

## **$A^2$ Term, Renormalization of Matter-Photon Interaction and Coherent States in Matter-Photon Systems**

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A non-relativistic quantum theory of matter-photon interaction is formulated, within the framework of the dipole approximation, by employing a canonical transformation to diagonalize the  $A^2$  term plus the free photon Hamiltonian. The matter-photon interaction is thereby expressed rigorously in a renormalized  $\mathbf{A}\cdot\mathbf{p}$  form. Using this form for the Dicke model of two-level atoms interacting with a single-mode radiation field, we examine a possibility of the occurrence of stationary coherent states which could arise for a photon and atomic polarization as the ground state of the system, deducing a condition for such coherent states. The condition, expressed as an inequality which imposes a stable nontrivial Bloch angle of the uniform atomic polarization, is identical with that for the occurrence of a second order phase transition, and indicates that the non-existence of a second order phase transition pointed out by Rzażewski et al. is in fact a consequence of the present treatment of the  $A^2$ -term renormalization. However, a fulfillment of the condition is recovered, if exchange-type atom-atom interaction is taken into account in the matter system. We also present a method to construct an effective Hamiltonian of atom-atom interaction which is equivalent to the above Dicke model in the thermodynamic limit.

### § 1. Introduction

With the advancement of quantum optics, nonlinear optics<sup>1)</sup> and laser physics,<sup>2)</sup> much attention has been paid recently on the coherence and cooperative properties of matter-photon systems.<sup>3)</sup> In the non-relativistic regime the Hamiltonian of a matter-photon system is generally written as<sup>\*)</sup>

$$H = \sum_i \left[ \sum_l (1/2m) \{ \mathbf{p}_{il} + e\mathbf{A}(\mathbf{r}_{il}) \}^2 + V_i(\{ \mathbf{r}_i \}) \right] + H_p + H_{aa}. \quad (1.1)$$

Here  $\mathbf{r}_{il}$  and  $\mathbf{p}_{il}$  are the coordinate and the momentum, respectively, of the  $l$ -th electron bound to the nucleus of an atom at a site  $\mathbf{R}_i$  by the potential  $V_i(\{ \mathbf{r}_i \})$ ,<sup>\*\*)</sup>  $m$  and  $-e$  the mass and the charge of the electron, respectively, and  $\mathbf{A}(\mathbf{r}_{il})$  is the Coulomb-gauge vector potential evaluated at the site  $\mathbf{r}_{il}$ . The symbols  $H_p$  and  $H_{aa}$  represent the free photon Hamiltonian and the Hamiltonian describing atom-atom interactions, respectively. Historically, the Hamiltonian for the matter-photon interaction has been treated in various approximate ways. Broadly speak-

\*) We use natural units to put  $\hbar=c=1$ .

\*\*) The symbol  $\{ \mathbf{r}_i \}$  denotes a set of the coordinates of electrons bound to the  $i$  atom.

ing, this may be classified into two types,  $\mathbf{A} \cdot \mathbf{p}$  form and  $\mathbf{E} \cdot \mathbf{r}$  form. The former is obtained by neglecting the  $A^2$  term in Eq. (1.1), while the latter is derived by applying a canonical transformation to express the matter-photon interaction entirely in terms of electric and magnetic fields, rather than of the vector potential, and by neglecting a self-energy term appearing therein.<sup>4), 5), \*</sup> For matter systems, a two-level-atom model has been used extensively to take into account essential features of nonlinearity in atomic excitations.<sup>6)</sup> Also employed here conventionally has been an approximation to neglect direct atom-atom interactions. It has been recognized for some time in the field of quantum optics, laser physics and others that such approximate Hamiltonians, of which the Dicke model Hamiltonian is most typical,<sup>6), 7)</sup> provide simple but fruitful starting points for the study of the coherence and cooperative properties of matter-photon systems.<sup>2), 3), 6)</sup>

Recently, however, Rzażewski et al. have shown that the Dicke model Hamiltonian with the  $A^2$  term included fails to yield a second order phase transition,<sup>8)</sup> a result incompatible with the conclusions reached by Hepp and Lieb<sup>9)</sup> and by many others.<sup>10)~16)</sup> This work appears to have aroused renewed attention to the role played by the  $A^2$  term which has hitherto been neglected in quantum optics and nonlinear optics. On the other hand, the  $A^2$  term has long been recognized to play a dominant role in solid-state plasma.<sup>17)</sup> Also, this term was taken into account previously by Agranovich<sup>18)</sup> and by Hopfield<sup>19)</sup> in their theories of linear polaritons.

The purpose of the present paper is: (i) to formulate a quantum theory of matter-photon interaction which contains an attempt to fully take into account the  $A^2$  term in the non-relativistic regime and (ii) to examine the effects of this term and of direct atom-atom interactions for the occurrence of stationary coherent states in matter-photon systems. The formulation is done by employing a canonical transformation to diagonalize the  $A^2$  term plus the free photon Hamiltonian and by using the atomic operator formalism<sup>20)</sup> to take into account the multi-level nature of the atomic spectra. The theory so developed enables us to express the effect of the  $A^2$  term rigorously in a renormalized  $\mathbf{A} \cdot \mathbf{p}$  form.

In a previous paper the present authors and Sugimoto studied the coherence properties of the Dicke model Hamiltonian to show that under a strong coupling condition the ground state is characterized by the simultaneous appearance of a stationary coherent state of a photon and that due to atomic polarization.<sup>21)</sup> To illustrate the role played by the  $A^2$  term, the same type of stationary coherent states in the Dicke model Hamiltonian with direct atom-atom interaction as well as the  $A^2$  term included are studied in this paper, without resorting to the use of the rotating-wave approximation to treat the matter-photon interaction. In doing this discussion is given on the condition for the occurrence of such coherent states, and it is shown to be identical with that of a second order phase transition. It

\*<sup>2)</sup> In the conventional  $\mathbf{E} \cdot \mathbf{r}$  form the effect of the  $A^2$  term is only partially taken into account. For a detailed discussion see, for example, Refs. 4) and 5).

is shown that the result obtained by Rzażewski et al.<sup>8)</sup> is a natural outcome of the renormalization of the matter-photon interaction. It is also shown that the appearance of the stationary coherent states becomes possible, if there exist exchange-type atom-atom interactions in the matter system.

In the next section the atomic operator formalism and the dipole approximation are employed to treat Eq. (1.1). In § 3 a canonical transformation is applied to diagonalize the *A*<sup>2</sup> term plus the free photon Hamiltonian. Section 4 is devoted to study of a stationary photon coherent state and that due to atomic polarization in the model matter-photon system. In § 5 another canonical transformation is introduced by eliminating the matter-photon interaction to recast it in the form of effective atom-atom interactions resulting from virtual exchange of photons.

### § 2. Atomic operators and dipole approximation

We consider a matter-photon system governed by the Hamiltonian (1.1). We assume for the sake of simplicity that the matter system is composed of identical one-electron atoms, omitting the subscripts *l* and *i* attached to the electron operators and the potential *V<sub>i</sub>*, respectively, and that the electron is spin-less. It is also assumed here that the electronic states of the matter system composed of atoms are well described by the Heitler-London scheme, in which the overlapping of atomic wave functions on neighbouring atoms is negligible in the first order approximation. We rewrite Eq. (1.1) as

$$H = H_a + H_p + H_{ap} + H_{aa}. \tag{2.1}$$

Here

$$H_a = \sum_i \{ (p_i^2/2m) + V(\mathbf{r}_i) \} \equiv \sum_i H_a(i), \quad H_p = \sum_{\mathbf{k}j} \omega(\mathbf{k}) a_{\mathbf{k}j} a_{\mathbf{k}j}^\dagger \tag{2.2}$$

and

$$H_{ap} = \sum_i (e/m) \mathbf{A}(\mathbf{r}_i) \cdot \mathbf{p}_i + \sum_i (e^2/2m) A(\mathbf{r}_i)^2 \equiv H_{ap}^{(1)} + H_{ap}^{(2)} \tag{2.3}$$

are the Hamiltonian for the atomic system, the free photon field and the matter-photon interaction, respectively. The quantities *a<sub>kj</sub>* and *a<sub>kj</sub><sup>†</sup>* are annihilation and creation operators of a photon of energy  $\omega(\mathbf{k})$  specified by the wave vector  $\mathbf{k}$  and the polarization index *j* (*j*=1, 2). In terms of the photon operators the vector potential  $\mathbf{A}(\mathbf{r}_i)$  is expressed in the form<sup>\*)</sup>

$$\begin{aligned} \mathbf{A}(\mathbf{r}_i) = & \sum_{\mathbf{k}j} [2\pi/\omega(\mathbf{k}) V]^{1/2} \mathbf{e}_{\mathbf{k}j} \{ \exp(i\mathbf{k} \cdot \mathbf{r}_i) a_{\mathbf{k}j} \\ & + \exp(-i\mathbf{k} \cdot \mathbf{r}_i) a_{\mathbf{k}j}^\dagger \} \equiv \sum_{\mathbf{k}j} \{ \mathbf{A}_{\mathbf{k}j}(\mathbf{r}_i) a_{\mathbf{k}j} + \mathbf{A}_{\mathbf{k}j}^*(\mathbf{r}_i) a_{\mathbf{k}j}^\dagger \}, \end{aligned} \tag{2.4}$$

where

\*) The asterisk denotes the complex conjugate.

