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Perturbation theory of super-radiance
I. Super-radiant emission

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Abstract. Perturbation theory is used to show that super-radiance as envisaged by Dicke, in which the radiation rate goes as N^2, the square of the number of atoms, can be interpreted in the same terms as classical diffraction theory. The rate goes as N^2 from samples small in three dimensions compared with a wavelength but goes essentially as N from larger samples. There is a natural distinction between coherent and incoherent contributions to the total radiation rate from larger samples. We contrast the two cases of large samples prepared in simple Dicke states and samples prepared (supposedly by an incident pulse) in Dicke states phased with a vector $\boldsymbol{k}$. The incoherent rates Γ_{inc} are essentially the same in the two cases: contributions to them are isotropic or almost isotropic. The coherent rates are different: the coherent rate for phased states simply exhibits features of propagating plane waves; the coherent rate from simple Dicke states describes directed spatially coherent emission controlled solely by the sample geometry. It does not follow in this case that Γ_{coh} necessarily dominates so that Γ_{inc} can be neglected. The coherent radiation rate may be trapped whilst in large enough samples Γ_{inc} simply exceeds Γ_{coh}. Dimensions for which $\Gamma_{inc} \simeq \Gamma_{coh}$ could define a maximum coherence length. However, contributions to Γ_{coh} are directed and there is no maximum coherence length for such directed emission. There is a limited amount of evidence in support of the view that a totally inverted dielectric evolves incoherently rather than coherently.

1. Introduction

The theory of the coherent spontaneous emission process now called super-radiance is due to Dicke (1954); for N two-level atoms on the same site the radiation rate is $N\Gamma_0$ if all atoms are excited but is $\frac{1}{4}N^2\Gamma_0$ if as many atoms are excited as unexcited: Γ_0 is the Einstein A coefficient. Comparable results are reported in a flood of recent papers (for example, Agarwal 1970, 1971a, b, 1973, Bonifacio and Preparata 1970, Bonifacio and Schwendimann 1970, Bonifacio *et al* 1971a, b, Bonifacio 1973, Bullough 1973). Despite this it is perhaps still not universally appreciated that the gross enhancement of the rate in the second case can be interpreted as the non-physical limit of an application of elementary diffraction theory; that enhancement is a small sample result already contained—albeit in one rather particular sense—in classical theory; and that it becomes substantially reduced in the case of macroscopic dielectrics. The time evolution of the emission process requires closed solutions to the full equation of motion; some have been reported (Bullough 1973) and we shall report others elsewhere. There is also a number of comparable solutions in the literature cited above but almost all of these are overtly or covertly small sample results, or else the number of modes is so restricted that it becomes difficult to assess the conclusions. For extended samples nobody has extracted all the information implicit in Dicke's original first-order perturbation theory. This paper reports the results of such a study. The argument is particularly simple and instructive and seems worth reporting if only for these reasons.

Both in the equation of motion and transition rate theories it proves useful to exhibit the photon propagator explicitly. We work in the dipole approximation so that in the simplest form in which atoms interact only through their radiation fields this proves to be the transverse part of the tensor propagator

$$\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) \equiv (\nabla\nabla + k_s^2 \mathbf{U}) \exp(\mathrm{i}k_s R) R^{-1}. \tag{1}$$

In this $\mathbf{U}$ is the unit tensor, $R = |\boldsymbol{x}-\boldsymbol{x}'|$ and the two-level atoms resonate at frequency $\omega_s = ck_s$. The atoms also interact through their Coulomb interactions $\nabla\nabla R^{-1}$ and this is included by using the whole propagator (1). This longitudinal Coulomb interaction does not appear in the radiation rates, but it shifts the energy levels of small partially inverted samples and it shifts the energy levels of the macroscopic partially inverted isotropic dielectrics we also consider by, amongst other things, a Lorentz field term. It is unimportant to order e^2 in the non-physical limit in which all atoms occupy the same site. This is because, as we shall show, in the following paper (part II) the limit as $R \to 0$ of such dipole–dipole interactions is not the self-interaction of point systems. The limits as $R \to 0$ of the small sample radiation rates are the rates obtained by Dicke, but the limits of the small sample level shifts are not the corresponding shifts for point systems.

The interaction is a sum of terms $-\boldsymbol{\mu}^{(i)} . \boldsymbol{e}(\boldsymbol{x}_i)$ where $\boldsymbol{\mu}^{(i)}$ is the dipole operator of the ith atom and $\boldsymbol{e}(\boldsymbol{x}_i)$ is the transverse field operator evaluated at that particular atomic site. The corresponding operator density $-\boldsymbol{\mu}(\boldsymbol{x}) . \boldsymbol{e}(\boldsymbol{x})$ is more convenient since these sites do not appear in the field operator $\boldsymbol{e}(\boldsymbol{x})$. This operator is the linear combination of annihilation and creation operators

$$\boldsymbol{e}(\boldsymbol{x}) = \mathrm{i}\left(\frac{2\pi\hbar}{V_q}\right)^{1/2} \sum_{\boldsymbol{k},\lambda} \hat{\boldsymbol{\epsilon}}_{\boldsymbol{k},\lambda}(ck)^{1/2}\{a_{\boldsymbol{k},\lambda} \exp(\mathrm{i}\boldsymbol{k} . \boldsymbol{x}) - a^\dagger_{\boldsymbol{k},\lambda} \exp(-\mathrm{i}\boldsymbol{k} . \boldsymbol{x})\} \tag{2}$$

in which λ ($=1$ or 2) is a polarization index, $\hat{\boldsymbol{\epsilon}}_{\boldsymbol{k},\lambda}$ is a unit polarization vector and V_q is the volume for quantization. We find $\boldsymbol{e}(\boldsymbol{x})$ actually becomes eliminated from the theory by concealment in the propagator $\mathbf{F}$ which is a c number. The operator density

$$\boldsymbol{\mu}(\boldsymbol{x}) \equiv e x_{0s} \hat{\boldsymbol{u}} \sigma_x(\boldsymbol{x}) \tag{3a}$$

where $x_{0s} \equiv \langle 0|x|s\rangle = x_{s0}$ is the dipole x matrix element, $\hat{\boldsymbol{u}}$ is a unit vector along x and $\sigma_x(\boldsymbol{x})$ is the total x component of spin density:

$$\sigma_x(\boldsymbol{x}) \equiv \sum_{i=1}^{N} \sigma_x^{(i)} \delta(\boldsymbol{x}-\boldsymbol{x}_i). \tag{3b}$$

