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Perturbation theory of super-radiance
II. Cooperative and non-cooperative level shifts

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Abstract. We extend the perturbation theory of super-radiant emission to derive a number of results on the level shifts of N atom systems. These also divide into 'coherent' and 'incoherent' parts. The incoherent part divides further into a generalized Lamb shift which is not cooperative, and an interatomic term which is. The former proves to be the only shift which can depend on the presence of ambient free photons. These results for the generalized Lamb shift support the view that a dynamical neoclassical theory of the Lamb shift is unacceptable. The theory is compared with the pseudoboson theories of linear dielectrics. The pseudoboson theory of an inverted dielectric (the 'amplifier') should be intrinsically unstable unlike the pseudoboson theory of the refractive index which is concerned with the 'attenuator'.

1. Introduction

In part I (Saunders and Bullough 1973, to be referred to as I) we showed that the radiation rates from extended samples prepared initially in the 'simple' Dicke states labelled by r, m had natural coherent and incoherent parts Γ_{coh} and Γ_{inc}. The label r is the co-operation number introduced by Dicke (1954): $m = \frac{1}{2}(N_+ - N_-)$ where $N_\pm$ are the numbers of two-level atoms in their upper (lower) states. Both the rates Γ_{coh} and Γ_{inc} are cooperative but reduce substantially in extended systems. We also displayed comparable results for the 'phased' Dicke states labelled by r, m and a wavevector $\boldsymbol{k}_0$. We have checked these order e^2 results for the radiation rates by calculating the complex energies of the system to this order. The real parts of these energies can also be separated into coherent and incoherent parts in a natural way; but only the shifts due to interactions between separated atoms cooperate. We indicate the results.

2. The complex energy shifts

For extended samples in simple Dicke states $|r, m\rangle$ we find the following generalization of the Feynman (1961) expression for the self-energy of the electron (compare also the all order in e^2 result for the ground state energy of a molecular fluid given by Bullough 1969):

$$\Delta E = -\frac{\hbar}{4\pi \mathrm{i}} \operatorname{Tr} \int_0^\infty \mathrm{d}\omega \int_V \mathrm{d}\boldsymbol{x} \int_{V'} \mathrm{d}\boldsymbol{x}' \times \{(n_k+1)(\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega) \cdot \boldsymbol{\Pi}_{\mathrm{T}}(\boldsymbol{x}, \boldsymbol{x}'; \omega) - \mathrm{cc}) + n_k(\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega) \cdot \boldsymbol{\Pi}_{\mathrm{T}}^*(\boldsymbol{x}, \boldsymbol{x}'; \omega) - \mathrm{cc})\}. \quad (1)$$

In this expression $k = \omega c^{-1}$ and n_k is the occupation number for ambient photons with wavenumber k: for simplicity this is supposed to be isotropic, that is the same for all directions $\boldsymbol{k}$. The photon propagator $\mathbf{F}$ is given by equation (1) of I. The matter propagator $\boldsymbol{\Pi}_T$ is the time-ordered propagator

$$\boldsymbol{\Pi}_T(\boldsymbol{x}, \boldsymbol{x}'; \omega) = -\hbar^{-1} \sum_s \left(\frac{1}{\omega_{ls} - \omega + i\delta} + \frac{1}{\omega_{ls} + \omega + i\delta} \right) \langle l|\boldsymbol{\mu}(\boldsymbol{x})|s\rangle \langle s|\boldsymbol{\mu}(\boldsymbol{x}')|l\rangle. \tag{2}$$

It applies to a system of N atoms each with an arbitrary number of levels:

$$\boldsymbol{\mu}(\boldsymbol{x}) = \sum_{i=1}^{N} \boldsymbol{\mu}^{(i)} \delta(\boldsymbol{x} - \boldsymbol{x}_i),$$

the total dipole operator; $|l\rangle$ is the unperturbed state of energy E_l, $|s\rangle$ is any one of the complete set of unperturbed states labelled by s and of energy E_s, $\omega_{ls} \equiv \hbar^{-1}(E_l - E_s)$. As usual δ is an infinitesimal of positive sign.

There is a natural complex energy obtainable from (1). This is just one half of (1) as it is written when the complex conjugates (cc's) in it are dropped. Then there is a natural radiation rate which is $-2\hbar^{-1}$ times the imaginary part of this shift. However, there is an ambiguity in the choice of imaginary parts of the integral: we can for example take $\mathrm{Re}\,\mathbf{F} \,.\, \boldsymbol{\Pi}_T^*$ or $-\mathrm{Re}\,\mathbf{F}^* \,.\, \boldsymbol{\Pi}_T$ from the bracket multiplied by n_k in (1). The correct choice is motivated by the conditions (*a*) that we want a radiation rate, that is a rate of increase of photons (*b*) that the emission term in $(n_k + 1)$ increases the number (positive rate) and that the induced absorption term in n_k decreases it (negative rate). The prescription above has this property as case (i) below, for example, shows. Thus we can identify the total imaginary part obtained this way as Γ_{tot}, the total radiation rate when the atomic system is in the state $|l\rangle$.

For N two-level atoms the uncoupled system is in general degenerate: for example, the set of $N_+ = (m + \frac{1}{2}N)$ atom excitations is $N!/N_+!N_-!$ degenerate (observe $N_+ + N_- = N$). Appropriate states for degenerate perturbation theory are the simple Dicke states $|r, m\rangle$ for small samples and the phased Dicke states $|r, m; \boldsymbol{k}\rangle$ for translationally invariant systems. In what follows we shall investigate level shifts for both sets of states. Since we now restrict the calculation to two-level atom systems,

$$\boldsymbol{\mu}(\boldsymbol{x}) = e x_{0s} \hat{\boldsymbol{u}} \sum_{i=1}^{N} \sigma_x^{(i)} \delta(\boldsymbol{x} - \boldsymbol{x}_i)$$

exactly as in equation (3) of I†.

