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Microscopic basis of thermal superradiance

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Abstract

Electromagnetic superradiant field coherence exists in a condensed matter system if the electromagnetic field oscillators undergo a mean displacement. Transitions into thermal states with ordered superradiant phases have been shown to theoretically exist in Dicke–Preparata models. The theoretical validity of these models for condensed matter has been called into question due to non-relativistic diamagnetic terms in the electronic Hamiltonian. The microscopic bases of Dicke–Preparata thermal superradiance for realistic macroscopic systems are explored in this work. The impossibility of 'diaelectric' correlations in condensed matter systems (via the Landau–Lifshitz theorem) provides a strong theoretical basis for understanding the physical reality of condensed matter thermodynamic superradiant phases.

1. Introduction

In quantum electrodynamic (QED) theory, the magnetic field $\boldsymbol{B}$ and the Maxwell displacement field $\boldsymbol{D}$ are non-commuting quantum fields; i.e. in Gaussian units

$$[D_i(\boldsymbol{r}), B_j(\boldsymbol{r}')] = -4\pi \mathrm{i}\hbar c \epsilon_{ijk} \partial_k \delta(\boldsymbol{r} - \boldsymbol{r}'). \tag{1}$$

The electromagnetic field in a condensed matter system is said to be coherent if and only if (on average) $\overline{\boldsymbol{B}} \neq 0$ or $\overline{\boldsymbol{D}} \neq 0$ or both situations hold true. In terms of the electromagnetic oscillator modes, there is QED coherence [1] when one or more of the long wavelength field oscillators exhibit a spontaneous non-zero mean displacement (even in the absence externally applied fields). Ferromagnets and ferroelectrics clearly exhibit such spontaneous electromagnetic field ordering on a macroscopic scale. Microscopic QED coherence may also occur on the smaller length scales of atomic and molecular physics as well as on the length scale of mesoscopic coherent domains. For example, atomic or molecular magnetic moments clearly exhibit finite magnetic fields $\overline{\boldsymbol{B}}(\boldsymbol{r}) \neq 0$ in a spatial domain surrounding the moments. Dicke models of

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thermal superradiance have been extensively studied [2–23] and constitute an important class of ordered electromagnetic field coherent states.

The Dicke Hamiltonian [24] for a single photon oscillator may be written in the form

$$H_{Dicke} = \tfrac{1}{2}(P^2 + \omega_\infty^2 Q^2) - f\,Q + H_{electronic}. \tag{2}$$

The first term on the right-hand side of equation (2) represents a single photon oscillator mode. The electronic states are described by the Hamiltonian $H_{electronic}$. In the Dicke model, $H_{electronic}$ describes a set of two (energy) level molecules. The oscillator force operator f is determined by the electronic dipole moment operators of the molecules. The existence of a superradiant phase transition has been rigorously proven [25, 26] for the strong coupling Dicke Hamiltonian in equation (2). The superradiant phase transition may be understood by computing the shift in the photon oscillator frequency from ω_∞ to ω_0 due to the dipole interaction. To see how this comes about, consider the propagator for the oscillator coordinate

$$\mathcal{D}(\zeta) = \frac{\mathrm{i}}{\hbar}\int_0^\infty \mathrm{e}^{\mathrm{i}\zeta t}\langle[Q(t), Q(0)]\rangle\,\mathrm{d}t \qquad \text{where } \mathrm{Im}(\zeta) > 0. \tag{3}$$

If there were no interaction between the oscillator and the electronic degrees of freedom, then the propagator would have the form

$$\mathcal{D}_0(\zeta) = \left(\frac{1}{\omega_\infty^2 - \zeta^2}\right) \qquad \text{so that } \mathcal{D}_0(0) = \frac{1}{\omega_\infty^2}. \tag{4}$$

When there is coupling between the oscillator and the electronic degrees of freedom, the resulting damping function $\Gamma(\omega)$ introduces into the propagator a 'polarization part'

$$\Pi(\zeta) = \frac{2}{\pi}\int_0^\infty \left(\frac{\omega^2\Gamma(\omega)}{\omega^2 - \zeta^2}\right)\mathrm{d}\omega \tag{5}$$

which describes both damping and oscillator frequency shift information; i.e. the full propagator reads

$$\mathcal{D}(\zeta) = \left(\frac{1}{\omega_\infty^2 - \zeta^2 - \Pi(\zeta)}\right) \qquad \text{so that } \mathcal{D}(0) = \frac{1}{\omega_0^2}. \tag{6}$$

