{p}} (b_{2\mathbf{p}}^{+} b_{2\mathbf{p}} - b_{1\mathbf{p}}^{+} b_{1\mathbf{p}}), \tag{2.11}$$

$$H' = -\sum_{k,l} D_{k\lambda} P_{-k\lambda} , \qquad (2\cdot 12)$$

$$H'' = 2\pi (1-\beta) \sum_{\mathbf{k}\lambda} P_{\mathbf{k}\lambda} P_{-\mathbf{k}\lambda} . \tag{2.13}$$

The kinetic energy of the translational motion of the atoms is not explicitly written, because we assume that  $T < \varepsilon_0$ . If this term cannot be ignored, the second term of (2·11) must be replaced by

$$\sum_{\mathbf{p}} \left[ \left\{ \frac{\varepsilon_{\mathbf{0}}}{2} + \varepsilon(\mathbf{p}) \right\} b_{2\mathbf{p}}^{+} b_{2\mathbf{p}} + \left\{ -\frac{\varepsilon_{\mathbf{0}}}{2} + \varepsilon(\mathbf{p}) \right\} b_{1\mathbf{p}}^{+} b_{1\mathbf{p}} \right]. \tag{2.14}$$

If we let  $\beta=1$  and take account of only a single rotating wave, the present Hamiltonian  $(2\cdot11)\sim(2\cdot13)$  reduces to that of the Dicke model.

### § 3. Collective mode in normal phase

Before proceeding to study a superradiant phase let us examine the instability in the system at the normal phase by showing the catastrophic behaviour of the collective mode. Let the propagator of the transverse electromagnetic field<sup>6)</sup>

$$F_{\lambda}(\mathbf{k}, \tau) = -\langle TD_{\mathbf{k}\lambda}(\tau)D_{-\mathbf{k}\lambda}\rangle. \tag{3.1}$$

the equation for which is given by

$$F = (1 - F^{(0)}\Pi)^{-1}F^{(0)}, \qquad (3\cdot2)$$

where

$$F_{\lambda}^{(0)}(\mathbf{k}, \omega_m) = -4\pi (ck)^2 \{\omega_m^2 + (ck)^2\}^{-1},$$

$$\omega_m = 2m\pi i/T. \tag{3.3}$$

 $\Pi$  is the polarization part defined by

$$\Pi_{\lambda}(\mathbf{h}, \tau) = -\langle TP_{\mathbf{h}\lambda}(\tau)P_{-\mathbf{h}\lambda}\rangle_{\text{irred}}. \tag{3.4}$$

The equation for  $\Pi$  is written as

$$= \Pi_{\lambda}^{(0)} + \Pi_{\lambda}^{(0)} \qquad (3.5)$$

or by solving it

$$\Pi_{\lambda}(\boldsymbol{k}, \omega_{m}) = \{1 - 4\pi(1 - \beta)\Pi_{\lambda}^{(0)}(\boldsymbol{k}, \omega_{m})\}^{-1}\Pi_{\lambda}^{(0)}(\boldsymbol{k}, \omega_{m}). \tag{3.6}$$

The second term of the r. h. s. of Eq. (3.5) arises due to the interaction H'' (2.13). Substituting Eq. (3.6) into Eq. (3.2), we have

$$F = F^{(0)} \{ \Pi^{(0)^{-1}} - 4\pi (1-\beta) \} / \{ \Pi^{(0)^{-1}} - F^{(0)} - 4\pi (1-\beta) \}.$$
 (3.7)

In the lowest order approximation  $\Pi^{(0)}$  is calculated from

$$\Pi_{\lambda}^{(0)}(\boldsymbol{k}, \omega_{m}) = d_{\lambda} \underbrace{\sum_{1}^{2}}_{1} d_{\lambda} + d_{\lambda} \underbrace{\sum_{2}^{1}}_{2} d_{\lambda}$$

$$= T d_{\lambda}^{2} \underbrace{\sum_{\boldsymbol{p}\nu_{n}}}_{1} \{G_{2}(\boldsymbol{p}, \nu_{n}) G_{1}(\boldsymbol{p} - \boldsymbol{k}, \nu_{n} - \omega_{m})$$