We find the following:

(i) N atoms on the same site. The emission rate is

$$\Gamma_{em} = \Gamma_0 (n_{k_s} + 1)(r + m)(r - m + 1), \tag{3a}$$

the absorption rate is (rate of increase of photons)

$$\Gamma_{abs} = -\Gamma_0 n_{k_s} (r - m)(r + m + 1) \tag{3b}$$

and the total radiation rate is

$$\Gamma_{tot} = \Gamma_0 \{2 n_{k_s} m + (r + m)(r - m + 1)\}. \tag{3c}$$

If no free photons at frequency ω_s are available, Γ_{tot} reduces to the spontaneous emission

† Later we use the notation (I.3) for this, for example.

rate (I.9); if photons at frequency ω_s are available the spontaneous emission is coherent (that is cooperative) but the total of the induced processes is not.

The (real) energy level shift is

$$\Delta E = -2m\Delta E'_{\mathrm{B}} \tag{4a}$$

where

$$\Delta E'_{\mathrm{B}} = \frac{e^2x_{0s}^2}{3\pi c^3}\left\{2P\int_0^\infty n_k\omega^3\left(\frac{1}{\omega_s+\omega}+\frac{1}{\omega_s-\omega}\right)\mathrm{d}\omega\right\}+\Delta E_{\mathrm{B}}, \tag{4b}$$

and ΔE_{B} is the Bethe (1947) level shift

$$\Delta E_{\mathrm{B}} = \frac{2}{3\pi}e^2x_{0s}^2\frac{\omega_s^3}{c^3}\ln\left(\frac{m_ec^2}{\hbar\omega_s}\right) \tag{4c}$$

providing the integral in ω is cut off at the Compton frequency. The quantity $\Delta E'_{\mathrm{B}}$ is the generalized Lamb shift for the two-level atom reported by Bullough and Caudrey (1971) obtained by rather different methods. It also agrees with the perturbation theory of Knight (1972). The result (4*a*) shows that the Lamb shift is not cooperative or coherent: in this it shows a significant difference from the spontaneous emission.

(ii) Two atoms on different sites. In the Dicke states $|1, \pm1\rangle$ the shifts are $\pm2\Delta E'_{\mathrm{B}}$. In the symmetric (+) and antisymmetric (−) Dicke states the shifts are respectively

$$\begin{aligned}\Delta E &= \mp e^2x_{0s}^2\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x}_1,\boldsymbol{x}_2;\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\\ &= \mp\tfrac{2}{3}k_s^3e^2x_{0s}^2\{\mathrm{j}_{-1}(k_sR)\mathbf{U}+\tfrac{1}{2}\mathrm{j}_{-3}(k_sR)(3\hat{\boldsymbol{R}}\hat{\boldsymbol{R}}-\mathbf{U})\}:\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}.\end{aligned} \tag{5}$$

In this $\boldsymbol{R}=\boldsymbol{x}_1-\boldsymbol{x}_2$ and $\hat{\boldsymbol{R}}$ is a unit vector along $\boldsymbol{R}$: the j_n are spherical Bessel functions diverging as R^{-n} at the origin. These results agree with those of Stephen (1964) even though $n_{k_s}\neq0$: the energy shift does not depend on the presence of free photons at this order in e^2 and will not do so at higher orders as the discussion in (iii) below makes plain.

Note that the single particle Lamb shifts have cancelled: these constitute the shift $\Delta E = 2m\Delta E'_{\mathrm{B}}$ of (i) with $m=0$. The correct value of (5) when $\boldsymbol{x}_1=\boldsymbol{x}_2$ is therefore zero and the limit $\boldsymbol{x}_1\to\boldsymbol{x}_2$ is not defined. In the physical situation overlap becomes important before $\boldsymbol{x}_1=\boldsymbol{x}_2$.

The radiation rates are

$$(n_{k_s}+1)(\Gamma_0\pm2e^2x_{0s}^2\hbar^{-1}\,\mathrm{Im}\,\mathbf{F}(\boldsymbol{x}_1,\boldsymbol{x}_2;\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}) \tag{6}$$

from the Dicke state $|1,1\rangle$ to states $|1,0\rangle$ and $|0,0\rangle$. The total rate when the atoms are in the state $|1,1\rangle$ is $(n_{k_s}+1)\Gamma_0$. The rate from $|1,0\rangle$ to $|1,-1\rangle$ is (6) with the positive sign. The rate from $|1,0\rangle$ to $|1,1\rangle$ is the positive sign in

$$-n_{k_s}(\Gamma_0\pm2e^2x_{0s}^2\hbar^{-1}\,\mathrm{Im}\,\mathbf{F}(\boldsymbol{x}_1,\boldsymbol{x}_2;\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}). \tag{7}$$

The total radiation rate when the atoms are in the state $|1,0\rangle$ is just the bracket with the plus sign in (6): it does not depend on n_{k_s}. The total radiation rate when the system is in the state $|0,0\rangle$ is just the bracket with the minus sign in (6) which does not depend on n_{k_s}. When $n_{k_s}=0$, the rates are spontaneous rates downwards only and agree with (I.7).

(iii) N atoms on different sites. There is a rate downwards which multiplies (I.7) by $(n_{k_s}+1)$; there is a rate upwards which multiplies this by $-n_{k_s}$ with m also changed to $-m$. These rates describe the $\Delta r=0$ transitions. The rates (I.19) with $\Delta r=\pm1$

generalize in the same way. The analysis into coherent and incoherent parts is unchanged. In particular the radiation rate from the state $|\frac{1}{2}N, \frac{1}{2}N\rangle$ is precisely $(n_{k_s}+1)N\Gamma_0$.

The energy shifts are of considerable interest. A general result is that the shift of the Dicke state $|r, m\rangle$ with $r = \frac{1}{2}N$ is

$$\Delta E = -\tfrac{1}{2}e^2 x_{0s}^2 \left\{ \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)+(\frac{1}{2}N+m+1)(\frac{1}{2}N-m)}{N^2} - \frac{(\frac{1}{2}N+m)(\frac{1}{2}N+m-1)+(\frac{1}{2}N-m-1)(\frac{1}{2}N-m)}{N^2(N-1)} \right\} \times \left(n^2 \int_V \mathrm{d}\boldsymbol{x} \int_{V'} \mathrm{d}\boldsymbol{x}' g(R)\, \mathrm{Re}\, \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \right) - 2m\Delta E'_{\mathrm{B}}. \tag{8}$$

$\Delta E'_{\mathrm{B}}$ is the generalized Lamb shift given by (4*b*). Note that only this depends on the presence of free photons. Hence the interparticle propagator is precisely $\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s)$ whether there are free photons or not. This result is to be expected since $\mathbf{F}$ is the Green function for the time Fourier transformed operator Maxwell wave equation and must be independent of the initial states (compare Bullough 1973, § 1): on the other hand the operator self-field which is capable of yielding both the spontaneous emission and Lamb shift correctly is not carried by this photon propagator (cf Bullough 1973 and remarks by (26) below).