The renormalized frequency ω_0 is determined by the oscillator strength sum rule [25]

$$\omega_\infty^2 = \omega_0^2 + \frac{2}{\pi}\int_0^\infty \Gamma(\omega)\,\mathrm{d}\omega \qquad \text{where } \Gamma(\omega) \geqslant 0. \tag{7}$$

The oscillator-frequency-dependent damping function, denoted by $\Gamma(\omega)$ and caused by the random force f, lowers the square of the frequency from ω_∞^2 to ω_0^2. If the damping is sufficiently strong, then $\omega_0^2 < 0$ and the oscillator becomes unstable about the old equilibrium position $\bar{Q}_{old} = 0$. A new equilibrium position is reached $\bar{Q}_{new} \neq 0$ which describes the superradiant ordered state. There is little doubt that strong damping can produce a superradiant ordered phase if the Dicke Hamiltonian is employed.

Nevertheless, the validity of the Dicke model Hamiltonian as a description of real condensed matter systems has been brought into question [27–31]. At the centre of the objections is the notion that the conventional non-relativistic electronic Hamiltonian [32–35] has both linear and quadratic terms in the vector potential $\boldsymbol{A}$. Some have asserted [27–29] that the quadratic terms in $\boldsymbol{A}$ stabilize the oscillator and thereby eliminate a phase transition into a coherent superradiant state. In a gauge invariant form, the terms quadratic in $\boldsymbol{A}$ are physically terms quadratic in the magnetic field $\boldsymbol{B} = \mathrm{curl}\,\boldsymbol{A}$, i.e. the diamagnetic terms. Diamagnetic inequalities have been invoked which claim [31] to prove the absence of the oscillator instability. Quite apart from the fact that the rigorous diamagnetic inequality has been proved only for spin zero bosons [36] and *not* for spin one half electrons, the fact that diamagnetism does

sometimes occur in electronic systems by no means implies that all oscillator modes are stable. The essential and physically correct ideas have been explored by Landau and Lifshitz [37]. The conventional QED interaction Hamiltonian does indeed contain terms quadratic in the magnetic field $\boldsymbol{B}$ but *only linear interaction terms* couple into the Maxwell displacement field $\boldsymbol{D}$. It is this *electric, strictly linear coupling* which is taken realistically into account in Dicke–Preparata models. The superradiant ordered phases in the Maxwell displacement field ($\overline{\boldsymbol{D}} \neq 0$) are *not* adversely affected by weak diamagnetic couplings since the coupling into $\boldsymbol{D}$ remains strictly linear.

Based on the above physical arguments and the nature of the linear and quadratic couplings, Landau and Lifshitz proved the following important theorem for materials described by a dielectric constant ε (wherein $\overline{\boldsymbol{D}} = \varepsilon\overline{\boldsymbol{E}}$) and a magnetic permeability μ (wherein $\overline{\boldsymbol{B}} = \mu\overline{\boldsymbol{H}}$):

Theorem 1. *The magnetic permeability* $\mu > 0$ *can be either paramagnetic* $\mu > 1$ *or diamagnetic* $\mu < 1$. *The dielectric constant must be paraelectric* $\varepsilon > 1$. *There is no 'diaelectricity' in condensed matter systems, i.e. the range* $1 > \varepsilon > 0$ *is strictly forbidden.*

The above theorem and the generalizations to be discussed below provide a strong theoretical basis for the physical reality of the thermal superradiant phase.

Our purpose is to discuss the theoretical microscopic bases of Dicke–Preparata models as applied to realistic macroscopic condensed matter systems. In particular, we shall examine in detail the linear (in $\boldsymbol{D}$ and $\boldsymbol{B}$) and quadratic (only in $\boldsymbol{B}$) terms in the interaction. In section 2 we describe the conventional QED Hamiltonian. The electronic degrees of freedom are considered to be non-relativistic. A canonical transformation is introduced which allows for a precise microscopic definition for the transverse Maxwell displacement field $\boldsymbol{D} = \boldsymbol{E} + 4\pi\boldsymbol{P}$. In section 3 the notion of diabatic and adiabatic changes in electrodynamic processes will be defined. Dissipation is introduced as a diabatic (i.e. non-adiabatic) process. In section 4, the statistical thermodynamics of the electric dipole moment interactions will be discussed and a generalization of the Landau–Lifshitz theorem 1 will be proved. The diabatic dissipation will be related to the thermodynamic dielectric response of the medium. In section 5, it will be shown how the instability of the thermodynamic dielectric response function is the signature of a transition into a superradiant phase.