$$+ G_{1}(\boldsymbol{p} + \boldsymbol{k}, \nu_{n} + \omega_{m}) G_{2}(\boldsymbol{p}, \nu_{n})\}, \tag{3.8}$$

where  $G_s(\mathbf{p}, \nu_n)$  is a propagator of free atoms at the level s=1, 2,

$$G_s(\mathbf{p}, \nu_n) = \frac{1}{i\nu_n \mp \varepsilon_0/2}, \qquad \mp \text{ for } s = \begin{cases} 2\\1 \end{cases},$$

$$\nu_n = 2\pi i \left(n + \frac{1}{2}\right) / T. \tag{3.9}$$

After summing over p and  $\nu_n$  we obtain

$$\Pi_{\lambda}^{(0)}(\mathbf{h}, \omega_{m}) = -\frac{2nd^{2}\varepsilon_{0}}{\omega_{m}^{2} + \varepsilon_{0}^{2}} \operatorname{th} \frac{\varepsilon_{0}}{2T}. \tag{3.10}$$

Substituting Eq.  $(3 \cdot 10)$  into Eq.  $(3 \cdot 7)$ , we obtain

$$F_{\lambda}(\mathbf{k}, \ \omega_{m}) = -\frac{4\pi (ck)^{2} Z_{1}}{\omega_{m}^{2} z_{1} + (ck)^{2} Z_{2}}, \qquad (3.11)$$

where

$$Z_1 = \omega_m^2 + \varepsilon_0^2 + (\gamma - 1) \nu_\lambda \varepsilon_0 \text{ th } \frac{\varepsilon_0}{2T}$$
,

$$Z_2 = \omega_m^2 + \varepsilon_0^2 - \nu_\lambda \varepsilon_0 \text{ th } \frac{\varepsilon_0}{2T}$$
 (3.12)

with  $\nu_{\lambda} = 8\pi\beta d_{\lambda}^{2} n$  and  $\gamma = \beta^{-1}$ .

After analytical continuation of the Matsubara frequency  $i\omega_m \rightarrow \omega$  in Eq. (3·11) we find the dispersion relation for the collective mode

$$(\omega_{k\lambda}^{+})^{2} = Q_{k\lambda}^{2} \pm \left[ Q_{k\lambda}^{4} - (ck)^{2} \varepsilon_{0} \left\{ \varepsilon_{0} - \nu_{\lambda} \operatorname{th} \frac{\varepsilon_{0}}{2T} \right\} \right]^{1/2}, \tag{3.13}$$

where

$$Q_{k\lambda}^2 = \frac{1}{2} \left\{ \varepsilon_0^2 + (ck)^2 + (\gamma - 1)\varepsilon_0 \nu_\lambda \operatorname{th} \frac{\varepsilon_0}{2T} \right\}. \tag{3.14}$$

So long as the coupling is weak;  $\nu_{\lambda} < \varepsilon_0$ , the spectrum given by Eq. (3·3) displays a usual polariton form at any temperature. However, if the coupling is sufficiently strong;  $\nu_{\lambda} > \varepsilon_0$ , it behaves in a very different way: the lower branch  $\omega_{k\lambda}$  becomes soft as the temperature is lowered and tends to zero at the temperature  $T_c$  which is given by

$$\frac{\nu_{\lambda}}{\varepsilon_0} \text{ th } \frac{\varepsilon_0}{2 T_c} = 1 , \qquad (3.15)$$

just as the case of the Dicke model.<sup>6)</sup> Near  $T_c(T \gtrsim T_c)$  the spectrum behaves as

$$(\omega_{\mathbf{k}\lambda}^+)^2 \cong \varepsilon_0^2 + (ck)^2 - \alpha_{\mathbf{k}\lambda}^2 (T - T_c),$$

$$(\omega_{\mathbf{k}\lambda}^{-})^{2} \simeq \alpha_{\mathbf{k}\lambda}^{2} (T - T_{c}), \tag{3.16}$$

where

$$\alpha_{\mathbf{k}\lambda} = \frac{\varepsilon_0 ck\sqrt{\nu_{\lambda}}}{2 T_c \left\{\gamma \varepsilon_0^2 + (ck)^2\right\}} \left(1 - \frac{\varepsilon_0^2}{\nu_{\lambda}^2}\right)^{1/2}.$$
 (3.17)

At  $T < T_c$ ,  $\omega_-^2$  is negative, so that the system becomes unstable.

