

### Coherence domains in matter interacting with radiation

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#### Abstract

The coupling of the electromagnetic field to matter polarization (dipole interaction) is examined in order to assess the possibility of setting up a coherent state as envisaged by G. Preparata (*QED Coherence in Matter*, World Sci (1995)). It is found that coherence domains may set up in matter, their phases being arranged in a periodic lattice, as a consequence of, basically, a two-level interaction, which leads to a long-range ordered state, governed by a macroscopic occupation of both the photon state and the two levels. The non-linear equations of motion are solved for the new, non-perturbational ground-state, which is energetically favourable, providing the coupling strength exceeds a critical value. The elementary excitations with respect to this ground-state are derived, their energy being non-trivially affected by interaction. The "thermodynamics" of the coherent phase is computed and the super-radiant phase transition is re-derived in this context. Except for the general suggestion of coherence, the present results differ appreciably from Preparata's, *loc cit.*

**Introduction.** We investigate herein the possibility of setting up coherence domains in matter interacting with electromagnetic radiation. This idea was originally suggested by Preparata,[1] who presented several speculations about possible consequences of such a state on various physical phenomena. The idea was also related to the lasing mechanism and the super-radiance phenomenon.[2]-[9]

We show herein that the coupling between electromagnetic radiation and matter polarization (dipole interaction) may lead to coherence domains, whose phases are arranged in a periodic lattice, involving a two-level state of matter, providing the coupling exceeds a certain critical value. The coherent state is characterized by a macroscopic occupation of both the photon state and the two levels. The ground-state and the elementary excitations are derived for such coherent domains. The solution has a non-perturbational character. The energy of the ground-state is negative, as for a bound state, involving a formation enthalpy for the coherence domain. The elementary excitations are affected in a non-trivial way by interaction, thus providing a most direct way of probing the existence of such a coherent state in matter. The "thermodynamics" of the coherent phase is computed and the super-radiant phase transition is re-derived in this context.

**Radiation field.** As it is well-known, the electromagnetic field is described by the vector potential

$$\mathbf{A}(\mathbf{r}) = \sum_{\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar c^2}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})a_{\alpha\mathbf{k}}e^{i\mathbf{k}\mathbf{r}} + \mathbf{e}_\alpha^*(\mathbf{k})a_{\alpha\mathbf{k}}^*e^{-i\mathbf{k}\mathbf{r}}] \quad (1)$$

in the standard Fourier representation, with the transverse gauge  $div\mathbf{A} = 0$ , where  $\hbar$  is Planck's constant,  $c$  is the velocity of light,  $V$  is the volume,  $\omega_k = ck$  is the frequency and  $\mathbf{e}_\alpha(\mathbf{k})$  are the polarization vectors,  $\mathbf{e}_\alpha(\mathbf{k})\mathbf{k} = 0$ ,  $\mathbf{e}_\alpha(\mathbf{k})\mathbf{e}_\beta^*(\mathbf{k}) = \delta_{\alpha\beta}$  ( $\alpha, \beta = \pm 1$ ),  $\mathbf{e}_{-\alpha}(-\mathbf{k}) = \mathbf{e}_\alpha^*(\mathbf{k})$ . The charge density and the scalar potential are set equal to zero. The electric and magnetic field are given by  $\mathbf{E} = -(1/c)\partial\mathbf{A}/\partial t$  and, respectively,  $\mathbf{H} = curl\mathbf{A}$ , and three Maxwell's equations are satisfied:  $curl\mathbf{E} = -\frac{1}{c}\partial\mathbf{H}/\partial t$ ,  $div\mathbf{H} = 0$ ,  $div\mathbf{E} = 0$ . The classical lagrangian of the radiation field reads

$$\begin{aligned} L_f &= \frac{1}{8\pi} \int d\mathbf{r} (E^2 - H^2) = \\ &= \sum_{\alpha\mathbf{k}} \frac{\hbar}{4\omega_k} (\dot{a}_{\alpha\mathbf{k}}\dot{a}_{-\alpha-\mathbf{k}} + \dot{a}_{\alpha\mathbf{k}}^*\dot{a}_{-\alpha-\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}}\dot{a}_{\alpha\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}}^*\dot{a}_{\alpha\mathbf{k}}) - \\ &- \sum_{\alpha\mathbf{k}} \frac{\hbar\omega_k}{4} (a_{\alpha\mathbf{k}}a_{-\alpha-\mathbf{k}} + a_{\alpha\mathbf{k}}^*a_{-\alpha-\mathbf{k}}^* + a_{\alpha\mathbf{k}}a_{\alpha\mathbf{k}}^* + a_{\alpha\mathbf{k}}^*a_{\alpha\mathbf{k}}) . \end{aligned} \quad (2)$$

The interaction lagrangian is given by

$$L_{int} = \frac{1}{c} \int d\mathbf{r} \cdot \mathbf{j}\mathbf{A} = \sum_{\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{j}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{j}(\mathbf{k})a_{\alpha\mathbf{k}}^*] , \quad (3)$$

where  $\mathbf{j}(\mathbf{k})$  is the Fourier transform of the current density,

$$\mathbf{j}(\mathbf{r}) = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k})e^{i\mathbf{k}\mathbf{r}} , \quad (4)$$

with  $div\mathbf{j} = 0$  (continuity equation). The Euler-Lagrange equations for the lagrangian  $L_f + L_{int}$  lead to the wave equation with sources

$$\ddot{a}_{\alpha\mathbf{k}} + \ddot{a}_{-\alpha-\mathbf{k}}^* + \omega_k^2 (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) = \sqrt{\frac{8\pi\omega_k}{\hbar}} \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{j}(\mathbf{k}) , \quad (5)$$

which is the fourth Maxwell's equation  $curl\mathbf{H} = (1/c)\partial\mathbf{E}/\partial t + 4\pi\mathbf{j}/c$ .

