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1979 J. Phys. A: Math. Gen. 12 1591

(http://iopscience.iop.org/0305-4470/12/9/025)

J. Phys. A: Math. Gen., Vol. 12, No. 9, 1979. Printed in Great Britain

# On polariton instability and thermodynamic phase transition in a photon–matter system

Motoomi Yamanoi

Department of Electrical Engineering, Meijo University, Tempaku-Ku, Nagoya, Japan

Received 13 December 1978, in final form 30 January 1979

**Abstract.** It is shown that the rotating wave approximation in the minimal coupling Hamiltonian with the $A^2$ term leads to an incorrect energy spectrum of the polariton mode: in particular, to a spurious polariton instability which should not occur for a photon–matter system consisting of radiation and atoms without the Coulomb interaction. Thus the counter-rotating term in the presence of the $A^2$ term plays a crucial role in the instability property of the system. The super-radiant phase transition in the Dicke model is shown to be accompanied by polariton instability, and therefore by instability of the ground state of the photon–matter system. The rotating wave approximation, as well as the neglect of the $A^2$ term, should therefore *not* be made for the study of the thermodynamic phase transition and instability properties of the ground state of the system.

## 1. Introduction

In general, a truncated Hamiltonian gives an approximate energy spectrum and an approximate equation of motion which differs from the original correct one. Therefore its validity is limited to particular usage. In quantum optics, the counter-rotating term, the $A^2$ term and the $P^2$ term are often truncated in the basic Hamiltonian in order to simplify the calculation. If the $A^2$ term is omitted, however, the resulting field equation for the vector potential $\boldsymbol{A}(\boldsymbol{r}, t)$ contains an extra term which makes the state $\boldsymbol{A} = 0$ unstable (Yamanoi 1976). Similarly, if the $P^2$ term in the dipole coupling Hamiltonian is omitted, an extra term $P(t)$ for polarisation is introduced in the equation of motion, making the state $\boldsymbol{P} = 0$ unstable (Yamanoi and Takatsuji 1977). In other words, the consequence of these extra terms is to make the polariton mode with eigenfrequency $\omega$ soft ($\omega \to 0$) and finally unstable ($\omega^2 < 0$) under some threshold condition (polariton instability). However, the above-mentioned instability does not represent the true dynamical property of the photon–matter system, because this is originated entirely in the truncation of the $A^2$ or the $P^2$ term.

Since the polariton mode is a small oscillation of fields $\boldsymbol{A}$ and $\boldsymbol{P}$ about the state $\boldsymbol{A} = \boldsymbol{P} = 0$ (the ground state in which the photon is absent and all atoms are in their lowest states), the polariton instability means the instability of the ground state of the photon–matter system, indicating the appearance of a new ground state ($\boldsymbol{A} \neq 0$, $\boldsymbol{P} \neq 0$) with energy lower than the normal ground state ($\boldsymbol{A} = \boldsymbol{P} = 0$).

Only when we consider a situation where each of these extra terms does not play any crucial role, we can use the approximate Hamiltonian without the $A^2$ or the $P^2$ term to study electromagnetic phenomena which have nothing to do with the instability of the ground state of the photon–matter system.

0305-4470/79/091591+13$01.00  

In the study of laser oscillation and super-radiance, one considers the behaviour of the amplitude of an electromagnetic wave whose frequencies are assumed not to be softened; in other words one is not concerned here with the polariton instability or the instability of the ground state. This is the reason why the truncated Hamiltonian (the Dicke Hamiltonian, for example) has worked well; i.e., a good agreement between theory and experiment in the study of laser oscillation and super-radiance. On the other hand, if we consider phenomena where instability of the ground state ($\omega^2 < 0$) or the softening ($\omega \to 0$) plays a crucial role, e.g. the thermodynamic phase transition, this truncation should not be made. Thus we should note that there exists a limit to the applicability of the truncated Hamiltonian.

The Dicke model Hamiltonian $H_D$, in which both the counter-rotating term (CR term) and the $A^2$ term are truncated, has often been successfully used to study the electromagnetic interaction between radiation and atoms. This has mainly been used to describe the resonant interaction in low atomic concentration (gas), occurring in a state far from thermal equilibrium in which the softening of the photon frequency is not concerned.

Recently, Hepp and Lieb (1973) studied the thermodynamics of the $H_D$ and found a second-order phase transition in it, which they called the super-radiant phase transition (SPT). Then the CR term was taken into account by Carmichael *et al* (1973), who showed that the critical property is not changed qualitatively, but that the CR term makes the SPT easier. Both the CR and the $A^2$ terms were taken into account by Razaewski *et al* (1975). They showed that in this case the SPT is forbidden to occur and concluded that in reality the SPT is impossible in the photon–matter system consisting of radiation and two-level atoms without the Coulomb interaction (system D).

