

Coherent interaction between matter and radiation

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email: apoma@theory.nipne.ro**Abstract**

The coupling of the electromagnetic field to the 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 indeed that coherence domains may set up in matter, their phases 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-perturbative ground-state, which is energetically favourable. In this respect it differs from the well-known lasing mechanism. The elementary excitations with respect to this ground-state are derived, their energy being non-trivially changed by interaction. The "thermodynamics" of the coherent phase is computed and the super-radiant phase transition is re-derived in this context. Apart from the general suggestion of coherence, the present results differ appreciably from Preparata's, *loc cit*, who chose to employ rather sloppy techniques. It is difficult to assess at this moment all the various physical implications of the present model, but the application of such a treatment to particular physical problems like superfluidity, superconductivity, ferromagnetism, Mossbauer effect, Weber's coherent scattering, water anomalies, nuclear forces, etc are promising and worth of further investigation.

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, neither well-founded nor convincingly worked out, about the possible consequence of such a state on various physical phenomena like superfluidity, superconductivity, ferromagnetism, Mossbauer effect, Weber's coherent scattering, water anomalies, nuclear forces, etc. The idea was also originally related to the lasing mechanism and the super-radiance phenomenon.²

We shown here that the coupling between electromagnetic radiation and matter polarization (dipole interaction) may lead to coherent domains involving a two-level state of matter, providing the coupling exceeds a certain critical value. The phases of the coherence doamins are arranged in a periodic lattice. 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 energy of the ground-state is negative, as for a bound

¹G. Preparata, *QED Coherence in Matter*, World Sci (1995).

²R. H. Dicke, Phys. Rev. **93** 99 (1954); W. E. Lamb, Jr., Phys. Rev. **134** A1429 (1964); K. Hepp and E. H. Lieb, Ann. Phys. **76** 360 (1973); Phys. Rev. **A8** 2517 (1973).

state, involving a formation enthalpy for the coherence domains. The "thermodynamics" of the coherent phase is computed and the super-radiant phase transition is re-derived in this context.

This is a new state of matter, which may be termed coherent matter. It is difficult at this moment to envisage all the possible consequences of such a state, though the coherent scattering seems to be very promising.

Electromagnetic field. As it is well-known, the lagrangian of the electromagnetic field is

$$L_f = \frac{1}{8\pi} \int d\mathbf{r} (E^2 - H^2) , \quad (1)$$

where \mathbf{E} is the electric field and \mathbf{H} is the magnetic field. These fields are given by

$$\mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{A}}{\partial t} - \text{grad}\varphi , \quad \mathbf{H} = \text{curl}\mathbf{A} , \quad (2)$$

where c denotes the velocity of light and \mathbf{A} and φ are the electromagnetic potentials. Under these conditions, the fields satisfy automatically the first pair of Maxwell's equations ($\text{curl}\mathbf{E} = -\frac{1}{c}\partial\mathbf{H}/\partial t$, $\text{div}\mathbf{H} = 0$), while the variation of the action $S_f = \int dt L_f$ with respect to the potentials gives the second pair of Maxwell's equations ($\text{curl}\mathbf{H} = \frac{1}{c}\partial\mathbf{E}/\partial t$, $\text{div}\mathbf{E} = 0$). If, in addition, the potentials obey the Lorentz gauge

$$\frac{1}{c} \frac{\partial \varphi}{\partial t} + \text{div}\mathbf{A} = 0 , \quad (3)$$

then they satisfy the wave equation.

In the presence of a charge density ρ and a current density $\mathbf{j} = \mathbf{v}\rho$ (which obey the continuity equation $\partial\rho/\partial t + \text{div}\mathbf{j} = 0$), where \mathbf{v} denotes the velocity of the charge, the interacting term

$$L_{int} = \frac{1}{c} \int d\mathbf{r} \cdot \mathbf{j}\mathbf{A} - \int d\mathbf{r} \rho\varphi \quad (4)$$

must be added to the lagrangian. The same scheme of variation of the action leads to the Maxwell's equations

$$\text{curl}\mathbf{H} = \frac{1}{c}\partial\mathbf{E}/\partial t + \frac{4\pi}{c}\mathbf{j} , \quad \text{div}\mathbf{E} = 4\pi\rho \quad (5)$$

in the presence of matter and to the wave equations

$$\frac{1}{c^2} \frac{\partial^2}{\partial t^2} \varphi - \Delta\varphi = 4\pi\rho , \quad \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{A} - \Delta\mathbf{A} = \frac{4\pi}{c}\mathbf{j} \quad (6)$$

with sources.

We adopt herein the radiation gauge $\varphi = 0$ (as for neutral atomic matter, where $\rho = 0$ but $\mathbf{j} \neq 0$ and $\text{div}\mathbf{j} = 0$), which, by (3), gives the transversality condition $\text{div}\mathbf{A} = 0$. We represent the vector potential as

$$\mathbf{A} = \sum_{\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar c^2}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})a_{\alpha\mathbf{k}}e^{i\mathbf{k}\mathbf{r}} + \mathbf{e}_\alpha^*(\mathbf{k})a_{\alpha\mathbf{k}}^*e^{-i\mathbf{k}\mathbf{r}}] , \quad (7)$$

where \hbar is Planck's constant, V denotes the volume, $\omega_k = ck$ are the frequencies of the free field (equation (6) with $\mathbf{j} = 0$), α is the polarization label ($\alpha = \pm 1$) and $\mathbf{e}_\alpha(\mathbf{k})$ denote the polarization vectors, $\mathbf{e}_\alpha(\mathbf{k})\mathbf{k} = 0$ (transversality condition), $\mathbf{e}_\alpha(\mathbf{k})\mathbf{e}_\beta^*(\mathbf{k}) = \delta_{\alpha\beta}$ (two independent polarizations). We represent the polarization vectors as $\mathbf{e}_+(\mathbf{k}) = \mathbf{e}_xe^{i\chi}$, $\mathbf{e}_-(\mathbf{k}) = \mathbf{e}_ye^{i\chi}$, where χ is an arbitrary phase and $\mathbf{e}_{x,y}$ the two (real) unit vectors perpendicular to \mathbf{k} directed along the z -axis, and note that $\mathbf{e}_+(-\mathbf{k}) = \mathbf{e}_ye^{-i\chi} = \mathbf{e}_-^*(\mathbf{k})$, $\mathbf{e}_-(-\mathbf{k}) = \mathbf{e}_xe^{-i\chi} = \mathbf{e}_+^*(\mathbf{k})$.