As it is well-known, the classical hamiltonian formalism can be established according to the usual rules, leading to the hamiltonian  $H = H_f + H_{int}$ , where  $H_f = (1/8\pi) \int d\mathbf{r} (E^2 + H^2)$  and  $H_{int} = -L_{int}$ . Similarly, the quantization scheme proceeds as usually, with  $a_{\alpha\mathbf{k}}$ ,  $a_{\alpha\mathbf{k}}^*$  annihilation and creation operators,  $[a_{\alpha\mathbf{k}}, a_{\beta\mathbf{k}'}^*] = \delta_{\alpha\beta}\delta_{\mathbf{k}\mathbf{k}'}$ ,  $[a_{\alpha\mathbf{k}}, a_{\beta\mathbf{k}'}] = 0$ , and  $a_{\alpha\mathbf{k}} \sim e^{-i\omega_k t}$ .

**Interacting matter and radiation.** We consider a set of  $N$  independent, non-relativistic, identical particles (atoms or molecules) labelled by  $i = 1, \dots, N$ , and write the hamiltonian corresponding to their internal degrees of freedom as

$$H_s = \sum_i H_s(i) . \quad (6)$$

We make no assumption about their positions, nor about their centre-of-mass motion. We introduce a set of orthonormal eigenfunctions  $\varphi_n(i)$ , such as

$$H_s(i)\varphi_n(j) = \varepsilon_n\delta_{ij} , \quad \int d\mathbf{r}\varphi_n^*(i)\varphi_m(j) = \delta_{ij}\delta_{nm} , \quad (7)$$

where  $\varepsilon_n$  is the energy level of the  $n$ -state. We construct also a set of normalized eigenfunctions

$$\psi_n = \sum_i c_{ni}\varphi_n(i) \quad (8)$$

for the whole ensemble,

$$H_s \psi_n = \varepsilon_n \psi_n , \quad (9)$$

where the coefficients  $c_{ni}$  are such as to satisfy the normalization condition,

$$\sum_i |c_{ni}|^2 = 1 . \quad (10)$$

Since the particles are identical the coefficients  $c_{ni}$  are of the form  $c_{ni} = e^{i\theta_{ni}}/\sqrt{N}$ , where  $\theta_{ni}$  are some undetermined phases, so we may write the wavefunctions as

$$\psi_n = \frac{1}{\sqrt{N}} \sum_i e^{i\theta_{ni}} \varphi_n(i) . \quad (11)$$

We notice that any  $n$ -state with wavefunction  $\psi_n$  can be occupied by any number of particles, up to  $N$ . Therefore, we introduce the field operator

$$\Psi = \sum_n b_n \psi_n \quad (12)$$

and assume boson-like commutation relations for the operators  $b_n$ ,  $[b_n, b_m^*] = \delta_{nm}$ ,  $[b_n, b_m] = 0$ , for large, macroscopic values of the number of particles

$$N = \sum_n b_n^* b_n . \quad (13)$$

The lagrangian of this ensemble of particles can be represented as

$$L_s = \frac{1}{2} \int d\mathbf{r} (\Psi^* \cdot i\hbar \partial \Psi / \partial t - i\hbar \partial \Psi^* / \partial t \cdot \Psi) - \int d\mathbf{r} \Psi^* H_s \Psi , \quad (14)$$

or

$$L_s = \frac{1}{2} \sum_n i\hbar [b_n^* \dot{b}_n - \dot{b}_n^* b_n] - \sum_n \varepsilon_n b_n^* b_n , \quad (15)$$

and the hamiltonian is given by

$$H_s = \sum_n \varepsilon_n b_n^* b_n . \quad (16)$$

The corresponding equation of motion  $i\hbar \dot{b}_n = \varepsilon_n b_n$  is Schrodinger's equation. It is worth noting that the same equation is obtained for  $b_n$  viewed as classical variables.

The current density associated with this ensemble of particles can be written as

$$\mathbf{j}(\mathbf{r}) = \sum_i \mathbf{J}(i) \delta(\mathbf{r} - \mathbf{r}_i) = \frac{1}{V} \sum_{i\mathbf{k}} \mathbf{J}(i) e^{-i\mathbf{k}\mathbf{r}_i} e^{i\mathbf{k}\mathbf{r}} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} , \quad (17)$$

where  $\mathbf{r}_i$  is the position of the  $i$ -th particle and  $\mathbf{J}(i)$  is the current density of this particle. Now, making use of equations (11) and (12), it is easy to see that the interaction lagrangian given by equation (3) can be written as

$$L_{int} = \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_{\mathbf{k}}}} [\mathbf{e}_{\alpha}(\mathbf{k}) \mathbf{I}_{mn}^*(\mathbf{k}) a_{\alpha\mathbf{k}} + \mathbf{e}_{\alpha}^*(\mathbf{k}) \mathbf{I}_{nm}(\mathbf{k}) a_{\alpha\mathbf{k}}^*] b_n^* b_m , \quad (18)$$

where

$$\mathbf{I}_{nm}(\mathbf{k}) = \frac{1}{N} \sum_i \mathbf{J}_{nm}(i) e^{-i(\theta_{ni} - \theta_{mi})} e^{-i\mathbf{k}\mathbf{r}_i} \quad (19)$$

and  $\mathbf{J}_{nm}(i)$  are the matrix elements of the  $i$ -th particle current density. These vectors have the same magnitude, since the particles are identical, but their directions depend in general on particle. The equation of motion (Schrodinger's equation) is given now by

$$i\hbar\dot{b}_n = \varepsilon_n b_n - \sum_{m\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^*] b_m, \quad (20)$$

while the wave equation (5) (Maxwell's equation) for the electromagnetic field becomes

$$\ddot{a}_{\alpha\mathbf{k}} + \ddot{a}_{-\alpha-\mathbf{k}}^* + \omega_k^2 (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) = \sum_{nm} \sqrt{\frac{8\pi\omega_k}{V\hbar}} \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})b_n^* b_m. \quad (21)$$