2. QED Hamiltonian

If we choose a vector potential in the Coulomb gauge,

$$\boldsymbol{B} = \operatorname{curl}\boldsymbol{A}, \qquad \operatorname{div}\boldsymbol{A} = 0, \tag{8}$$

then the QED Hamiltonian of interest in the work which follows has the form

$$H_{QED} = \frac{1}{8\pi}\int (|\boldsymbol{E}'|^2 + |\boldsymbol{B}|^2)\,\mathrm{d}^3\boldsymbol{r} + H[\boldsymbol{A}]. \tag{9}$$

$\boldsymbol{E}'$ is the operator which denotes the *transverse part* of the electric field. When operating on a wavefunction Ψ, the transverse electric field operator may be written as

$$\boldsymbol{E}'(\boldsymbol{r})\Psi = 4\pi\mathrm{i}\hbar c\left(\frac{\delta\Psi}{\delta\boldsymbol{A}(\boldsymbol{r})}\right) = 4\pi\mathrm{i}\hbar c\operatorname{curl}\left(\frac{\delta\Psi}{\delta\boldsymbol{B}(\boldsymbol{r})}\right). \tag{10}$$

$H[\boldsymbol{A}]$ denotes the Hamiltonian (including Coulomb interactions) of the charged particles. The Coulomb Hamiltonian when $\boldsymbol{B} = 0$ has the form

$$H[\boldsymbol{A} = 0] = H_{Coul} = -\sum_j \frac{\hbar^2}{2m}\Delta_j - \sum_a \frac{\hbar^2}{2M_a}\Delta_a + U, \tag{11}$$

where the total potential energy for the electrons and nuclei in the condensed matter system is given by

$$U = e^2\left(\sum_{j<k}\frac{1}{r_{jk}} + \sum_{a<b}\frac{Z_a Z_b}{R_{ab}} - \sum_{ja}\frac{Z_a}{|\boldsymbol{r}_j - \boldsymbol{R}_a|}\right). \tag{12}$$

For $\boldsymbol{A} \neq 0$,

$$H[\boldsymbol{A}] = -\sum_j \frac{(\hbar c\nabla_j - \mathrm{i}e\boldsymbol{A}_j)^2}{2mc^2} - \sum_a \frac{(\hbar c\nabla_a - \mathrm{i}Z_a|e|\boldsymbol{A}_a)^2}{2M_a c^2} + H_s + U, \tag{13}$$

where $\boldsymbol{A}_j = \boldsymbol{A}(\boldsymbol{r}_j)$, $\boldsymbol{A}_a = \boldsymbol{A}(\boldsymbol{R}_a)$ and the interaction of the magnetic field with the particle spins is given by

$$H_s = -\left(\frac{ge}{2mc}\right)\sum_j \boldsymbol{s}_j \cdot \boldsymbol{B}(\boldsymbol{r}_j) - \sum_a\left(\frac{g_a Z_a|e|}{2M_a c}\right)\boldsymbol{S}_a \cdot \boldsymbol{B}(\boldsymbol{R}_a). \tag{14}$$

Let us consider a basis in which the Coulomb Hamiltonian is diagonal; i.e.

$$H[\boldsymbol{A}=0]\psi_j = H_{Coul}\psi_j = W_j[\boldsymbol{A}=0]\psi_j \qquad \text{where } j = 0, 1, 2, \ldots. \tag{15}$$

In the Coulomb basis, one may define the matrix elements

$$H_{jk}[\boldsymbol{A}] = (\psi_k, H[\boldsymbol{A}]\psi_j) \tag{16}$$

so that the Hamiltonian in equation (13) has the equivalent matrix representation

$$H[\boldsymbol{A}] = \begin{pmatrix} H_{00}[\boldsymbol{A}] & H_{01}[\boldsymbol{A}] & H_{02}[\boldsymbol{A}] & \ldots \\ H_{10}[\boldsymbol{A}] & H_{11}[\boldsymbol{A}] & H_{12}[\boldsymbol{A}] & \ldots \\ H_{20}[\boldsymbol{A}] & H_{21}[\boldsymbol{A}] & H_{22}[\boldsymbol{A}] & \ldots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \tag{17}$$

In principle, the Hamiltonian can be brought to diagonal form by a unitary transformation

$$W[\boldsymbol{A}] = U^{\dagger}[\boldsymbol{A}]H[\boldsymbol{A}]U[\boldsymbol{A}] \qquad \text{where } U^{\dagger}[\boldsymbol{A}] = U^{-1}[\boldsymbol{A}] \tag{18}$$

and

$$W[\boldsymbol{A}] = \begin{pmatrix} W_0[\boldsymbol{A}] & 0 & 0 & \ldots \\ 0 & W_1[\boldsymbol{A}] & 0 & \ldots \\ 0 & 0 & W_2[\boldsymbol{A}] & \ldots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \tag{19}$$