It should be noted that within the above argument the critical temperature  $T_c$  depends only on  $\lambda$  through  $d_{\lambda}$ , but not on  $\mathbf{k}$ . To speak more realistically, however, effects other than we have considered would have caused some dependence on  $\mathbf{k}$ . Indeed, we can show that the kinetic energy of the atoms which have been ignored gives rise to a term proportional to  $k^2$ , thus in place of Eq. (16)

$$\omega_{-}^{2} \simeq \alpha_{k\lambda} (T - T_c) + \mu k^{2}. \tag{3.18}$$

As a consequence, one can see that the mode with k=0 is made to be the most unstable, implying the new phase below  $T_c$  to be characterized by spacially uniform condensate.

#### § 4. Thermodynamic properties of superradiant phase

Along the line of ideas mentioned in the previous sections we introduce for the superradiant phase the Bose condensation field of the collective mode with k=0, which is taken as uniform and static. They are expressed as nonzero averages of

$$-i\lim_{k\to 0} \sqrt{2\pi ck} \langle a_{k\lambda_s} \rangle = \alpha \quad \text{and} \quad \langle P_{0\lambda_s} \rangle = P , \qquad (4\cdot 1)$$

where an arbitrariness of the direction  $\lambda$  is removed by fixing  $\lambda = \lambda_s$ . As will be shown later, the self-consistently determined  $\alpha$  and P differ from each other only by a constant factor, so that either  $\alpha$  or P can be regarded as the order parameter of the superradiant phase.

Now, the mean field treatment of the original Hamiltonian  $(2 \cdot 11) \sim (2 \cdot 13)$  under the prescription  $(4 \cdot 1)$  leas to a new effective Hamiltonian

$$H^{\text{eff}} = \frac{|\alpha|^2}{2} - 2\pi (1 - \beta) P^2 + \frac{\varepsilon_0}{2} \sum_{\mathbf{p}} (b_{2\mathbf{p}}^+ b_{2\mathbf{p}} - b_{1\mathbf{p}}^+ b_{1\mathbf{p}}) + \frac{\mathcal{\Delta}}{2} \sum_{\mathbf{p}} (b_{1\mathbf{p}}^+ b_{2\mathbf{p}} + b_{2\mathbf{p}} b_{1\mathbf{p}}), \tag{4.2}$$

where

$$\Delta = 4\{\operatorname{Re}\alpha + 2(1-\beta)P\}. \tag{4.3}$$

The quadratic Hamiltonian  $(4\cdot2)$  is diagonalized by the global canonical transformation<sup>10)</sup>

$$\beta_{1p} = \cos \theta b_{1p} - \sin \theta b_{2p}$$
,

$$\beta_{2\mathbf{p}} = \sin \theta b_{1\mathbf{p}} - \cos \theta b_{2\mathbf{p}} \,, \tag{4.4}$$

where

$$\cos 2\theta = \varepsilon_0/\varepsilon$$
 and  $\sin 2\theta = \Delta/\varepsilon$  (4.5)

with

$$\varepsilon = \sqrt{\varepsilon^2 + \Delta^2} \ . \tag{4.6}$$

Thereby the Hamiltonian  $(4\cdot2)$  is transformed as

$$H^{\text{eff}} = \frac{|\alpha|^2}{2\pi} - 2\pi (1-\beta)P^2 + \sum_{p} \frac{\varepsilon}{2} (\beta_{2p}^+ \beta_{2p} - \beta_{1p}^+ \beta_{1p}). \tag{4.7}$$

The other solution where  $\cos \theta = 1$ ,  $\sin \theta = 0$  which corresponds to  $\alpha$  and P = 0 is also allowed. However, this is a trivial solution which corresponds to a normal phase and is energetically unstable below  $T_c$ .