It is convenient to write down also the interaction hamiltonian  $H_{int} = -L_{int}$  in the interaction representation  $b_n \rightarrow b_n e^{-i\varepsilon_n t/\hbar}$ ,  $a_{\alpha\mathbf{k}} \rightarrow a_{\alpha\mathbf{k}} e^{-i\omega_k t}$ ; it reads

$$\begin{aligned} H_{int} = & - \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} e^{\frac{i}{\hbar}(\varepsilon_n - \varepsilon_m - \hbar\omega_k)} + \\ & + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^* e^{\frac{i}{\hbar}(\varepsilon_n - \varepsilon_m + \hbar\omega_k)}] b_n^* b_m. \end{aligned} \quad (22)$$

One can easily recognize in equation (22) the excitation and dis-excitation processes with absorption or emission of photons, and the well-known form of the interaction hamiltonian suitable for perturbation calculations.

**Coherence. Coherence domains.** Making use of equation (19) the interaction lagrangian given by equation (18) can be written as

$$L_{int} = \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} F_{nm}(\alpha\mathbf{k}) (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) b_n^* b_m, \quad (23)$$

where

$$F_{nm}(\alpha\mathbf{k}) = \frac{1}{N} \sum_i \mathbf{e}_\alpha(\mathbf{k})\mathbf{J}_{nm}(i) e^{i\mathbf{k}\mathbf{r}_i - i(\theta_{ni} - \theta_{mi})}. \quad (24)$$

For any pair  $(nm)$  of energy levels, we represent the position  $\mathbf{r}_i$  of any particle  $i$  as  $\mathbf{r}_i = \mathbf{R}_p + \mathbf{r}_{pi}$ , where the vectors  $\mathbf{R}_p$  define a spatial lattice characterized by the set of integers  $p = (p_1, p_2, p_3)$  and  $\mathbf{r}_{pi}$  are restricted to the first Wigner-Seitz cell of such a lattice. The lattice  $\mathbf{R}_p$  is chosen such that the magnitudes of its shortest reciprocal vectors  $\mathbf{k}_r$ ,  $r = 1, 2, 3$ , are equal with the magnitude of the relevant wavevectors  $\mathbf{k}$ , *i.e.* those wavevectors which satisfy  $\hbar\omega_k = \varepsilon_n - \varepsilon_m > 0$ . It is easy to see that only a cubic and a trigonal (rhombohedral) symmetry is thus allowed. For instance, a cubic lattice is characterized in this case by a periodicity length  $\lambda = 2\pi/k$ , where  $k$  is the magnitude of the relevant wavevector. A similar periodicity length (different from  $\lambda$ ) occurs for the rhombohedral lattice. We limit the relevant wavevectors  $\mathbf{k}$  to this finite set of basic reciprocal vectors, for which  $\mathbf{k}_r \mathbf{R}_p = 2\pi \times integer$ . Equation (24) becomes then

$$F_{nm}(\alpha\mathbf{k}_r) = \frac{1}{N} \sum_{pi} \mathbf{e}_\alpha(\mathbf{k}_r)\mathbf{J}_{nm}(i) e^{i\mathbf{k}_r \mathbf{r}_{pi} - i(\theta_{ni} - \theta_{mi})}, \quad (25)$$

where the summation over  $p$  stands for all the elementary cells in the spatial lattice .

The summation of the phase factors in equation (25), over label  $i$  in the Wigner-Seitz cell, is in general vanishing, in view of the randomness of such phase factors. We note that there is both a spatial phase  $\mathbf{k}_r \mathbf{r}_{pi}$  in equation (25) and an internal phase  $\theta_{ni} - \theta_{mi}$ , leaving aside the various orientations of the current density  $\mathbf{J}_{nm}(i)$  with respect to the polarization vector  $\mathbf{e}_\alpha(\mathbf{k})$ . However, we can define a subset of  $N_{nm}(\alpha\mathbf{k}_r)$  particles such that their phases  $\theta_{ni}$  fulfill the condition

$$\mathbf{k}_r \mathbf{r}_{pi} - (\theta_{ni} - \theta_{mi}) = K \ , \quad (26)$$

where  $K$  is a constant. We can see that these sub-sets of particles are disjoint, *i.e.* if a particle satisfies condition (26) for a given  $\mathbf{k}_r$  it does not satisfy it for a different  $\mathbf{k}_r$ . In addition, any particle belongs to a well-determined pair ( $nm$ ). It is also reasonable to assume that all the particles  $N_{nm}(\alpha\mathbf{k}_r)$  have their current density  $\mathbf{J}_{nm}(i)$  aligned with the polarization vector  $\mathbf{e}_\alpha(\mathbf{k}_r)$ , *i.e.*  $\mathbf{e}_\alpha(\mathbf{k}_r)\mathbf{J}_{nm}(i) = J_{nm}$ . Under these circumstances, up to a phase factor  $\exp(iK)$ , equation (25) gives  $F_{nm}(\alpha\mathbf{k}_r) = J_{nm}N_{nm}(\alpha\mathbf{k}_r)/N$ . It is reasonable to assume in addition the completeness of the partition operated by condition (26), *i.e.*  $\sum_{(nm)\alpha\mathbf{k}_r} N_{nm}(\alpha\mathbf{k}_r) = N$ .