In virtue of gauge invariance

$$W[\boldsymbol{A}] \equiv W[\boldsymbol{B}]. \tag{20}$$

The unitary transformation can be employed to transform the total Hamiltonian in equation (9) into the adiabatic representation

$$\mathcal{H} = U^{\dagger}[\boldsymbol{A}]H_{QED}U[\boldsymbol{A}]. \tag{21}$$

When acting on the electric field in equation (10), the transformation defines the Maxwell displacement field $\boldsymbol{D}$ and polarization $\boldsymbol{P}$ via

$$U^{\dagger}[\boldsymbol{A}]\boldsymbol{E}'U[\boldsymbol{A}] = \boldsymbol{E} = \boldsymbol{D} - 4\pi\boldsymbol{P}, \tag{22}$$

where

$$\boldsymbol{D}(\boldsymbol{r}) = 4\pi\mathrm{i}\hbar c\left(\frac{\delta}{\delta\boldsymbol{A}(\boldsymbol{r})}\right) = 4\pi\mathrm{i}\hbar c\,\mathrm{curl}\left(\frac{\delta}{\delta\boldsymbol{B}(\boldsymbol{r})}\right), \tag{23}$$

and

$$\boldsymbol{P}(\boldsymbol{r}) = -\mathrm{i}\hbar c U^{\dagger}[\boldsymbol{A}]\left(\frac{\delta U[\boldsymbol{A}]}{\delta \boldsymbol{A}(\boldsymbol{r})}\right) = \mathrm{i}\hbar c\left(\frac{\delta U^{\dagger}[\boldsymbol{A}]}{\delta \boldsymbol{A}(\boldsymbol{r})}\right)U[\boldsymbol{A}]. \tag{24}$$

Equations (9), (10) and (21)–(24) imply the adiabatic Hamiltonian representation

$$\mathcal{H} = \frac{1}{8\pi}\int (|\boldsymbol{D} - 4\pi\boldsymbol{P}|^2 + |\boldsymbol{B}|^2)\,\mathrm{d}^3\boldsymbol{r} + W[\boldsymbol{A}], \tag{25}$$

which may be conveniently written as

$$\mathcal{H} = \frac{1}{8\pi}\int |\boldsymbol{D}|^2\,\mathrm{d}^3\boldsymbol{r} - \int \boldsymbol{D}\cdot\boldsymbol{P}\,\mathrm{d}^3\boldsymbol{r} + H'[\boldsymbol{A}]. \tag{26}$$

In equation (26),

$$H'[\boldsymbol{A}] = \frac{1}{8\pi}\int |\boldsymbol{B}|^2\,\mathrm{d}^3\boldsymbol{r} + W[\boldsymbol{A}] + 2\pi\int |\boldsymbol{P}|^2\,\mathrm{d}^3\boldsymbol{r}. \tag{27}$$

Finally, the transverse current operator in the adiabatic representation of equations (18) and (19) is given by

$$\boldsymbol{J}(\boldsymbol{r}) = -cU^{\dagger}[\boldsymbol{A}]\frac{\delta H[\boldsymbol{A}]}{\delta \boldsymbol{A}(\boldsymbol{r})}U[\boldsymbol{A}]. \tag{28}$$

The notion of 'diabatic damping' is associated with a decomposition of the current equation (28) into adiabatic and non-adiabatic parts.

3. Damping and electrical conductivity

For the unitary transformation in equation (18) we have the differential identity

$$U^{\dagger}\frac{\delta H}{\delta \boldsymbol{A}}U = \frac{\delta}{\delta \boldsymbol{A}}(U^{\dagger}HU) - U^{\dagger}HUU^{\dagger}\frac{\delta U}{\delta \boldsymbol{A}} - \frac{\delta U^{\dagger}}{\delta \boldsymbol{A}}UU^{\dagger}HU, \tag{29}$$

which together with equations (24) and (28) yields the current decomposition

$$\boldsymbol{J}(\boldsymbol{r}) = -c\frac{\delta W[\boldsymbol{A}]}{\delta \boldsymbol{A}(\boldsymbol{r})} + \frac{\mathrm{i}}{\hbar}[W[\boldsymbol{A}], \boldsymbol{P}(\boldsymbol{r})]. \tag{30}$$