Note also that the generalized Lamb shift is not cooperative whilst the shift depending on Re $\mathbf{F}$ certainly is. This cooperative shift vanishes for the top and bottom states $|\frac{1}{2}N, \pm\frac{1}{2}N\rangle$ and is symmetric in m. The generalized Lamb shift is the only shift in the top and bottom states and is antisymmetric in m. In the limit in which all atoms occupy the same site the limit of the shift in terms of Re $\mathbf{F}$ is undefined and should be rejected as spurious as we noted already in case (ii): the key point is that atoms cannot overlap and this is described in (8) by $g(R)$ which vanishes as $R \to 0$.

On the other hand since Re $\mathbf{F}$ is long range and $g(R) \to 1$ as $R \to \infty$ it is necessary to split this shift into 'coherent' and 'incoherent' parts by splitting the integral in (8) as follows:

$$n^2 \int_V \mathrm{d}\boldsymbol{x} \int_{V'} \mathrm{d}\boldsymbol{x}' g(R)\, \mathrm{Re}\, \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} = n^2 V \int (g(R)-1)\, \mathrm{Re}\, \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\, \mathrm{d}\boldsymbol{R} + n^2 \int_V \int_{V'} \mathrm{d}\boldsymbol{x}\, \mathrm{d}\boldsymbol{x}'\, \mathrm{Re}\, \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}}. \tag{9}$$

The first integral on the right side is obtained by appeal to the fact that its value does not depend on $\boldsymbol{x}$ except for a surface layer of points $\boldsymbol{x}$ within about a correlation distance of the surface of V. This is the 'incoherent' shift. The remaining 'coherent' part depends explicitly on the form of V.

First of all the integral over $\boldsymbol{x}'$ in the 'coherent' part double integral is undefined for points $\boldsymbol{x}'$ close to $\boldsymbol{x}$. It is therefore defined as the conditionally convergent integral obtained by extracting a small sphere of vanishingly small radius about $\boldsymbol{x}$. The argument is already familiar from refractive index theory (Rosenfeld 1951, Born and Wolf 1959,

Bullough 1968). It follows that this integral is

$$\frac{4\pi}{3}V+\int_V \mathrm{d}\boldsymbol{x}(\nabla\nabla+k_s^2\mathbf{U}):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\int_{V'}\frac{\exp(\mathrm{i}k_s|\boldsymbol{x}-\boldsymbol{x}'|)}{|\boldsymbol{x}-\boldsymbol{x}'|}\mathrm{d}\boldsymbol{x}'. \tag{10}$$

The term $\frac{4}{3}\pi V$ is equivalent to the effect of a Lorentz dielectric cavity field and shifts the energy appropriately: we shall assign it to the 'incoherent' part of the shift for in this way we can make a comparison with refractive index theory in the linear regime.

The double integral when evaluated for the slab of width c and axis normal to $\hat{\boldsymbol{u}}$, the direction of the atomic dipole x matrix elements, reduces to

$$-4\pi V+4\pi V\frac{\sin k_s c}{k_s c} \tag{11}$$

(with the understanding that $V = Ac$ and that the cross section is very large). The details of the calculations leading to both (10) and (11) are given in the appendix to this paper. With these results the total shift for the slab is therefore†

$$\begin{aligned}\Delta E = {} & -\tfrac{1}{2}e^2x_{0s}^2\left(\frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)+(\frac{1}{2}N-m+1)(\frac{1}{2}N-m)}{N}\right.\\ & \left.-\frac{(\frac{1}{2}N+m)(\frac{1}{2}N+m-1)+(\frac{1}{2}N-m-1)(\frac{1}{2}N-m)}{N(N-1)}\right)\\ & \times\left(-\frac{8\pi}{3}n+4\pi n\frac{\sin k_s c}{k_s c}+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\right)\\ & -2m\Delta E'_{\mathrm{B}}.\end{aligned} \tag{12}$$

The cooperative part of the shift is extensive, that is of order N, only for $m \simeq 0$. For $m = 0$ precisely

$$\Delta E = -\frac{N}{4}e^2x_{0s}^2\left(-\frac{8\pi}{3}n+4\pi n\frac{\sin k_s c}{k_s c}+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\right) \tag{13}$$

with terms O(1) neglected. The cooperative part of the shift vanishes for $m = \pm\frac{1}{2}N$ as noted. For $m = \pm\frac{1}{2}N\mp 1$ the cooperative shift is O(1) whilst the Lamb shift is extensive:

$$\begin{aligned}\Delta E = {} & -e^2x_{0s}^2\left(-\frac{8\pi}{3}n+4\pi n\frac{\sin k_s c}{k_s c}+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\right)\\ & \mp(N-2)\Delta E'_{\mathrm{B}}.\end{aligned} \tag{14}$$

The energy spacings $\mp\hbar\omega_s$ (say) between the perturbed states $|\frac{1}{2}N, \pm\frac{1}{2}N\mp 1\rangle$ and $|\frac{1}{2}N, \pm\frac{1}{2}N\rangle$ are

$$\begin{aligned}\mp\hbar\omega'_s = {} & \mp\hbar\omega_s\pm 2\Delta E'_{\mathrm{B}}-e^2x_{0s}^2\\ & \times\left(-\frac{8\pi}{3}n+4\pi n\frac{\sin k_s c}{k_s c}+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\right).\end{aligned} \tag{15}$$

† Compare Friedberg *et al* (1971).