Condition (26) tells that the phases of the internal motion of the  $i$ -th particle is correlated to the position of that particle. It implies a long-range order in a cooperative phenomenon, where the phase of the internal motion "feels" the particle position. Equation (26) may be taken as the basic condition for coherence. We call such an ensemble of particles which satisfies condition (26) a lattice of coherence domains. Since, typically, the wavelength  $\lambda_r = 2\pi/k_r \gg a$ , where  $a$  is the mean inter-particle distance, we can see that for particles located near the centre of the Wigner-Seitz cell we may take  $\theta_{ni} - \theta_{mi} \simeq 0$  and  $K = 0$ , while for particles located near the boundaries of the Wigner-Seitz cell the phases are such as  $\theta_{ni} - \theta_{mi}$  get non-vanishing values, such as to preserve the constant value  $K = 0$ .

It is easy to see that for various pairs ( $nm$ ) we have a superposition of such lattices of coherence domains. Similarly, these lattices can also be one- or two-dimensional. For instance, a one-dimensional lattice of coherence domains looks like a set of parallel sheets (layered structure), with the relevant periodicity length  $\lambda$ . A two-dimensional lattice of coherence domains looks like a set of parallel threads, with a corresponding periodicity.

Here we restrict ourselves to the ground-state of the ensemble of particles, labelled by  $n = 0$ , and the first excited state  $n = 1$ , *i.e.* to only one pair (01). We assume a macroscopic occupation for these states, which means to use  $c$ -numbers  $\beta_{0,1}$  for their operators  $b_{0,1}$ . As it is well-known, the occupation number has not definite values anymore in this case, while its conjugate phase is well defined. These are coherent states defined by  $b_{0,1}|\beta_{0,1}\rangle = \beta_{0,1}|\beta_{0,1}\rangle$ . [10] Under these circumstances the interaction reduces to the contribution arising from those photons which satisfy the conservation of energy  $\varepsilon_1 - \varepsilon_0 = \hbar\omega_0$ , where  $\omega_0 = ck_0$ . As it was said above, we limit these wavevectors to the basic reciprocal vectors  $\mathbf{k}_r$  of the coherence lattice, of magnitude  $k_r = k_0 = 2\pi/\lambda_0$ . Their operators  $a_{\alpha\mathbf{k}_r}$ ,  $k_r = k_0$ , are then replaced by  $c$ -numbers  $\alpha$ , the same for any polarization  $\alpha$  and any  $k_r$ . There is no particular reason to have an anisotropy or a polarization dependence for these photon modes. It is easy to see that the interaction lagrangian given by equation (23) becomes then

$$L_{int} = \sqrt{\frac{2\pi\hbar}{V\omega_0}} J_{01} (\alpha + \alpha^*) (\beta_1^* \beta_0 + \beta_1 \beta_0^*) \ , \quad (27)$$

where we have taken  $J_{01} = J_{10}$ . A similar replacement of the field operators by  $c$ -numbers is made in the field lagrangian given by equation (2) and in the particles lagrangian given by equation (15). The summation over  $\alpha\mathbf{k}_r$ ,  $k_r = k_0$ , in the field lagrangian  $L_f$  gives a factor 12, for a three-dimensional lattice. This factor can be absorbed in the photon operators, so we can write down

the "classical" lagrangian

$$\begin{aligned}
 L_f &= \frac{\hbar}{4\omega_0} (\dot{\alpha}^2 + \dot{\alpha}^{*2} + 2|\dot{\alpha}|^2) - \frac{\hbar\omega_0}{4} (\alpha^2 + \alpha^{*2} + 2|\alpha|^2) \ , \\
 L_s &= \frac{1}{2}i\hbar \left( \beta_0^*\dot{\beta}_0 - \dot{\beta}_0^*\beta_0 + \beta_1^*\dot{\beta}_1 - \dot{\beta}_1^*\beta_1 \right) - (\varepsilon_0 |\beta_0|^2 + \varepsilon_1 |\beta_1|^2) \ , \\
 L_{int} &= \frac{g}{\sqrt{N}} (\alpha + \alpha^*) (\beta_0\beta_1^* + \beta_1\beta_0^*) \ ,
 \end{aligned} \tag{28}$$

where the coupling constant is given by

$$g = \sqrt{\pi\hbar/6a^3\omega_0} J_{01} \ . \tag{29}$$

It is worth noting that while the field lagrangian  $L_f$  in equation (28) is the classical lagrangian, the particles lagrangian  $L_s$  and the interaction lagrangian  $L_{int}$  in equations (28) are "classical" only with respect to the second-quantization (field operators), while they preserve their quantum character with respect to the "first quantization".

In order to have some numerical estimates, we may take as a typical value for the energy difference  $\varepsilon_1 - \varepsilon_0 = \hbar\omega_0 = 10eV$ , which corresponds to a photon wavelength  $\lambda_0 = 10^3\text{\AA}$ . This wavelength is much longer than the typical inter-particle distance  $a$ . We can obtain an estimate of the coupling constant  $g$  by representing the matrix element  $J_{01}$  of the current density as  $J_{01} \sim qv \sim qa_0\omega_0 = d\omega_0$ , where  $q$  denotes a charge moving with velocity  $v$  inside each particle with a characteristic radius  $a_0$ ,  $d$  being the corresponding dipole moment.<sup>1</sup> Taking  $q = e$  (the electron charge) we get

$$g = \sqrt{\pi\hbar\omega_0(e^2/6a_0)}(a_0/a)^{3/2} \ , \tag{30}$$

which gives  $g \sim 0.8eV$  for  $\hbar\omega_0 = 10eV$ ,  $a_0 = 0.53\text{\AA}$  (the Bohr radius) and  $a \sim 3\text{\AA}$ . For one- and two-dimensional coherence lattices this coupling constant increases by factors  $\sqrt{3}$  and respectively  $\sqrt{3/2}$ , as a result of the factor  $\sum_{\alpha\mathbf{k}_r}$  in front of the field lagrangian  $L_f$  (this factor is 8 for a two-dimensional lattice and 4 a one-dimensional lattice).