One may employ the gauge invariant $\boldsymbol{B} = \mathrm{curl}\,\boldsymbol{A}$ in the form (see equation (20)) of an adiabatic current

$$\boldsymbol{J}_{adiabatic}(\boldsymbol{r}) = -c\frac{\delta W[\boldsymbol{A}]}{\delta \boldsymbol{A}(\boldsymbol{r})} = -c\,\mathrm{curl}\frac{\delta W[\boldsymbol{B}]}{\delta \boldsymbol{B}(\boldsymbol{r})} = c\,\mathrm{curl}\,\boldsymbol{M}. \tag{31}$$

The gauge invariant adiabatic magnetization has a form which follows from equations (19) and (31); i.e.

$$\boldsymbol{M} = -\begin{pmatrix} \delta W_0[\boldsymbol{B}]/\delta \boldsymbol{B} & 0 & 0 & \dots \\ 0 & \delta W_1[\boldsymbol{B}]/\delta \boldsymbol{B} & 0 & \dots \\ 0 & 0 & \delta W_2[\boldsymbol{B}]/\delta \boldsymbol{B} & \dots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \tag{32}$$

The dissipative (or 'diabatic') part of the current operator is given by

$$\boldsymbol{J}_d(\boldsymbol{r}) = \frac{\mathrm{i}}{\hbar}[W, \boldsymbol{P}(\boldsymbol{r})] = \dot{\boldsymbol{P}}(\boldsymbol{r}). \tag{33}$$

Equation (30) then takes the conventional form having both magnetization and polarization parts; i.e.

$$\boldsymbol{J}(\boldsymbol{r}) = c\,\mathrm{curl}\boldsymbol{M}(\boldsymbol{r}) + \dot{\boldsymbol{P}}(\boldsymbol{r}). \tag{34}$$

Although equation (34) is well known in classical electrodynamics, we have given the proof from a fully QED viewpoint.

The diabatic part of the current in equation (33) describes the dissipation via the non-local transverse electrical conductivity tensor $\sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+)$. The microscopic expression for the transverse conductivity is determined by the fluctuation dissipation theorem

$$\sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta) = \frac{1}{\hbar} \int_0^\beta \int_0^\infty \mathrm{e}^{\mathrm{i}\zeta t} \langle J_{d,j}(\boldsymbol{r}', -\mathrm{i}\lambda) J_{d,i}(\boldsymbol{r}, t) \rangle \, \mathrm{d}t \, \mathrm{d}\lambda. \tag{35}$$

Note

(i) The complex frequency obeys Im $\zeta > 0$.
(ii) The thermal average $\langle \cdots \rangle$ is over the charged particle degrees of freedom.
(iii) The time variation of operators in equation (35) employs the Hamiltonian $H'[\boldsymbol{A}]$ in equation (27).
(iv) Finally,

$$\beta = \left(\frac{\hbar}{k_B T} \right). \tag{36}$$

For the transverse dielectric properties of charged particles, one employs the electric susceptibility

$$\chi_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta) = \frac{\mathrm{i}}{\hbar} \int_0^\infty \mathrm{e}^{\mathrm{i}\zeta t} \langle [P_i(\boldsymbol{r}, t), P_j(\boldsymbol{r}', 0)] \rangle \, \mathrm{d}t. \tag{37}$$

Integrating equation (37) by parts yields

$$-\mathrm{i}\zeta \, \chi_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta) = h_{ij}(\boldsymbol{r}, \boldsymbol{r}') + \frac{\mathrm{i}}{\hbar} \int_0^\infty \mathrm{e}^{\mathrm{i}\zeta t} \langle [\dot{P}_i(\boldsymbol{r}, t), P_j(\boldsymbol{r}', 0)] \rangle \, \mathrm{d}t, \tag{38}$$

where the equal time commutator contribution is given by

$$h_{ij}(\boldsymbol{r}, \boldsymbol{r}') = \frac{\mathrm{i}}{\hbar} \langle [P_i(\boldsymbol{r}), P_j(\boldsymbol{r}')] \rangle. \tag{39}$$

Employing the Kubo–Martin–Schwinger [38–41] condition

$$\mathrm{i}\langle [\dot{P}_i(\boldsymbol{r}, t), P_j(\boldsymbol{r}', 0)] \rangle = \int_0^\beta \langle \dot{P}_j(\boldsymbol{r}', -\mathrm{i}\lambda) \dot{P}_i(\boldsymbol{r}, t) \rangle \, \mathrm{d}\lambda \tag{40}$$

and equation (33) in (35) yields a simple relationship between the conductivity and the susceptibility

$$-\mathrm{i}\zeta \, \chi_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta) = \sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta) + h_{ij}(\boldsymbol{r}, \boldsymbol{r}'). \tag{41}$$

The dissipative part of the conductivity is determined by

$$\mathrm{Re}\{\sigma_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+)\} = \omega \, \mathrm{Im}\{\chi_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+)\}. \tag{42}$$

The quantum mechanical Hall conductivity contribution $h_{ij}(\boldsymbol{r}, \boldsymbol{r}')$ from the transverse polarization is determined by the commutation relation in equation (39).