The free energy of the system with Hamiltonian (4.7) is

$$F = \frac{|\alpha|^2}{2} - 2\pi (1 - \beta) P^2 - \frac{n}{2} \varepsilon - Tn \ln(1 + e^{\varepsilon/T}). \tag{4.8}$$

The order parameter  $\alpha$  is determined by making the free energy minimum:  $\partial F/\partial \alpha = 0$ , leading to

$$\alpha = 2\pi n d \frac{\Delta}{\varepsilon} \operatorname{th} \frac{\varepsilon}{2T} . \tag{4.9}$$

By the definition  $(4 \cdot 1)$  and Eq.  $(4 \cdot 9)$  one obtains for P

$$P = -\alpha/2\pi = -nd\frac{\Delta}{\varepsilon} \operatorname{th} \frac{\varepsilon}{2T}, \qquad (4\cdot 10)$$

thus, for the "gap parameter" (4.3)

$$\Delta = 4\beta d_{\lambda}\alpha \ . \tag{4.11}$$

The value of P given by Eq. (4·10) also makes the free energy minimum with respect to P. Eliminating  $\alpha$  from Eqs. (4·9) and (4·11), one obtains the "gap equation"

$$\frac{\varepsilon}{v} = \text{th} \frac{\varepsilon}{2T}$$
, (4·12)

where

$$\nu = 8\pi\beta d^2 n$$
,  $(d = d_{\lambda s})$ 

Equation (4.12) has a real unique solution for  $\varepsilon$  only if

$$\varepsilon_0 < \nu_\lambda$$
 (4·13)

Now, by making use of Eqs.  $(4 \cdot 10)$  and  $(3 \cdot 12)$  the free energy becomes

$$F = \frac{\Delta^2}{4\nu} - Tn \ln(2\cosh \varepsilon/2T). \tag{4.14}$$

From this the entropy is calculated as

$$S = -\frac{n\varepsilon^2}{2T\nu} \operatorname{th} \frac{\varepsilon}{2T} + n \ln(2\cosh \varepsilon/2T), \tag{4.15}$$

and the specific heat as

$$C = \frac{n\varepsilon^2}{4T^2} \frac{\operatorname{sech}^2 \varepsilon / 2T}{1 - \frac{\nu}{2T} \operatorname{sech}^2 \varepsilon / 2T}.$$
 (4·16)

It must be compared with the specific heat of the normal phase

$$C_n = \frac{n\varepsilon_0^2}{4T^2} \operatorname{sech}^2 \varepsilon_0 / 2T , \qquad (4.17)$$

which is obtained from Eq. (4·14) by making  $\Delta = 0$ . Therefore, the specific heat is discontinuous at  $T = T_c$  with a jump

$$\frac{C}{C_n} = \left(1 - \frac{\nu}{2T_c} \operatorname{sech}^2 \frac{\varepsilon_0}{2T_c}\right)^{-1} > 1, \qquad (4 \cdot 18)$$

whereas the entropy is continuous at  $T = T_c$ . The free energy (4·14) is always lower than that of the normal phase as long as  $T < T_c$ .

At the ground state<sup>10)</sup> (T=0) the gap equation (4·12) reduces to

$$\varepsilon = \nu$$
, i. e.,  $\Delta^2 = \nu^2 - \varepsilon_0^2$ . (4.19)

After a little manipulation one obtains the condensation energy of the superradiant ground state as

$$E - E_n = -\frac{n}{4\nu} (\nu - \varepsilon_0)^2 < 0.$$
 (4.20)

Near  $T_c$  we can expand the free energy (4·14) with respect to  $\Delta$ . The result is the Landau free energy, well approximated by

$$F = F_0 + \alpha (T - T_c) \Delta^2 + \frac{\beta_\bullet}{2} \Delta^4, \qquad (4.21)$$

where

$$F_0 = -T_c n \ln(2 \cosh \varepsilon_0 / 2 T_c), \tag{4.22}$$

$$\alpha = \frac{n}{8T_c^2} \operatorname{sech}^2 \frac{\epsilon_0}{2T_c}, \tag{4.23}$$

$$\beta = \frac{n}{8\varepsilon_0^3} \left( \operatorname{th} \frac{\varepsilon_0}{2T_c} - \frac{\varepsilon_0}{2T_c} \operatorname{sech}^2 \frac{\varepsilon_0}{2T_c} \right). \tag{4.24}$$