On the contrary, Gilmore and Bowden (1976, 1977a, b) and Orszag (1977) argue that it is possible for the SPT to occur in the system D. In their mathematical formulation use was made of the Hamiltonian without the CR term, while the $A^2$ term is retained. Thus, in the present state of things, two contradictory arguments exist on the problem of the possibility of the SPT in the system D. This discrepancy, however, originates entirely in the problem as to what form of interaction Hamiltonian should be used to describe the electromagnetic interaction: more concretely in the problem of the validity of the RWA, for the study of the thermodynamic phase transition.

The purpose of the present paper is to examine the validity of the RWA in the Hamiltonian with the $A^2$ term for a correct mathematical formulation of the thermodynamic phase transition. In the next section we establish the relation between polariton instability and the SPT by studying the instability property of various model Hamiltonians for which critical properties have been studied by the thermodynamical argument. It will be shown that the SPT is always accompanied by the polariton instability ($\omega^2 < 0$). Accordingly the onset of the polariton instability may be required for the SPT to occur. Thus the problem of the possibility of the SPT in the system D is reduced to that of the polariton instability in the same system. In § 3, the stability of the system D is shown rigorously, through the polariton dispersion relation $\omega^2(k)$ evaluated by taking into account the $A^2$ term, the CR term, and the many levels of the atom, instead of two levels. The reason for considering the many-level atom when taking into account the CR term is as follows. Consider for example the response of an atom (with resonance frequencies $\Omega_1, \Omega_2 \ldots$) to external radiation of frequency $\omega (\sim \Omega_1)$. The most dominant term in the response is the resonant term $1/(\omega - \Omega_1)$. The CR term in the Hamiltonian gives rise to a factor $1/\omega + \Omega_1)$. If this smaller term is taken into account, one should also take into account another term of small magnitude $1/(\omega - \Omega_2)$ arising

from off-resonance of another level, if $|1/(\omega-\Omega_2)| \sim |1/(\omega+\Omega_1)|$. Thus, taking into account the CR term requires many more than two levels. The rigorous result of $\omega^2(k)$ will show that the polariton instability does not occur in the system D, i.e. $\omega^2(k)>0$. In § 4, we study the effect of the RWA on the polariton instability and the dispersion relation. It will be shown that the RWA in the Hamiltonian with the $A^2$ term leads to an incorrect eigenfrequency of the polariton; that is, the RWA leads to a spurious polariton instability $\omega^2(k)<0$, by which the occurrence of the SPT is made possible even in the system D. In conclusion, the truncation of the CR term in the presence of the $A^2$ term leads to incorrect results for the study of thermodynamic phase transitions and the instability properties of the ground state in the photon–matter system.

## 2. Relation between polariton instability and the SPT

In the study of the thermodynamic phase transition in the photon–matter system consisting of single-mode radiation and $N$ two-level atoms without the Coulomb interaction, four types of Hamiltonian have been used which are summarised as follows:

$$H = a^+a + \sum_{j=1}^{N} (\epsilon/2)\sigma_j^z + N^{-1/2} \sum_{j=1}^{N} [\lambda(a^+\sigma_j^- + a\sigma_j^+) + \lambda'(a^+\sigma_j^+ + a\sigma_j^-)] + \kappa(a+a^+)^2 \tag{2.1}$$

where $a$ and $a^+$ are photon annihilation and creation operators respectively; $\sigma_j^z$, $\sigma_j^{\pm} = (\sigma_j^x \pm i\sigma_j^y)/2$ are Pauli operators for the $j$th atom. In equation (2.1) photon energy is normalised to unity, $\epsilon$ is the energy difference between two levels; and the coupling constant $\lambda'$ for the counter-rotating term (CR term) is assumed to be arbitrary. The last term is the $A^2$ term. Throughout this paper we use units such that $\hbar = c = 1$. The Hamiltonian (2.1) includes all of the case mentioned in § 1. The strong coupling condition for each case, derived by thermodynamical considerations, is given by

(i) Hepp and Lieb (1973) ($\lambda' = 0$, $\kappa = 0$):

$$\epsilon < \lambda^2. \tag{2.2a}$$

(ii) Carmichael *et al* (1973) ($\lambda' = \lambda$, $\kappa = 0$):

$$\epsilon < (2\lambda)^2. \tag{2.2b}$$

(iii) Rzazewski *et al* (1975) ($\lambda' = \lambda$, $\kappa \neq 0$):

$$\epsilon(1+4\kappa) < (2\lambda)^2. \tag{2.2c}$$

(iv) Gilmore and Bowden (1976) and Orszag (1977) ($\lambda' = 0$, $\kappa \neq 0$):

$$\epsilon < \lambda^2. \tag{2.2d}$$

In the following we show that these conditions are just the same as those for the polariton instability; (polariton eigenfrequency)$^2 < 0$. To this end, we evaluate the eigenfrequency of the polariton (elementary excitation in the photon–matter system). It can be obtained as the pole of $G(\omega)$ which is the Fourier transform of the double time-retarded photon Green's function defined by Zubarev (1960):

$$G(t) = -i\theta(t)\langle[a(t), a^+]\rangle.$$

The equation of motion for $G(\omega) = \langle\langle a : a^+ \rangle\rangle$ is given by

$$\omega\langle\langle a : a^+ \rangle\rangle = \langle [a, a^+] \rangle + \langle\langle [a, H] : a^+ \rangle\rangle.$$