Making use of this representation for the vector potential \mathbf{A} we get

$$\frac{1}{8\pi} \int d\mathbf{r} E^2 = \sum_{\alpha\mathbf{k}} \frac{\hbar}{4\omega_k} (\dot{a}_{\alpha\mathbf{k}}\dot{a}_{-\alpha-\mathbf{k}} + \dot{a}_{\alpha\mathbf{k}}^*\dot{a}_{-\alpha-\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}}\dot{a}_{\alpha\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}}^*\dot{a}_{\alpha\mathbf{k}}) \quad (8)$$

and

$$\frac{1}{8\pi} \int d\mathbf{r} B^2 = \sum_{\alpha\mathbf{k}} \frac{\hbar\omega_k}{4} (a_{\alpha\mathbf{k}}a_{-\alpha-\mathbf{k}} + a_{\alpha\mathbf{k}}^*a_{-\alpha-\mathbf{k}}^* + a_{\alpha\mathbf{k}}a_{\alpha\mathbf{k}}^* + a_{\alpha\mathbf{k}}^*a_{\alpha\mathbf{k}}) \quad (9)$$

which help in constructing the lagrangian L_f given by (1).

Similarly, making use of the Fourier transform

$$\mathbf{j} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} \quad (10)$$

for the current density, with $\mathbf{j}^*(-\mathbf{k}) = \mathbf{j}(\mathbf{k})$, we get the interacting part of the lagrangian given by equation (4) as

$$L_{int} = \sum_{\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{\omega_k}} [\mathbf{e}_{\alpha}(\mathbf{k})\mathbf{j}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_{\alpha}^*(\mathbf{k})\mathbf{j}(\mathbf{k})a_{\alpha\mathbf{k}}^*] . \quad (11)$$

By (4), we can see that, in the radiation gauge, this interaction part of the action is equivalent with the dipolar interaction.

The variation of the action $\int dt(L_f + L_{int})$ with respect to $a_{\alpha\mathbf{k}}$ leads to the equation of motion

$$\ddot{a}_{\alpha\mathbf{k}} + \ddot{a}_{-\alpha-\mathbf{k}}^* + \omega_k^2 (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) = \sqrt{\frac{8\pi\omega_k}{\hbar}} \mathbf{e}_{\alpha}^*(\mathbf{k})\mathbf{j}(\mathbf{k}) , \quad (12)$$

which is Maxwell's equation (6) within the representation given by equation (7). It is worth noting that this equation of motion is equally valid both in the classical and quantum case ($[a_{\alpha\mathbf{k}}, a_{\beta\mathbf{k}'}^*] = \delta_{\alpha\beta}\delta_{\mathbf{k}\mathbf{k}'}$, $[a_{\alpha\mathbf{k}}, a_{\beta\mathbf{k}}] = 0$).

From the lagrangian $L_f + L_{int}$ written in the a 's representation we can construct the hamiltonian, either classically or quantally, by introducing the momentum $p_{\alpha\mathbf{k}} = \delta L_f / \delta \dot{a}_{\alpha\mathbf{k}} = (\hbar/2\omega_k) (\dot{a}_{-\alpha-\mathbf{k}} + \dot{a}_{\alpha\mathbf{k}}^*)$. It is given by

$$H_f = \frac{1}{8\pi} \int d\mathbf{r} (E^2 + H^2) \quad (13)$$

and $H_{int} = -L_{int}$ as expected, and leads to the same equation of motion (12).

Matter. The lagrangian of a non-relativistic particle with mass m and charge q in an electromagnetic field is given by

$$L_{mf} = \frac{1}{2}mv^2 + \frac{q}{c}\mathbf{v}\mathbf{A} \quad (14)$$

(in the radiation gauge). We can see that the interacting part of the lagrangian in equation (14) is the same as the interacting part L_{int} given by equation (4) (since $\mathbf{j} = \mathbf{v}\rho = q\mathbf{v}\delta(\mathbf{r})$). Therefore, the source terms in equations of motion (12) for the electromagnetic field will not be modified on adding the matter lagrangian, as it should be.

The equation of motion for the particle is readily obtained from (14) as Lorentz's force³

$$\frac{d}{dt}(m\mathbf{v}) = q\mathbf{E} + \frac{q}{c}\mathbf{v} \times \mathbf{H} . \quad (15)$$

³But using the identity $grad(\mathbf{a}\mathbf{b}) = (\mathbf{a}grad)\mathbf{b} + (\mathbf{b}grad)\mathbf{a} + \mathbf{a} \times curl\mathbf{b} + \mathbf{b} \times curl\mathbf{a}$. The fully relativistic equation of motion replaces $m\mathbf{v}$ in (15) by $m\mathbf{v}/\sqrt{1-v^2/c^2}$.

The momentum of the particle is given by $\mathbf{P} = m\mathbf{v} + q\mathbf{A}/c$, and the hamiltonian reads $H_{mf} = (\mathbf{P} - q\mathbf{A}/c)^2/2m$, leading, of course, to the same equation of motion for the particle. We can write

$$H_{mf} = P^2/2m - \frac{q}{mc}\mathbf{P}\mathbf{A} + \frac{q^2}{2mc^2}A^2, \quad (16)$$

and may view the last two terms in equation (16) as the interacting part of the hamiltonian. If we compute the contribution of this interaction to the equation of motion of the electromagnetic field,⁴ we find a term of the form $-\mathbf{P} + q\mathbf{A}/c$, which, by using $\mathbf{P} = m\mathbf{v} + q\mathbf{A}/c$, reads $-m\mathbf{v}$. This shows again that indeed, the interaction is governed by velocity \mathbf{v} , as in equation (12). Therefore, we consider the non-interacting part of matter, to which we add the free electromagnetic field and the interaction as given by equation (4) or (11).