For completeness we quote the complete hamiltonian for N two-level atoms coupled by both the radiation field and the dipole–dipole Coulomb interaction, although we shall make little explicit reference to this hamiltonian later. This hamiltonian is

$$H = \sum_{\boldsymbol{k},\lambda} \hbar ck(a^\dagger_{\boldsymbol{k},\lambda} a_{\boldsymbol{k},\lambda} + \tfrac{1}{2}) + \tfrac{1}{2}\hbar\omega_s \sum_{\boldsymbol{x}} \sigma_z(\boldsymbol{x}) - \tfrac{1}{2} \sum_{\boldsymbol{x}} \sum_{\boldsymbol{x}'} \boldsymbol{\mu}(\boldsymbol{x})\boldsymbol{\mu}(\boldsymbol{x}') : \nabla\nabla|\boldsymbol{x}-\boldsymbol{x}'|^{-1} - \sum_{\boldsymbol{x}} \boldsymbol{\mu}(\boldsymbol{x}) . \boldsymbol{e}(\boldsymbol{x}). \tag{4}$$

The 'summations' over $\boldsymbol{x}$ and $\boldsymbol{x}'$ are to be interpreted as integrations; self-interactions are to be ignored in the dipole–dipole term.

The operator $\sigma_z(\boldsymbol{x})$ is the z component of spin density:

$$\sigma_z(\boldsymbol{x}) = \sum_{i=1}^{N} \sigma_z^{(i)}\delta(\boldsymbol{x}-\boldsymbol{x}_i). \tag{5}$$

A spin density $\sigma_y(\boldsymbol{x})$ similarly defined completes the spin algebra. The commutation relations are then

$$\boldsymbol{\sigma}(\boldsymbol{x}) \times \boldsymbol{\sigma}(\boldsymbol{x}') = 2\mathrm{i}\boldsymbol{\sigma}(\boldsymbol{x})\delta(\boldsymbol{x}-\boldsymbol{x}') \tag{6a}$$

in general; but if (and only if) all atoms occupy the same site $\boldsymbol{x}_0$ (say) a number density $\delta(\boldsymbol{x}-\boldsymbol{x}_0)$ factors from each component of spin density and only the commutation relations for total spin

$$\boldsymbol{\sigma} \times \boldsymbol{\sigma} = 2\mathrm{i}\boldsymbol{\sigma} \tag{6b}$$

are relevant.

It is convenient in the first instance to take as basis states the Dicke states $|r, m\rangle$. The two-level atom is a pseudospin system and r (Dicke's cooperation number) is total pseudospin; $m = \frac{1}{2}(N_+ - N_-)$ where $N_\pm$ are the numbers of atoms excited (+) or unexcited (−). Plainly $0 \leqslant r \leqslant \frac{1}{2}N$; $|m| \leqslant r$. The 2^N states $|r, m\rangle$ form a complete basis.

There is an outgoing condition on $\mathbf{F}$; therefore only the *emission* of photons is acceptable and to order e^2 the selection rules for m are $\Delta m = -1$. For all atoms on the same site, r is a good quantum number: thus $\Delta r = 0$ and $\Delta m = -1$ only, in this case. For spatially separated atoms r is not good. A simple application of the Wigner–Eckart theorem shows that the selection rules are now $\Delta r = 0, \pm 1$; $\Delta m = -1$.

In order to describe an extended system Dicke also introduced states $|r, m; \boldsymbol{k}\rangle$ labelled by a wavevector $\boldsymbol{k}$. For fixed $\boldsymbol{k}$ the set $|r, m; \boldsymbol{k}\rangle$ with $0 \leqslant r \leqslant \frac{1}{2}N$; $|m| \leqslant r$ forms a complete basis. Transitions between states of the same $\boldsymbol{k}$ have the same selection rules $\Delta r = 0, \pm 1$; $\Delta m = -1$. We need to distinguish between the states $|r, m\rangle$ and the states $|r, m; \boldsymbol{k}\rangle$ and we call the former 'simple' Dicke states, the latter 'phased' Dicke states: $|r, m; \boldsymbol{0}\rangle \equiv |r, m\rangle$. It is usually supposed the $|r, m; \boldsymbol{k}\rangle$ can be excited by a pulse with wavevector $\boldsymbol{k}$.

We first consider radiating transitions between *simple* Dicke states in a spatially extended system: we consider the case $\Delta r = 0$ first.

2. Perturbation theory of super-radiance: $\Delta r = 0$ transitions

From the dipole interaction $-\Sigma_{\boldsymbol{x}}\, \boldsymbol{\mu}(\boldsymbol{x}) \cdot \boldsymbol{e}(\boldsymbol{x})$ of § 1 we find by a straightforward application of transition rate theory that for separated atoms, and up to order e^2, the radiation rate Γ for the transitions $r = \frac{1}{2}N$, $\Delta r = 0$, $\Delta m = -1$ is the following:

$$\Gamma = (\tfrac{1}{2}N + m)(\tfrac{1}{2}N - m + 1)2e^2x_{0s}^2\hbar^{-1}\left(\int_V \int_{V'} \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \times \sum_{i=1}^{N}\sum_{j=1}^{N} \delta(\boldsymbol{x}-\boldsymbol{x}_i)\delta(\boldsymbol{x}'-\boldsymbol{x}_j)\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}\boldsymbol{x}\right) \tag{7}$$

where $V = V'$ bounds the region occupied by the atomic sites $\boldsymbol{x}_i, \boldsymbol{x}_j$; $\Gamma_0 \equiv \frac{4}{3}e^2x_{0s}^2\hbar^{-1}k_s^3$ is again the Einstein A coefficient.