There is an analogous result for the spacing between the phased Dicke states $|\frac{1}{2}N, \pm\frac{1}{2}N-1; \boldsymbol{k}\rangle$ and $|\frac{1}{2}N; \pm\frac{1}{2}N\rangle$:

$$\Delta E = -\tfrac{1}{2}e^2x_{0s}^2\left\{\frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)+(\frac{1}{2}N-m+1)(\frac{1}{2}N-m)}{N}\right.$$
$$\left.-\frac{(\frac{1}{2}N+m)(\frac{1}{2}N+m-1)+(\frac{1}{2}N-m-1)(\frac{1}{2}N-m)}{N(N-1)}\right\}$$
$$\times\left\{\frac{4\pi}{3}n+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\cos\boldsymbol{k}\,.\,\boldsymbol{R}\,\mathrm{d}\boldsymbol{R}\right.$$
$$\left.+\tfrac{1}{2}nV\hbar(e^2x_{0s}^2)^{-1}\,\mathrm{Re}\,\{I(\boldsymbol{k},k_s)\}\right\}-2m\Delta E_B'. \tag{16}$$

The quantity $I(\boldsymbol{k},k_s)$ is evaluated in the appendix to this paper by the methods used in refractive index theory: the region V is taken to be the slab $-\frac{1}{2}c \leqslant z \leqslant \frac{1}{2}c$ with $\boldsymbol{k}$ along the slab axis and $\hat{\boldsymbol{u}}$ orthogonal to this. There is a difficulty associated with waves reflected from the surface $z = \frac{1}{2}c$ (when $\boldsymbol{k}$ is along the positive z axis) which is analysed in this appendix. Within a prescription deliberately chosen to eliminate reflected waves the real part of $I(\boldsymbol{k},k_s)$ proves to be given by

$$\tfrac{1}{2}nV^2\hbar(ex_{0s})^{-2}\,\mathrm{Re}\,\{I(\boldsymbol{k},k_s)\} = \frac{4\pi nk_s}{k-k_s}-\frac{4\pi nk_s}{c}\frac{\sin c(k-k_s)}{(k-k_s)^2} \tag{17}$$

and this determines the magnitude of the coherent part of the level shift. In the case of a resonant pulse described by excitation by the Dicke state $|r,m;\boldsymbol{k}_s\rangle$ the quantity (17) is, however, undefined and we then define it as its limit as $k \to k_s$. In this case the coherent part of the shift vanishes since the limit of (17) does: it does not vanish if reflected waves are included, however, and the correction due to this can be deduced from the appendix: we ignore this correction here. With this understanding the shift (16) reduces to the incoherent shift there, and in particular the analogue of (15) for the energy spacings adjacent to the top and bottom states becomes

$$\mp\hbar\omega_s' = \mp\hbar\omega_s\pm 2\Delta E_B'-e^2x_{0s}^2$$
$$\times\left\{\frac{4\pi}{3}n+n\int(g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\cos\boldsymbol{k}_s\,.\,\boldsymbol{R}\,\mathrm{d}\boldsymbol{R}\right\}. \tag{18}$$

An essentially identical result can be obtained in linear refractive index theory as we now show.

3. Connections with linear theory

It is well known that linear refractive index theory is equivalent to assuming that atoms behave as oscillators of natural frequencies equal to their excitation frequencies. These excitation frequencies are excitations from the ground state. The theory is equivalent to restricting the whole system of N atoms to single particle excitations. A linearly independent set of these N states can be labelled by distinct wavevectors $\boldsymbol{k}$. These states are those created and annihilated by operators analogous to the operator of (I.22)

now normalized to create and annihilate normalized single atom excitations. These operators are

$$\sigma_{\pm}(\boldsymbol{k}) \equiv \frac{1}{\sqrt{N}} \sum_i \exp(\mp i\boldsymbol{k} \cdot \boldsymbol{x}_i)\sigma_{\pm}^{(i)}. \tag{19}$$

We can suppose N so large that $\boldsymbol{k}$ can be chosen as any vector. The operators (19) satisfy the commutation relations

$$[\sigma_+(\boldsymbol{k}), \sigma_-(\boldsymbol{k}')] = \frac{2}{N} \sum_{i=1}^{N} \exp\{-i(\boldsymbol{k}-\boldsymbol{k}') \cdot \boldsymbol{x}_i\}\sigma_z^{(i)}. \tag{20}$$

The expectation value in the Dicke states $|r, m\rangle$ has the ensemble average

$$\left\langle \frac{2}{N} \sum_{i=1}^{N} \langle r, m|\sigma_z^{(i)}|r, m\rangle \right\rangle_{\text{ave}} \delta_{\boldsymbol{k}\boldsymbol{k}'} = \left\langle \frac{2m}{N} \right\rangle_{\text{ave}} \delta_{\boldsymbol{k}\boldsymbol{k}'}. \tag{21}$$

The expectation value in the Dicke state $|r, m; \boldsymbol{k}_0\rangle$ has the same ensemble average and the result is independent of $\boldsymbol{k}_0$. The right hand side of (21) is $O(2m/N)$ in terms of the argument developed in I. For $m = -\frac{1}{2}N+1$ the ensemble average of the expectation value of the commutator is $-\delta_{\boldsymbol{k}\boldsymbol{k}'} + O(N^{-1})$. To this extent the $\sigma_{\pm}(\boldsymbol{k})$ are boson operators. Similarly for $m = \frac{1}{2}N-1$ the $\sigma_{\pm}(\boldsymbol{k})$ are boson operators; but note now that $\sigma_-(\boldsymbol{k})$ (and not $\sigma_+(-\boldsymbol{k})$) creates a pseudoboson with momentum $\hbar\boldsymbol{k}$ and this pseudoboson consists of one particle de-excitations taken with reference to the fully inverted state. There are thus at least two pseudoboson theories which are those in which transitions take place between the ground state $|\frac{1}{2}N, -\frac{1}{2}N\rangle$ and states of one particle excitations or take place between the fully inverted state $|\frac{1}{2}N, \frac{1}{2}N\rangle$ and states of one particle de-excitations. We shall call these pseudoboson theories for the 'attenuator' and for the 'amplifier' respectively. Refractive index theory is normally concerned with the attenuator but can apparently be extended to include the amplifier.