Note that the response functions discussed above are defined with respect to an applied magnetic field $\boldsymbol{B} = \mathrm{curl}\, \boldsymbol{A}$. Magneto-conductivity in σ_{ij} and the Faraday effect in χ_{ij} are implicitly included in the above considerations. For superradiance described by a coherent field $\overline{\boldsymbol{D}} \neq 0$ one averages over magnetic field fluctuations. The mean magnetic field (on macroscopic length scales) obeys $\overline{\boldsymbol{B}} = 0$. Let us now consider the details of the magnetic field fluctuations.

4. Statistical thermodynamics

The QED free energy F of a condensed matter system can be written as

$$\mathcal{G} = -k_B T \ln\{\mathrm{Tr}\, \mathrm{e}^{-\mathcal{H}/k_B T}\} \tag{43}$$

where $\mathcal{H}$ is given in equation (26). The complete trace over quantum states includes electromagnetic field degrees of freedom as well as charged particle degrees of freedom. Thus

$$\mathrm{Tr}\, \mathrm{e}^{-\mathcal{H}/k_B T} = \mathrm{Tr}_{(\boldsymbol{D},\boldsymbol{A})}(\mathrm{Tr}_{(Charged)}\, \mathrm{e}^{-\mathcal{H}/k_B T}). \tag{44}$$

If one treats the electromagnetic field trace in the quasi-classical limit of a functional integral

$$\mathrm{Tr}_{(\boldsymbol{D},\boldsymbol{A})}(\cdots) \to \int \prod_{\boldsymbol{r}} \left(\frac{\mathcal{D}\boldsymbol{D}(\boldsymbol{r})\mathcal{D}\boldsymbol{A}(\boldsymbol{r})}{8\pi\hbar c} \right)(\cdots), \tag{45}$$

then equations (26) and (43)–(45) imply the functional representation

$$\exp\left(-\frac{\mathcal{G}}{k_B T}\right) = \int \exp\left(-\frac{\mathcal{F}_{tot}[\boldsymbol{D}]}{k_B T}\right) \prod_{\boldsymbol{r}} (\mathcal{D}\boldsymbol{D}(\boldsymbol{r})). \tag{46}$$

The total free energy functional of the Maxwell displacement field obeys

$$\mathcal{F}_{tot}[\boldsymbol{D}] = \frac{1}{8\pi} \int |\boldsymbol{D}|^2 \, \mathrm{d}^3\boldsymbol{r} + \mathcal{F}[\boldsymbol{D}], \tag{47}$$

where

$$\exp\left(-\frac{\mathcal{F}[\boldsymbol{D}]}{k_B T}\right) = \mathrm{Tr}_{(Charged)} \int \exp\left(-\frac{H''[\boldsymbol{A}, \boldsymbol{D}]}{k_B T}\right) \prod_{\boldsymbol{r}} \left(\frac{\mathcal{D}\boldsymbol{A}(\boldsymbol{r})}{8\pi\hbar c} \right) \tag{48}$$

and

$$H''[\boldsymbol{A}, \boldsymbol{D}] = H'[\boldsymbol{A}] - \int \boldsymbol{P}(\boldsymbol{r}, [\boldsymbol{A}]) \cdot \boldsymbol{D}(\boldsymbol{r}) \, \mathrm{d}^3\boldsymbol{r}. \tag{49}$$

The coupling into the Maxwell displacement field in the Hamiltonian equation (49) is linear. Equations (48) and (49) imply that the free energy $\mathcal{F}[\boldsymbol{D}]$ is a convex upward functional. For the second functional derivative

$$\tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}]) = -\left(\frac{\delta^2 \mathcal{F}[\boldsymbol{D}]}{\delta D_i(\boldsymbol{r}) \delta D_j(\boldsymbol{r}')} \right) \tag{50}$$

we have the following:

Theorem 2. *The spectrum of the zero frequency susceptibility* $\{\tilde{\chi}_\lambda[\boldsymbol{D}]\}$ *defined by the eigenvalue equation*

$$\sum_j \int \tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}]) \xi^j_\lambda(\boldsymbol{r}') \, \mathrm{d}^3\boldsymbol{r}' = \tilde{\chi}_\lambda[\boldsymbol{D}] \xi^i_\lambda(\boldsymbol{r})$$

obeys $\tilde{\chi}_\lambda[\boldsymbol{D}] \geqslant 0$ *for all* λ.