The propagator $\mathbf{F}$ appears in a natural way in the perturbation theory as the Fourier transform of the time-ordered propagator

$$\mathrm{i}\hbar^{-1}\langle|T\boldsymbol{e}(\boldsymbol{x}, t)\boldsymbol{e}(\boldsymbol{x}', t')|\rangle$$

in which $e(x, t)$ is a Heisenberg operator for the free field and $|\,\rangle$ is the initial photon state. We plan to discuss the way in which this propagator **F** emerges in the theory in greater detail in another paper.

The propagator **F** has imaginary part

$$\mathrm{Im}\,\mathbf{F} = \tfrac{2}{3}k_s^3\{\mathbf{U}\mathrm{j}_0(k_sR)+\tfrac{1}{2}(3\hat{\boldsymbol{R}}\hat{\boldsymbol{R}}-\mathbf{U})\mathrm{j}_2(k_sR)\}\,; \tag{8}$$

$\hat{\boldsymbol{R}}$ is a unit vector along $\boldsymbol{R} \equiv \boldsymbol{x}'-\boldsymbol{x}$ and j_0 and j_2 are spherical Bessel functions. Since $\mathrm{j}_0(0) = 1$ and $\mathrm{j}_2(0) = 0$, we find the result of Dicke

$$\Gamma = \Gamma_0(\tfrac{1}{2}N+m)(\tfrac{1}{2}N-m+1) \tag{9}$$

when all atoms occupy the same site. When they do not the system develops spatial coherence if the atoms occupy a periodic lattice: otherwise there is coherence to the extent that the atomic sites are completely uncertain and *un*correlated inside V. For the sake of example we consider a macroscopic homogeneous isotropic sample of volume V which exhibits the local order which can be described by an atomic pair correlation function $g(R)$. If the order is merely local $g(R) \to 1$ as $R \equiv |\boldsymbol{x}'-\boldsymbol{x}| \to \infty$. This feature will survive even though in principle $g(R)$ depends on the state $|r, m\rangle$.

We take an appropriate ensemble average of (4) and define†

$$\left\langle \sum_{i=1}^{N} \delta(\boldsymbol{x}-\boldsymbol{x}_i) \right\rangle_{\mathrm{ave}} \equiv n, \qquad \left\langle \sum_{i\neq j} \delta(\boldsymbol{x}-\boldsymbol{x}_i)\delta(\boldsymbol{x}'-\boldsymbol{x}_j) \right\rangle_{\mathrm{ave}} \equiv n^2 g(R). \tag{10}$$

For simplicity we identify n with NV^{-1} although this is not true for the grand ensemble, for example. There is now a natural way to separate the radiation rate Γ into two parts: an 'incoherent' part Γ_{inc} and a 'coherent' part Γ_{coh}. These (again $r = \frac{1}{2}N$) are

$$\Gamma_{\mathrm{inc}} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N^2}\left(N\Gamma_0+2Nn\frac{e^2x_{0s}^2}{\hbar}\int (g(R)-1)\,\mathrm{Im}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\right) \tag{11a}$$

$$\Gamma_{\mathrm{coh}} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N^2}n^2\left(\frac{2e^2x_{0s}^2}{\hbar}\int_V\int_{V'} \mathrm{Im}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{x}\,\mathrm{d}\boldsymbol{x}'\right). \tag{11b}$$

We look at Γ_{inc} first.

The pair correlation length $g(R)$ is sensibly 1 for all $R > l$, a 'correlation length'. This is why for macroscopic samples (but only for these) the integral in (11*a*) can be taken over all space and does not depend on the shape of V. But this means that Γ_{inc} does not depend on the shape of V. Providing $l \ll k_s^{-1}$, (8) means moreover that

$$\Gamma_{\mathrm{inc}} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N}\Gamma_0\left(1+n\int (g(R)-1)\,\mathrm{d}\boldsymbol{R}\right). \tag{12}$$

In the grand ensemble, for example, the Ornstein–Zernike relation

$$\left(1+n\int (g(R)-1)\,\mathrm{d}\boldsymbol{R}\right) = n\kappa k_{\mathrm{B}}T \tag{13}$$

applies. Here κ is the isothermal compressibility at temperature T; k_{B} is Boltzmann's constant. For a dense fluid $\kappa k_{\mathrm{B}}T \sim 10^{-26}$ and $n \sim 10^{23}$; for an ideal gas $n\kappa k_{\mathrm{B}}T \equiv 1$. Thus for $m \simeq 0$, $\Gamma_{\mathrm{inc}} \simeq \frac{1}{4}N\Gamma_0$, not $\frac{1}{4}N^2\Gamma_0$ and may be as small as $10^{-3}N\Gamma_0$. The incoherent rate controlled by Γ_{inc} is thus drastically reduced by 'coherence narrowing'

† We do not consider the case in which the two-level atoms adopt different orientations so that the vectors $\hat{\boldsymbol{u}}$ must do so and we do not average over these directions.

for the total radiation rate is now controlled by the coherent part $\Gamma_{\rm coh}$ if this is large enough.