Consequently, the gap  $\Delta$  varies with temperature as

$$\Delta^2 = \alpha (T_c - T)/\beta$$
 for  $T \lesssim T_c$ . (4.25)

## § 5. Electrodynamics of superradiant phase

As was anticipated in § 1 the condensation fields  $\alpha$  and P must be static in time. To verify this statement we return to the original Hamiltonian  $(2 \cdot 11) \sim (2 \cdot 13)$  and examine the Heisenberg equation of motion for  $a_{k\lambda}$  and  $P_{k\lambda}$ . First, one obtains

$$i\dot{a}_{k\lambda} = [a_{k\lambda}, H] = cka_{k\lambda} + i\sqrt{2\pi ck}P_{-k\lambda}.$$
 (5.1)

On multiplying both sides of Eq. (5·1) by  $\sqrt{2\pi ck}$  and taking the limit  $k \to 0$  for its average, we obtain

$$i\dot{\alpha} = ck(\alpha + 2\pi P). \tag{5.2}$$

By virtue of Eq. (4·10) the terms in the bracket of the r.h.s. of Eq. (5·2) are cancelled out to zero. In the same way, we obtain for  $P_{0\lambda}$ 

$$i\dot{P}_{0\lambda} = [P_{0\lambda}, H] = -\varepsilon_0 d_0 \sum_{p} (b_{2p}^+ b_{1p}^- - b_{1p}^+ b_{2p}^-)$$

$$= -\varepsilon_0 d_0 \cos 2\theta \sum_{p} (\beta_{2p}^+ \beta_{1p}^- - \beta_{1p}^+ \beta_{2p}^-). \tag{5.3}$$

In the average the r.h.s. of Eq.  $(5\cdot3)$  also vaninshes since there is no term diagonal in  $\beta$ , hence,  $\dot{P}=0$ . Therefore, in the superradiant phase we have not only the homogeneous but also static electric induction and electric polarization. The equilibrium value of them are given by

$$D = 2\alpha = 4\pi n d_0 \frac{\Delta}{\varepsilon} \operatorname{th} \frac{\varepsilon}{2T} = -4\pi P . \qquad (5.4)$$

The electric field itself vanishes  $E = D - 4\pi P = 0$  by virtue of  $D = 2\alpha$  and Eq. (3·10), as it should be. The features derived above are well interpreted as a ferroelectricity.<sup>7)</sup>

Now, we examine the behaviour of the new collective mode in the superradiant phase in the same way as in § 3. To this end it is usefull to rewrite the Hamiltonian  $(2 \cdot 11) \sim (2 \cdot 13)$  in terms of  $\beta$ -operators by Eq.  $(4 \cdot 4)$ :

$$H_0 = \sum_{\mathbf{k},\lambda} ck a_{\mathbf{k}\lambda}^+ a_{\mathbf{k}\lambda} + \frac{\varepsilon}{2} \sum_{\mathbf{p}} (\beta_{2\mathbf{p}}^+ \beta_{2\mathbf{p}} - \beta_{1\mathbf{p}}^+ \beta_{1\mathbf{p}}) + \text{const} .$$
 (5.5)

In  $H'(2\cdot 12)$  and  $(2\cdot 13)$ ,  $P_{k\lambda}$  is replaced by

$$P_{\mathbf{k}\lambda} = d_{\lambda} \sum_{\mathbf{p}} \frac{\varepsilon_0}{\varepsilon} (\beta_{2\mathbf{p}}^+ \beta_{1\mathbf{p}+\mathbf{k}} + \beta_{1\mathbf{p}+\mathbf{k}}^+ \beta_{2\mathbf{p}}) + \frac{\mathcal{L}}{\varepsilon} (\beta_{2\mathbf{p}}^+ \beta_{2\mathbf{p}+\mathbf{k}} - \beta_{1\mathbf{p}}^+ \beta_{1\mathbf{p}+\mathbf{k}}). \tag{5.6}$$