Since the usual refractive index theory is concerned with one particle excitations above the ground state we can expect that the shift of the resonance frequency in that theory will be close to or even identically equal to the frequency $+\omega_s'$ in (18). There is however a significant difference between refractive index theory and the theory of the N atom system considered so far. The phased Dicke states $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ are approximate eigenstates of the coupled system and are *approximate* normal modes for one particle excitations labelled by their wavevectors $\boldsymbol{k}$. However within linear theory the exact normal modes of the coupled matter photon system are modes labelled by $\boldsymbol{k}$ of definite frequency $\omega(k)$ determined by k. In the case of a finite system of volume V an external field of wavenumber ωc^{-1} excites a particular one of these modes with wavenumber $m(\omega)\omega c^{-1}$: $m(\omega)$ is defined to be the refractive index and, since $m(\omega)\omega = ck$ this is an implicit relation for $\omega(k)$. At the same time the surface of V plays a subtle role. It largely extinguishes the external field inside V according to the optical extinction theorem of Ewald (1912) and has the effect of refracting the wavevector inside V according to Snell's law. This spatially coherent behaviour has the effect of eliminating the coherent part of the shift in (16), that is the part depending on (17): indeed one should include the reflected wave in (17) (see appendix) and it then eliminates this also by generating the appropriate reflected waves at the surfaces of V. In this theory a unique $\boldsymbol{k}$ is thus excited inside V by a system of waves with definite wavevectors $\boldsymbol{k}_0$ outside V; $\boldsymbol{k}$ is preserved therefore and the incoherent part of the shift contains the mode function $\exp(i\boldsymbol{k} \cdot \boldsymbol{x})$.

We find (cf Bullough 1968) that the refractive index $m(\omega)$ satisfies the dispersion relation

$$m^2(\omega)-1 = \frac{4\pi n\alpha(\omega)}{1-\frac{4}{3}\pi n\alpha(\omega)-n\alpha(\omega)R(\omega)}. \tag{22}$$

The quantity $R(\omega)$ is a cluster expansion of which the leading terms have the form

$$\Bigg(\int (g_2(R)-1+n^{-1}\delta(\boldsymbol{R}))\exp(im(\omega)\boldsymbol{k}_0\,.\,\boldsymbol{R})\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}:\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega)\,\mathrm{d}\boldsymbol{R}$$

$$+n\alpha(\omega)\int\int (g_3(\boldsymbol{R},\boldsymbol{R}')-g_2(R)g_2(R'))\exp\{im(\omega)\boldsymbol{k}_0\,.\,(\boldsymbol{R}+\boldsymbol{R}')\}$$

$$\times\hat{\boldsymbol{u}}\,.\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega)\,.\,\mathbf{F}(\boldsymbol{x}',\boldsymbol{x}'';\omega)\,.\,\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\,\mathrm{d}\boldsymbol{R}'$$

$$+\alpha(\omega)\int g_2(R)\hat{\boldsymbol{u}}\,.\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega)\,.\,\mathbf{F}(\boldsymbol{x}',\boldsymbol{x};\omega)\,.\,\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}\Bigg); \tag{23}$$

$g_2(R)$ is the two-atom correlation function previously called $g(R)$ and defined in (I.10); $g_3(\boldsymbol{R},\boldsymbol{R}')$ is an analogous three-atom correlation function; the vector $\boldsymbol{k}_0$ has magnitude $k_0=\omega c^{-1}$ and is in a direction determined by that of the wavevector of the external field. The quantity $\alpha(\omega)$ is the polarizability which for two-level atoms reduces to

$$\alpha(\omega) = 2e^2x_{0s}^2\hbar^{-1}\omega_s(\omega_s^2-\omega^2)^{-1}. \tag{24}$$

The form (24) allows us to rationalize (22) up to two-body correlations in the form

$$m^2(\omega)-1 = \frac{8\pi ne^2x_{0s}^2\omega_s\hbar^{-1}}{\omega_s^2-\omega^2-\frac{8}{3}\pi ne^2x_{0s}^2\omega_s\hbar^{-1}-2e^2x_{0s}^2\omega_s\hbar^{-1}nR(\omega)}. \tag{25}$$

As long as the shift is small enough the resonance occurs at

$$\omega_s' \simeq \omega_s-\tfrac{4}{3}\pi ne^2x_{0s}^2\hbar^{-1}-e^2x_{0s}^2\hbar^{-1}nR(\omega_s) \tag{26}$$

which is then valid up to two-body correlations only. The expression (23) includes three-body correlations explicitly and has indeed been worked to all orders of correlation (Bullough and Hynne 1968, Bullough *et al* 1968). Up to three-body correlation (23) diverges at $\omega=\omega_s$ because $\alpha(\omega)$ diverges there. Thus it is necessary to rationalize by an extra factor $(\omega_s^2-\omega^2)$ and the shift (if this is small) can be obtained as the root of a quadratic in $(\omega_s-\omega)$.

There is the difficulty that all the terms multiplying $\alpha(\omega)$ in (23) depend mildly on the surface of V: this has a significant effect in the theory of fluorescence (Bullough and Hynne 1968, Bullough *et al* 1968), but we shall ignore this awkward problem here. The resonance is in principle also complicated by the fact that $R(\omega)$ depends on $m(\omega)$: we ignore this subtlety also.