With $n \simeq NV^{-1}$, $\Gamma_{\rm coh}$ depends on

$$\mathscr{D} \equiv \frac{2e^2x_{0s}^2}{V^2\hbar}\int_V\int_{V'} \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{x}\,\mathrm{d}\boldsymbol{x}'. \tag{14}$$

To evaluate this we use the identity

$$\frac{2e^2x_{0s}^2}{\hbar}\operatorname{Im}\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}} = \int \Gamma(\boldsymbol{k})\exp\{-\mathrm{i}\boldsymbol{k}\,.\,(\boldsymbol{x}-\boldsymbol{x}')\}\,\mathrm{d}\boldsymbol{k} \tag{15a}$$

where

$$\Gamma(\boldsymbol{k}) \equiv \frac{e^2x_{0s}^2k}{2\pi\hbar}(\mathbf{U}-\hat{\boldsymbol{k}}\hat{\boldsymbol{k}}):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\delta(k-\omega_sc^{-1}) \tag{15b}$$

(and $\hat{\boldsymbol{k}}$ is a unit vector along $\boldsymbol{k}$). For V we shall take the right-sided box defined by $-\frac{1}{2}a \leqslant x \leqslant \frac{1}{2}a$, $-\frac{1}{2}b \leqslant y \leqslant \frac{1}{2}b$, $-\frac{1}{2}c \leqslant z \leqslant \frac{1}{2}c$ (here c is *not* the velocity of light!). The integrals on $\boldsymbol{x}$ and $\boldsymbol{x}'$ if done first reduce $\mathscr{D}$ to

$$\mathscr{D} \equiv \int \Gamma(\boldsymbol{k})L(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k} \tag{16a}$$

where $L(\boldsymbol{k})$ is the Fraunhofer diffraction function

$$L(\boldsymbol{k}) \equiv \left(\frac{\sin\frac{1}{2}k_xa}{\frac{1}{2}k_xa}\right)^2\left(\frac{\sin\frac{1}{2}k_yb}{\frac{1}{2}k_yb}\right)^2\left(\frac{\sin\frac{1}{2}k_zc}{\frac{1}{2}k_zc}\right)^2. \tag{16b}$$

The following cases exemplify the consequences of this elementary piece of diffraction theory in super-radiance theory.

Case 1: the point system; a, b, c all tend to zero (with N fixed). We find $\mathscr{D} = \Gamma_0$ and $\Gamma_{\rm coh}$ is given by the Dicke result (9): $\Gamma_{\rm inc}$ formally vanishes for point systems. Obviously as $a, b, c \to 0$ with n fixed the rate vanishes. The point however is the old point that if $k_xa \ll 1$, $k_yb \ll 1$, $k_zc \ll 1$ and $N = n\times(abc)$, then the result (9) holds to neglect of $O(k_x^2a^2)$, etc.

Case 2: the slab of width c; $a, b \to \infty$, c = finite. We use

$$\left(\frac{\sin\frac{1}{2}k_xa}{\frac{1}{2}k_xa}\right)^2 \sim 2\pi a^{-1}\delta(k_x)$$

as $a \to \infty$ to find

$$\mathscr{D} = \frac{8\pi e^2x_{0s}^2}{\hbar}\frac{1}{V}\frac{(\sin\frac{1}{2}k_sc)^2}{\frac{1}{2}k_sc} \tag{17a}$$

and $\Gamma_{\rm coh}$ goes as N when $m \simeq 0$:

$$\Gamma_{\rm coh} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N}6\pi\Gamma_0nk_s^{-3}\frac{(\sin\frac{1}{2}k_sc)^2}{\frac{1}{2}k_sc}. \tag{17b}$$

For *small* k_sc the effective rate constant replacing $N\Gamma_0$ is

$$3\pi\Gamma_0nk_s^{-3}x(k_sc) = 4\pi ne^2x_{0s}^2\hbar^{-1}k_sc.$$

For thick slabs however $(\sin\frac{1}{2}k_sc)^2/\frac{1}{2}k_sc \simeq (k_sc)^{-1}$ which is substantially smaller than k_sc. Thus for $k_sc \sim 100$ and $n \sim 10^{18}$, for example, Γ_0 is replaced by a number also of order Γ_0. Further the slab has the Fabry–Perot character of totally trapping coherent radiation when $k_s c = 2\nu\pi$.

Case 3: for the long thin rod we find in any case the following: $a = b = 0$ and $c \to \infty$ means

$$L(\boldsymbol{k}) \sim 2\pi c^{-1}\delta(k_z) \tag{18a}$$

and

$$\Gamma_{\text{coh}} = (\tfrac{1}{2}N+m)(\tfrac{1}{2}N-m+1)\frac{3\pi\Gamma_0}{4k_s c}. \tag{18b}$$

This is super-radiant when $m \simeq 0$ because $V = 0$, but the super-radiance is reduced when $k_s c$ is very large. For a narrow rod of cross section A and volume V,

$$\Gamma_{\text{coh}} = \tfrac{1}{4}NnA\left(\frac{3\pi\Gamma_0}{4k_s}\right).$$

It is also evident from (15) that the radiation has the usual dipole radiation envelope—that is a doughnut with axis along $\hat{\boldsymbol{u}}$ (or x). In case 2 radiation only occurs in the two directions normal to the slab ($\boldsymbol{k}$ only lies along z). Notice further that (again for $m \simeq 0$) $\Gamma_{\text{coh}} \propto n^2Vc^{-1} = n^2A$ where A is the area of the slab which is supposed large. The intensity emitted normal to the slab is therefore proportional to n^2 and does not depend otherwise on N or V. In case 3, wavevectors $\boldsymbol{k}$ are normal to z and the usual factor $\{1-(\hat{\boldsymbol{u}}\,.\,\hat{\boldsymbol{k}})^2\}$ appears. Thus radiation is *normal* to the rod and the intensity does not depend on N and V simply through n^2.