The polarization part is now calculated with this new form of Hamiltonian to give

$$\widetilde{\Pi}_{\lambda}^{(0)}(\boldsymbol{k}, \omega_{m}) = d_{\lambda}^{2} \left(\frac{\varepsilon_{0}}{\varepsilon}\right)^{2} T \sum_{\boldsymbol{p}\nu_{n}} \{\widetilde{G}_{2}(\boldsymbol{p}, \nu_{n}) \widetilde{G}_{1}(\boldsymbol{p} - \boldsymbol{k}, \nu_{n} - \omega_{m}) + \widetilde{G}_{1}(\boldsymbol{p} + \boldsymbol{k}, \nu_{n} + \omega_{m}) \widetilde{G}_{2}(\boldsymbol{p}, \nu_{n})\} + d_{\lambda}^{2} \left(\frac{\Delta}{\varepsilon}\right)^{2} T \sum \{\widetilde{G}_{1}(\boldsymbol{p}, \nu_{n}) \widetilde{G}_{1}(\boldsymbol{p} - \boldsymbol{k}, \nu_{n} - \omega_{m}) + \widetilde{G}_{2}(\boldsymbol{p}, \nu_{n}) \widetilde{G}_{2}(\boldsymbol{p} - \boldsymbol{k}, \nu_{n} - \omega_{m})\}.$$
(5.7)

Here  $\tilde{G}_i$  is the propagator of a  $\beta_i$ -particle,  $\tilde{G}_i = 1/(\nu_n + \varepsilon/2)$ , i = 1, 2. The second term of Eq. (5·7) vanishes except for the frequency  $\omega_m = 0$ . Aside from this  $\delta \omega_{m,0}$  contribution<sup>6)</sup> we obtain from Eq. (5·7)

$$\Pi_{\lambda}^{(0)}(\boldsymbol{k},\,\omega_{m}) = -\frac{d_{\lambda}^{2}}{4\pi\beta d^{2}} \frac{\varepsilon_{0}^{2}}{\omega_{m}^{2} + \varepsilon^{2}}.$$
 (5.8)

The photon propagator is now given by the same form as Eq. (3.11) but now  $Z_1$  and  $Z_2$  are replaced by

$$\tilde{Z}_{1} = 1 + (\gamma - 1)\cos^{2}\theta \frac{\varepsilon_{0}^{2}}{\omega_{m}^{2} + \varepsilon^{2}},$$

$$\tilde{Z}_{2} = 1 - \cos^{2}\theta \frac{\varepsilon_{0}^{2}}{\omega_{m}^{2} + \varepsilon^{2}},$$
(5.9)

where  $\theta$  is the angle between d and  $d_{\lambda}$ . Thus, we find the new spectrum as

$$(\omega_{bi}^{\pm})^{2} = \tilde{Q}_{bi}^{2} \pm \{Q_{bi}^{4} - (ck)^{2}(\varepsilon^{2} - \varepsilon_{0}^{2}\cos^{2}\theta)\}^{1/2}, \qquad (5\cdot10)$$

where

$$\widetilde{\mathcal{Q}}_{\boldsymbol{k}\lambda}^2 = \frac{1}{2} \{ \varepsilon^2 + (ck)^2 + (\gamma - 1)\varepsilon_0^2 \cos^2 \theta \}. \tag{5.11}$$

The spectrum (5·10) is anisotropic through its dependence on  $\theta$ . For a region of small  $k \le \varepsilon_0$  it has the form

$$\omega_{+}^{2} \cong \varepsilon^{2} + (ck)^{2} \{1 + Z\},$$

$$\omega_{-}^{2} \cong (ck)^{2} Z,$$

$$(5 \cdot 12)$$

where a renormalization constant for the light velocity Z is given by

$$Z = \left\{ 1 - \left( \frac{\varepsilon_0}{\varepsilon} \cos \theta \right)^2 \right\} / \left\{ 1 + (\gamma - 1) \left( \frac{\varepsilon_0}{\varepsilon} \cos \theta \right)^2 \right\}. \tag{5.13}$$