Within these terms the essential points are these: the leading term in (23) at $\omega=\omega_s$ has the real part

$$\int (g(R)-1+n^{-1}\delta(\boldsymbol{R}))\cos m(\omega)\boldsymbol{k}_0\,.\,\boldsymbol{R}\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\,\mathrm{d}\boldsymbol{R}; \tag{27}$$

we use the previous notation for $g(R)$. The term in $(g(R)-1)$ combines with the Lorentz

field term in (26) to provide precisely the incoherent part of the shift in (18). The (incoherent) Lamb shift is here given by

$$e^2 x_{0s}^2 \int \operatorname{Re} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \delta(\boldsymbol{x} - \boldsymbol{x}') \, \mathrm{d}\boldsymbol{x}', \tag{28}$$

however. It formally reduces to the long time limit of the neoclassical dynamical shift of Stroud and Jaynes (1970). The pseudoboson model here disagrees with the quantum theory (Bullough and Caudrey 1971, Bullough 1973, Ackerhalt *et al* 1973). The real part of the remaining integrals in (23) appears to be providing three-body incoherent shifts of the same general type as (27) presumably associated with the shift of the excited state: the mode function $\cos m\boldsymbol{k}_0 \cdot (\boldsymbol{R} + \boldsymbol{R}')$ appears with these. The remaining two-body term appears to be an approximation to a contribution of the Casimir–Polder (1948) retarded pair interaction which is known (Bullough 1969) to shift the ground state. An important feature of the shift of this state alone, however, is that it is extensive and is linear in V or N (Bullough 1969). In contrast the one particle excitations shift in (18) is O(1) in the large bracket and is not extensive—presumably because one particle only is excited. The Lamb shift there is extensive however.

Next we notice (23) has an imaginary part and there is a 'radiation rate'. The incoherent rate associated with transitions $|\frac{1}{2}N, -\frac{1}{2}N+1\rangle \to |\frac{1}{2}N, -\frac{1}{2}N\rangle$ is seen from (I.11*a*) to be precisely

$$\Gamma_0 + n2e^2 x_{0s}^2 \hbar^{-1} \int (g(R) - 1) \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \, \mathrm{d}\boldsymbol{R}. \tag{29}$$

The corresponding rate between phased Dicke states $|\frac{1}{2}N; -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ and $|\frac{1}{2}N, -\frac{1}{2}N\rangle$ is

$$\Gamma_0 + n2e^2 x_{0s}^2 \hbar^{-1} \int (g(R) - 1) \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \exp\{\mathrm{i}\boldsymbol{k} \cdot (\boldsymbol{x} - \boldsymbol{x}')\} \, \mathrm{d}\boldsymbol{R}. \tag{30}$$

Likewise the rate between phased Dicke states $|\frac{1}{2}N, -\frac{1}{2}N+1, \alpha; \boldsymbol{k}\rangle$ and $|\frac{1}{2}N, -\frac{1}{2}N\rangle$ can be seen from comparison with (I.19*b*) to be precisely

$$(N-1)^{-1}(N-1)\Gamma_0 - n2e^2 x_{0s}^2 \hbar^{-1} \int (g(R) - 1) \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \exp\{\mathrm{i}\boldsymbol{k} \cdot (\boldsymbol{x} - \boldsymbol{x}')\} \, \mathrm{d}\boldsymbol{R} \tag{31}$$

and is dominated by the term in Γ_0. In the transition (30) the outgoing photons have wavevectors in the direction $\hat{\boldsymbol{k}}$ of $\boldsymbol{k}$; in (31) the outgoing photons change $\hat{\boldsymbol{k}}$. However from $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ only (30) is possible. Because of the invariance of $(g(R)-1)\mathbf{F}$ under $\boldsymbol{R} \equiv \boldsymbol{x} - \boldsymbol{x}' \to -\boldsymbol{R}$ the mode function can be replaced by $\cos \boldsymbol{k} \cdot (\boldsymbol{x} - \boldsymbol{x}') = \cos \boldsymbol{k} \cdot \boldsymbol{R}$. The rate from the leading term in (23) is actually

$$\tfrac{1}{2}\left(\Gamma_0 + 2e^2 x_{0s}^2 \hbar^{-1} n \int (g(R) - 1) \exp(\mathrm{i}m(\omega)\boldsymbol{k}_0 \cdot \boldsymbol{R}) \operatorname{Im} \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \, \mathrm{d}\boldsymbol{R}\right). \tag{32}$$

The factor $\frac{1}{2}$ is to be expected in the resonance width. Otherwise (32) coincides with (30), with the identification $\boldsymbol{k} \equiv m(\omega)\boldsymbol{k}_0$, as we expect.

There is a curious aspect of super-radiance associated with (32): the rate described by it is intensive (O(1) in N) but the fluorescence described by it is extensive. This can easily be seen by observing that $\tau \equiv 2\omega c^{-1} \operatorname{Im}(m(\omega))$ is the extinction coefficient for the intensity of the mode with wavevector $m(\omega)\boldsymbol{k}_0$ emitting scattered photons of frequency

ω. This is directly related to the number of photons per second of this frequency scattered from unit volume which is $I_0\tau/\hbar\omega : I_0$ is essentially the initial intensity of the mode in V. The total number of such photons leaving the volume V per second is then $VI_0\tau/\hbar\omega$. From (32) and (25) this is proportional to

$$Vn\left(\Gamma_0 + 2e^2x_{0s}^2\hbar^{-1}n\int (g(R)-1)\exp(im\boldsymbol{k}_0\,.\,\boldsymbol{R})\,\mathrm{Im}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s)\,\mathrm{d}\boldsymbol{R}\right). \tag{33}$$

The extra factor Vn compared with (32) means that the incoherent fluorescence is super-radiant in the same way that (I.11a), for example, is super-radiant when the excitation number $m \simeq 0$ (allowing for introduction of the mode function, (I.11a) coincides with (33) up to a factor $\frac{1}{4}$, when but only when $m \simeq 0$). Indeed (compare Bullough 1970b) if we recognize the '1' in $(g(R)-1)$ as a consequence of isolation of the coherent part we see that when the N atoms in V are condensed to occupy a region small compared with a wavelength the fluorescence goes as N^2 rather than N. The total fluorescence is always proportional to the input intensity I_0 of the excited pulse.

The 'coherent part' is of course the coherent transmission of intensity in the direction of $\boldsymbol{k}_0$. A resonant mode with $\omega = \omega_s$ does not have wavenumber k_s however. Its wavenumber is $m(\omega_s)\omega_s c^{-1}$ and is determined by the dispersion relation (25). Apparently in consequence (since this is where the mathematics develops differently from the resonant radiation rate theory of I) the coherent rate from the slab is proportional to the area of the slab and the input intensity I_0. It is not extensive (proportional to $N = nV$) and it does not increase linearly (like (I.25)) with the width c of the slab†.