In the linear approximation and long-wavelength limit, we may use the following approximations:

$$\langle\langle \sigma_j^z a : a^+ \rangle\rangle \sim \langle \sigma^z \rangle_0 \langle\langle a : a^+ \rangle\rangle$$

$$\langle\langle \sigma_j^- : a^+ \rangle\rangle \sim \langle\langle \sigma_k^- : a^+ \rangle\rangle = \langle\langle \sigma^- : a^+ \rangle\rangle, \ (j \neq k)$$

where

$$\langle \sigma^z \rangle_0 = -\tanh(\epsilon/2kT).$$

In these approximations the coupled algebraic equation for the four Green's functions is obtained in the form:

$$\begin{pmatrix} (\omega - 1 - 2\kappa) & -2\kappa & -\lambda N^{1/2} & -\lambda' N^{1/2} \\ 2\kappa & (\omega + 1 + 2\kappa) & \lambda' N^{1/2} & \lambda N^{1/2} \\ \lambda \langle \sigma^z \rangle_0 / N^{1/2} & \lambda' \langle \sigma^z \rangle_0 / N^{1/2} & (\omega - \epsilon) & 0 \\ -\lambda' \langle \sigma^z \rangle_0 / N^{1/2} & -\lambda \langle \sigma^z \rangle_0 / \sqrt{N} & 0 & (\omega + \epsilon) \end{pmatrix} \begin{pmatrix} G \\ H \\ I \\ J \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \tag{2.3}$$

where $G = \langle\langle a : a^+ \rangle\rangle$, $H = \langle\langle a^+ : a^+ \rangle\rangle$, $I = \langle\langle \sigma^- : a^+ \rangle\rangle$, $J = \langle\langle \sigma^+ : a^+ \rangle\rangle$. From equation (2.3) we get $G(\omega)$, whose denominator is the determinant $D(\omega^2)$ of the matrix in equation (2.3). Thus the eigenfrequency is a solution of the secular equation given by $D(\omega^2)^2 = 0$; that is

$$\omega^4 + \omega^2[2(\tilde{\lambda}^2 - \tilde{\lambda}'^2) - \epsilon^2 - 1 - 4\kappa] + (1 + 4\kappa)\epsilon^2 + 2\epsilon(1 + 2\kappa)(\tilde{\lambda}^2 + \tilde{\lambda}'^2) - 8\kappa\epsilon\tilde{\lambda}\tilde{\lambda}' + (\tilde{\lambda}^2 - \tilde{\lambda}'^2)^2 = 0 \tag{2.4}$$

where $\tilde{\lambda} = \lambda \langle \sigma^z \rangle_0^{1/2}$, $\tilde{\lambda}' = \lambda' \langle \sigma^z \rangle_0^{1/2}$.

Let the two solutions of equation (2.4) be $\omega_+^2$ and $\omega_-^2$ corresponding to the upper branch and the lower branch respectively. The instability condition ($\omega^2 \leqslant 0$) is given by $D(\omega^2 = 0) \leqslant 0$; that is

$$(1 + 4\kappa)\epsilon^2 + 2\epsilon(1 + 2\kappa)(\tilde{\lambda}^2 + \tilde{\lambda}'^2) - 8\kappa\epsilon\tilde{\lambda}\tilde{\lambda}' + (\tilde{\lambda}^2 - \tilde{\lambda}'^2)^2 \leqslant 0. \tag{2.5}$$

Let us consider the four cases mentioned before.

(i) ($\lambda' = 0$, $\kappa = 0$). Equation (2.5) reduces to

$$(\epsilon + \tilde{\lambda}^2)^2 \leqslant 0. \tag{2.6a}$$

(ii) ($\lambda' = \lambda$, $\kappa = 0$). Equation (2.5) reduces to $\epsilon + (2\tilde{\lambda})^2 \leqslant 0$, i.e.

$$\epsilon/(2\lambda)^2 \leqslant \tanh(\epsilon/2kT).$$

At the absolute zero ($T = 0$), the condition of $\omega_-^2 < 0$ becomes

$$\epsilon/(2\lambda)^2 < 1 \text{ (strong coupling condition).} \tag{2.6b}$$

At finite temperatures, the critical temperature $T_c$ of softening, $\omega_-^2 = 0$, is determined by the gap equation

$$\epsilon/(2\lambda)^2 = \tanh(\epsilon/2kT_c).$$

(iii) ($\lambda' = \lambda$, $\kappa \neq 0$). Equation (2.5) reduces to

$$(1 + 4\kappa)\epsilon + (2\tilde{\lambda})^2 \leqslant 0.$$

The strong coupling condition is then given by

$$(1+4\kappa)\epsilon < (2\lambda)^2 \tag{2.6c}$$

which cannot be satisfied (Rzazewski *et al* 1975) because, expressing $\tilde{\lambda}^2$ and $\kappa$ in terms of atomic parameters ($d$ = matrix element of dipole moment, $\rho$ = density of the atoms, $e$ and $m$ are respectively charge and mass of electron),