**Equations of motion.** Making use of the lagrangian given by equation (28) we get the equations of motion

$$\begin{aligned}
 \ddot{A} + \omega_0^2 A &= \frac{2\omega_0 g}{\hbar\sqrt{N}} (\beta_0\beta_1^* + \beta_1\beta_0^*) \ , \\
 i\hbar\dot{\beta}_0 &= \varepsilon_0\beta_0 - \frac{g}{\sqrt{N}} A\beta_1 \ , \\
 i\hbar\dot{\beta}_1 &= \varepsilon_1\beta_1 - \frac{g}{\sqrt{N}} A\beta_0 \ ,
 \end{aligned} \tag{31}$$

where  $A = \alpha + \alpha^*$ . The corresponding hamiltonian reads

$$\begin{aligned}
 H_f &= \frac{\hbar}{4\omega_0} \dot{A}^2 + \frac{\hbar\omega_0}{4} A^2 \ , \\
 H_s &= \varepsilon_0 |\beta_0|^2 + \varepsilon_1 |\beta_1|^2 \ , \\
 H_{int} &= -\frac{g}{\sqrt{N}} A (\beta_0\beta_1^* + \beta_1\beta_0^*) \ .
 \end{aligned} \tag{32}$$

It is easy to see, by making use of the equations of motion (31), that it is conserved,

$$H_f + H_s + H_{int} = E \ , \tag{33}$$

<sup>1</sup>This corresponds to the dipole approximation, which, in the non-relativistic limit leaves aside the spin and the so-called diamagnetic contributions to the current density.

where  $E$  is the energy. The number of particles is also conserved: from equations (31) we get easily

$$|\beta_0|^2 + |\beta_1|^2 = N , \quad (34)$$

corresponding to equation (13). Similarly, by making use of equations (31) we get straightforwardly another conservation law, given by

$$\frac{\hbar}{4\omega_0} \left( \dot{A}^2 + \omega_0^2 A^2 \right) - \frac{g}{\sqrt{N}} A (\beta_0 \beta_1^* + \beta_1 \beta_0^*) + \frac{(\varepsilon_1 - \varepsilon_0)}{2} (|\beta_1|^2 - |\beta_0|^2) = Q , \quad (35)$$

where  $Q$  is a constant energy; it can be checked out without difficulty that this is not an independent conservation law; it amounts to  $E - N(\varepsilon_1 + \varepsilon_0)/2 = Q$ .

The stationary solutions of equations (31) are obtained by putting  $\beta_{0,1} = B_{0,1} e^{i\theta}$ ; the equations of motion become

$$\begin{aligned} \ddot{A} + \omega_0^2 A &= \frac{4\omega_0 g}{\hbar\sqrt{N}} B_0 B_1 , \\ i\hbar\dot{B}_0 - \hbar\dot{\theta}B_0 &= \varepsilon_0 B_0 - \frac{g}{\sqrt{N}} A B_1 , \\ i\hbar\dot{B}_1 - \hbar\dot{\theta}B_1 &= \varepsilon_1 B_1 - \frac{g}{\sqrt{N}} A B_0 \end{aligned} \quad (36)$$

The last two equations tell that  $B_{0,1}$  and  $\dot{\theta} = \Omega$  are constant in time and the particular solution of the first equation (36) is

$$A = \frac{4g}{\hbar\omega_0\sqrt{N}} B_0 B_1 . \quad (37)$$

Now it is easy to find out the solutions:

$$\begin{aligned} A &= \frac{2g}{\hbar\omega_0} \sqrt{N} \left[ 1 - (\hbar\omega_0/2g)^4 \right]^{1/2} , \\ B_0^2 &= \frac{1}{2} N \left[ 1 + (\hbar\omega_0/2g)^2 \right] , \\ B_1^2 &= \frac{1}{2} N \left[ 1 - (\hbar\omega_0/2g)^2 \right] , \end{aligned} \quad (38)$$

and frequency

$$\Omega = \omega_0 \left[ -\frac{1}{2} + \frac{2g^2}{\hbar^2\omega_0^2} \right] , \quad (39)$$

where  $\varepsilon_1 - \varepsilon_0 = \hbar\omega_0$  has been used and  $\varepsilon_0$  was put equal to zero.

We can see that the ensemble of atomic particles and the associated electromagnetic field can be put into a coherent state, the occupation amplitudes oscillating with frequency  $\Omega$ , providing the critical condition

$$g > g_{cr} = \hbar\omega_0/2 \quad (40)$$

is fulfilled. Making use of equation (30), this condition reads  $a < a_0 (e^2/\hbar\omega_0 a_0)^{1/3}$ , which is rather a strong condition. For realistic values of  $a$  it requires much lower values of the excitation energy  $\hbar\omega_0$  than those assumed here for the sake of a numerical example.

The total energy of the coherence domain is given by

$$E = -\frac{g^2}{\hbar\omega_0} N \left[ 1 - (\hbar\omega_0/2g)^2 \right]^2 = -\hbar\Omega B_1^2 . \quad (41)$$

It is lower than the non-interacting ground-state energy  $N\varepsilon_0 = 0$ . It may be viewed as the formation enthalpy of the coherence domains. It must be emphasized that this effect of setting

up a coherence in matter is different from the lasing effect, precisely by this formation enthalpy. Rather, the picture emerging from the solution given here resembles to some extent a quantum phase transition. The coupled ensemble of matter and radiation is unstable for a macroscopic occupation of the atomic quantum states and the associated photon states.

Obviously, the coherence solutions obtained here are non-perturbational; they are not analytic in the coupling constant  $g$ . It is worth noting that the stationary solutions given by equations (38) and (39) can also be obtained by minimizing the hamiltonian (32) with the constraint  $B_0^2 + B_1^2 = N$  given by equation (34).