The theory of refractive index just sketched appears to describe the one problem in which a phased Dicke state $|r, m; \boldsymbol{k}\rangle$ can be explicitly excited by an incident 'pulse' and the problem solved essentially exactly. The arguments of this section show that the state excited is the state $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ where $\boldsymbol{k}$ is fixed in magnitude and direction by the frequency and direction of an incident plane wave. They show that the excitation process is an essential part of a proper physical description of the excitation of this state and show moreover that perturbation theory (which does not distinguish k from k_s on resonance) is not sufficient adequately to describe the propagation of the coherent part of the radiation. It is difficult to determine at this stage what features of this dielectric theory will survive in the super-radiance region. There is a close correspondence between the one particle excitation region $m = -\frac{1}{2}N+1$ and the regions of larger m because the photon propagator $\mathbf{F}$ performs directly comparable functions for all values of m. But the $m \simeq 0$ region is a nonlinear region and in this region pulses with resonant carriers satisfying the free field dispersion relation $k_s = \omega_s c^{-1}$ are possible (eg McCall and Hahn 1969).

We now look at the $m \simeq \frac{1}{2}N$ region where the pseudoboson theory of the amplifier could apply. It is important to notice that there is a profound dissymmetry between the two pairs of states $|\frac{1}{2}N, -\frac{1}{2}N\rangle$ with $|\frac{1}{2}N, -\frac{1}{2}N+1\rangle$ and $|\frac{1}{2}N, \frac{1}{2}N\rangle$ with $|\frac{1}{2}N, \frac{1}{2}N-1\rangle$

† According to (I.25) the rate from $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}_0\rangle$ is (on resonance) $6\pi\Gamma_0 nk_s^{-3}x(\frac{1}{2}k_s c) = 4\pi e^2x_{0s}^2\hbar^{-1}nk_s c$ and the intensity is $O(A^{-1})$ where A is the area of the slab. It is therefore negligible compared with the driven coherent intensity which is I_0 corrected only by transmission and reflexion coefficients at the two surfaces of the slab. In the super-radiant region $m \simeq 0$ (I.25) shows that the rate from $|\frac{1}{2}N, m; \boldsymbol{k}_0\rangle$ to $|\frac{1}{2}N, m-1; \boldsymbol{k}_0\rangle$ is $N\pi e^2x_{0s}^2\hbar^{-1}nk_s c$ and the intensity is $\pi e^2x_{0s}^2\hbar^{-1}k_s n^2c^2$. It is interesting to note for comparison that the driven intensity in the pseudoboson theory ($m \simeq -\frac{1}{2}N$) is created by electric fields scattered by a dipole field proportional to $n\alpha$ (where α is given by (24)): it might appear that in this very different region of m the driven intensity is then proportional to n^2 also; but summation of the coherent contributions from the dipole fields eliminates this n^2 dependence.

and thus, by implication, with the pseudoboson theories for the attenuator and the amplifier. From (21) the pseudoboson theory for the attenuator applies to the amplifier with $\alpha(\omega)$ (which depends on $-2mN^{-1}$) changed in sign. This changes the sign of the real energy shift so that the resonance shifts to

$$\hbar\omega_s' = \hbar\omega_s + e^2x_{0s}^2\left(\tfrac{4}{3}\pi n + n\int (g(R)-1)\,\mathrm{Re}\,\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\cos(m(\omega_s)\boldsymbol{k}_s\,.\,\boldsymbol{R})\,\mathrm{d}\boldsymbol{R}\right). \tag{34}$$

The Lamb shift based on (28) is neoclassical and also changes sign. In other respects (34) agrees identically with the shift of the energy spacing between $|\frac{1}{2}N,\frac{1}{2}N\rangle$ and $|\frac{1}{2}N,\frac{1}{2}N-1;\boldsymbol{k}\rangle$ providing $m(\omega_s)=1$.

However, pseudoboson theories of this type exclude the damping and this is apparently not symmetric. The radiative rate between $|\frac{1}{2}N,-\frac{1}{2}N+1;\boldsymbol{k}\rangle$ and $|\frac{1}{2}N,-\frac{1}{2}N\rangle$ is precisely (30) and no other transitions are possible: the fluorescence width is therefore (32) as found. But the radiation rate out of the states $|\frac{1}{2}N,\frac{1}{2}N\rangle$ is precisely $N\Gamma_0$ and not just† (30) because of the $(N-1)$ additive rates of the form (31). The rate from $|\frac{1}{2}N,\frac{1}{2}N\rangle$ to $|\frac{1}{2}N,\frac{1}{2}N-1;\boldsymbol{k}\rangle$ is certainly (30) and agrees with (32) whilst the pseudoboson theory of the amplifier also predicts a fluorescence width (which is a damping width) which is (32).

Thus we have found that in addition to the transition $|\frac{1}{2}N,\frac{1}{2}N\rangle$ to $|\frac{1}{2}N,\frac{1}{2}N-1;\boldsymbol{k}\rangle$ in which matter states may take on the wavevector $\boldsymbol{k}$, there are the transitions (31) which take the same wavevector $\boldsymbol{k}$ but which may change the direction of the outgoing photon direction from $\hat{\boldsymbol{k}}$. These mean that the *total* rate is $N\Gamma_0$—independent of $\boldsymbol{k}$ as it surely should be—and the radiation is both isotropic and independent of the sample geometry. It is intuitive now that the pseudoboson theory of the amplifier is a correct description of the coherent part associated with the driving frequency ω resonant or not of an external field; then within linear theory there must be added the spontaneous emission $N\Gamma_0$ which is $O(N)$ compared with (30). The linear theory is then unstable and evolves rapidly out of the linear regime: thus the pseudoboson theory of the amplifier misses an essential feature.

It seems appropriate to conclude from this that it cannot be correct to develop a theory of super-radiant emission from an extended totally inverted dielectric in terms of a single wavevector $\boldsymbol{k}$ whatever the sample geometry. Equation of motion methods indicate the same feature but the relative significance of different $\boldsymbol{k}$ in this theory is not yet determined.