$$\mathbf{A}_{kj}(\mathbf{r}) = [2\pi/\omega(\mathbf{k})V]^{1/2} \mathbf{e}_{kj} \exp(i\mathbf{k} \cdot \mathbf{r}_i) \quad \text{with} \quad \mathbf{e}_{kj} \cdot \mathbf{e}_{kj'} = \mathcal{A}(j, j'), \quad (2.5)$$

in which  $\mathbf{e}_{kj}$  is the unit vector denoting the polarization direction,  $V$  the normalization volume and  $\mathcal{A}$  the Kronecker delta.

We assume that the eigenvalue problem for the free atom

$$H_a(i) |\alpha_i\rangle = \epsilon_\alpha |\alpha_i\rangle \quad (2.6)$$

has been solved. Here the label  $\alpha_i$  is envisaged as running over all eigenfunctions of  $H_a(i)$  for the  $i$  atom.\*<sup>1</sup> Let us introduce an atomic operator by the equation<sup>20</sup>

$$\sigma_{i\alpha\beta} = |\alpha_i\rangle \langle \beta_i| \quad \text{with} \quad \sum_\alpha \sigma_{i\alpha\alpha} = I, \quad (2.7)$$

where  $I$  is an identity operator. Physically, the operator  $\sigma_{i\alpha\beta}$  destroys an atom in the state  $|\beta_i\rangle$  at the site  $\mathbf{R}_i$  and re-creates it in the state  $|\alpha_i\rangle$ . The multiplication rule obeyed by the  $\sigma$ 's is given by

$$\sigma_{i\alpha\beta} \sigma_{i\gamma\delta} = \sigma_{i\alpha\delta} \mathcal{A}(\beta, \gamma). \quad (2.8)$$

By virtue of the completeness of the states  $|\alpha_i\rangle$ , any operator  $X_i$  affecting only electrons in the  $i$  atom can be expressed in terms of  $\sigma_{i\alpha\beta}$  according to

$$X_i = \sum_{\alpha\beta} \langle \alpha_i | X_i | \beta_i \rangle \sigma_{i\alpha\beta} = \sum_{\alpha\beta} X_{\alpha\beta} \sigma_{i\alpha\beta}. \quad (2.9)$$

The first of Eqs. (2.2) is then rewritten as

$$H_a = \sum_{i\alpha} \epsilon_\alpha \sigma_{i\alpha\alpha}. \quad (2.10)$$

Similarly, the atom-atom interaction Hamiltonian  $H_{aa}$ , which is assumed to be expressed in the form of multi-pole or exchange-type interaction, is taken to be of the form

$$H_{aa} = (1/2) \sum_{ij} \sum_{\alpha\beta\gamma\delta} L_{\alpha\beta,\gamma\delta}(ij) \sigma_{i\alpha\beta} \sigma_{j\gamma\delta}. \quad (2.11)$$

Explicit expressions for the  $L$ 's are obtained once the form of the inter-atomic interaction is specified.

In treating the matter-photon interaction we adopt the dipole approximation, which amounts to evaluating the vector potential  $\mathcal{A}(\mathbf{r}_i)$  at the position  $\mathbf{R}_i$  of the nucleus of the  $i$  atom. As is well known, the use of this approximation is justified if the orbital radius of the electron in the atom is much smaller than the relevant transition wavelengths. By the use of this approximation the Hamiltonian  $H_{ap}^{(2)}$  can be expressed entirely in terms of the photon operators. Thus it is convenient to incorporate  $H_{ap}^{(2)}$  into the free photon Hamiltonian  $H_p$  to write<sup>\*\*</sup>)

$$\begin{aligned} H_p + H_{ap}^{(2)} \\ = (1/2) \sum_{\mathbf{k}\mathbf{k}'j'j} \{ B(\mathbf{k}j, \mathbf{k}'j') a_{\mathbf{k}j}^\dagger a_{\mathbf{k}'j'}^\dagger + 2C(\mathbf{k}j, \mathbf{k}'j') a_{\mathbf{k}j}^\dagger a_{\mathbf{k}'j'} + B^*(\mathbf{k}j, \mathbf{k}'j') a_{\mathbf{k}j} a_{\mathbf{k}'j'} \}, \end{aligned} \quad (2.12)$$

\*<sup>1</sup>) We will often omit the index  $i$  attached to the label  $\alpha_i$  to avoid the use of complicated symbols whenever appropriate to do so.

\*\*<sup>2</sup>) We omit any constant factor appearing in the Hamiltonian.

where

$$B(\mathbf{k}j, \mathbf{k}'j') = W(\mathbf{k}, \mathbf{k}') \mathbf{e}_{\mathbf{k}j} \cdot \mathbf{e}_{\mathbf{k}'j'} \rho(\mathbf{k} + \mathbf{k}') \quad \text{with} \quad W(\mathbf{k}, \mathbf{k}') = \omega_p^2 / 2 [\omega(\mathbf{k})\omega(\mathbf{k}')]^{1/2}, \tag{2.13a}$$

$$C(\mathbf{k}j, \mathbf{k}'j') = W(\mathbf{k}, \mathbf{k}') \mathbf{e}_{\mathbf{k}j} \cdot \mathbf{e}_{\mathbf{k}'j'} \rho(\mathbf{k} - \mathbf{k}') + \omega(\mathbf{k}) \mathcal{J}(\mathbf{k}, \mathbf{k}') \mathcal{J}(j, j'). \tag{2.13b}$$

In the above equations

$$\rho(\mathbf{k}) = (1/N) \sum_{\mathbf{r}_i} \exp(-i\mathbf{k} \cdot \mathbf{R}_i) \quad \text{and} \quad \omega_p = (4\pi N e^2 / V m)^{1/2} \equiv (4\pi n e^2 / m)^{1/2} \tag{2.14}$$

are, respectively, the Fourier transform of the atomic number density normalized to unity and the plasma frequency, in which  $N$  and  $n = N/V$  are the total number and the average number density, respectively, of the atoms in the matter system. In terms of the  $a$ 's and the  $\sigma$ 's the interaction Hamiltonian  $H_{ap}^{(1)}$  is rewritten as

$$\begin{aligned} H_{ap}^{(1)} &= -iN^{-1/2} \sum_{i\alpha\beta} \sum_{\mathbf{k}j} \lambda_{\alpha\beta\mathbf{k}j} \sigma_{i\alpha\beta} \{ \exp(i\mathbf{k} \cdot \mathbf{R}_i) a_{\mathbf{k}j} + \exp(-i\mathbf{k} \cdot \mathbf{R}_i) a_{\mathbf{k}j}^\dagger \} = - \sum_i \mathbf{J}_i \cdot \mathbf{A}_i \\ &= \sum_{i\alpha\beta} \sum_{\mathbf{k}j} \sigma_{i\alpha\beta} \mathbf{J}_{\alpha\beta} \cdot (\mathbf{A}_{i\mathbf{k}j} a_{\mathbf{k}j} + \mathbf{A}_{i\mathbf{k}j}^* a_{\mathbf{k}j}^\dagger) \quad \text{with} \quad \mathbf{A}_i \equiv \mathbf{A}(\mathbf{R}_i) \quad \text{and} \quad \mathbf{A}_{i\mathbf{k}j} \equiv \mathbf{A}_{\mathbf{k}j}(\mathbf{R}_i). \end{aligned} \tag{2.15}$$