$$\tilde{\lambda}^2 = -2\pi\rho\epsilon^2 d^2 \tanh(\epsilon/2kT), \qquad \kappa = 2\pi\rho e^2/2m$$

and using the inequality derived from the TRK sum rule $e^2/2m > \epsilon d^2$, we have

$$(1+4\kappa)\epsilon + (2\tilde{\lambda})^2 > \epsilon + 8\pi\epsilon^2 d^2 \rho(1-\tanh \epsilon/2kT) > 0.$$

(iv) ($\lambda' = 0$, $\kappa \neq 0$). Equation (2.5) reduces to

$$(\epsilon + \tilde{\lambda}^2)[(1+4\kappa)\epsilon + \tilde{\lambda}^2] \leqslant 0.$$

The second factor on the left-hand side is positive definite:

$$0 < (1+4\kappa)\epsilon + (2\tilde{\lambda})^2 < (1+4\kappa)\epsilon + \tilde{\lambda}^2.$$

However, the first term can become negative: $\epsilon + \tilde{\lambda}^2 \leqslant 0$, i.e.

$$\epsilon/\lambda^2 \leqslant \tanh(\epsilon/2kT)$$

from which the strong coupling condition is obtained as

$$\epsilon < \lambda^2 \tag{2.6d}$$

and the gap equation is

$$\epsilon/\lambda^2 = \tanh \epsilon/2kT_c.$$

These strong coupling conditions for each case are just the same as equations (2.2) except equation (2.6*a*).

Furthermore, the gap equations obtained above are just the same as those obtained by thermodynamic considerations. We notice that the instability condition given by equation (2.6*a*) cannot be satisfied. This means that under the strong coupling condition the eigenfrequency does not become purely imaginary but becomes negative (negative energy polariton). To see this, we begin with the original Hamiltonian (2.1) with $\lambda' = \kappa = 0$:

$$H = a^+ a + (\epsilon/2) \sum_{j=1}^{N} \sigma_j^z + (\lambda/N^{1/2}) \sum_{j=1}^{N} (a^+ \sigma_j^- + a\sigma_j^+). \tag{2.7}$$

Shifting the zero of energy to the lowest energy level, and introducing the cooperative operator defined by

$$b_k^+ = (1/N^{1/2}) \sum_{j=1}^{N} \sigma_j^+ \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_j) \tag{2.8a}$$

$$b_k = (1/N^{1/2}) \sum_{j=1}^{N} \sigma_j^- \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_j) \tag{2.8b}$$

we rewrite equation (2.7) as

$$H = a^+ a + \epsilon \sum_{\boldsymbol{k}} b_{\boldsymbol{k}}^+ b_{\boldsymbol{k}} + \lambda(a^+ b + a b^+) \tag{2.9}$$

where

$$b = b_{k=0}, \qquad b^+ = b^+_{k=0}.$$

Since we are considering low excited states, the operators $b_k$ and $b^+_k$ may be regarded as Bose operators:

$$[b_k, b^+_{k'}] = -(1/N)\sum_{j=1}^{N} \sigma_j^z \exp i(k'-k)\cdot R_j \sim \delta_{k,k'} \qquad \text{(for low excited states).}$$

In equation (2.9) we retain only the mode with $k = 0$ for the matter field, since another mode with $k \neq 0$ does not interact with the long-wavelength single-mode radiation. Then in the long-wavelength limit and in low excited states above the normal ground state, equation (2.9) reduces to

$$H = a^+a + \epsilon b^+b + \lambda(a^+b + ab^+)$$
$$= \omega_- a^+_{in}a_{in} + \omega_+ b^+_{in}b_{in},$$

where

$$\omega_- = \tfrac{1}{2}\{1+\epsilon-[(1-\epsilon)^2+4\lambda^2]^{1/2}\}$$
$$\omega_+ = \tfrac{1}{2}\{1+\epsilon+[(1-\epsilon)^2+4\lambda^2)^{1/2}\}$$
$$a_{in} = a\cos\theta + b\sin\theta$$
$$b_{in} = b\cos\theta - a\sin\theta \qquad (\tan 2\theta = 2\lambda/(1-\epsilon)).$$

Therefore we see that under the condition given by

$$\epsilon < \lambda^2 \tag{2.10}$$

the polariton energy of the lower branch becomes negative ($\omega_+ < 0$), showing the instability of the ground state. Then equation (2.10) is the strong coupling condition for case 1.

From these examples we may conclude that the SPT is accompanied by the polariton instability, and that whereas the nonlinearity is required to determine the structure in the ordered phase, the criterion of the possibility of the phase transition is given by the linear theory, for which a rigorous answer can be obtained.