The quantization of matter requires the presence of the momentum \mathbf{P} in the hamiltonian H_{mf} . We can write it as

$$H_{mf} = P^2/2m - \frac{q}{c}\mathbf{v}\mathbf{A} - \frac{q^2}{2mc^2}A^2, \quad (17)$$

where the term linear in \mathbf{A} is the same as the interaction given in L_{int} . In order to estimate the interactions in equation (17) we may employ the Lienard-Wiechert potential for a charge moving with velocity \mathbf{v} . We have $A \sim qv/cr$ at distance r , so the interaction term linear in A in (17) is $\sim (q^2/r)(v/c)^2$, while the interaction term quadratic in A in (17) is $\sim (q^2/mc^2r)(q^2/r)(v/c)^2$. We may see that for atomic matter we may safely neglect the interaction term quadratic in A . To the same approximation to the non-relativistic dynamics, the quantization brings yet another interaction, $-(\hbar q/mc)\mathbf{H}\mathbf{s}$, which implies the magnetic momentum (\mathbf{s} is the particle spin). It is easy to see that it can be estimated as $\sim (\hbar^2/ma^2E)^{1/2}(q^2/r)(v/c)^2$, where a is the mean inter-particle distance and E is an atomic transition energy. This interaction can also be neglected, though, in some cases, it may bring contributions comparable with those brought by the term linear in \mathbf{A} . In any case it may be included in the interaction term linear in \mathbf{A} , as the spin current.

We consider a set of N independent, identical atomic particles labelled by i , and write their internal hamiltonian as

$$H_m = \sum_i H_m(i). \quad (18)$$

We introduce a set of orthonormal eigenfunctions $\varphi_n(i)$, such as

$$H_m(i)\varphi_n(j) = \varepsilon_n\delta_{ij}, \quad \int d\mathbf{r}\varphi_n^*(i)\varphi_m(j) = \delta_{ij}\delta_{nm} \quad (19)$$

and construct a set of normalized eigenfunctions

$$\varphi_n = \sum_i c_{ni}\varphi_n(i) \quad (20)$$

for the whole ensemble,

$$H_m\varphi_n = \varepsilon_n\varphi_n, \quad (21)$$

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

$$\sum_i |c_{ni}|^2 = 1. \quad (22)$$

⁴In the quantum case the $\mathbf{P}\mathbf{A}$ -term must be symmetrized.

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

$$\varphi_n = \frac{1}{\sqrt{N}} \sum_i e^{i\theta_{ni}} \varphi_n(i) . \quad (23)$$

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

$$\Psi = \sum_n b_n \varphi_n \quad (24)$$

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

$$N = \sum_n b_n^* b_n . \quad (25)$$

The lagrangian of this ensemble of particles can be represented as

$$L_m = \frac{1}{2} \int d\mathbf{r} (\Psi^* \cdot i\hbar \partial \Psi / \partial t - i\hbar \partial \Psi^* / \partial t \cdot \Psi) - \int d\mathbf{r} \Psi^* H_m \Psi , \quad (26)$$

or

$$L_m = \frac{1}{2} \sum_n i\hbar [b_n^* \dot{b}_n - \dot{b}_n^* b_n] - \sum_n \varepsilon_n b_n^* b_n , \quad (27)$$

where

$$H_m = \sum_n \varepsilon_n b_n^* b_n \quad (28)$$

is the hamiltonian. They lead to the equations of motion

$$i\hbar \dot{b}_n = \varepsilon_n b_n , \quad (29)$$

which is Schrodinger's equation. It is worth noting that the same equation is obtained for b_n viewed as classical variables.

Since the charge density is given by

$$\rho = \sum_i q_i \delta(\mathbf{r} - \mathbf{r}_i) \quad (30)$$

for an ensemble of point-like charges q_i placed at positions \mathbf{r}_i , the current density can be written as

$$\mathbf{j} = \frac{1}{V} \sum_{i\mathbf{k}} q_i \mathbf{v}_i e^{-i\mathbf{k}\mathbf{r}_i} \cdot e^{i\mathbf{k}\mathbf{r}} = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \mathbf{j}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} . \quad (31)$$

Here we have assumed that \mathbf{r}_i gets an additional degree of freedom, $\mathbf{r}_i \rightarrow \mathbf{r}_i + \mathbf{u}_i$, and the velocity \mathbf{v}_i corresponds to $\dot{\mathbf{u}}_i$. We write therefore the Fourier transform of the current density as

$$\mathbf{j}(\mathbf{k}) = \frac{1}{\sqrt{V}} \sum_i \mathbf{J}(i) e^{-i\mathbf{k}\mathbf{r}_i} , \quad (32)$$

where $\mathbf{J}(i) = q_i \mathbf{v}_i$ is the current of the i -th particle. Now, it is easy to see that the interacting part L_{int} of the lagrangian given by equation (11) becomes

$$L_{int} = \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^*] b_n^* b_m, \quad (33)$$

where

$$\mathbf{I}_{nm}(\mathbf{k}) = \frac{1}{N} \sum_i \mathbf{J}_{nm}(i) e^{-i(\theta_{ni} - \theta_{mi})} e^{-i\mathbf{k}\mathbf{r}_i} \quad (34)$$

with $\mathbf{J} = q\mathbf{v}$ the current for each particle. The equations of motion (29) become

$$i\hbar\dot{b}_n = \varepsilon_n b_n - \sum_{m\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^*] b_m, \quad (35)$$

while the equation of motion (12) for the electromagnetic field reads now

$$\ddot{a}_{\alpha\mathbf{k}} + \ddot{a}_{-\alpha-\mathbf{k}}^* + \omega_k^2 (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) = \sum_{nm} \sqrt{\frac{8\pi\omega_k}{V\hbar}} \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k}) b_n^* b_m. \quad (36)$$