It is also worth noting that the electromagnetic potential given by equation (1) for  $a_{\alpha\mathbf{k}_r} = \alpha$ ,  $k_r = k_0$ , does not depend on the time. Consequently, the electric field is vanishing in the coherence domains. The magnetic field is not vanishing, in general. The vector potential  $\mathbf{A}(\mathbf{r})$  given by equation (1) exhibits spatial oscillations according to the reciprocal vectors  $\mathbf{k}_r$ . The magnetic field may attain high values, depending on the coupling strength  $g$ . Typically, the magnitude of the magnetic field is of the order of  $\sqrt{\hbar\omega_0/a^3}$ . For  $\hbar\omega_0 = 10eV$  and  $a \sim 3\text{\AA}$  this field may be as high as  $\sim 10^6Gs$ . Following Preparata,[1] we may speculate that such a magnetic field might be a good candidate for the Weiss's molecular field of ferromagnetism.

The polarization

$$\mathbf{P} = \frac{1}{V} \sum_i \mathbf{p}(i) \quad (42)$$

of the coherence domains, where  $\mathbf{p}(i)$  is the dipole momentum of the  $i$ -th particle, can easily be calculated by using equations (11), (12) and (38); we get

$$\begin{aligned} \mathbf{P} &= \frac{1}{VN} \sum_i \mathbf{p}(i) [\beta_0^* \beta_1 e^{-i(\theta_{0i} - \theta_{1i})} + \beta_1^* \beta_0 e^{i(\theta_{0i} - \theta_{1i})}] = \\ &= \frac{1}{V} \sum_i \mathbf{p}(i) \cos(\theta_{1i} - \theta_{0i}) [1 - (\hbar\omega_0/2g)^4]^{1/2}, \end{aligned} \quad (43)$$

where  $\mathbf{p}(i) = \mathbf{p}_{01}(i) = \mathbf{p}_{10}(i)$ . In general, without particular assumptions about  $\mathbf{p}(i)$ , the phase summation in equation (43) vanishes and the polarization is zero. It is easy to see for instance that an external field which would modulate the distribution of the dipole momenta  $\mathbf{p}(i)$  with a periodicity corresponding to the reciprocal vectors  $\mathbf{k}_r$  may give rise to a non-vanishing polarization, in view of the coherence condition (26).

**Elementary excitations of the coherence domain.** We change the coordinates in the lagrangian given by equations (28) according to  $A \rightarrow A + \delta A$ ,  $\beta_{0,1} \rightarrow \beta_{0,1} + \delta\beta_{0,1}$ , where  $\delta\beta_{0,1} = (\delta B_{0,1} + iB_{0,1}\delta\theta_{0,1}) e^{i\Omega t}$ . The first-order variation of the lagrangian gives the equations of motion (31), so we are left with the second-order variation of the lagrangian, where  $\delta A$ ,  $\delta\beta_{0,1}$  are viewed as the new coordinates. In addition, we impose the conservation of the number of particles  $B_0\delta B_0 + B_1\delta B_1 = 0$ . With this constraint we get the variation of the lagrangian

$$\begin{aligned} \delta L_f &= \frac{\hbar}{4\omega_0} \delta \dot{A}^2 - \frac{\hbar\omega_0}{4} \delta A^2, \\ \delta L_s &= \hbar B_1 \left[ \delta B_1 (\delta \dot{\theta}_0 - \delta \dot{\theta}_1) - \delta \dot{B}_1 (\delta \theta_0 - \delta \theta_1) \right] - \\ &- (\hbar\Omega N/B_0^2 + \hbar\omega_0) \delta B_1^2 - \hbar\Omega B_0^2 \delta \theta_0^2 - \hbar(\Omega + \omega_0) B_1^2 \delta \theta_1^2, \\ \delta L_{int} &= \frac{2g}{\sqrt{N}} \frac{B_0^2 - B_1^2}{B_0} \delta A \delta B_1 - \frac{2g}{\sqrt{N}} \frac{AB_1}{B_0} \delta B_1^2 + \frac{2g}{\sqrt{N}} AB_0 B_1 \delta \theta_0 \delta \theta_1. \end{aligned} \quad (44)$$

The hamiltonian can readily be obtained from equation (44). It is convenient to introduce the coupling strength  $\lambda = 2g/\hbar\omega_0$  ( $\lambda > 1$ ) and to make use of equations (38) and (39). The hamiltonian



can then be expressed as

$$\begin{aligned} \delta H = & \frac{\hbar}{4\omega_0} \delta \dot{A}^2 + \frac{\hbar\omega_0}{4} \left( \delta A - \frac{2\sqrt{N}}{\lambda B_0} \delta B_1 \right)^2 + \\ & + 2\hbar\omega_0 (\lambda^2 - 1) \delta B_1^2 + \hbar\omega_0 N \frac{\lambda^4 - 1}{4\lambda^2} (\delta\theta_0 - \delta\theta_1)^2, \end{aligned} \quad (45)$$

which tells, first, that the relevant phase coordinate is  $\delta\varphi = \delta\theta_0 - \delta\theta_1$  and, second, that the coordinates  $\delta A$ ,  $\delta B_1$  and  $\delta\varphi$  are associated with the elementary excitations (excited states).