Notice that the spatial coherence and directed emission occur because the system is supposed prepared in a simple Dicke state $|\tfrac{1}{2}N, m\rangle$. It is controlled by the surface geometry of V and is symmetrically directed for symmetric V. This kind of coherent emission was not discussed by Dicke (1954) (or in Dicke 1964). Unfortunately it is still an open question whether states with $r = \tfrac{1}{2}N$ and $m \simeq 0$ can ever be reached with sufficient occupation in an extended system: we show in the next section that starting from total inversion in the state $|\tfrac{1}{2}N, \tfrac{1}{2}N\rangle$, for example, the system tends to change r: it thus tends to evolve incoherently rather than coherently.

3. Transitions when $\Delta r \neq 0$

We use an additional quantum number α to distinguish the $N-1$ different but degenerate unperturbed states with fixed m and definite total spin $r = \tfrac{1}{2}N-1$. The rate Γ from the state $|\tfrac{1}{2}N, m\rangle$ to any one of the $N-1$ states $|\tfrac{1}{2}N-1, m-1, \alpha\rangle$ is (to the approximation using (13))

$$\Gamma(|\tfrac{1}{2}N, m\rangle \to |\tfrac{1}{2}N-1, m-1, \alpha\rangle) = \frac{(\tfrac{1}{2}N+m)(\tfrac{1}{2}N+m-1)}{N(N-1)^2}(N\Gamma_0 - \Gamma_0 n\kappa k_B T - N\mathscr{D}). \tag{19a}$$

Note that the incoherent part in $\Gamma_0 n\kappa k_B T$ is O(1) compared with the part $N\Gamma_0$ and that the coherent part is O(1) compared with this also.

Next one has the general result for rates changing r and m that

$$\Gamma(|r, m, \alpha\rangle \to |r+1, m-1, \beta\rangle) = \Gamma(|r+1, -m+1, \beta\rangle \to |r, -m, \alpha\rangle).$$

Thus

$$\Gamma(|\tfrac{1}{2}N-1, m, \alpha\rangle \rightarrow |\tfrac{1}{2}N, m-1\rangle) = \Gamma(|\tfrac{1}{2}N, -m+1\rangle \rightarrow |\tfrac{1}{2}N-1, -m, \alpha\rangle)$$

and

$$\Gamma(|\tfrac{1}{2}N-1, m, \alpha\rangle \rightarrow |\tfrac{1}{2}N, m-1\rangle) = \frac{(\tfrac{1}{2}N-m)(\tfrac{1}{2}N-m+1)}{N(N-1)^2}(N\Gamma_0 - \Gamma_0 n\kappa k_B T - N\mathscr{D}). \qquad (19b)$$

Equations (19) together demonstrate that when $m \sim 0$, all rates to and from $r = \frac{1}{2}N$ involving change of r are incoherent to relative order $O(1/N)$.

We can now consider transitions from the totally inverted state where $m = r = \frac{1}{2}N$. It follows from (11) that the rate to $|r, m-1\rangle$ is $O(1) \times \Gamma_0$; however, the extended system will prefer to make transitions $\Delta r = -1$ simply because there are $N-1$ ways of doing this. The rate to each of these is Γ_0 and the total rate $(N-1)\Gamma_0$. The total rate should therefore be approximately $N\Gamma_0$ and should be incoherent since the coherent part is $O(1)$. In fact since the Dicke state $|\frac{1}{2}N, \frac{1}{2}N\rangle$ is a product of single particle states it is easily proved outside perturbation theory that the total rate is precisely $N\Gamma_0$ and is totally incoherent. The same result emerges in perturbation theory to order e^2: (19*a*) shows that the coherent part is identically eliminated.

In the $m \sim 0$ region the total rate for $\Delta r = \pm 1$ transitions is still of order $N\Gamma_0$ and adds to Γ_{inc} which by (12) is already of order $N\Gamma_0$. There is no contribution to Γ_{coh} which alone depends on surface geometry and is still given by (11*b*).

This result means that the $m \sim 0$ region is coherent if and only if Γ_{coh} actually dominates there: in contrast the $m \sim \frac{1}{2}N$ region is strictly quantal and always incoherent. A number of authors, notably Bonifacio, Schwendimann and Haake for example have noticed *classical* features in the $m \sim 0$ region: in particular these authors note small quantal fluctuations near $m \simeq 0$: they are concerned with the long rod prepared by an incident pulse in phased states with $m \simeq 0$. Our conclusions for systems prepared in simple Dicke states are that relative fluctuations in the spatial coherence are smallest in the $m \sim 0$ region and there are two points here: first, it can never be correct to isolate the coherent part Γ_{coh} of the radiation rate and reject Γ_{inc} in the region $m \simeq \frac{1}{2}N$ since Γ_{coh} actually vanishes there; second, even when $m \simeq 0$ in a large macroscopic system it may not be correct to ignore Γ_{inc} since Γ_{coh} may still not dominate. If $n \gtrsim 10^{18}$, $k_s^{-1} \simeq 10^{-5}$ cm and $k_s c \simeq 1$, $\Gamma_{coh} \sim 5 \times 10^3 \Gamma_0 N$ for the slab when $m \simeq 0$; but if the slab is 100 reciprocal wavenumbers thick, $0 \leqslant \Gamma_{coh} \leqslant 5 \times 10^{-1} \Gamma_0 N$ and the slab is scarcely coherent or classical.

This feature allows us to define a maximum 'coherence length' for states $|\frac{1}{2}N, m\rangle$ which is in some ways analogous to the 'maximum cooperation' length for phased Dicke states introduced by Arecchi and Courtens (1970). This coherence length depends on the geometry of the sample, however. For the slab, in particular, $\Gamma_{inc} \sim N\Gamma_0$ and $\Gamma_{inc} \leqslant \Gamma_{coh}$ for widths less than about 100 reciprocal wavenumbers thick. This length is rather less than the number for the rod quoted by Arecchi and Courtens (0·1 cm for $2\pi k_s^{-1} = 7 \times 10^{-5}$ cm and ruby with 0·05% Cr^{3+} concentration). Moreover this line of analysis shows no evidence for the spiking of pulses postulated by Arecchi and Courtens (although within low order perturbation theory this might be expected): Γ_{coh} remains coherent however large the system is, and is merely dominated by Γ_{inc} as increasing size reduces its magnitude.