It is worthwhile writing explicitly the energy of the interacting matter and radiation. The full hamiltonian reads $H = H_m + H_f + H_{int}$, where

$$\begin{aligned} H_m &= \sum_n \varepsilon_n b_n^* b_n, \\ H_f &= \sum_{\alpha\mathbf{k}} \frac{\hbar}{4\omega_k} (\dot{a}_{\alpha\mathbf{k}} \dot{a}_{-\alpha-\mathbf{k}} + \dot{a}_{\alpha\mathbf{k}}^* \dot{a}_{-\alpha-\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}} \dot{a}_{\alpha\mathbf{k}}^* + \dot{a}_{\alpha\mathbf{k}}^* \dot{a}_{\alpha\mathbf{k}}) + \\ &+ \sum_{\alpha\mathbf{k}} \frac{\hbar\omega_k}{4} (a_{\alpha\mathbf{k}} a_{-\alpha-\mathbf{k}} + a_{\alpha\mathbf{k}}^* a_{-\alpha-\mathbf{k}}^* + a_{\alpha\mathbf{k}} a_{\alpha\mathbf{k}}^* + a_{\alpha\mathbf{k}}^* a_{\alpha\mathbf{k}}) = \\ &= \sum_{\alpha\mathbf{k}} \frac{\hbar\omega_k}{2} (a_{\alpha\mathbf{k}} a_{\alpha\mathbf{k}}^* + a_{\alpha\mathbf{k}}^* a_{\alpha\mathbf{k}}), \\ H_{int} &= - \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^*] b_n^* b_m. \end{aligned} \quad (37)$$

The equation of motion for $a_{\alpha\mathbf{k}}$ reads

$$i\hbar\dot{a}_{\alpha\mathbf{k}} = [a_{\alpha\mathbf{k}}, H] = \hbar\omega_k a_{\alpha\mathbf{k}} - \sum_{nm} \sqrt{\frac{2\pi\hbar}{V\omega_k}} \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k}) b_n^* b_m, \quad (38)$$

which is equivalent with equation (36).

We have already assumed $a_{\alpha\mathbf{k}} \rightarrow a_{\alpha\mathbf{k}} e^{-i\omega_k t}$ in deriving the quantized field hamiltonian. It is convenient to make a similar assumption $b_n \rightarrow b_n e^{-i\varepsilon_n t/\hbar}$ for the operators of the particles quanta. Under these circumstances the equations of motion become

$$\begin{aligned} i\hbar\dot{b}_n &= - \sum_{m\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{mn}^*(\mathbf{k})a_{\alpha\mathbf{k}} e^{\frac{i}{\hbar}(\varepsilon_n - \varepsilon_m - \hbar\omega_k)} + \\ &+ \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^* e^{\frac{i}{\hbar}(\varepsilon_n - \varepsilon_m + \hbar\omega_k)}] b_m, \\ i\hbar\dot{a}_{\alpha\mathbf{k}} &= - \sum_{nm} \sqrt{\frac{2\pi\hbar}{V\omega_k}} \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k}) e^{\frac{i}{\hbar}(\varepsilon_n - \varepsilon_m + \hbar\omega_k)} \cdot b_n^* b_m, \end{aligned} \quad (39)$$

and the interaction hamiltonian reads

$$H_{int} = - \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} [\mathbf{e}_\alpha(\mathbf{k})\mathbf{I}_{nm}^*(\mathbf{k})a_{\alpha\mathbf{k}}e^{\frac{i}{\hbar}(\varepsilon_n-\varepsilon_m-\hbar\omega_k)} + \mathbf{e}_\alpha^*(\mathbf{k})\mathbf{I}_{nm}(\mathbf{k})a_{\alpha\mathbf{k}}^*e^{\frac{i}{\hbar}(\varepsilon_n-\varepsilon_m+\hbar\omega_k)}] b_n^* b_m . \quad (40)$$

Now it is easy to recognize the interaction picture of the electromagnetic field with particles quanta, amenable to perturbation theory for calculating transition amplitudes, as in absorption or radiation of photons, or radiation scattering on matter.

Macroscopic occupation. Coherence. The transition amplitudes involving highly-excited states are small. The most relevant transitions generated by the interaction hamiltonian (40) are those between the ground-state and the lowest-excited states of the matter. Following Preparata⁵ we examine herein whether a macroscopic occupation of such states, and of the corresponding photon states, is favourable.

First, let a, a^* be the destruction and creation operators for bosonic states $|n\rangle$, with occupation number n , such as $a|0\rangle = 0, a|n\rangle = \sqrt{n}|n-1\rangle, a^*|n\rangle = \sqrt{n+1}|n+1\rangle, [a, a^*] = 1$, the number of quanta being given by $N = a^*a, N|n\rangle = n|n\rangle$. We may represent a as $a = e^{i\theta}\sqrt{N}$ and get $[a, a^*] = e^{i\theta}Ne^{-i\theta} - N = 1$; hence, $[N, \theta] = 1$, or $N = i\partial/\partial\theta$. The quantal states have a definite occupation number n , so their phase is completely undetermined. The classical, macroscopically occupied states have a determined phase and a completely undetermined occupation number. They can be represented as $|\alpha\rangle = \sum_n C_n |n\rangle$, and from $a|\alpha\rangle = \alpha|\alpha\rangle$, we get the coefficients C_n . Such a normalized state reads⁶

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_n \frac{\alpha^n}{\sqrt{n!}} |n\rangle , \quad (41)$$

and the (mean) number of quanta in state $|\alpha\rangle$ is $N = |\alpha|^2$; its mean square deviation is \sqrt{N} . Therefore, the q -number a can be viewed as a c -number α , which is a complex number whose modulus is the square root of the occupation number N . Such macroscopically occupied states are called coherent states because they have a definite phase; they imply an off-diagonal order. For such coherent states we denote the photon operators a by α and the particles states operators b by β .

Coherence domains. Making use of equation (34) the interaction lagrangian given by equation (33) can be written as

$$L_{int} = \sum_{nm\alpha\mathbf{k}} \sqrt{\frac{2\pi\hbar}{V\omega_k}} F_{nm}(\alpha\mathbf{k}) (a_{\alpha\mathbf{k}} + a_{-\alpha-\mathbf{k}}^*) b_n^* b_m , \quad (42)$$

where

$$F_{nm}(\alpha\mathbf{k}) = \frac{1}{N} \sum_i \mathbf{e}_\alpha(\mathbf{k}) \mathbf{J}_{nm}(i) e^{i\mathbf{k}\mathbf{r}_i - i(\theta_{ni} - \theta_{mi})} . \quad (43)$$

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

⁵G. Preparata, *loc cit.*

⁶R. J. Glauber, *Phys. Rev.* **131** 2766 (1963).