The equations of motions corresponding to the lagrangian given by equations (44) can be written as

$$\begin{aligned} B_0 \left( \delta \ddot{A} + \omega_0^2 \delta A \right) - \frac{2\omega_0^2 \sqrt{N}}{\lambda} \delta B_1 &= 0, \\ \omega_0 N \lambda^2 \delta B_1 - B_0^2 B_1 \delta \dot{\varphi} - \frac{\omega_0 \sqrt{N}}{2\lambda} B_0 \delta A &= 0, \end{aligned} \quad (46)$$

$$\omega_0 N \frac{\lambda^4 - 1}{4\lambda^2} \delta \varphi + B_1 \delta \dot{B}_1 = 0.$$

Their solutions are of the form  $(\delta A, \delta B_1, \delta\varphi)e^{i\omega t}$ , where the frequencies  $\omega$  are given by

$$\omega_{1,2}^2 = \frac{1}{2} \omega_0^2 \left[ \lambda^4 + 1 \pm \sqrt{(\lambda^4 - 1)^2 + 4} \right]. \quad (47)$$

The excitations energies correspond to the frequencies  $\Omega_{1,2} = \Omega \pm \omega_{1,2}$ . In the weak coupling limit these frequencies behave as  $\omega_1 \simeq \sqrt{2}\omega_0$  and  $\omega_2 \simeq \sqrt{\lambda^2 - 1}\omega_0$  ( $\Omega_{1,2} \simeq \omega_{1,2}$ ). In this limit the solution corresponding to the former frequency is  $\delta A \simeq -2\delta B_1 \simeq -i\sqrt{N}(\lambda^2 - 1)\delta\varphi$ , while the one corresponding to the second frequency is  $\delta A \simeq 2\delta B_1 \simeq i\sqrt{2N}\delta\varphi$ . Since for the former solution  $\delta A$  and  $\delta B_1$  vanish in the limit  $\lambda \rightarrow 1$ , while  $\delta\varphi$  is non-vanishing, we may call this elementary excitation "phason". As for the second solution, since all coordinates are non-vanishing, we may call it "amplitudon". Although this terminology is reminiscent of the well-known dynamics of the charge-density waves,[11, 12] the analogy is insubstantial to a large extent.

**"Thermodynamics" of the coherent phase.** In the limit of low temperatures the thermodynamics is controlled by the coherent ground-state energy given by equation (41); the elementary excitations derived above bring no thermodynamical contribution. We can compute directly the partition function  $Z = \text{tr} \exp[\beta(\mu N - H)]$ , where  $\beta = 1/T$  is the inverse of the temperature,  $\mu$  is the chemical potential and the hamiltonian  $H$  is given by equations (32) with  $|\beta_0|^2 + |\beta_1|^2 = N$ . The trace is computed by  $\int d\beta_{0x} d\beta_{0y} \dots$ , where  $\beta_0 = \beta_{0x} + i\beta_{0y}$ , etc. In the thermodynamical limit we get

$$Z \simeq \int d\rho \cdot \frac{e^{\beta N \mu \rho}}{\sqrt{\hbar\omega_0 (\hbar\omega_0 - \mu) - 4g^2 \rho}} \simeq e^{\beta N \mu \hbar\omega_0 (\hbar\omega_0 - \mu) / 4g^2} \quad (48)$$

for  $\mu < 0$ . The thermodynamic potential is given by  $\Omega = N\mu\hbar\omega_0 (\hbar\omega_0 - \mu) / 4g^2$ . We can see that the coherent phase is perfectly ordered, with a vanishing entropy. The chemical potential  $\mu = \hbar\omega_0/2 - 2g^2/\hbar\omega_0 < 0$  implies  $g > \hbar\omega_0/2$ , which is the critical condition given by equation (40). The energy (and free energy) is given by  $E = \Omega + \mu N = -N\hbar\omega_0 (\hbar\omega_0/4g - g/\hbar\omega_0)^2$ , which coincides with the ground-state energy given by equation (41).

**Super-radiant phase transition.** The coherent state described herein is characterized by a macroscopic occupation of the photon state and the two levels. It is indeed known that matter coupled to radiation may suffer an instability toward a super-radiant state at some critical temperature, depending on the coupling constant.[6]-[9]

We start with the quantum hamiltonian written as

$$\begin{aligned} H_f &= \hbar\omega_0 \sum_{\mu} (a_{\mu}^* a_{\mu} + 1/2) , \quad H_s = \hbar\omega_0 b_1^* b_1 , \\ H_{int} &= -\frac{1}{\sqrt{N}} (G b_1^* b_0 + G^* b_0^* b_1) , \end{aligned} \quad (49)$$

where  $\mu$  stands for the pair  $\alpha\mathbf{k}_r$ ,  $G = \sum_{\mu} g_{\mu} a_{\mu}$  and  $g_{\mu} = \sqrt{2\pi\hbar/V\omega_0} J_{01} N(\mu)/\sqrt{N}$ . This is known as the Dicke-Preparata hamiltonian.[7]-[9] Here we follow closely the analysis given in Ref. 6.

First we introduce the spin operators

$$\begin{aligned} S_z &= b_0^* b_0 - b_1^* b_1 = \sum_i (b_{0i}^* b_{0i} - b_{1i}^* b_{1i}) = \sum_i s_{zi} , \\ S_+ &= b_0^* b_1 = \sum_i b_{0i}^* b_{1i} = \sum_i s_{+i} , \\ S_- &= b_1^* b_0 = \sum_i b_{1i}^* b_{0i} = \sum_i s_{-i} , \end{aligned} \quad (50)$$

where  $s$ 's are Pauli matrices. The trace over  $b$ 's in the partition function  $Z = \text{tr} \exp(-\beta H)$ , where  $H = H_f + H_s + H_{int}$ , can then be represented as

$$\text{tr}_b \exp(-\beta H_s - \beta H_{int}) = e^{-\beta\hbar\omega_0 N/2} (\text{tr} e^{\mathbf{h}\mathbf{s}})^N , \quad (51)$$

where

$$h_x = \frac{\beta}{2\sqrt{N}} (G^* + G) , \quad h_y = \frac{i\beta}{2\sqrt{N}} (G^* - G) , \quad h_z = \beta\hbar\omega_0/2 . \quad (52)$$