There is however a crucial difference between Γ_{coh} and Γ_{inc}: Γ_{coh} summarizes all that radiation emitted in directions determined solely by the macroscopic geometry of the sample; Γ_{inc} on the other hand is independent of that geometry and the contributions to it are essentially isotropic showing angular dependence only insofar as the radiation is dipole radiation from vector dipole matrix elements in the chosen direction $\hat{\boldsymbol{u}}$.

To see this observe that (15*a*) means that

$$2ne^2x_{0s}^2\hbar^{-1}\int(n^{-1}\delta(\boldsymbol{R})+g(R)-1)\,\mathrm{Im}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\mathrm{d}\boldsymbol{R}$$

$$=n\int\Gamma(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k}\int(n^{-1}\delta(\boldsymbol{R})+g(R)-1)\exp(\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R})\,\mathrm{d}\boldsymbol{R}$$

$$=n\int\Gamma(\boldsymbol{k})\,\mathrm{d}\boldsymbol{k}\int(n^{-1}\delta(\boldsymbol{R})+g(R)-1)\frac{\sin kR}{kR}\,\mathrm{d}\boldsymbol{R}. \tag{20}$$

This quantity appears in the large bracket in Γ_{inc} in (11*a*): the single particle contributions there are now shown to be due to a self-interaction described by self-correlation functions $\delta(\boldsymbol{R})$. The radiation rate into solid angle $\mathrm{d}\Omega(\hat{\boldsymbol{k}})$ for the transitions $\Delta r = 0$, $\Delta m = -1$ is therefore proportional to

$$\frac{ne^2x_{0s}^2}{2\pi\hbar}k_s^3(\mathbf{U}-\hat{\boldsymbol{k}}\hat{\boldsymbol{k}}):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\Omega(\hat{\boldsymbol{k}})\int(n^{-1}\delta(\boldsymbol{R})+g(R)-1)\frac{\sin k_sR}{k_sR}\,\mathrm{d}\boldsymbol{R}. \tag{21}$$

The integral of this over all solid angles $\Omega(\hat{\boldsymbol{k}})$ reduces again to (20).

The result (21) means that the radiation is dipole radiation identical with point source radiation as far as the envelope goes: the integral in (21) does not depend on $\hat{\boldsymbol{k}}$. When $k_sl \ll 1$ since $(\sin k_sR)/k_sR \equiv \mathrm{j}_0(k_sR) = 1$ for all $R < l$, the integral reduces to κk_BT within the terms leading to (12) with (13), for example; but even when $k_sl \gtrsim 1$ the envelope is unchanged except for scale. Thus the envelope is indeed essentially isotropic and Γ_{coh} can be isolated from it by selecting directions in which this spatially coherent part of the radiation is dominant. There will be such directions for the slab, for example, if the area of the slab is big enough. In these directions there is no maximum coherence length.

4. Transitions between phased Dicke states

We have so far considered transitions between 'simple' Dicke states $|r, m\rangle$. The significance of the results depends on a capacity to put the system in one of these states. It may be easier to put the extended system into a phased Dicke state by exciting it with an optical pulse which has a carrier wave with wavevector $\boldsymbol{k}$. In this form however the problem becomes a problem in optical pulse propagation: it is not possible to describe the optical excitation of an extended system and the *subsequent* decay of that excitation: the processes of excitation and decay are inextricably linked in the propagation process. A pulse has a unique wavevector $\boldsymbol{k}$ if it behaves like a plane wave with this wavevector. Outside the linear region $\boldsymbol{k}$ is in general only locally constant. Nevertheless for small enough dielectrics there may be pulses which vary in time but which behave like $\exp(\pm\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{x})$ at all points in space inside the dielectric. This type of pulse underlies the work of Bonifacio *et al* (1971a, b) for example.

For this type of pulse the phased Dicke states $|r, m; \boldsymbol{k}\rangle$ form a basis in which since $\boldsymbol{k}$ is fixed r is a constant of the motion. However even in the evolution of such a pulse it is very doubtful whether total occupation of a single Dicke state $|r, m; \boldsymbol{k}\rangle$ occurs. The only exception is the weak field case where the state $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ can be excited. This is discussed in detail in § 3 of the paper II.

We believe the significance of phased Dicke states has been misunderstood. Nevertheless the states were studied by Dicke (1954) and most subsequent authors and we look at them here. Following Dicke we introduce states $|r, m; \boldsymbol{k}_0\rangle$ labelled by an 'incident' wavevector $\boldsymbol{k}_0$. These states are created by the action of the creation operator

$$\sigma_+(\boldsymbol{k}_0) \equiv \sum_{i=1}^{N} \sigma_+^{(i)} \exp(-\mathrm{i}\boldsymbol{k}_0 \cdot \boldsymbol{x}_i) \tag{22}$$

acting on, for example, the ground state (which is a simple Dicke state) $|\frac{1}{2}N, -\frac{1}{2}N\rangle$. We find that for transitions involving fixed $\boldsymbol{k}_0$ the sole effect is to replace $\boldsymbol{k}$ by $\boldsymbol{k}_0$ in the exponent in the integrand of (15*a*)†. Equivalently the propagator $\mathsf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s)$ is replaced by the phased propagator $\mathsf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) \exp\{-\mathrm{i}\boldsymbol{k}_0 \cdot (\boldsymbol{x}'-\boldsymbol{x})\}$. In consequence the coherent transition rate $|\frac{1}{2}N, m; \boldsymbol{k}_0\rangle \rightarrow |\frac{1}{2}N, m-1; \boldsymbol{k}_0\rangle$ in which a photon of frequency ω_s is emitted in the direction $\hat{\boldsymbol{k}}$ into $\mathrm{d}\boldsymbol{k}$ is $\Gamma_{\hat{\boldsymbol{k}}}\, \mathrm{d}\boldsymbol{k}$ where

$$\Gamma_{\hat{\boldsymbol{k}}} = (\tfrac{1}{2}N+m)(\tfrac{1}{2}N-m+1)\Gamma(\boldsymbol{k})L(\boldsymbol{k}-\boldsymbol{k}_0). \tag{23}$$