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 λ) occurs for the rhombohedral lattice. We limit the relevant wavevectors \mathbf{k} to this finite set of basic reciprocal vectors, for which $\mathbf{k}_r \mathbf{R}_p = 2\pi \times \text{integer}$. Equation (43) becomes then

$$F_{nm}(\alpha \mathbf{k}_r) = \frac{1}{N} \sum_{pi} \mathbf{e}_\alpha(\mathbf{k}_r) \mathbf{J}_{nm}(i) e^{i\mathbf{k}_r \mathbf{r}_{pi} - i(\theta_{ni} - \theta_{mi})} , \quad (44)$$

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

The summation over i in the Wigner-Seitz cell of the remaining phase factors in equation (44) 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 (44) 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 θ_{ni} fulfill the condition

$$\mathbf{k}_r \mathbf{r}_{pi} - (\theta_{ni} - \theta_{mi}) = K , \quad (45)$$

where K is a constant. We can see that these sub-sets of particles are disjoint, *i.e.* if a particle satisfies condition (45) 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 $\exp(K)$, equation (44) 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 (45), *i.e.* $\sum_{(nm)\alpha \mathbf{k}_r} N_{nm}(\alpha \mathbf{k}_r) = N$.

Condition (45) is a strong condition, which 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 (45) may be taken as the basic condition for coherence. We call such an ensemble of particles which satisfies condition (45) 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 λ . 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 $b_{0,1}$. 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 α , the same for any polarization α and any k_r . There is no particular reason to have an anisotropy or a polarization dependence for these

relevant photon modes. It is easy to see that the interaction lagrangian given by equation (42) becomes then

$$L_{int} = \sqrt{\frac{2\pi\hbar}{V\omega_0}} J_{01} (\alpha + \alpha^*) (\beta_1^* \beta_0 + \beta_1 \beta_0^*) \quad , \quad (46)$$

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 equations (8) and (9) and in the particles lagrangian given by equation (27). 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

$$L_f = \frac{\hbar}{4\omega_0} (\dot{\alpha}^2 + \dot{\alpha}^{*2} + 2|\dot{\alpha}|^2) - \frac{\hbar\omega_0}{4} (\alpha^2 + \alpha^{*2} + 2|\alpha|^2) \quad ,$$

$$L_m = \frac{1}{2}i\hbar \left(\beta_0^* \dot{\beta}_0 - \dot{\beta}_0^* \beta_0 + \beta_1^* \dot{\beta}_1 - \dot{\beta}_1^* \beta_1 \right) - (\varepsilon_0 |\beta_0|^2 + \varepsilon_1 |\beta_1|^2) \quad , \quad (47)$$

$$L_{int} = \frac{g}{\sqrt{N}} (\alpha + \alpha^*) (\beta_0 \beta_1^* + \beta_1 \beta_0^*) \quad ,$$

where the coupling constant is given by

$$g = \sqrt{\pi\hbar/6a^3\omega_0} J_{01} \quad . \quad (48)$$

Is worth noting that the interaction lagrangian given by equation (47) differs from its "quantum" (field-theoretical) counterpart by the non-vanishing overlapping of the coherent states, which allows both α and α^* in the "elementary" interaction processes. It is also worth noting that while the field lagrangian L_f in equation (47) is the classical lagrangian, the particles lagrangian L_m and the interaction lagrangian L_{int} in equations (47) 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.⁷ Taking $q = e$ (the electron charge) we get

$$g = \sqrt{\pi\hbar\omega_0(e^2/6a_0)} (a_0/a)^{3/2} \quad , \quad (49)$$

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 .

Equations of motion. Making use of the lagrangian given above we get the equations of motion

$$\ddot{\alpha} + \ddot{\alpha}^* + \omega_0^2 (\alpha + \alpha^*) = \frac{2\omega_0 g}{\hbar\sqrt{N}} (\beta_0 \beta_1^* + \beta_1 \beta_0^*) \quad (50)$$

and

$$i\hbar\dot{\beta}_0 = \varepsilon_0 \beta_0 - \frac{g}{\sqrt{N}} (\alpha + \alpha^*) \beta_1 \quad ,$$

$$i\hbar\dot{\beta}_1 = \varepsilon_1 \beta_1 - \frac{g}{\sqrt{N}} (\alpha + \alpha^*) \beta_0 \quad . \quad (51)$$

⁷This 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.

We rewrite these equations of motion with the real quantity $A = \alpha + \alpha^*$,

$$\begin{aligned}\ddot{A} + \omega_0^2 A &= \frac{2\omega_0 g}{\hbar\sqrt{N}} (\beta_0\beta_1^* + \beta_1\beta_0^*) , \\ i\hbar\dot{\beta}_0 &= \varepsilon_0\beta_0 - \frac{g}{\sqrt{N}} A\beta_1 , \\ i\hbar\dot{\beta}_1 &= \varepsilon_1\beta_1 - \frac{g}{\sqrt{N}} A\beta_0 .\end{aligned}\tag{52}$$

The corresponding hamiltonian reads

$$\begin{aligned}H_f &= \frac{\hbar}{4\omega_0} \dot{A}^2 + \frac{\hbar\omega_0}{4} A^2 , \\ H_m &= \varepsilon_0 |\beta_0|^2 + \varepsilon_1 |\beta_1|^2 , \\ H_{int} &= -\frac{g}{\sqrt{N}} A (\beta_0\beta_1^* + \beta_1\beta_0^*) .\end{aligned}\tag{53}$$

It is conserved,

$$H_f + H_m + H_{int} = E ,\tag{54}$$

where E is the energy. The number of particles is also conserved: from equations (52) we get easily

$$|\beta_0|^2 + |\beta_1|^2 = N .\tag{55}$$

Making use of equations (52) we get straightforwardly another conservation law, given by

$$\frac{\hbar}{4\omega_0} (\dot{A}^2 + \omega_0^2 A^2) - \frac{g}{\sqrt{N}} A (\beta_0\beta_1^* + \beta_1\beta_0^*) + \frac{(\varepsilon_1 - \varepsilon_0)}{2} (|\beta_1|^2 - |\beta_0|^2) = Q ,\tag{56}$$