Here

$$\lambda_{\alpha\beta\mathbf{k}j} = \epsilon_{\alpha\beta} d_{\alpha\beta\mathbf{k}j} [2\pi/\omega(\mathbf{k})]^{1/2} n^{1/2} \quad \text{with} \quad \lambda_{\beta\alpha\mathbf{k}j} = -\lambda_{\alpha\beta\mathbf{k}j}^* \tag{2.16}$$

and

$$\mathbf{J}_{\alpha\beta} = -e \langle \alpha_i | d\mathbf{r}_i / dt | \beta_i \rangle = i\epsilon_{\alpha\beta} \boldsymbol{\mu}_{\alpha\beta} \tag{2.17}$$

are, respectively, a constant characterizing atom-photon interaction in the  $\mathbf{A} \cdot \mathbf{p}$  form and the matrix element of the current operator  $\mathbf{J}_i = -e(d\mathbf{r}_i/dt) = d\boldsymbol{\mu}_i/dt$  due to the electron bound to the  $i$  atom. In the above equations the quantities  $\epsilon_{\alpha\beta}$ ,  $\boldsymbol{\mu}_{\alpha\beta}$  and  $d_{\alpha\beta\mathbf{k}j}$  are defined by

$$\epsilon_{\alpha\beta} = \epsilon_\alpha - \epsilon_\beta, \quad \boldsymbol{\mu}_{\alpha\beta} = \langle \alpha_i | \boldsymbol{\mu}_i | \beta_i \rangle = \langle \alpha_i | -e\mathbf{r}_i | \beta_i \rangle, \quad d_{\alpha\beta\mathbf{k}j} = \boldsymbol{\mu}_{\alpha\beta} \cdot \mathbf{e}_{\mathbf{k}j}. \tag{2.18}$$

It has been assumed that the dipole moment operator  $\boldsymbol{\mu}_i = -e\mathbf{r}_i$  has no diagonal matrix element for any of the  $\alpha$ 's. Combining Eq. (2.17) with the commutation relation obeyed by  $\mathbf{r}_i$  and  $\mathbf{p}_i$  gives the well-known sum-rule:<sup>22)</sup>

$$\sum_{\beta} 2m\epsilon_{\alpha\beta} \mu_{\alpha\beta}^u \mu_{\beta\alpha}^u / e^2 = \mathcal{J}(u, v), \quad (u, v = x, y, z) \tag{2.19}$$

where  $\mu_{\alpha\beta}^u$  is the  $u$  component of the  $\alpha\beta$  element of the dipole moment  $\boldsymbol{\mu}_i$ .

### § 3. Diagonalization of the $A^2$ term plus the free photon Hamiltonian

For the sake of simplicity, we limit our discussion henceforth to the case in which all the atoms in the matter system are arranged on periodic lattice sites.

Here the quantity  $\rho(\mathbf{k})$  in Eqs. (2.13) and (2.14) becomes  $\rho(\mathbf{k}) = \mathcal{A}(\mathbf{k}, \mathbf{G})$ , where  $\mathbf{G}$  is the reciprocal lattice vector. We can neglect the optical Umklapp process, thus putting  $\mathbf{G} = 0$ , since the wavelength of relevant photons is much longer than the lattice constant. In this specific case Eq. (2.12) reduces to

$$H_p + H_{ap}^{(2)} = \sum_{\mathbf{k}j} [\{\omega(\mathbf{k}) + [\omega_p^2/2\omega(\mathbf{k})]\} a_{\mathbf{k}j}^+ a_{\mathbf{k}j} + [\omega_p^2/4\omega(\mathbf{k})] (a_{\mathbf{k}j}^+ a_{-\mathbf{k}j}^+ + a_{\mathbf{k}j} a_{-\mathbf{k}j})]. \quad (3.1)$$

Let  $b_{\mathbf{k}j}$  and  $b_{\mathbf{k}j}^+$  be new Bose operators defined by the Bogoliubov transformation<sup>23)</sup>

$$a_{\mathbf{k}j} = \xi_{\mathbf{k}j} b_{\mathbf{k}j} - \eta_{\mathbf{k}j} b_{-\mathbf{k}j}^+ \quad \text{and} \quad a_{\mathbf{k}j}^+ = \xi_{\mathbf{k}j} b_{\mathbf{k}j}^+ - \eta_{\mathbf{k}j} b_{-\mathbf{k}j} \quad (3.2)$$

with

$$\xi_{\mathbf{k}j}^2 - \eta_{\mathbf{k}j}^2 = 1, \quad (3.3)$$

where the  $\xi$ 's and  $\eta$ 's are real. Then, Eq. (3.1) can be diagonalized as follows:

$$H_p + H_{ap}^{(2)} = \sum_{\mathbf{k}j} \Omega(\mathbf{k}) b_{\mathbf{k}j}^+ b_{\mathbf{k}j} \quad (3.4)$$

with

$$[b_{\mathbf{k}j}, b_{\mathbf{k}'j'}^+] = \mathcal{A}(\mathbf{k}, \mathbf{k}') \mathcal{A}(j, j'), \quad [b_{\mathbf{k}j}, b_{\mathbf{k}'j'}] = [b_{\mathbf{k}j}^+, b_{\mathbf{k}'j'}^+] = 0, \quad (3.5)$$

where

$$\Omega(\mathbf{k}) = [k^2 + \omega_p^2]^{1/2} \quad (3.6)$$

is the eigenfrequencies of quasi-photons corresponding to the new operators  $b_{\mathbf{k}j}$  and  $b_{\mathbf{k}j}^+$ . Diagonalization of the right-hand side of Eq. (2.12) for a general case of arbitrary spatial configurations of atoms in the matter system is done in the Appendix.

By the use of the transformation (3.2) the interaction Hamiltonian, the vector potential and other field variables can be expressed entirely in terms of the new Bose operators  $b_{\mathbf{k}j}$  and  $b_{\mathbf{k}j}^+$ . Combining Eqs. (2.4) and (3.2), we get

$$\mathbf{A}(\mathbf{r}) = \sum_{\mathbf{k}j} [2\pi/\Omega(\mathbf{k}) V]^{1/2} \mathbf{e}_{\mathbf{k}j} \{ \exp(i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j} + \exp(-i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j}^+ \}, \quad (3.7)$$

where we have used the relation

$$\xi_{\mathbf{k}j} - \eta_{\mathbf{k}j} = [\omega(\mathbf{k})/\Omega(\mathbf{k})]^{1/2}. \quad (3.8)$$

Similarly, we get for the electric field  $\mathbf{E}(\mathbf{r})$  and the magnetic field  $\mathbf{B}(\mathbf{r})$ :

$$\mathbf{E}(\mathbf{r}) = i \sum_{\mathbf{k}j} [2\pi\Omega(\mathbf{k})/V]^{1/2} \mathbf{e}_{\mathbf{k}j} \{ \exp(i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j} - \exp(-i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j}^+ \}, \quad (3.9)$$

$$\mathbf{B}(\mathbf{r}) = i \sum_{\mathbf{k}j} [2\pi/\Omega(\mathbf{k}) V]^{1/2} (\mathbf{k} \times \mathbf{e}_{\mathbf{k}j}) \{ \exp(i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j} - \exp(-i\mathbf{k} \cdot \mathbf{r}) b_{\mathbf{k}j}^+ \}. \quad (3.10)$$

It is of interest to note that Eqs. (3.7), (3.9) and (3.10) are identical with Eq. (2.4) and the conventional expressions for  $\mathbf{E}(\mathbf{r})$  and  $\mathbf{B}(\mathbf{r})$  written in terms of the  $a$ 's, provided  $a_{\mathbf{k}j}$ ,  $a_{\mathbf{k}j}^+$  and  $\omega(\mathbf{k})$  are replaced by  $b_{\mathbf{k}j}$ ,  $b_{\mathbf{k}j}^+$  and  $\Omega(\mathbf{k})$ , respectively.