We therefore find the following: the δ function in $\Gamma(\boldsymbol{k})$ requires $k = k_s$; if the pulse is nonresonant, so that $k \neq k_s$, $L(\boldsymbol{k}-\boldsymbol{k}_0)$ has a nonvanishing argument and the previous geometrically dependent results are regained with appropriate changes in the effective wavevector. These results are now controlled by the geometry of V and by $\boldsymbol{k}_0$ however: they are no longer symmetrical if V is and are directed by $\boldsymbol{k}_0$.

In contrast, if the pulse is resonant so that the incident wavenumber is k_s and $\boldsymbol{k}_0$ and $\boldsymbol{k}$ have the same direction, $L(\boldsymbol{k}-\boldsymbol{k}_0)$ is unity for all a, b, c and we regain the result of Dicke (1954):

$$\Gamma_{\hat{\boldsymbol{k}}} = (\tfrac{1}{2}N+m)(\tfrac{1}{2}N-m+1)\Gamma(\boldsymbol{k}). \tag{24}$$

This applies to arbitrary extended systems and appears to mean that, when $m \simeq 0$, $\Gamma_{\hat{\boldsymbol{k}}} \simeq \frac{1}{4}N^2\Gamma(\boldsymbol{k})$ for any geometry.

However, experience with linear dielectric theory shows that travelling plane wave pulses with wavevectors $\boldsymbol{k}_0$ satisfying the free field dispersion relation $\omega = ck_0$ are possible at best only on resonance (as here) and further that perturbation theory yields erroneous results of the same qualitative character as is exhibited by (24). We can already see that (24) requires some delicacy of interpretation by adopting a different point of view. We first let $a, b \rightarrow \infty$ to construct the infinite slab: we find that when $\boldsymbol{k}_0$ is normal to the slab, so that the pulse is both normally incident and resonant, only a factor $\{(\sin\frac{1}{2}(k_s-k_0)c)/\frac{1}{2}(k_s-k_0)c\}^2$ is unity for all c, all photons are emitted in the direction of $\boldsymbol{k}_0$, and the total rate is

$$\Gamma_{\mathrm{coh}} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N} 6\pi\Gamma_0 n k_s^{-3}(\tfrac{1}{2}k_s c). \tag{25}$$

This result has reduced the N^2 dependence near $m = 0$ to N dependence even though it changes the small sample form of (17*b*) to one applicable for all slab widths c. The

† More precisely this is the sole effect in a prescription rejecting reflected waves. The situation is analysed in the appendix to the following paper (part II).

effective rate constant replacing $N\Gamma_0$ is now $4\pi n e^2 x_{0s}^2 \hbar^{-1} k_s c$ in exact agreement with the constant used by Bonifacio *et al* (1971a, b) for long rods bounded only by their 'maximum cooperation length'.

It might still appear from (25) that the emitted intensity $\Gamma_{\mathrm{coh}} A^{-1} (\propto n^2 c^2)$ can be made as large as one wishes simply by increasing the width c and that this intensity diverges with c. However, a comparable difficulty appears in linear dielectric theory and is there resolved by recognition that the excitation mechanism is significant as is also the spatial dispersion. The second point requires solution of the problem outside perturbation theory. At this stage, therefore, rather than invoke the ideas of maximum cooperation number and maximum cooperation length as they were first introduced by Arecchi and Courtens (1970), we shall cautiously take the view that the results (24) and (25) merely indicate that perturbation theory and the restriction to a single fixed $\boldsymbol{k}$ cannot adequately describe plane wave pulse propagation. Equation of motion methods support this view but contain difficulties still not elucidated.

Taking (25) on its merits we see that it relies on the following result for $L(\boldsymbol{k}-\boldsymbol{k}_0)$:

$$L(\boldsymbol{k}-\boldsymbol{k}_0) = \frac{4\pi^2}{ab}\delta(k_x - k_{0x})\delta(k_y - k_{0y})\left(\frac{\sin\frac{1}{2}(k_z - k_{0z})c}{\frac{1}{2}(k_z - k_{0z})c}\right)^2. \tag{26}$$

Since $k = k_s$, $k_z = k_{0z}$—because of the two δ functions—whenever $k_0 = k_s$†. Thus if the pulse is resonant the term in c vanishes from (26) and the radiation is in the direction of the incident pulse. When the pulse is not resonant the large bracket does not vanish and there is evidently actual refraction, obeying Snell's law, at the boundary of the slab. These are typical features of resonant and nonresonant plane wave pulse propagation. An explicit connection with linear dielectric theory is established in part II for $|m| \simeq \frac{1}{2}N$. The situation in the interesting $m \simeq 0$ region is of course more complicated and difficult and is not yet wholly understood.

With $\boldsymbol{k}_0$ fixed there is still an incoherent contribution to the radiation rate. This is almost identical with the incoherent contribution to transitions between simple Dicke states with $\Delta r = 0$. To calculate it it is sufficient to replace the integral in (21) by

$$\int (n^{-1}\delta(\boldsymbol{R}) + g(R) - 1)\frac{\sin k_\theta R}{k_\theta R}\,\mathrm{d}\boldsymbol{R} \tag{27}$$

in which $k_\theta = 2k_s \sin\frac{1}{2}k_s\theta$ and θ is the angle between the directions of $\hat{\boldsymbol{k}}_0$ and the outgoing photon direction $\hat{\boldsymbol{k}}$. Providing $k_s \gtrsim 1$, the envelope of outgoing radiation shows a small dissymmetry identical with the dissymmetry associated with a large correlation distance l in linear optical scattering theory. It applies whether $\boldsymbol{k}_0$ is resonant or not since k_θ in (27) simply changes slightly (cf, eg, Rosenfeld 1951).