It is easy to establish the equality  $\text{tr} e^{\mathbf{h}\mathbf{s}} = 2 \cosh h$ , where  $h = \beta (G^* G/N + \hbar^2 \omega_0^2/4)^{1/2}$ . The partition function can now be written as

$$Z = e^{-\beta\hbar\omega_0(N+s)/2} \text{tr} \left\{ e^{-\beta\hbar\omega_0 \sum_{\mu} a_{\mu}^* a_{\mu}} \left[ 2 \cosh \beta (G^* G/N + \hbar^2 \omega_0^2/4)^{1/2} \right]^N \right\} , \quad (53)$$

where  $s = \sum_{\mu}$ . We can see easily that there exists a unitary transformation  $A$ ,  $a_{\mu} = A_{\mu\nu} c_{\nu}$ , which diagonalizes the quadratic form  $G^* G = \sum_{\mu\nu} g_{\mu} g_{\nu} a_{\mu}^* a_{\nu}$ , while preserving the diagonal form  $\sum_{\mu} a_{\mu}^* a_{\mu}$ . It has only one non-vanishing eigenvalue

$$G_0^2 = \sum_{\mu} g_{\mu}^2 = \frac{2\pi\hbar}{V\omega_0} J_{01}^2 \sum_{\mu} N^2(\mu)/N , \quad (54)$$

corresponding to one photon mode denoted by  $c$ . We take  $N(\mu) = N/s$ , and get  $G_0^2 = g^2$ , where  $g$  is given by equation (29) (for  $s = 12$ ). We keep now in the partition function only the contributions which are relevant in the thermodynamical limit, and get

$$Z \simeq e^{-\beta\hbar\omega_0 N/2} \text{tr} \left\{ e^{-\beta\hbar\omega_0 c^* c + N \ln \left[ 2 \cosh \beta (g^2 c^* c/N + \hbar^2 \omega_0^2/4)^{1/2} \right]} \right\} \quad (55)$$

The trace in this equation is computed in the classical limit, where the temperature is much higher than all the relevant energy scales (*e.g.*,  $\beta\hbar\omega_0 \ll 1$ ). We get

$$Z \simeq e^{-\beta\hbar\omega_0 N/2} \int_0^{\infty} dx \cdot e^{-N\phi(x)} , \quad (56)$$

where

$$\phi(x) = \beta \hbar \omega_0 x - \ln \left[ 2 \cosh \beta (g^2 x + \hbar^2 \omega_0^2 / 4)^{1/2} \right] . \quad (57)$$

The main contribution to the integral in equation (56) comes from the minimum value of the function  $\phi(x)$  (Laplace's method), located at  $x_0$  given by

$$\frac{2\hbar\omega_0}{g^2} \sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4} = \tanh \beta \sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4} . \quad (58)$$

This equation has no solution for  $g < \hbar\omega_0$ , at any temperature ( $x_0 = 0$ ). For  $g > \hbar\omega_0$ , there exists a critical temperature  $T_c$  given by  $\hbar^2 \omega_0^2 / g^2 = \tanh \beta_c \hbar \omega_0 / 2$  (or  $\beta_c \simeq 2\hbar\omega_0 / g^2$ ), such that for temperatures higher than  $T_c$  equation (58) has no solution ( $x_0 = 0$ ), while for  $T < T_c$  it has a non-vanishing solution. In the former case the ensemble of particles is in the normal state, with a free energy per particle given by

$$f_0 = \hbar\omega_0 / 2 - \beta^{-1} \ln [2 \cosh \beta \hbar \omega_0 / 2] \quad (59)$$

(interaction-free ensemble). For  $T$  slightly below  $T_c$  we expand equation (58) in powers of  $\sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4} - \hbar\omega_0 / 2$  and get

$$x_0 \simeq \frac{1}{2} (1 - T/T_c)^{1/2} . \quad (60)$$

Now it is easy to get the free energy per particle

$$f \simeq f_0 - \frac{\hbar\omega_0}{4} (1 - T/T_c)^2 . \quad (61)$$

As one can see, the entropy is continuous at the critical temperature, while the specific heat has a discontinuity,  $C = C_0 + \hbar\omega_0 / 2T_c$ . The transition is of the second kind, with the order parameter the photon occupation number. Indeed, it is easy to compute the mean occupation number for photons, which vanishes for  $T > T_c$  and is proportional to  $x_0$  given by equation (60) for  $T < T_c$ . It is worth noting that the super-radiant transition is described by a quantum hamiltonian, while the coherent phase obeys a classical dynamics. This accounts also for the difference in the two critical conditions  $g > \hbar\omega_0 / 2$  and  $g > \hbar\omega_0$ .

**Conclusion.** In conclusion we may say that the interaction of matter with electromagnetic interaction may lead to coherence domains, whose phases are arranged in a periodic lattice, governed basically by a two-level state, providing the coupling constant is greater than a critical value. The coherence domains are made possible by a spatial arrangement in a regular lattice of the phases of the internal motion of the particles, according to the coherence condition (26). These coherence domains are characterized by a macroscopic occupation of the quantum states. The non-linear equations of motion have been solved for the coherent ground state and the elementary excitations have been identified. The solution is a non-perturbational one, the radiation frequency being renormalized in an appreciable way. Perhaps the most direct experimental proof for the existence of such a coherent state is the identification of such elementary excitations which are non-trivially renormalized in comparison with the radiation frequencies. The "thermodynamics" of the coherent phase is computed and the super-radiant transition is re-derived in this context.

A non-trivial generalization of the present approach should address the issue of several level pairs ( $nm$ ). The equations of motion (31) become then matricial equations, and getting their solution is a more difficult task.

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