Using the same procedure, we can reduce Eqs. (2·15) to

$$H_{\text{ap}}^{(1)} = -iN^{-1/2} \sum_{i\alpha\beta} \sum_{kj} \tilde{\lambda}_{\alpha\beta kj} \sigma_{i\alpha\beta} \{ \exp(i\mathbf{k} \cdot \mathbf{R}_i) b_{kj} + \exp(-i\mathbf{k} \cdot \mathbf{R}_i) b_{kj}^+ \}, \quad (3\cdot11)$$

where

$$\tilde{\lambda}_{\alpha\beta kj} = \epsilon_{\alpha\beta} d_{\alpha\beta kj} [2\pi/\Omega(\mathbf{k})]^{1/2} n^{1/2}. \quad (3\cdot12)$$

As in the case of the field variables, the interaction constant  $\tilde{\lambda}_{\alpha\beta kj}$  is also identical in form with the bare interaction constant  $\lambda_{\alpha\beta kj}$ , provided  $\Omega(\mathbf{k})$  is replaced by  $\omega(\mathbf{k})$ . It is therefore seen that the diagonalization of the  $A^2$  term plus the free photon Hamiltonian leads to the renormalization of the atom-photon interaction constants in the  $\mathbf{A} \cdot \mathbf{p}$  form. From the result thus obtained, we can express the total Hamiltonian  $H$  in terms of the new photon operators and the atomic operators in the form

$$H = \sum_{kj} \Omega(\mathbf{k}) b_{kj}^+ b_{kj} + \sum_{i\alpha} \epsilon_{\alpha} \sigma_{i\alpha\alpha} + (1/2) \sum_{ij} \sum_{\alpha\beta\gamma\delta} L_{\alpha\beta,\gamma\delta}(ij) \sigma_{i\alpha\beta} \sigma_{j\gamma\delta} - iN^{-1/2} \sum_{i\alpha\beta} \sum_{kj} \tilde{\lambda}_{\alpha\beta kj} \sigma_{i\alpha\beta} \{ \exp(i\mathbf{k} \cdot \mathbf{R}_i) b_{kj} + \exp(-i\mathbf{k} \cdot \mathbf{R}_i) b_{kj}^+ \}. \quad (3\cdot13)$$

In this renormalized form, equations obeyed by the field variables are immediately obtained by using Eq. (3·6). For the vector potential  $\mathbf{A}(\mathbf{r})$  and the electric field  $\mathbf{E}(\mathbf{r})$  we get the equations

$$\{ \Delta - (\partial^2/\partial t^2) - \omega_p^2 \} \mathbf{A}(\mathbf{r}) = -4\pi \mathbf{J}(\mathbf{r}), \quad (3\cdot14)$$

$$\{ \Delta - (\partial^2/\partial t^2) - \omega_p^2 \} \mathbf{E}(\mathbf{r}) = 4\pi (\partial^2 \mathbf{P}(\mathbf{r})/\partial t^2). \quad (3\cdot15)$$

Here

$$\mathbf{J}(\mathbf{r}) = \sum_i' \mathbf{J}_i \mathcal{A}(\mathbf{r} - \mathbf{R}_i) \quad \text{and} \quad \mathbf{P}(\mathbf{r}) = \sum_i' \boldsymbol{\mu}_i \mathcal{A}(\mathbf{r} - \mathbf{R}_i) \quad (3\cdot16)$$

are the current density and the electric polarization density, respectively, at the site  $\mathbf{r}$ . In Eqs. (3·16) the prime on the summation symbol denotes a sum over the atomic sites in a unit volume. The physical meaning of the new photon operators is understood by noting that neglect of the right-hand sides of Eqs. (3·14) and (3·15) gives the well-known expression for the dispersion of electromagnetic waves in a plasma:<sup>17)</sup>

$$\omega^2 = \Omega(\mathbf{k})^2 = k^2 + \omega_p^2. \quad (3\cdot17)$$

We close this section by giving the following remarks: The  $A^2$  term was taken into account previously by Agranovich<sup>17)</sup> and by Hopfield<sup>18)</sup> in their theories of the dispersion of polaritons. These workers, however, limited from the outset their discussion to linear excitations in matter systems, which amount here to replacing the atomic operators  $\sigma_{i\alpha\beta}$  by Bose-type exciton operators  $B_i$  and  $B_i^+$  with  $[B_i, B_j^+] = \mathcal{A}(i, j)$ ,  $[B_i, B_j] = [B_i^+, B_j^+] = 0$ . The total Hamiltonian of the matter-photon system studied by these workers can be written in the form

$$H = \sum_{kj} [ \{ \omega(\mathbf{k}) + [\omega_p^2/2\omega(\mathbf{k})] \} a_{kj}^+ a_{kj} + [\omega_p^2/4\omega(\mathbf{k})] (a_{kj}^+ a_{-kj}^+ + a_{kj} a_{-kj}) ]$$

$$\begin{aligned}
 & + \sum_i [\epsilon B_i^+ B_i + \sum_j K(ij) (B_i^+ B_j + B_i B_j^+ + B_i^+ B_j^+ + B_i B_j)] \\
 & - iN^{-1/2} \sum_i \sum_{kj} (\lambda_{ikj} B_i^+ - \lambda_{ikj}^* B_i) \{ \exp(i\mathbf{k} \cdot \mathbf{R}_i) a_{kj} + \exp(-i\mathbf{k} \cdot \mathbf{R}_i) a_{kj}^+ \}. \quad (3.18)
 \end{aligned}$$

Here  $\epsilon$  is the excitation energy of the atom and  $K(ij)$  denotes the dipole-dipole interaction energy between the  $i$  and  $j$  atoms. It is seen that replacing the photon operators  $a_{kj}$  and  $a_{kj}^+$  and the interaction constants  $\lambda_{\alpha\beta kj}$  by  $b_{kj}$ ,  $b_{kj}^+$  and  $\tilde{\lambda}_{\alpha\beta kj}$ , respectively, gives Eq. (3.13) with the atomic operators  $\sigma_{i\alpha\beta}$  replaced by the Bose-type exciton operators  $B_i$  and  $B_i^+$ . Introducing the Fourier transform  $B_k = N^{-1/2} \times \sum_i \exp(i\mathbf{k} \cdot \mathbf{R}_i) B_i$  and  $B_k^+ = N^{-1/2} \sum_i \exp(-i\mathbf{k} \cdot \mathbf{R}_i) B_i^+$  of the exciton operators and employing a canonical transformation similar to Eq. (A.1), these workers succeeded in diagonalizing the Hamiltonian (3.18) as a whole.<sup>20)</sup>

#### § 4. Stationary coherent states in a matter-photon system

As an illustrative example to elucidate the role played by the  $A^2$  term, we study stationary coherent states in a matter-photon system. For this purpose, we consider a specific model of a matter-photon system composed of identical two-level atoms interacting with a single-mode radiation field with a photon of energy  $\Omega(\mathbf{k}) \equiv \Omega$  and momentum  $\mathbf{k}$ .\*) We also assume that the wave function of the ground state and that of the excited state of the atom specified by the indices  $\alpha=0$  and 1, respectively, are real. We rewrite the atomic operators in terms of the Pauli operators as

$$\begin{aligned}
 \sigma_{i10} &= \sigma_i^+, \quad \sigma_{i01} = \sigma_i^-, \quad \sigma_{i11} = \sigma_i^z + (1/2), \quad \sigma_{i00} = (1/2) - \sigma_i^z, \\
 (\sigma_{i10} + \sigma_{i01})/2 &= \sigma_i^x, \quad (\sigma_{i10} - \sigma_{i01})/2i = \sigma_i^y, \quad (\sigma_{i11} - \sigma_{i00})/2 = \sigma_i^z, \quad (4.1)
 \end{aligned}$$

together with abbreviations

$$\begin{aligned}
 \epsilon_{10} &= \epsilon, \quad \boldsymbol{\mu}_{10} = \boldsymbol{\mu}_{01} = \boldsymbol{\mu}, \quad \mathbf{J}_{10} = \mathbf{J}_{01} = i\epsilon \boldsymbol{\mu}, \\
 \tilde{\lambda}_{10kj} &= \tilde{\lambda} = \epsilon d (2\pi/\Omega)^{1/2} n^{1/2}, \quad \mathbf{e}_{kj} = \mathbf{e}_1, \quad d = \boldsymbol{\mu} \cdot \mathbf{e}_1. \quad (4.2)
 \end{aligned}$$

We note in passing that by the use of Eq. (2.9) the atomic dipole moment operator  $\boldsymbol{\mu}_i$  can be expressed as follows:

$$\boldsymbol{\mu}_i = \boldsymbol{\mu}_{10} \sigma_{i10} + \boldsymbol{\mu}_{01} \sigma_{i01} = 2\boldsymbol{\mu} \sigma_i^x. \quad (4.3)$$

Then, on the assumption that atom-atom interactions are described by multi-pole or exchange-type interactions, Eq. (3.13) can be rewritten as

$$\begin{aligned}
 H &= \Omega b^+ b + \epsilon \sum_i \sigma_i^z + 2\tilde{\lambda} N^{-1/2} \sum_i \sigma_i^y \{ \exp(i\mathbf{k} \cdot \mathbf{R}_i) b + \exp(-i\mathbf{k} \cdot \mathbf{R}_i) b^+ \} \\
 & + 2 \sum_{ij} \{ K_1(ij) \sigma_i^x \sigma_j^x + K_2(ij) \sigma_i^y \sigma_j^y + K_3(ij) \sigma_i^z \sigma_j^z \}, \quad (4.4)
 \end{aligned}$$

where  $K_n(ij)$  ( $n=1, 2, 3$ ) are constants of the atom-atom interaction.