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

$$\begin{aligned}\ddot{A} + \omega_0^2 A &= \frac{4\omega_0 g}{\hbar\sqrt{N}} B_0 B_1 , \\ i\hbar\dot{B}_0 - \hbar\dot{\theta}B_0 &= \varepsilon_0 B_0 - \frac{g}{\sqrt{N}} A B_1 , \\ i\hbar\dot{B}_1 - \hbar\dot{\theta}B_1 &= \varepsilon_1 B_1 - \frac{g}{\sqrt{N}} A B_0\end{aligned}\tag{57}$$

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 in (57) is

$$A = \frac{4g}{\hbar\omega_0\sqrt{N}} B_0 B_1 .\tag{58}$$

Now it is easy to find out the solutions

$$\begin{aligned}A &= \frac{2g}{\hbar\omega_0} \sqrt{N} [1 - (\hbar\omega_0/2g)^4]^{1/2} , \\ B_0^2 &= \frac{1}{2} N [1 + (\hbar\omega_0/2g)^2] , \\ B_1^2 &= \frac{1}{2} N [1 - (\hbar\omega_0/2g)^2] ,\end{aligned}\tag{59}$$

and frequency

$$\Omega = \omega_0 \left[-\frac{1}{2} + \frac{2g^2}{\hbar^2\omega_0^2} \right] , \quad (60)$$

where $\varepsilon_1 - \varepsilon_0 = \hbar\omega_0$ has been used and ε_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 Ω , providing the critical condition

$$g > g_{cr} = \hbar\omega_0/2 \quad (61)$$

is fulfilled. The total energy of the coherence domain is given by

$$E = -\frac{g^2}{\hbar\omega_0} N \left[1 - (\hbar\omega_0/2g)^2 \right]^2 = -\hbar\Omega B_1^2 . \quad (62)$$

It is lower than the non-interacting ground-state energy $N\varepsilon_0 = 0$. It may be viewed as the enthalpy of formation for the coherence domain. 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. The coupled ensemble of matter and radiation is unstable for a macroscopic occupation of the atomic quantum states and the associated photon states. From equations (62) and (60) we may see that $(\hbar\Omega/2) \left[1 - (\hbar\omega_0/2g)^2 \right]$ can be viewed as the chemical potential associated to the number N of particles.

Obviously, the coherence solutions obtained here are non-perturbative; they are not analytic in the coupling constant g . It is worth noting that the stationary solutions given by equations (59) and (60) can also be obtained by minimizing the hamiltonian (54) with the constraint $B_0^2 + B_1^2 = N$ given by equation (55).

It is also worth noting that the electromagnetic potential given by equation (7) 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 (7) 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 $\sim 10^6Gs$. We may speculate that such a magnetic field might be a good candidate for the Weiss's molecular field of ferromagnetism.

The polarization

$$\mathbf{P} = \frac{1}{V} \sum_i \mathbf{p}(i) \quad (63)$$

of the coherence domains, where $\mathbf{p}(i)$ is the dipole momentum of the i -th particle, can easily be calculated by using equations (23), (24) and (59); we get

$$\begin{aligned} \mathbf{P} &= \frac{1}{VN} \sum_i \mathbf{p}(i) \left[\beta_0^* \beta_1 e^{-i(\theta_{0i} - \theta_{1i})} + \beta_1^* \beta_0 e^{i(\theta_{0i} - \theta_{1i})} \right] = \\ &= \frac{1}{V} \sum_i \mathbf{p}(i) \cos(\theta_{1i} - \theta_{0i}) \left[1 - (\hbar\omega_0/2g)^4 \right]^{1/2} , \end{aligned} \quad (64)$$

where $\mathbf{p}(i) = \mathbf{p}_{01}(i) = \mathbf{p}_{10}(i)$. In general, without particular assumptions on $\mathbf{p}(i)$, the phase summation in equation (64) 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 (45).

Elementary excitations of the coherence domains. We change the coordinates in the lagrangian given by equations (47) 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 (52), so we are left with the second-order variation of the lagrangian, where δA , $\delta\beta_{0,1}$ 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

$$\begin{aligned} \delta L_f &= \frac{\hbar}{4\omega_0} \delta \dot{A}^2 - \frac{\hbar\omega_0}{4} \delta A^2 , \\ \delta L_m &= \hbar B_1 \left[\delta B_1 \left(\delta \dot{\theta}_0 - \delta \dot{\theta}_1 \right) - \delta \dot{B}_1 (\delta \theta_0 - \delta \theta_1) \right] - \\ &- (\hbar\Omega N/B_0^2 + \hbar\omega_0) \delta B_1^2 - \hbar\Omega B_0^2 \delta \theta_0^2 - \hbar(\Omega + \omega_0) B_1^2 \delta \theta_1^2 , \\ \delta L_{int} &= \frac{2g}{\sqrt{N}} \frac{B_0^2 - B_1^2}{B_0} \delta A \delta B_1 - \frac{2g}{\sqrt{N}} \frac{AB_1}{B_0} \delta B_1^2 + \frac{2g}{\sqrt{N}} AB_0 B_1 \delta \theta_0 \delta \theta_1 . \end{aligned} \quad (65)$$

The hamiltonian can readily be obtained from equation (65). It is convenient to introduce the coupling strength $\lambda = 2g/\hbar\omega_0$ ($\lambda > 1$) and to make use of equations (59) and (60). We get the hamiltonian

$$\begin{aligned} \delta H &= \frac{\hbar}{4\omega_0} \delta \dot{A}^2 + \frac{\hbar\omega_0}{4} \left(\delta A - \frac{2\sqrt{N}}{\lambda B_0} \delta B_1 \right)^2 + \\ &+ 2\hbar\omega_0 (\lambda^2 - 1) \delta B_1^2 + \hbar\omega_0 N \frac{\lambda^4 - 1}{4\lambda^2} (\delta \theta_0 - \delta \theta_1)^2 , \end{aligned} \quad (66)$$

which tells, first, that the relevant phase coordinate is $\delta\varphi = \delta\theta_0 - \delta\theta_1$ and, second, that the coordinates δA , δB_1 and $\delta\varphi$ are associated with the elementary excitations (excited states).