At temperature near  $T_c$ , Z is expanded with respect to  $\Delta$  and is estimated as

$$Z \simeq \sin^2 \theta + \cos^2 \theta \frac{\alpha}{\varepsilon_0^2 \beta} (T_c - T),$$
 (5.14)

where Eq. (4.35) was used. Substituting Eq. (5.14) into Eq. (5.12), one finds that the law for the softening of  $\omega_{k\lambda}$  displays a crossover as

$$\omega_{k\lambda}^{-} \cong \begin{cases} ck \left\{ \sin^2 \theta + \cos^2 \theta \frac{\alpha}{2\varepsilon_0^2 \beta} (T_c - T) \right\} & \text{for } T_c - T < \frac{\varepsilon_0^2 \beta}{\alpha} \tan^2 \theta \\ \frac{ck}{\varepsilon_0} \sqrt{\frac{\alpha}{\beta} (T - T_c)} & \text{for } T_c - T > \frac{\varepsilon_0^2 \beta}{\alpha} \tan^2 \theta \end{cases}$$
 (5.15)

A little manipulation shows that our result  $(5\cdot10)$  is just the same as that of E.  $K.^{8)}$  derived by another way.

Finally, let us sketch the behaviour of the transverse susceptibility<sup>6)</sup> which is defined by

$$\gamma_{\lambda}(\mathbf{h}, \tau) = -\langle TP_{\mathbf{h}\lambda}(\tau)P_{-\mathbf{h}\lambda}\rangle. \tag{5.16}$$

The equation for it is obtained as

$$= \prod_{\lambda}^{(0)}(\boldsymbol{k}, \omega_m)[1 + \{F_{\lambda}^{(0)}(\boldsymbol{k}, \omega_m) + 4\pi(1-\beta)\}\chi_{\lambda}(\boldsymbol{k}, \omega_m)]. \tag{5.17}$$

Thus, we have

$$\chi(\mathbf{k}, \omega_m) = \frac{1}{\prod_{\lambda}^{(0)^{-1}}(\mathbf{k}, \omega_m) - \{F_{\lambda}^{(0)}(\mathbf{k}, \omega_m) + 4\pi(1-\beta)\}}.$$
 (5.18)

Comparing this with Eq. (3.7), we know that  $\chi$  must have the same poles as  $F_{\lambda}(\mathbf{k}, \omega_m)$ , as given by Eq. (3.13) for  $T > T_c$  and Eq. (5.10) for  $T < T_c$ .

# § 6. Conclusions

The foregoing analysis shows that the superradiant phase transition is not a special feature of the Dicke model but also present in a natural generalization of models, i.e. the atoms interacting with an infinite number of modes of radiation field. With respect to the spectrum of the collective mode our results support the previous ones<sup>8)</sup> which were derived by a different method.

Within our crude approximation our results about the thermodynamic properties of the E. K. model coincide with those of the Dicke model, while the electrodynamic collective motion for our case behaves in a different way. The point comes about from the truncation to a rotating field in the Dicke model.

Althogh the Dicke model can be solved exactly at arbitrary temperature in the limit of a large number n asymptotically at least, our model allows us to carry out an analysis at most only approximately. In particular, we note that the present mean field treatment dose not apply to a critical region near  $T_c$ .

Finally, we briefly examine the implication of the strong coupling condition (3 ·13) for the equilibrium superradiance to be realized. If we suppose the separation of the atomic levels to be of an electronic origin, we have  $\varepsilon_0 \sim e^2/a_B$  and  $d \sim e a_B$  as an order of magnitude, where  $a_B$  is the atomic Bohr radius. The local field parameter  $\beta \sim 0[1]$  (=4 $\pi$ /3 for an isotropic medium). Therefore, Eq. (3·13) means that

$$4\pi n a_{\mathsf{B}}^{3} \gtrsim 1 , \qquad (6 \cdot 1)$$

i. e., the atomic density should be of the order of a condensed phase. Here, we note that the experiments<sup>14)</sup> thus far reported have been performed at gas phases with lower densities rather than required by Eq. (5·1) where they have concentrated their attention to the non-equilibrium superradiance as originally proposed by Dicke.<sup>15)</sup> We think that one of candidates for observation of an equilibrium superradiance may be molecular crystals with a comparably large polarizability in a low frequency region.

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