\*) We omit the indices  $k$  and  $j$  attached to quantities or operators pertaining to the photon.



Let  $|\beta\rangle = |r, \psi\rangle$  and  $|\theta_i, \varphi_i\rangle$  be the coherent state of the photon mode<sup>253</sup> and that of the Pauli-spin operator  $\sigma_i = (\sigma_i^x, \sigma_i^y, \sigma_i^z)$  characterized by angles  $\theta_i$  and  $\varphi_i$  in the spherical polar coordinate.<sup>261~281</sup> Here  $r$  and  $\psi$  are action and angle variables which are related to the eigenvalue  $\beta$  of the photon operator  $b$  by  $\beta = r \exp(i\psi)$ . The coherent states of the whole spin system are denoted by  $|\{\theta\}, \{\varphi\}\rangle$ . We evaluate the expectation value  $\langle H \rangle = \langle r, \psi, \{\theta\}, \{\varphi\} | H | r, \psi, \{\theta\}, \{\varphi\} \rangle$  of Eq. (4.4) with respect to the photon and spin or atomic coherent states  $|r, \psi, \{\theta\}, \{\varphi\}\rangle$ . By the use of the relations

$$\begin{aligned} \langle r, \psi | b | r, \psi \rangle &= r \exp(i\psi), & \langle r, \psi | b^\dagger | r, \psi \rangle &= r \exp(-i\psi), \\ \langle \theta_i, \varphi_i | \sigma_i^x | \theta_i, \varphi_i \rangle &= (1/2) \sin \theta_i \cos \varphi_i, & \langle \theta_i, \varphi_i | \sigma_i^y | \theta_i, \varphi_i \rangle &= (1/2) \sin \theta_i \sin \varphi_i, \\ \langle \theta_i, \varphi_i | \sigma_i^z | \theta_i, \varphi_i \rangle &= (1/2) \cos \theta_i, \end{aligned} \tag{4.5}$$

we then obtain.

$$\begin{aligned} \langle H \rangle &= \Omega r^2 + (\epsilon/2) \sum_i \cos \theta_i + (2\tilde{\lambda} N^{-1/2}) r \sum_i \sin \theta_i \sin \varphi_i \cos(\psi + \mathbf{k} \cdot \mathbf{R}_i) \\ &+ (1/2) \sum_{ij} \{ K_1(ij) \sin \theta_i \sin \theta_j \cos \varphi_i \cos \varphi_j \\ &+ K_2(ij) \sin \theta_i \sin \theta_j \sin \varphi_i \sin \varphi_j + K_3(ij) \cos \theta_i \cos \theta_j \}. \end{aligned} \tag{4.6}$$

The expectation value  $\langle H \rangle$  of the Hamiltonian is stationary with respect to the variation of  $r, \psi, \theta_i, \varphi_i$  for

$$\partial \langle H \rangle / \partial r = \partial \langle H \rangle / \partial \psi = 0; \quad \partial \langle H \rangle / \partial \theta_i = \partial \langle H \rangle / \partial \varphi_i = 0 \text{ for all } i. \tag{4.7}$$

Let a solution of Eqs. (4.7) for which  $\langle H \rangle$  is minimum be  $r_0, \psi_0, \theta_{i0}, \varphi_{i0}$ . We are concerned here only with those  $\theta_{i0}$  and  $\varphi_{i0}$  which are independent of the site index  $i$ ; such a solution is considered to yield the lowest of the minimum of  $\langle H \rangle$  to which we pay particular attention (hereafter we omit the index  $i$  for  $\theta_{i0}$  and the  $\varphi_{i0}$ ). Physically, this corresponds to a uniform atomic polarization. A simple calculation gives

$$\left. \begin{aligned} \psi_0 + \mathbf{k} \cdot \mathbf{R}_i = 0 \\ \varphi_0 = 3\pi/2 \end{aligned} \right\} \text{ or } \left. \begin{aligned} \psi_0 + \mathbf{k} \cdot \mathbf{R}_i = \pi \\ \varphi_0 = \pi/2 \end{aligned} \right\} \text{ for all } i, \tag{4.8a}$$

$$I_0 = N^{1/2} (\tilde{\lambda}/\Omega) \sin \theta_0, \quad \theta_0 = \cos^{-1}(-1/g), \tag{4.8b}$$

where

$$g = (4\tilde{\lambda}^2/\epsilon\Omega) + 2(K_3/\epsilon) - 2(K_2/\epsilon) \quad \text{with} \quad K_n = \sum_j K_n(ij). \quad (n=1, 2, 3) \tag{4.9}$$

The solution (4.8) can exist only when the condition

$$\mathbf{k} = 0 \tag{4.10a}$$

and

$$|g| > 1 \tag{4.10b}$$

is satisfied. Inserting the fourth of Eqs. (4·2) into Eq. (4·9), we can rewrite the condition (4·10b) as

$$|g| = |[2m\epsilon d^2/e^2 \{1 + (\omega/\omega_p)\}] + [2(K_3 - K_2)/\epsilon]| > 1. \quad (4\cdot11)$$

On the other hand the sum rule (2·19) imposes the condition

$$2m\epsilon d^2/e^2 < 1, \quad (4\cdot12)$$

since we are concerned here only with two states among many atomic eigenstates. Thus, it is seen that the condition (4·10b) or (4·11) cannot be satisfied when the direct atom-atom interaction is absent. This is identical with the result obtained Rzażewski et al.<sup>8)</sup> The presence of the term  $2(K_3 - K_2)/\epsilon$  here, however, does not necessarily invalidate inequality (4·11), provided  $K_2 < 0$  and  $K_3 > 0$ . Since we get  $K_3 > 0$  in almost all cases of physical interest<sup>29)</sup> and the dipole-dipole interaction energy here is expressible entirely in terms of the  $\sigma^x$ 's, it is seen from the result obtained above that in order for inequality (4·11) to be satisfied, an exchange-type, rather than dipole-dipole, interaction with  $K_2 < 0$  must exist.

To see the physical meaning of the solution (4·8), let us consider the expectation values of the vector potential  $\mathbf{A}(\mathbf{R}_i)$ , the electric field  $\mathbf{E}(\mathbf{R}_i)$  and the current density operator  $\mathbf{J} = (1/V) \sum_i \mathbf{J}(\mathbf{R}_i) = -(2\epsilon\boldsymbol{\mu}/V) \sum_i \sigma_i^y$  at the stationary point  $r = r_0$ ,  $\psi = \psi_0$ ,  $\theta_i = \theta_0$  and  $\varphi_i = \varphi_0$ . Let us put  $\langle r_0, \psi_0 | \mathbf{A}(\mathbf{R}_i) | r_0, \psi_0 \rangle = \mathbf{A}_0(\mathbf{R}_i)$ ,  $\langle r_0, \psi_0 | \mathbf{E}(\mathbf{R}_i) | r_0, \psi_0 \rangle = \mathbf{E}_0(\mathbf{R}_i)$  and  $\langle \{\theta_0\}, \{\varphi_0\} | \mathbf{J} | \{\theta_0\}, \{\varphi_0\} \rangle = \mathbf{J}_0$ . Then, applying Eqs. (3·7) and (3·9) to the case of the single photon mode and using Eqs. (4·2) and (4·8), we get

$$\mathbf{A}_0(\mathbf{R}_i) = \pm 4\pi n\epsilon (\mathbf{e}_1 \cdot \boldsymbol{\mu}) \sin \theta_0 \mathbf{e}_1 / \Omega^2 \equiv \mathbf{A}_0, \quad \mathbf{E}_0(\mathbf{R}_i) = 0 \quad \text{and} \quad \mathbf{J}_0 = \pm n\epsilon \sin \theta_0 \boldsymbol{\mu}. \quad (4\cdot13)$$