The equations of motions corresponding to the lagrangian given by equations (65) can be written as

$$\begin{aligned} B_0 \left(\delta \ddot{A} + \omega_0^2 \delta A \right) - \frac{2\omega_0^2 \sqrt{N}}{\lambda} \delta B_1 &= 0 , \\ \omega_0 N \lambda^2 \delta B_1 - B_0^2 B_1 \delta \dot{\varphi} - \frac{\omega_0 \sqrt{N}}{2\lambda} B_0 \delta A &= 0 , \\ \omega_0 N \frac{\lambda^4 - 1}{4\lambda^2} \delta \varphi + B_1 \delta \dot{B}_1 &= 0 . \end{aligned} \quad (67)$$

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

$$\omega_{1,2}^2 = \frac{1}{2} \omega_0^2 \left[\lambda^4 + 1 \pm \sqrt{(\lambda^4 - 1)^2 + 4} \right] . \quad (68)$$

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 δA and δB_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,⁸ 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 (62); the elementary

⁸P. A. Lee, T. M. Rice and P. W. Anderson, Solid State Commun. **14** 703 (1974); G. Gruner, Revs. Mod. Phys. **60** 1129 (1988).

excitations derived above bring no thermodynamical contribution. We can compute directly the partition function $Z = tr \exp [\beta (\mu N - H)]$, where $\beta = 1/T$ is the inverse of the temperature, μ is the chemical potential and the hamiltonian H is given by equations (53) 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

$$Z \simeq \int d\rho \cdot \frac{e^{\beta N \mu \rho}}{\sqrt{\hbar\omega_0 (\hbar\omega_0 - \mu) - 4g^2 \rho}} \simeq e^{\beta N \mu \hbar\omega_0 (\hbar\omega_0 - \mu) / 4g^2} \quad (69)$$

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 (61). 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 (62).

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.⁹

We start with the quantum hamiltonian written as

$$H_f = \hbar\omega_0 \sum_{\mu} (a_{\mu}^* a_{\mu} + 1/2) \quad , \quad H_s = \hbar\omega_0 b_1^* b_1 \quad , \quad (70)$$

$$H_{int} = -\frac{1}{\sqrt{N}} (G b_1^* b_0 + G^* b_0^* b_1) \quad ,$$

where μ 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.

First we introduce the spin operators

$$S_z = b_0^* b_0 - b_1^* b_1 = \sum_i (b_{0i}^* b_{0i} - b_{1i}^* b_{1i}) = \sum_i s_{zi} \quad ,$$

$$S_+ = b_0^* b_1 = \sum_i b_{0i}^* b_{1i} = \sum_i s_{+i} \quad , \quad (71)$$

$$S_- = b_1^* b_0 = \sum_i b_{1i}^* b_{0i} = \sum_i s_{-i} \quad ,$$

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

$$tr_b \exp (-\beta H_s - \beta H_{int}) = e^{-\beta \hbar\omega_0 N/2} (tr e^{\mathbf{h}\mathbf{s}})^N \quad , \quad (72)$$

where

$$h_x = \frac{\beta}{2\sqrt{N}} (G^* + G) \quad , \quad h_y = \frac{i\beta}{2\sqrt{N}} (G^* - G) \quad , \quad h_z = \beta \hbar\omega_0 / 2 \quad . \quad (73)$$

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

$$Z = e^{-\beta \hbar\omega_0 (N+s)/2} tr \left\{ e^{-\beta \hbar\omega_0 \sum_{\mu} a_{\mu}^* a_{\mu}} \left[2 \cosh \beta (G^* G / N + \hbar^2 \omega_0^2 / 4)^{1/2} \right]^N \right\} \quad , \quad (74)$$

⁹Y. K. Wang and F. T. Hioe, *Phys. Rev.* **A7** 831 (1973); S. Sivasubramanian, A. Widom and Y. N. Srivastava, *Int. J. Mod. Phys.* **B15** 537 (2001); S. Sivasubramanian, A. Widom and Y. N. Srivastava, *Mod. Phys.* **B16** 1201 (2002); S. Sivasubramanian, A. Widom and Y. N. Srivastava, *J. Phys.: Cond. Matt.* **15** 1109 (2003).

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

$$G_0^2 = \sum_{\mu} g_{\mu}^2 = \frac{2\pi\hbar}{V\omega_0} J_{01}^2 \sum_{\mu} N^2(\mu)/N, \quad (75)$$

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 (49) (for $s = 12$). We keep now in the partition function only the contributions which are relevant in the thermodynamical limit, and get

$$Z \simeq e^{-\beta\hbar\omega_0 N/2} \text{tr} \left\{ e^{-\beta\hbar\omega_0 c^* c + N \ln \left[2 \cosh \beta (g^2 c^* c / N + \hbar^2 \omega_0^2 / 4)^{1/2} \right]} \right\} \quad (76)$$

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

$$Z \simeq e^{-\beta\hbar\omega_0 N/2} \int_0^{\infty} dx \cdot e^{-N\phi(x)}, \quad (77)$$

where

$$\phi(x) = \beta\hbar\omega_0 x - \ln \left[2 \cosh \beta (g^2 x + \hbar^2 \omega_0^2 / 4)^{1/2} \right]. \quad (78)$$

The main contribution to the integral in equation (77) comes from the minimum value of the function $\phi(x)$ (Laplace's method), located at x_0 given by

$$\frac{2\hbar\omega_0}{g^2} \sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4} = \tanh \beta \sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4}. \quad (79)$$

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 (79) 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

$$f_0 = \hbar\omega_0 / 2 - \beta^{-1} \ln [2 \cosh \beta \hbar\omega_0 / 2] \quad (80)$$

(interaction-free ensemble). For T slightly below T_c we expand equation (79) in powers of $\sqrt{g^2 x_0 + \hbar^2 \omega_0^2 / 4} - \hbar\omega_0 / 2$ and get

$$x_0 \simeq \frac{1}{2} (1 - T/T_c)^{1/2}. \quad (81)$$

Now it is easy to get the free energy per particle

$$f \simeq f_0 - \frac{\hbar\omega_0}{4} (1 - T/T_c)^2. \quad (82)$$

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 (81) 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 $g > \hbar\omega_0$.

Miscellanea.