## 3. Stability of a system consisting of many-level atoms and a multimode radiation field

We saw in the preceding section that the SPT is accompanied by a polariton instability in the photon–matter system. This relation may be interpreted as follows. As mentioned in § 1, the instability of a polariton means the instability of the ground state, which indicates the appearance of a new ground state ($A \neq 0$, $P \neq 0$) with a lower energy than the normal ground state ($A = 0$, $P = 0$), hence allowing the SPT to occur. Thus the problem of the possibility of the SPT is reduced to the problem of the polariton instability, which can be solved rigorously using the relation between wave-vector $k$ and eigenfrequency $\omega$ given by $k^2 = \epsilon(\omega)\omega^2$ (e.g. see Landau and Lifshitz 1963).

In this section, we study the linear instability property of the system D consisting of radiation and atoms without any direct interaction. We begin by treating the atom as a two-level system. In this case, $\omega^2(k)$ is given by

$$k^2 = \epsilon(\omega)\omega^2 = [1 + 4\pi 2\epsilon\rho d_{12}^2/(\epsilon^2 - \omega^2)]\omega^2 \tag{3.1}$$

where $\rho$ is the number density of atoms and $d_{12}$ is a dipole matrix element. The schematic curve of $\omega^2(k^2)$ obtained from equation (3.1) is shown by curve C in figure 2, which shows $\omega^2(k^2) > 0$ for all modes; that is, all polaritons are stable. This is consistent with the result obtained by Rzazewski *et al* that the SPT does not occur in the system D.

Equation (3.1) with $e^2/m$ instead of $2\epsilon d_{12}^2$ can be derived by Lorentz's Theory (Lorentz 1909). A quantum mechanical treatment has been given by Hopfield (1958), using the second quantised Hamiltonian. In his formulation, both the CR and the $A^2$ terms are included in the Hamiltonian; only for these can equation (3.1) be derived. However, making the two-level approximation and at the same time taking into account the CR term are not consistent procedures. The reason is, as mentioned in § 1, the inclusion of the CR term requires one more level than the two under consideration. To remove this shortcoming we should take a many-level atom when the CR term is taken into account. Then the number of interaction terms between atoms and radiation is increased. It may be plausible to expect that many more interaction terms make the SPT easier to occur. Then the question arises: Can we expect onset of the instability if many levels are taken into account although it is impossible in the two-level atom? This question will be answered by considering many modes and many levels at the same time.

In addition there exists a reason to consider many modes of radiation. In this connection we first note that the approximation of the two levels for the atom and the single mode for radiation is intended to describe the situation in which strong single-mode radiation is present, which interacts resonantly with the atom. Only for such a situation is the two-level atom and single-mode photon a meaningful approximation. In considering the onset of the thermodynamic phase transition, however, we should start from the normal thermodynamic branch, in which a strong particular mode has not yet grown up, but in which many modes are excited with small oscillations about the mean zero value. Thus we should consider many modes of radiation and a many-level atom.

Guided by these considerations we next study the linear instability property of the system rigorously by taking into account many modes, many levels, the CR and the $A^2$ terms. This can be effected easily by extension of equation (3.1) to the case of many-level atoms. With a replacement of the dielectric function in equation (3.1) by the dielectric function of many-level atoms (e.g. Loudon 1972), the required dispersion relation is obtained in the form

$$k^2 = \left[1 + 4\pi \sum_i 2\Omega_i \rho d_{i1}^2/(\Omega_i^2 - \omega^2)\right]\omega^2. \tag{3.2}$$

The schematic curve of $\omega^2(k^2)$ obtained from equation (3.2) is depicted in figure 1, which shows that even the lowest branch is stable, and that the stability obtained in the two-level approximation is not changed although the number of energy levels has increased. Therefore, we may use the two-level approximation for the atom to study the instability property of the photon–matter system without introducing an error by this approximation.

## 4. Influence of the RWA on the polariton eigenfrequencies and the instability property

The RWA has been used often with the approximation of two-level atoms and single-mode radiation. A physical argument to justify it may be as follows. As mentioned in § 1, inclusion of the CR term requires more energy levels than two. Conversely, if one restricts oneself within the two-level approximation, (i.e. taking into account only the

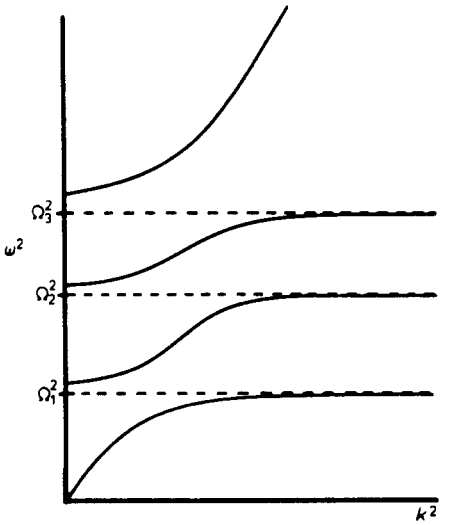


**Figure 1.** Polariton dispersion curve for a case in which each atom has three resonance frequencies $\Omega_1$, $\Omega_2$ and $\Omega_3$. If the direct interaction between the atoms is neglected, the resonance frequencies remain constant as the atomic density is increased.