In order to prove a generalization of the Landau and Lifshitz theorem 1, one must investigate the second functional derivatives

$$\eta_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}]) = 4\pi \left(\frac{\delta^2 \mathcal{F}_{tot}[\boldsymbol{D}]}{\delta D_i(\boldsymbol{r}) \delta D_j(\boldsymbol{r}')} \right) = \Delta_{ij}(\boldsymbol{r} - \boldsymbol{r}') - 4\pi \tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}]), \tag{51}$$

where equation (47) has been invoked and the transverse delta function is defined as

$$\Delta_{ij}(\boldsymbol{r}) = \int \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\left(\delta_{ij} - \frac{k_i k_j}{|\boldsymbol{k}|^2}\right)\frac{\mathrm{d}^3\boldsymbol{k}}{(2\pi)^3},$$
$$= \left(\frac{2}{3}\right)\delta_{ij}\delta(\boldsymbol{r}) + \left(\frac{1}{4\pi}\right)\left(\frac{3r_i r_j - r^2\delta_{ij}}{r^5}\right). \tag{52}$$

The spectrum of the zero frequency transverse dielectric constants $\{\varepsilon_\lambda[\boldsymbol{D}]\}$ is defined by the eigenvalue equation

$$\sum_j \int \eta_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}])\xi_\lambda^j(\boldsymbol{r}')\,\mathrm{d}^3\boldsymbol{r}' = \left(\frac{1}{\varepsilon_\lambda[\boldsymbol{D}]}\right)\xi_\lambda^i(\boldsymbol{r}), \tag{53}$$

and obeys

$$\varepsilon_\lambda[\boldsymbol{D}] = \left[\frac{1}{1 - 4\pi\,\tilde{\chi}_\lambda[\boldsymbol{D}]}\right]. \tag{54}$$

The thermodynamic stability condition for the free energy $\mathcal{F}_{tot}[\boldsymbol{D}]$ can be summarized by the following.

Theorem 3. *Thermodynamic stability for a Maxwell displacement field* $\boldsymbol{D}$ *requires that* $\varepsilon_\lambda[\boldsymbol{D}] > 0$ *for all* λ.

Proof. For a Maxwell field $\boldsymbol{D}$ to represent thermal equilibrium, one expects the free energy $\mathcal{F}_{tot}[\boldsymbol{D}]$ to be at a minimum. The second (functional) derivative conditions for achieving a thermodynamic free energy minimum are those stated in the theorem in virtue of equations (51) and (53). □

The central result of this section is the following *generalization* of the Landau–Lifshitz theorem 1.

Theorem 4. *Thermodynamic stability for a Maxwell displacement field* $\boldsymbol{D}$ *requires the paraelectric inequality* $\varepsilon_\lambda[\boldsymbol{D}] > 1$ *for all* λ. *Diaelectric behaviour* $(0 < \varepsilon_\lambda[\boldsymbol{D}] < 1)$ *is strictly forbidden.*

Proof. Theorem 4 follows directly from theorem 2, equation (54) and theorem 3. □

We now reconsider the dissipative properties of a condensed matter system *after* coherent averaging over magnetic field $\boldsymbol{B} = \mathrm{curl}\,\boldsymbol{A}$ fluctuations. We employ the functional averaging measure $\int(\cdots)\prod_r(\mathcal{D}\boldsymbol{A}/8\pi\hbar c)$ as in equations (47) and (48). For example, the new susceptibility

$$\tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', \zeta, [\boldsymbol{D}]) = \frac{\mathrm{i}}{\hbar}\int_0^\infty \mathrm{e}^{\mathrm{i}\zeta t}\langle\langle[P_i(\boldsymbol{r}, t),\, P_j(\boldsymbol{r}', 0)]\rangle\rangle\,\mathrm{d}t, \tag{55}$$

where the complete 'double averaging' $\langle\langle\cdots\rangle\rangle$ is over both charged particle motions and magnetic field fluctuations with the Maxwell displacement field $\boldsymbol{D}$ held fixed. The time-dependent operators in equation (55) are with respect to the Hamiltonian $H''[\boldsymbol{A}, \boldsymbol{D}]$ defined in equation (49). The dissipative motions of the condensed matter system are then described by

$$\mathrm{Re}\{\tilde{\sigma}_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+, [\boldsymbol{D}])\} = \omega\,\mathrm{Im}\{\tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+, [\boldsymbol{D}])\}. \tag{56}$$

Finally, the static susceptibility in equation (50) is the zero frequency limit

$$\tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', [\boldsymbol{D}]) \equiv \lim_{\omega\to 0}\mathrm{Re}\{\tilde{\chi}_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega + \mathrm{i}0^+, [\boldsymbol{D}])\}. \tag{57}$$

The thermodynamic stability test for a coherent electromagnetic superradiant state follows from theorem 2, equation (54) and theorem 4.