Here the plus (minus) sign corresponds to the first (second) of Eqs. (4·8a). It is seen that the vector potential  $\mathbf{A}_0(\mathbf{R}_i)$  is spatially uniform and the electric field  $\mathbf{E}_0(\mathbf{R}_i)$  vanishes. This result corresponds to the solution  $\psi_0 + \mathbf{k} \cdot \mathbf{R}_i = 0$  or  $\psi_0 + \mathbf{k} \cdot \mathbf{R}_i = \pi$  for all  $i$  in Eqs. (4·8a); such a solution can exist only for  $\mathbf{k} = 0$ . Combining the first and the third of Eqs. (4·13) gives

$$\Omega^2 \mathbf{e}_1 \cdot \mathbf{A}_0 = 4\pi \mathbf{e}_1 \cdot \mathbf{J}_0. \quad (4\cdot14)$$

We can understand this result as follows: The stationary point determined by Eqs. (4·7) corresponds to the appearance of a uniform coherent atomic polarization or a coherent current induced by a static vector potential or a Bose-condensed photon with  $\mathbf{k} = 0$ , while a coherent vector potential is produced by the coherent atomic polarization. It is seen from Eqs. (4·8) that the stationary coherent states which can exist simultaneously for photon and atomic polarization are characterized by

$$\langle I_0, \psi_0 | b | r_0, \psi_0 \rangle = O(N^{1/2}), \quad \langle \{\theta_0\}, \{\varphi_0\} | \sigma_i^x | \{\theta_0\}, \{\varphi_0\} \rangle = 0$$

and

$$\langle \{\theta\}, \{\varphi\} | \sigma_i^y | \{\theta\}, \{\varphi\} \rangle \neq 0. \tag{4.15}$$

The phase transition as discussed by Rzażewski et al.<sup>8)</sup> can be considered as the onset of nonvanishing of  $\langle r, \psi | b | r, \psi \rangle$  and  $\langle \{\theta\}, \{\varphi\} | \sigma_i^y | \{\theta\}, \{\varphi\} \rangle$ . The result (4.13) and (4.15) may probably preclude a possibility of a simultaneous appearance of a stationary photon coherent state and a stationary coherent state of atomic polarization due to dipole-dipole interaction which could arise if the  $\mathbf{E} \cdot \mathbf{r}$  form of matter-photon interaction is to be used. Let  $\mathbf{P}_0$  be the expectation value  $\langle \{\theta\}, \{\varphi\} | \mathbf{P} | \{\theta\}, \{\varphi\} \rangle$  at the stationary point  $\theta_i = \theta_0$  and  $\varphi_i = \varphi_0$  of the dipole moment operator  $\mathbf{P}$  defined by  $\mathbf{P} = (2\boldsymbol{\mu}/V) \sum_i \sigma_i^x$ . Then, in the case of the Dicke model Hamiltonian in which the  $\mathbf{E} \cdot \mathbf{r}$  form of the atom-photon interaction is used, it was shown in the previous paper<sup>2)</sup> that the electric field  $\mathbf{E}_0(\mathbf{R}_i)$  and  $\mathbf{P}_0$  become nonvanishing and related to each other by the relation  $\mathbf{E}_0 = -4\pi\mathbf{P}_0$ .\*) Stationary coherent states in such a case are characterized by  $\langle \{\theta\}, \{\varphi\} | \sigma_i^x | \{\theta\}, \{\varphi\} \rangle \neq 0$  as well as  $\langle r, \psi | b | r, \psi \rangle = O(N^{1/2})$ .

We are also concerned here with the expectation value of energy at the stationary state,  $\langle H \rangle_s$ , and that corresponding to the state in which all the atoms are in their ground state while no photon is present,  $\langle H \rangle_0$ . The former and the latter are obtained by inserting Eqs. (4.8) and the relation  $r=0$  and  $\cos \theta_i = \pi$  for all  $i$ , respectively, into Eq. (4.6) as

$$\langle H \rangle_s = - (N\epsilon/2) \{g + (1/g)\} + N(K_3/\epsilon) < - (N\epsilon/2) + N(K_3/\epsilon) = \langle H \rangle_0. \tag{4.16}$$

Here the inequality holds since inequality (4.10b) reduces to  $g > 1$  for  $K_2 < 0$  and  $K_3 > 0$ .

Thus we have shown that when atom-atom interactions are taken into account, matter-photon interaction can induce, under certain circumstances, a simultaneous appearance of a coherently polarized atomic state and a Bose-condensed photon as shown by Eqs. (4.8), with energy eigenvalue lower than that of the state in which all the atoms in the matter system are in their ground state while no photon is present. The role played by the  $A^2$  term is most easily seen by considering the case in which the direct atom-atom interaction is neglected. Here the condition for the appearance of the stationary coherent state is given by

$$4\lambda^2/\epsilon\omega > 1 \quad \text{or} \quad (8\pi\epsilon a^2/\omega^2)n > 1. \tag{4.17}$$

This is identical with the results obtained by Hepp and Lieb<sup>9)</sup> and by Wang and Hioe<sup>10)</sup> for the occurrence of the second order phase transition in the conventional Dicke model in which the  $A^2$  term is omitted. Such a condition can always be satisfied for a sufficiently large value of the atomic number density  $n = N/V$ , a result which is qualitatively different from the condition

\*) We have rewritten  $\mathbf{E}_0(\mathbf{R}_i)$  as  $\mathbf{E}_0$ , since in the conventional Dicke model the spatial dimension of the matter system is taken to be small compared with the wavelength of relevant photon.

$$4\tilde{\lambda}^2/\epsilon\omega > 1 \quad \text{or} \quad 2mc d^2/e^2 \{1 + (\omega/\omega_p)^2\} > 1. \tag{4.18}$$

This is the same as the condition obtained by Rzażewski et al.<sup>9)</sup> for the occurrence of the second order phase transition in the Dicke model with the  $A^2$  term included.\*<sup>3)</sup> Due to inequality (4.12), however, the relation (4.18), which is obtained from (4.11) by neglecting  $K_2$  and  $K_3$ , can never be satisfied.

**§ 5. Canonical transformation and effective atom-atom interaction**

It is shown that the condition (4.10) or (4.11) can also be derived from the following effective spin Hamiltonian:

$$H_{\text{eff}} = \epsilon \sum_i \sigma_i^z - (4\tilde{\lambda}^2/2N) \sum_{ij} \sigma_i^y \sigma_j^y + 2 \sum_{ij} \{K_1(ij) \sigma_i^x \sigma_j^x + K_2(ij) \sigma_i^y \sigma_j^y + K_3(ij) \sigma_i^z \sigma_j^z\}. \tag{5.1}$$

Namely, inequality (4.10) or (4.11) can be derived by demanding the condition for the expectation value  $\langle H_{\text{eff}} \rangle = \langle \{\theta\} \{\varphi\} | H_{\text{eff}} | \{\theta\} \{\varphi\} \rangle$  of  $H_{\text{eff}}$  with respect to the spin coherent states  $|\{\theta\} \{\varphi\}\rangle$  to be stationary:

$$\partial \langle H_{\text{eff}} \rangle / \partial \theta_i = \partial \langle H_{\text{eff}} \rangle / \partial \varphi_i = 0 \quad \text{for all } i. \tag{5.2}$$

As in the case of the previous section, we are concerned here only with those solutions  $\theta_{i0}$  and  $\varphi_{i0}$  of Eqs. (5.2) which are independent of the site index  $i$ . The equation  $\partial \langle H_{\text{eff}} \rangle / \partial \varphi_i = 0$  then gives the condition  $\sin \varphi \cos \varphi = 0$ . We are interested here in the solution

$$\cos \varphi = 0 \quad \text{or} \quad \varphi = \pi/2 \quad \text{or} \quad 3\pi/2, \tag{5.3}$$

which yields a non-vanishing contribution from the second term of Eq. (5.1). Equation (5.3) is identical with the second of Eqs. (4.8a). The equation  $\partial \langle H_{\text{eff}} \rangle / \partial \theta_i = 0$  then is identical with the second of Eqs. (4.8b). Thus Eq. (5.1) is equivalent to Eq. (4.4) for the coherent photon mode with  $\mathbf{k} = 0$ , as far as we are concerned with a uniform stationary atomic coherent state.