most dominant resonant term), one should neglect smaller effects due to the CR term. However, the situation exists in which the CR term contributes the same magnitude as the resonant term. An example is a low-frequency limit of response of the atoms to the radiation, because in this condition we have $1/(\Omega_1+\omega) = 1/(\Omega_1-\omega)$ as $\omega \to 0$. Since the SPT is also the phenomena in the static limit, we see that the CR term should not be regarded as a small effect in the formulation of the SPT. To see this, we study the contribution of the CR term in the Hamiltonian with the $A^2$ term to the instability property of the photon–matter system D, whose rigorous stability has been studied in § 3.

The Hamiltonian is given, if the CR term is neglected, by

$$H = \sum_k k a_k^+ a_k + \sum_{j=1}^N (\epsilon/2)\sigma_j^z + (N)^{-1/2} \sum_k \sum_{j=1}^N \lambda_k [\sigma_j^+ a_k \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j) - \sigma_j^- a_k^+ \exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j)]$$

$$+ \sum_k \kappa_k [a_k a_{-k} + a_k a_k^+ + a_k^+ a_k + a_k^+ a_{-k}^+] \tag{4.1}$$

where

$$\lambda_k = -\mathrm{i}\epsilon(2\pi\rho/k)^{1/2}(\boldsymbol{d}\cdot\boldsymbol{e}) \tag{4.2a}$$

$$\kappa_k = (e^2/2m)2\pi\rho/k. \tag{4.2b}$$

Changing the zero of energy to the lowest energy level, and introducing the collective operators $b_k^+$ and $b_k$ defined by equations (2.8), we rewrite equation (4.1) in the following form:

$$H = \sum_k k a_k^+ a_k + \epsilon \sum_k b_k^+ b_k + \sum_k \lambda_k [b_k^+ a_k - b_k a_k^+] + \sum_k \kappa_k [a_k a_{-k} + a_k a_k^+ + a_k^+ a_k + a_k^+ a_{-k}^+]. \tag{4.3}$$

To get the polariton dispersion relation $\omega^2(k)$, we linearise equation (4.3) by regarding

$b_k^+$ and $b_k$ as Bose operators, as done in § 2. In this linear approximation, the ensemble of two-level atoms is equivalent to a harmonic polarisation field. With the help of the relation (e.g. Milonni 1976) $e^2/m = 2\epsilon d_{12}^2$, we rewrite $\lambda_k$ and $\kappa_k$ in equations (4.2) as follows:

$$\lambda_k = -i\epsilon(\pi\epsilon\beta/k)^{1/2} \tag{4.4a}$$

$$\kappa_k = \pi\epsilon^2\beta/k \tag{4.4b}$$

where $\beta = 2\rho d_{12}^2/\epsilon$.

The Heisenberg equations of motion in the RWA are obtained in the form

$$i\frac{d}{dt}\begin{pmatrix} a_k \\ b_k \\ a_{-k}^+ \\ b_{-k}^+ \end{pmatrix} = \begin{pmatrix} k+2\kappa_k, & -\lambda_k & 2\kappa_k & 0 \\ \lambda_k & \epsilon & 0 & 0 \\ -2\kappa_k & 0 & -k-2\kappa_k & -\lambda_k \\ 0 & 0 & \lambda_k & -\epsilon \end{pmatrix}\begin{pmatrix} a_k \\ b_k \\ a_{-k}^+ \\ b_{-k}^+ \end{pmatrix} \tag{4.5}$$

where $\lambda_k$ and $\kappa_k$ are given by equations (4.4).

Assuming the time dependence of $\exp(-i\omega t)$ for all the operators, we obtain the following secular equation in case (iv) with the $A^2$ term but without the CR term:

$$\omega^4 - \omega^2(2\pi\beta\epsilon^3/k + k^2 + 4\pi\beta\epsilon^2 + \epsilon^2) + \epsilon^2k^2 + 2\pi\beta\epsilon^4 - 3\pi^2\beta^2\epsilon^6/k^2 = 0.$$

Note the appearance of a term which is singular at $k = 0$. This singular term, however, does not appear in the correct equation for case (iii) with both the CR and the $A^2$ terms, which is given by (Hopfield 1958):

$$\omega^4 - \omega^2(k^2 + 4\pi\beta\epsilon^2 + \epsilon^2) + k^2\epsilon^2 = 0.$$

Corresponding to case (ii) without the $A^2$ term but with the CR term, we have the secular equation in the form

$$\omega^4 - \omega^2(k^2 + \epsilon^2) + \epsilon^2k^2 - 4\pi\beta\epsilon^4 = 0$$

and for case (i) with neither the CR nor the $A^2$ terms, we have

$$\omega^2 - \omega(k+\epsilon) + k\epsilon - \pi\beta\epsilon^3/k = 0.$$

For each case, the dispersion curve, $\omega_-^2(k^2)$, of the lower branch is depicted schematically in figure 2. From the secular equations, the instability condition $\omega_-^2 \leqslant 0$ for each case is obtained:

(i):

$$k\epsilon \leqslant \pi\beta\epsilon^3/k = |\lambda_k|^2$$

(ii):

$$\epsilon^2k^2 - 4\pi\beta\epsilon^4 \leqslant 0, \qquad \text{i.e. } k\epsilon \leqslant |2\lambda_k|^2$$

(iii):

no instability

(iv):

$$\epsilon^2k^2 + 2\pi\beta\epsilon^4 - 3\pi^2\beta^2\epsilon^6/k^2 \leqslant 0, \qquad \text{i.e. } (\epsilon k - |\lambda_k|^2)(\epsilon k + 3|\lambda_k|^2) \leqslant 0.$$

If one mode is chosen and its photon energy is set at unity ($k = 1$), these conditions are just the same as that obtained in § 2. Note that all these instabilities are only spurious,

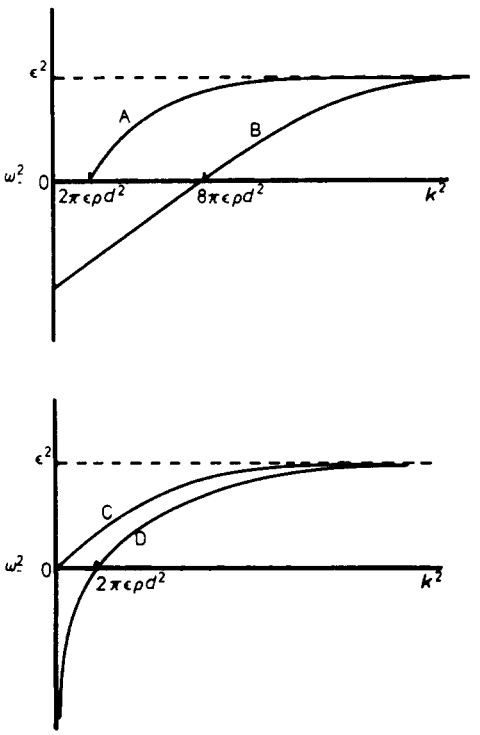


**Figure 2.** Polariton dispersion curve of the lower branch derived from Hamiltonian: A, without both the CR and the $A^2$ terms; B, without the $A^2$ term but with the CR term; C, with both the CR and the $A^2$ terms; D, with the $A^2$ term but without the CR term. The matter system with which radiation interacts is assumed to be an ensemble of two-level atoms without any direct mutual interaction. $\epsilon$ is the energy difference between the two levels; $d$ is the dipole matrix element, and $\rho$ is the number density of the atoms.

originating entirely in the neglect of the CR or the $A^2$ term. In particular we observe that, in the presence of the $A^2$ term, the CR term changes the instability property drastically: (compare curves C and D in figure 2).

Therefore we conclude that the truncation of the CR term or the $A^2$ term leads to an incorrect result of the polariton instability and therefore these truncations should not be done in the study of the thermodynamic phase transition.

## 5. Discussion (thermodynamics of minimal coupling Hamiltonian)

We have seen that if the full Hamiltonian in the minimal coupling theory is used, the soft-mode instability does not occur in the photon–matter system D within the electric dipole approximation, even if many-level atoms and multi-mode radiation are taken into account. As is well known, the dynamical property of the equilibrium phase transition is characterised by the appearance of the soft-mode instability. Thus non-existence of the soft-mode instability indicates that the SPT cannot possibly occur in the system D due only to the electromagnetic interaction within the electric dipole approximation. The same conclusion has been obtained from thermodynamic consideration by Bialynicki-Birula and Rzazewski (1978). In their argument, neither the

two-level nor the single-mode approximations are required, but the full minimal coupling Hamiltonian is used. Therefore their conclusion is a general consequence of the minimal coupling in the thermodynamic limit within the electric dipole approximation. In this section we study the thermodynamics of the minimal coupling and the nature of the thermodynamic phase transition in the photon–matter system D with the help of their argument.