Plasmons. We consider an ensemble of identical particles with mass m and charge q moving against a rigid, neutralizing background, as in a jellium model. The small disturbances of particles density are represented as $\delta n = -n \text{div} \mathbf{u}$, where n is the particles concentration and \mathbf{u} is a local displacement field. The hamiltonian of these density disturbances reads

$$H_p = \frac{nm}{2} \int d\mathbf{r} \dot{\mathbf{u}}^2(\mathbf{r}) + \frac{n^2}{2} \int d\mathbf{r} d\mathbf{r}' \varphi(\mathbf{r} - \mathbf{r}') \text{div} \mathbf{u}(\mathbf{r}) \text{div} \mathbf{u}(\mathbf{r}') , \quad (83)$$

where $\varphi(\mathbf{r}) = q^2/r$ is the Coulomb interaction. We introduce the Fourier transform

$$\mathbf{u}(r) = \frac{1}{\sqrt{V}} \sum_{\mathbf{k}} \mathbf{u}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}} , \quad (84)$$

$\mathbf{u}^*(-\mathbf{k}) = \mathbf{u}(\mathbf{k})$ and notice that only the longitudinal coordinates are relevant. Therefore, we use $\mathbf{u}(\mathbf{k}) = \mathbf{k}u(\mathbf{k})/k$, $u^*(-\mathbf{k}) = -u(\mathbf{k})$ and get

$$H_p = -\frac{nm}{2} \sum_{\mathbf{k}} \dot{u}(\mathbf{k}) \dot{u}(-\mathbf{k}) - 2\pi n^2 q^2 \sum_{\mathbf{k}} u(\mathbf{k}) u(-\mathbf{k}) , \quad (85)$$

which leads to the equation of motion

$$\ddot{u}(\mathbf{k}) + \omega_p^2 u(\mathbf{k}) = 0 , \quad (86)$$

where ω_p given by $\omega_p^2 = 4\pi n q^2 / m$ is the plasma frequency.

The quantization of the plasma hamiltonian proceeds in the usual way, by introducing the creation and annihilation boson-like operators $b^*(\mathbf{k})$, $b(\mathbf{k})$ through

$$\begin{aligned} u(\mathbf{k}) &= i\sqrt{\hbar/2nm\omega_p} (b_{\mathbf{k}} + b_{-\mathbf{k}}^*) , \\ \dot{u}(\mathbf{k}) &= -\sqrt{\hbar\omega_p/2nm} (b_{\mathbf{k}} - b_{-\mathbf{k}}^*) , \end{aligned} \quad (87)$$

which gives the hamiltonian

$$H_p = \frac{1}{2} \hbar\omega_p \sum_{\mathbf{k}} (b_{\mathbf{k}}^* b_{\mathbf{k}} + b_{\mathbf{k}} b_{\mathbf{k}}^*) . \quad (88)$$

We write down here also the lagrangian

$$\begin{aligned} L_p &= \frac{\hbar}{4\omega_p} \sum_{\mathbf{k}} \left(\dot{b}_{\mathbf{k}} \dot{b}_{-\mathbf{k}} + \dot{b}_{\mathbf{k}} \dot{b}_{\mathbf{k}}^* + \dot{b}_{-\mathbf{k}}^* \dot{b}_{-\mathbf{k}} + \dot{b}_{-\mathbf{k}}^* \dot{b}_{\mathbf{k}}^* \right) - \\ &\quad - \frac{\hbar\omega_p}{4} \sum_{\mathbf{k}} (b_{\mathbf{k}} b_{-\mathbf{k}} + b_{\mathbf{k}} b_{\mathbf{k}}^* + b_{-\mathbf{k}}^* b_{-\mathbf{k}} + b_{-\mathbf{k}}^* b_{\mathbf{k}}^*) , \end{aligned} \quad (89)$$

which leads to the equation of motion (86) written as

$$\ddot{b}_{\mathbf{k}} + \ddot{b}_{-\mathbf{k}}^* + \omega_p^2 (b_{\mathbf{k}} + b_{-\mathbf{k}}^*) = 0 . \quad (90)$$

The current density associated with the plasmons is $\mathbf{j} = nq\dot{\mathbf{u}}$. Obviously, there is no coupling of the form $\mathbf{j}\mathbf{A}$, between plasmons and photons, since \mathbf{j} is longitudinal and \mathbf{A} is transversal with respect to the wavevector \mathbf{k} , as it is well-known.

One may imagine a plasmonic model of atoms, molecules, molecular aggregates, much along the lines sketched above. Then, the particle states are indeed the plasmon excitations; the coupling is of the form abb^* , as derived in the preceding sections, and the relevant particles states are the ground-state and the excited plasma state, which would imply $\omega_0 = \omega_p$. The theory described afore applies, but it is merely a reformulation of the coupling through the current J in terms of the coupling through the plasma frequency. They also agree in order of magnitude.

Phonons. We may note that a coupling of the form ab is possible between transverse optical phonons in a crystal lattice and photons, where a stands for photons and b for the phonon states. The associated coherent state, which involves a macroscopic occupation of the photons and phonons states amounts to a static transverse distortion of the crystal lattice. Such a state was not yet investigated so far, to the knowledge of the present author.

An interesting situation occurs in the electron-phonon coupling in one dimension, or in layered structures with so-called nesting Fermi surface. There, the electron states are boson-like, according to the Bloch-Tomonaga theory, and we may be in the situation of a coherent state with ab -like coupling, where a corresponds to phonons and b to (bosonized) electronic states. This state is nothing else but the well-known Peierls distortion and Frohlich charge density.

Superfluidity and superconductivity. The coherence of the Bose particles like He^4 atoms in the superfluid state is well-known. We must stress here however that this coherence refers to the motion of the particles, in contrast to the internal motion of particles described herein. The former is quite possible, and is related to quite interesting phenomena related to the coherent scattering.

As regards the superconductivity, the phonon-mediated electronic pairs certainly exhibit a macroscopic occupation, as approximate boson-like excitations. However, to the knowledge of this author, there is yet no field mediating the pair interaction, such as to resemble the coherent theory described herein. Other various speculations regarding the high- T_c superconductivity or the cold fusion (mediated, supposedly, by an atomic plasma)¹⁰ are even farther away from any resemblance to a coherent state as described herein.

Conclusion. In conclusion we may say that the interaction of matter with electromagnetic interaction may lead to coherence domains, 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 (45). 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 phase 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 (52) become then matricial equations, and getting their solution is a more difficult task.

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¹⁰See G. Preaprata, *loc cit*