To study the physical meaning of this effective Hamiltonian, we apply a canonical transformation to Eq. (4.4) to eliminate the third term representing the matter-photon interaction. This is done by introducing new Bose operators  $\hat{b}$  and  $\hat{b}^+$  and new spin operators  $\hat{\sigma}_i^x, \hat{\sigma}_i^y, \hat{\sigma}_i^z$  by the equations<sup>30)</sup>

$$\left. \begin{aligned} \hat{b} &= \exp(-iS) b \exp(iS) = b + 2(\tilde{\lambda}/N^{1/2}Q) \sum_i \exp(-i\mathbf{k} \cdot \mathbf{R}_i) \sigma_i^y \\ \hat{b}^+ &= \exp(-iS) b^+ \exp(iS) = b^+ + 2(\tilde{\lambda}/N^{1/2}Q) \sum_i \exp(i\mathbf{k} \cdot \mathbf{R}_i) \sigma_i^y \\ \hat{\sigma}_i^x &= \exp(-iS) \sigma_i^x \exp(iS) = \cos \phi_i \sigma_i^x - \sin \phi_i \sigma_i^z \end{aligned} \right\}, \tag{5.4}$$

\*<sup>3)</sup> From the results obtained by Hepp and Lieb and by Rzażewski et al., it is expected that inequality (4.11) obtained in this paper is also the condition for the occurrence of the second order phase transition in the Dicke model with the direct atom-atom interaction as well as the  $A^2$  term included. This point will be studied elsewhere.

$$\left. \begin{aligned} \hat{\sigma}_i^y &= \exp(-iS)\sigma_i^y \exp(iS) = \sigma_i^y \\ \hat{\sigma}_i^z &= \exp(-iS)\sigma_i^z \exp(iS) = \sin \phi_i \sigma_i^x + \cos \phi_i \sigma_i^z \end{aligned} \right\} \quad (5.5)$$

Here the operator  $S$  and the angle  $\phi_i$  are defined by the equation

$$\begin{aligned} S &= (2\tilde{\lambda}/iN^{1/2}\Omega) \sum_i \{ \exp(-i\mathbf{k} \cdot \mathbf{R}_i) b^+ - \exp(i\mathbf{k} \cdot \mathbf{R}_i) b \} \sigma_i^y \\ &= (2\tilde{\lambda}/iN^{1/2}\Omega) \sum_i \{ \exp(-i\mathbf{k} \cdot \mathbf{R}_i) \hat{b}^+ - \exp(i\mathbf{k} \cdot \mathbf{R}_i) \hat{b} \} \sigma_i^y \equiv \sum_i \phi_i \sigma_i^y. \end{aligned} \quad (5.6)$$

The canonical transformation here is to introduce a displacement of the photon operators on the one hand and a rotation of the spin operators around the  $y$ -axis on the other. Insertion of Eqs. (5.4) and (5.5) into Eq. (4.4) gives an expression for the Hamiltonian written entirely in terms of the new Bose operators  $\hat{b}$  and  $\hat{b}^+$  and the new spin operators  $\hat{\sigma}_i^x, \hat{\sigma}_i^y, \hat{\sigma}_i^z$ :

$$\begin{aligned} H &= \Omega \hat{b}^+ \hat{b} + \sum_i \epsilon \cos \phi_i \hat{\sigma}_i^z - \sum_i \epsilon \sin \phi_i \hat{\sigma}_i^x - 4(\tilde{\lambda}^2/N\Omega) \sum_{ij} \exp[i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)] \sigma_i^y \sigma_j^y \\ &+ 2 \sum_{ij} \{ \hat{K}_1(ij) \hat{\sigma}_i^x \hat{\sigma}_j^x + K_2(ij) \hat{\sigma}_i^y \hat{\sigma}_j^y + \hat{K}_3(ij) \hat{\sigma}_i^z \hat{\sigma}_j^z + K'(ij) \hat{\sigma}_i^x \hat{\sigma}_j^z \}, \end{aligned} \quad (5.7)$$

where

$$\begin{aligned} \hat{K}_1(ij) &= K_1(ij) \cos \phi_i \cos \phi_j + K_3(ij) \sin \phi_i \sin \phi_j, \\ \hat{K}_3(ij) &= K_1(ij) \sin \phi_i \sin \phi_j + K_3(ij) \cos \phi_i \cos \phi_j, \\ K'(ij) &= 2 \{ K_1(ij) \cos \phi_i \sin \phi_j - K_3(ij) \sin \phi_i \cos \phi_j \} \end{aligned} \quad (5.8)$$

The Hamiltonian (5.7) has the following physical meaning: (i) The energy  $\epsilon \cos \phi_i$  in the new basis states undergoes fluctuations due to the factor  $\cos \phi_i$  being a function of the photon operators  $\hat{b}$  and  $\hat{b}^+$ . (ii) There arises a non-diagonal part of the energy of the atom represented by the factor  $\epsilon \sin \phi_i \hat{\sigma}_i^x$ . (iii) The inter-atomic interaction energies  $\hat{K}_1(ij)$  and  $\hat{K}_3(ij)$  undergo fluctuations, which give rise to the destruction of the phase coherence of the electronic wave functions. (iv) The fourth term in Eq. (5.7) represents effective atom-atom interaction of infinite range resulting from virtual exchange of the photon. The effect of the diagonalization of the  $A^2$  term here is seen by the fact that effective atom-atom interaction is also renormalized due to the presence of the factor  $\tilde{\lambda}$ .

We introduce the reduced Hamiltonian  $H_{\text{red}}$  by the equation

$$H_{\text{red}} = \langle \tilde{O} | H | \tilde{O} \rangle. \quad (5.9)$$

Here  $|\tilde{O}\rangle$  is the ground state of the new photon state defined by

$$\hat{b} |\tilde{O}\rangle = 0. \quad (5.10)$$

The expectation values of the terms such as  $\cos \phi_i, \sin \phi_i, \cos \phi_i \cos \phi_j, \sin \phi_i \sin \phi_j$ , etc. with respect to  $|\tilde{O}\rangle$  can be obtained by making repeated use of the identity<sup>30)</sup>

$$\exp(c_1 \hat{b}^+ + c_2 \hat{b}) = \exp(c_1 c_2 / 2) \exp(c_1 \hat{b}^+) \exp(c_2 \hat{b}) \quad (5.11)$$

to get the normal ordered form, where  $c_1$  and  $c_2$  are constants, and Eq. (5·10). A straightforward calculation leads to the following result:

$$\begin{aligned} \langle \tilde{O} | \cos \phi_i | \tilde{O} \rangle &= \exp(-g_0/2), \quad \langle \tilde{O} | \sin \phi_i | \tilde{O} \rangle = 0, \\ \langle \tilde{O} | \cos \phi_i \cos \phi_j | \tilde{O} \rangle &= \{ \exp(-g_0)/2 \} \{ \exp[g_0 \exp\{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)\}] \\ &\quad + \exp[-g_0 \exp\{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)\}] \}, \\ \langle \tilde{O} | \sin \phi_i \sin \phi_j | \tilde{O} \rangle &= \{ \exp(-g_0)/2 \} \{ \exp[g_0 \exp\{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)\}] \\ &\quad - \exp[-g_0 \exp\{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)\}] \}, \\ \langle \tilde{O} | \sin \phi_i \cos \phi_j | \tilde{O} \rangle &= \langle \tilde{O} | \cos \phi_i \sin \phi_j | \tilde{O} \rangle = 0, \end{aligned} \quad (5 \cdot 12)$$

where

$$g_0 = 4\tilde{\lambda}^2/N\Omega^2 = [2m\epsilon d^2/\epsilon^2 \{1 + (\omega/\omega_p)^2\}] (\epsilon/\Omega) (1/N). \quad (5 \cdot 13)$$