5. Thermodynamic stability

In the absence of external electric fields, the stable values of $\boldsymbol{D}$ are such as to minimize [37] the total free energy $\mathcal{F}_{tot}[\boldsymbol{D}]$. The free energy minimization condition implies a vanishing thermal electric field;

$$\boldsymbol{E}(\boldsymbol{r}) = 4\pi\left(\frac{\delta\mathcal{F}_{tot}[\boldsymbol{D}]}{\delta\boldsymbol{D}(\boldsymbol{r})}\right) = \boldsymbol{D}(\boldsymbol{r}) - 4\pi\langle\langle\boldsymbol{P}(\boldsymbol{r})\rangle\rangle = 0. \tag{58}$$

The first derivative equation (58) will in general have more than one possible solution with a non-zero Maxwell displacement field $[\boldsymbol{D}]$. The multiple solutions correspond to differing possibilities for superradiant coherent domains. A necessary condition for a true free energy minimum has been proved in theorem 4. Employing equation (54) we find the stability condition

$$0 < 4\pi\tilde{\chi}_\lambda[\boldsymbol{D}] < 1. \tag{59}$$

From (i) equation (55), (ii) the resulting dispersion relation

$$\tilde{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}',\zeta,[\boldsymbol{D}]) = \frac{2}{\pi}\int_0^\infty \frac{\mathrm{Im}\ \tilde{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}',\omega+\mathrm{i}0^+,[\boldsymbol{D}])\,\mathrm{d}\omega}{\omega^2-\zeta^2}$$

$$\tilde{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}',[\boldsymbol{D}]) \equiv \lim_{\zeta\to 0}\tilde{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}',\zeta,[\boldsymbol{D}]) = \frac{2}{\pi}\int_0^\infty \mathrm{Im}\ \tilde{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}',\omega+\mathrm{i}0^+,[\boldsymbol{D}])\frac{\mathrm{d}\omega}{\omega}, \tag{60}$$

(iii) theorem 2 and (iv) the stability equation (59), it follows that strong dissipation, i.e. substantial $\{\mathrm{Im}\,\tilde{\chi}(\omega+\mathrm{i}0^+)/\omega\}$, tends to yield thermodynamic instabilities.

Symmetry under parity transformations yields the solution $\boldsymbol{D} = 0$ corresponding to a 'normal' phase. If $4\pi\tilde{\chi}_\lambda[\boldsymbol{D}=0] > 1$ for some λ, then the normal phase is unstable. The true free energy minimum will arise for coherent superradiant domains with $[\boldsymbol{D} \neq 0]$. There may be many possible superradiant domain configurations as discussed above. Domain walls and/or normal phase regions may separate the superradiant domains wherein $\boldsymbol{D} = 4\pi\langle\langle\boldsymbol{P}(\boldsymbol{r})\rangle\rangle \neq 0$.

6. Conclusions

We have discussed a generalization of the theorem by Landau and Lifshitz whereby $\varepsilon \geqslant 1$ for materials described by $\boldsymbol{D} = \varepsilon\boldsymbol{E}$. The theorem and its generalization is *independent* of magnetic permeability μ when the material also obeys $\boldsymbol{B} = \mu\boldsymbol{H}$. Magnetic properties may be either paramagnetic or diamagnetic, but diaelectric properties of matter are ruled out by quantum statistical mechanical stability considerations.

The stability criteria are crucial for the understanding of stable superradiant domains. In particular, for all of the possible eigenmodes of the dielectric susceptibility, the generalized Landau–Lifshitz stability condition $\varepsilon_\lambda[\boldsymbol{D}] \geqslant 1$ is crucial for testing whether (or not) a superradiant domain is thermodynamically stable. Contrary to what appeared some time ago in the literature [29], the dielectric stability conditions are independent of possible diamagnetism. No approximations to the free energy functional $\mathcal{F}_{tot}[\boldsymbol{D}]$ have been required to reach important general conclusions, however further approximations *are required* for concrete computation.

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