Let us consider the distribution function of photon amplitude

$$P(\alpha) = \langle \delta(a - \alpha) \rangle = Z^{-1} \operatorname{Tr} \exp(-\beta H) \delta(a - \alpha) \tag{5.1}$$

where $a$ is the photon annihilation operator and $\beta = 1/kT$. The trace over the radiation field is effected by using the coherent state:

$$\begin{aligned} P(\alpha) &= Z^{-1} \operatorname{Tr}_M \int \langle \alpha' | \exp(-\beta H) \delta(a - \alpha) | \alpha' \rangle d^2\alpha' \\ &= Z^{-1} \operatorname{Tr}_M \langle \alpha | \exp(-\beta H) | \alpha \rangle \end{aligned} \tag{5.2}$$

where $\operatorname{Tr}_M$ means the trace over the variables of the matter system. Equation (5.2) can be rewritten in the form:

$$P(\alpha) = Z^{-1} \exp(-\beta F(\alpha)) \tag{5.3}$$

where $F(\alpha)$ is the constrained free energy defined by

$$\begin{aligned} \exp(-\beta F(\alpha)) &= \operatorname{Tr}\ (\alpha\text{: fixed})\ \operatorname{Tr}_M \exp(-\beta H) = \operatorname{Tr} \exp(-\beta H) \delta(a - \alpha) \\ &= \operatorname{Tr}_M \langle \alpha | \exp(-\beta H) | \alpha \rangle \end{aligned} \tag{5.4}$$

If $P(\alpha)$ has sharp maximum at $\alpha_0 \neq 0$, it indicates the appearance of an order parameter. In laser oscillation the order parameter varies in time as $\dot{\alpha}_0 \neq 0$, while in the state of thermal equilibrium an order parameter, if it can appear, is static: $\dot{\alpha}_0 = 0$.

Let us now consider the constrained free energy $F(A)$ defined by

$$\exp(-\beta F(A)) = \operatorname{Tr} \exp(-\beta H) \delta(\hat{A} - A) \tag{5.5}$$

where $\hat{A}$ is the operator of the vector potential. To evaluate the trace in equation (5.5) we use the approximation of Wang and Hioe (1973), according to which the photon operators $a$ and $a^+$ in the Boltzmann factor in equation (5.4) can be replaced by the $c$ numbers $\alpha$ and $\alpha^*$ in the thermodynamic limit.

Using this idea we may replace the operator $\hat{A}$ by the fixed $c$ number $A$ in the Boltzmann factor in equation (5.5). Thus we have

$$\exp(-\beta F(A)) = \operatorname{Tr}_M \exp(-\beta H(A)) \tag{5.6}$$

with

$$H(A) = \sum_{j=1}^{N} \{[\boldsymbol{p}_j - (e/c)\boldsymbol{A}(\boldsymbol{R}_j)]^2/2m + V(\boldsymbol{r}_j)\} + H_F \tag{5.7}$$

where the field energy $H_F$ and the vector potential $A$ are fixed $c$ numbers and $V(\boldsymbol{r}_j)$ is the potential energy of one electron in the $j$th atom. In the electric dipole approximation, the spatial dependence of the vector potential is neglected within each atom; i.e. the atom at site $\boldsymbol{R}_j$ is assumed to be driven only by an electric field

$$\boldsymbol{E}(\boldsymbol{R}_j) = -\frac{1}{c} \frac{\partial}{\partial t} \boldsymbol{A}(\boldsymbol{R}_j).$$

Then in equation (5.7) we have evaluated the vector potential, not at the electron position $\boldsymbol{r}_j$ but at the nuclear position $\boldsymbol{R}_j$.

To evaluate the trace over the atomic variables in equation (5.6), we use the fact that the trace is invariant under a similarity transformation:

$$\mathrm{Tr}_{\mathrm{M}} \exp(-\beta H(A)) = \mathrm{Tr}_{\mathrm{M}} \exp(-\mathrm{i}G) \exp(-\beta H(A)) \exp(\mathrm{i}G) \tag{5.8}$$

where

$$G = \sum_{j=1}^{N} (e/\hbar c)\boldsymbol{A}(\boldsymbol{R}_j) \cdot \boldsymbol{r}_j. \tag{5.9}$$

The generating function $G$ given by equation (5.9) has often been used to transform from the minimal coupling Hamiltonian to the dipole coupling one.

In general this transformation effects the shift of momenta, both of charged particles and of the radiation field, if both the atomic system and the radiation field are considered as constituting a single closed system. However, since the value of $A$ in (5.6) has been fixed, the effect of the transformation is to shift the momenta of electrons only.

Effecting the transformation

$$\exp(-\mathrm{i}G)H(A) \exp(\mathrm{i}G) = \sum_{j=1}^{N} [\boldsymbol{p}_j^2/2m + V(\boldsymbol{r}_j)] = H_0 \tag{5.10}$$

we see that the trace in equation (5.6) is independent of variables of the radiation field:

$$\mathrm{Tr}_{\mathrm{M}} \exp(-\beta H(A)) = \mathrm{Tr}_{\mathrm{M}} \exp(-\beta H_0) = \exp(-\beta F_0) \tag{5.11}$$

where $F_0$ is the free energy of the atomic system alone. This means that in the electric dipole approximation and in the thermodynamic limit the radiative interaction between atoms vanishes and then the SPT cannot occur. The relation (5.11) is a part of the no-go theorem concerning the super-radiant phase transition in atomic systems due to I Bialynicki-Birula and K Rzazewski, and this is consistent with our present result obtained from the dynamical viewpoint.

## Acknowledgments

The author expresses his sincere thanks to Professor T Usui for valuable discussion and assistance in preparing the manuscript.

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