5. Summary of results and conclusions

The 'super-radiant' radiation rate from point systems can be as large as $\frac{1}{4}N^2\Gamma_0$ where N is the number of atoms. We have shown in contrast that from extended systems in simple Dicke states $|r, m\rangle$ the rates never exceed $N\Gamma_0$. These rates have natural coherent and incoherent parts: the coherent rate consists of directed emission controlled by the sample geometry; the incoherent rate is isotropic. In the case of a dielectric slab the coherent emission is symmetric and normal to each surface. If the slab is wide enough

† Providing the reflected wave is ignored: see the analysis in the appendix to paper II.

the total incoherent rate exceeds the total coherent rate. There is therefore 'a maximum coherence length' for the width of the slab: beyond this the radiation is dominantly incoherent, but since coherent emission is directed it can be distinguished in the total emission however wide the slab. We have also examined comparable emission from the narrow rod.

Such coherent emission occurs predominantly in $\Delta r = 0$ transitions between simple Dicke states $|\frac{1}{2}N, m\rangle$: it requires placing the system in such a state initially. A totally inverted system prefers to make transitions with $\Delta r = -1$ and to emit incoherently. This appears also to be a general feature of simple Dicke states: thus we can expect an inverted system to tend to evolve incoherently with some admixture of coherent emission controlled only by the geometry.

Systems in phased Dicke states $|r, m; \boldsymbol{k}\rangle$ emit coherently but unsymmetrically in directions controlled by the wavevector $\boldsymbol{k}$. They also emit incoherently: the incoherent emission is almost isotropic but is partly controlled by $\boldsymbol{k}$. If the wavevector is resonant $k = \omega_s c^{-1}$ where ω_s is the atomic resonance. In this case the geometry does not control the direction of the coherent emission. If $k \neq \omega_s c^{-1}$, however, there are refraction effects at the surface of the system.

In the case of the slab super-radiance going as $\frac{1}{4}N^2\Gamma_0$ does not occur in the $m \simeq 0$ region. The rate goes as $N\Gamma_0$. However if $k = \omega_s c^{-1}$ it appears that by making the slab width large enough the coherent rate can be increased without limit. Apparently the coherent rate will always dominate over the incoherent rate if the slab is wide enough.

However, this result depends on placing the system in a Dicke state phased with a definite resonant wavevector $\boldsymbol{k} = \boldsymbol{k}_0$ initially. It is doubtful whether perturbation theory is applicable on resonance. Further it is doubtful whether such phased Dicke states can be reached by exciting the system with a resonant pulse with wavevector $\boldsymbol{k}_0$ as is usually assumed. Nevertheless the results for phased Dicke states show some features of resonant pulse propagation, although we conclude that they form in themselves an inadequate description of pulse propagation. For this last reason there is no need at this stage to postulate a maximum 'cooperation length' to bound the coherent rate from very thick slabs.

The distinction between super-radiance from large and small systems relies on interference–diffraction effects controlled by the propagator $\mathbf{F}$. These effects will be the same in character (but not in magnitude) whether $|m| \simeq \frac{1}{2}N$ or $|m| \simeq 0$.

A distinction is usually made between the coherent and incoherent excitation of a dielectric. In the former the off-diagonal elements of the reduced one-atom density matrix are nonzero in general and the atoms exhibit a nonvanishing expectation value of their dipole moments. This feature is also characteristic of both semiclassical radiation theory and the *neo*classical theory of Jaynes (eg Jaynes and Cummings 1963). It is possible to show to a good approximation from our hamiltonian (the hamiltonian (4)) that if a dielectric is placed in a Dicke state, simple or phased, the expectation value of the total dipole moment vanishes throughout the motion. The radiation is therefore incoherent in this sense.

Since $\mathbf{F}$ is a Green function of Maxwell's equations, whether these are read as c number or operator equations, the diffraction–interference features of the theory will be the same whether the expectation values of the dipole moments vanish or not. An important conclusion seems to be that the enhanced radiation rates going as N^2 from small samples occur whether the sample is 'coherently' excited, has a dipole moment, and hence is (almost) classical or whether it radiates quantally and incoherently from a definite Dicke state (simple or phased). Furthermore for extended samples like the slab of case 2, for example, in a simple Dicke state or the slab in a phased Dicke state interpreted as in (25) the emitted intensity is proportional to $n^2 = (NV^{-1})^2$ and this again seems to be a result independent of the presence of an atomic phase, a dipole moment, or off-diagonal elements of the density matrix. Thus observation of n^2 behaviour as recently reported by Brewer and Shoemaker (1971) in the photon echoes from $^{13}CH_3F$ would not seem to distinguish between these two situations.

Some of the qualitative content of the significance of diffraction–interference in modifying small sample super-radiance going as N^2 is already implicit in the work of Rehler and Eberly (1971). Their theory is an equation of motion theory using coherent Bloch states phased by an incident pulse: the propagation problem is ignored. The theory is decorrelated in all except self-correlations: this makes individual atoms emit quantally and incoherently but coherence effects between atoms are decorrelated and the system has a dipole. This illustrates the point that **F** has the same significance both in the correlated quantal theory developed in this paper and in a decorrelated neoclassical theory.

In the following paper (part II) we extend the perturbation theory to include level shifts and the effect of free photons on both these and the radiation rates. We also compare the results for $|m| = \frac{1}{2}N - 1$ with the results of equation of motion methods for linear dielectrics. This is the one case where a phased Dicke state can be excited by an external field: agreement is essentially complete.

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