The quantity  $g_0$  vanishes in the thermodynamic limit, namely,

$$g_0 \rightarrow 0 \text{ for } N \rightarrow \infty, \text{ with } n = N/V \text{ being kept constant} \quad (5 \cdot 13')$$

due to the inequality (4·12) and to the factor  $\epsilon/\Omega$  being at least of the order of unity. Combining Eqs. (5·7) ~ (5·10) and (5·12) and putting  $\mathbf{k} = 0$ , which is the condition for the first of the solution (4·8a) to exist, we get

$$\begin{aligned} H_{\text{red}} &= \sum_i \epsilon' \sigma_i^z - (4\tilde{\lambda}^2/N\Omega) \sum_{ij} \hat{\sigma}_i^y \hat{\sigma}_j^y \\ &\quad + 2 \sum_{ij} \{ K_1'(ij) \hat{\sigma}_i^x \hat{\sigma}_j^x + K_2'(ij) \hat{\sigma}_i^y \hat{\sigma}_j^y + K_3'(ij) \hat{\sigma}_i^z \hat{\sigma}_j^z \}, \end{aligned} \quad (5 \cdot 14)$$

where

$$\epsilon' = \epsilon \exp(-g_0/2), \quad (5 \cdot 15a)$$

$$K_1'(ij) = (1/2) \{1 + \exp(-g_0)\} K_1(ij) + \{1 - \exp(-g_0)\} K_3(ij), \quad (5 \cdot 15b)$$

$$K_3'(ij) = (1/2) \{1 - \exp(-g_0)\} K_1(ij) + \{1 + \exp(-g_0)\} K_3(ij). \quad (5 \cdot 15c)$$

We observe the relations

$$\begin{aligned} \epsilon' = \epsilon, \quad K_1'(ij) = K_1(ij) \quad \text{and} \quad K_3'(ij) = K_3(ij) \quad \text{for } N \rightarrow \infty \text{ and} \\ n = N/V = \text{const.} \end{aligned} \quad (5 \cdot 16)$$

Thus we arrive at the conclusion that in the thermodynamic limit the reduced Hamiltonian  $H_{\text{red}}$  is equivalent to the effective Hamiltonian  $H_{\text{eff}}$  given by (5·1), namely,

$$H_{\text{red}} = H_{\text{eff}} \text{ for } N \rightarrow \infty \text{ and } n = N/V = \text{const.} \quad (5 \cdot 17)$$

It is seen that due to the renormalization of the effective atom-atom interaction, which is of infinite range, combined effect of this and the direct atom-atom interaction of exchange type, which is of finite range, can induce the stationary coherent state under the condition (4·10) or (4·11). It is also of interest to note that the nonvanishing of the expectation value of the spin Hamiltonian which has a

form similar to Eq. (5·1) bears some resemblance to situations in the phase transition of liquid helium discussed previously by Matsubara and Matsuda.<sup>31)</sup>

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### Appendix

—Diagonalization of Eq. (2·21)—

Equation (2·12) can also be diagonalized exactly even when the spatial distribution of atoms in the matter system is taken to be arbitrary. Let  $b_s$  and  $b_s^+$  be new Bose operators defined by a Bogoliubov-Tyablikov-type transformation:<sup>32)</sup>

$$a_{kj} = \sum_s (\xi_{kjs} b_s - \eta_{-kjs}^* b_s^+), \quad a_{kj}^+ = \sum_s (\xi_{kjs}^* b_s^+ - \eta_{-kjs} b_s) \quad (A·1)$$

with

$$\begin{aligned} \sum_s (\xi_{kjs} \xi_{k'j's}^* - \eta_{-k'j's}^* \eta_{-kjs}^*) &= \mathcal{A}(\mathbf{k}, \mathbf{k}') \mathcal{A}(\gamma, \gamma'), \quad \sum_s (\xi_{kjs} \eta_{-k'j's}^* - \xi_{k'j's}^* \eta_{-kjs}^*) = 0, \\ \sum_{kj} (\xi_{kjs} \xi_{kjs}^* - \eta_{-kjs}^* \eta_{-kjs}^*) &= \mathcal{A}(s, s'), \quad \sum_s (\xi_{kjs} \eta_{-kjs} - \xi_{kjs}^* \eta_{-kjs}^*) = 0. \end{aligned} \quad (A·2)$$

Equation (2·12) can then be diagonalized as follows:

$$H_p + H_{ap}^{(2)} = \sum_s \Omega_s b_s^+ b_s \quad \text{with} \quad [b_s, b_s^+] = \mathcal{A}(s, s'), \quad [b_s, b_{s'}] = [b_s^+, b_{s'}^+] = 0. \quad (A·3)$$

Here the new photon eigenfrequencies  $\Omega = \Omega_s$  characterized by the index  $s$  is determined by the eigenvalue equation

$$\begin{aligned} \sum_{k'j'} \{C(\mathbf{k}j, \mathbf{k}'j') \xi_{k'j's} - B(\mathbf{k}j, \mathbf{k}'j') \eta_{-k'j's}\} &= \Omega \xi_{kjs}, \\ \sum_{k'j'} \{B^*(\mathbf{k}j, \mathbf{k}'j') \xi_{k'j's} - C^*(\mathbf{k}j, \mathbf{k}'j') \eta_{-k'j's}\} &= \Omega \eta_{-kjs}. \end{aligned} \quad (A·4)$$

By the use of Eqs. (2·13) and (2·14) Eqs. (A·4) are rewritten as

$$\begin{aligned} \mathbf{e}_{kj} \cdot \sum_{k'j'} W(\mathbf{k}, \mathbf{k}') \mathbf{e}_{k'j'} \{ \rho(\mathbf{k} - \mathbf{k}') \xi_{k'j's} - \rho(\mathbf{k} + \mathbf{k}') \eta_{-k'j's} \} &= \{ \Omega - \omega(\mathbf{k}) \} \xi_{kjs}, \\ \mathbf{e}_{kj} \cdot \sum_{k'j'} W(\mathbf{k}, \mathbf{k}') \mathbf{e}_{k'j'} \{ \rho^*(\mathbf{k} + \mathbf{k}') \xi_{k'j's} - \rho(\mathbf{k} - \mathbf{k}') \eta_{-k'j's} \} &= \{ \Omega + \omega(\mathbf{k}) \} \eta_{-kjs}. \end{aligned} \quad (A·5)$$

As in the case of Eqs. (3·7) and (3·11), the vector potential  $\mathbf{A}(\mathbf{r})$  and the interaction Hamiltonian  $H_{ap}^{(1)}$  etc. can be expressed entirely in terms of the new Bose operators  $b_s$  and  $b_s^+$  as follows:

$$\mathbf{A}(\mathbf{r}) = \sum_s \{ \mathbf{A}_s(\mathbf{r}) b_s + \mathbf{A}_s^*(\mathbf{r}) b_s^+ \}, \quad (A·6)$$

$$H_{ap}^{(1)} = - \sum_i \mathbf{A}_i \cdot \mathbf{J}_i = - \sum_{i\alpha\beta} \sum_s \sigma_{i\alpha\beta} \mathbf{J}_{\alpha\beta} \cdot (\mathbf{A}_{is} b_s + \mathbf{A}_{is}^* b_s^+), \quad (A·7)$$

where

$$\mathbf{A}_s(\mathbf{r}) = \sum_{\mathbf{k}j} [2\pi/\omega(\mathbf{k}) V]^{1/2} \mathbf{e}_{\mathbf{k}j} \exp(i\mathbf{k} \cdot \mathbf{r}) (\xi_{\mathbf{k}js} - \eta_{\mathbf{k}js}). \quad (\text{A} \cdot 8)$$

In this general case total Hamiltonian of the matter-photon system can be expressed in the form

$$H = \sum_s \Omega_s b_s^\dagger b_s + \sum_{i\alpha} \epsilon_\alpha \sigma_{i\alpha\alpha} + (1/2) \sum_{ij} \sum_{\alpha\beta\gamma\delta} L_{\alpha\beta,\gamma\delta}(ij) \sigma_{i\alpha\beta} \sigma_{j\gamma\delta} - \sum_{i\alpha\beta} \sum_s \sigma_{i\alpha\beta} \mathbf{J}_{\alpha\beta} \cdot (\mathbf{A}_{is} b_s + \mathbf{A}_{is}^* b_s^\dagger). \quad (\text{A} \cdot 9)$$

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