Some of the results of these two papers seem intuitively obvious; but our purpose in investigating the perturbation theory in depth is to exhibit them explicitly and as far as possible quantitatively. We cannot add to the results in the text of this paper, part II, by summarizing them here. The main conclusions of the paper are summarized in the abstract to it.

Appendix

The radiation rates calculated in both paper I and this paper, II, depend on the imaginary part of the integral

$$I(\boldsymbol{k},k_s) \equiv 2e^2x_{0s}^2\hbar^{-1}V^{-2}\int_V \mathrm{d}\boldsymbol{x}\int_{V'}\mathrm{d}\boldsymbol{x}'\mathbf{F}(\boldsymbol{x},\boldsymbol{x}';\omega_s):\hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\exp\{\mathrm{i}\boldsymbol{k}\,.\,(\boldsymbol{x}-\boldsymbol{x}')\}. \tag{A.1}$$

† As shown in I individual rates with $m\to m-1$ and $-m+1\to -m$ are identical.

The two important cases were $k = 0$ (k is the magnitude of $\boldsymbol{k}$) and $k = k_s \equiv \omega_s c^{-1}$. The case $k = 0$ occurs for rates between simple Dicke states $|r, m\rangle$; the case $k \neq 0$ occurs for transitions between phased Dicke states $|r, m; \boldsymbol{k}\rangle$ (we sometimes used $\boldsymbol{k}_0$ for the vector $\boldsymbol{k}$ in I and do so again whenever we need to emphasize that $\boldsymbol{k}$ is the matter wavevector as opposed to an outgoing photon wavevector); the case $k = k_s$ in particular is the case of resonant transitions between phased Dicke states also preserving $\boldsymbol{k}$.

The energy shifts treated in this paper, II, depend on the real part of the integral $I(\boldsymbol{k}, k_s)$. There is a difficulty in interpreting the complex valued integral within the context of the perturbation theory of these two papers which we now elucidate.

The integral $I(\boldsymbol{k}, k_s)$ is invariant under $\boldsymbol{k} \to -\boldsymbol{k}$. To see this interchange $\boldsymbol{x}$ and $\boldsymbol{x}'$ and observe that $\mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s)$ is invariant under the interchange. If the orders of integration are now reversed $I(\boldsymbol{k}, k_s)$ is transformed to $I(-\boldsymbol{k}, k_s)$. It follows that $I(\boldsymbol{k}, k_s)$ is equally

$$I(\boldsymbol{k}, k_s) = 2e^2 x_{0s}^2 V^{-2} \hbar \int_V \mathrm{d}\boldsymbol{x} \int_{V'} \mathrm{d}\boldsymbol{x}' \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}} \cos\{\boldsymbol{k} \cdot (\boldsymbol{x} - \boldsymbol{x}')\}. \quad \text{(A.2)}$$

Either (A.1) or (A.2) may be evaluated by using Green's theorem to evaluate the integral over (say) $\boldsymbol{x}'$ first. The steps are first to get (from (A.1) for example) the result (and compare (II.10))

$$\int_{V'} \mathrm{d}\boldsymbol{x}' \mathbf{F}(\boldsymbol{x}, \boldsymbol{x}'; \omega_s) \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}') \, \mathrm{d}\boldsymbol{x}'$$
$$= \tfrac{4}{3}\pi \mathbf{U} \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}) + (\nabla\nabla + k_s^2 \mathbf{U}) \int_{V'} \frac{\exp(\mathrm{i}k_s |\boldsymbol{x} - \boldsymbol{x}'|)}{|\boldsymbol{x} - \boldsymbol{x}'|} \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}') \, \mathrm{d}\boldsymbol{x}'. \quad \text{(A.3)}$$

The argument is that the integral over $\boldsymbol{x}'$ is conditionally convergent so that a vanishingly small sphere centred on $\boldsymbol{x}$ must be extracted from the region V' of integration. The tensor operator is then transferred outside the integral at the expense of the additional term $\frac{4}{3}\pi \mathbf{U} \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x})$ (the 'Lorentz field' term). Once the operator is outside the integral the region V' can be closed up because the remaining integral is well defined.

The 'Lorentz field' term contributes $\frac{4}{3}\pi V^{-1} 2e^2 x_{0s}^2 \hbar^{-1}$ to the integral $I(\boldsymbol{k}, k_s)$. As noted below (II.10) we classify this term as part of the 'incoherent' shift (it is certainly real). We do not consider it further in this appendix.

We evaluate the remaining part of (A.3) for the parallel-sided slab

$$-\tfrac{1}{2}c \leqslant z \leqslant \tfrac{1}{2}c, \qquad x^2 + y^2 \leqslant R^2$$

where $R \to \infty$. We consider only the case in which $\boldsymbol{k}$ lies along the z axis: for definiteness we suppose it in the positive z direction. The integral is evaluated in the refractive index theory (Bullough 1962, 1970a). The case in which $\boldsymbol{k}$ is not parallel to the z axis has been treated by Darwin (1924) also within refractive index theory and is reported in Born and Wolf (1959).

From Bullough (1962, 1970a) we have (with $R = |\boldsymbol{x} - \boldsymbol{x}'|$)

$$(\nabla\nabla + k_s^2 \mathbf{U}) \int_{V'} \exp(\mathrm{i}k_s |\boldsymbol{x} - \boldsymbol{x}'|) \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}') \, \mathrm{d}\boldsymbol{x}'$$
$$= \frac{4\pi(k_s^2 \mathbf{U} - \boldsymbol{k}\boldsymbol{k})}{k^2 - k_s^2} \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}) + \frac{(\nabla\nabla + k_s^2 \mathbf{U})}{k^2 - k_s^2}$$
$$\times \left(\int_\Sigma \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}') \, \mathrm{d}\boldsymbol{S} \cdot \nabla\{(\exp \mathrm{i}k_s R) R^{-1}\} \right.$$
$$\left. - (\exp \mathrm{i}k_s R) R^{-1} \, \mathrm{d}\boldsymbol{S} \cdot \nabla \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}') \right). \quad \text{(A.4)}$$

The surface integral is taken over the surface Σ of the slab considered to be simply the surfaces $z = -\frac{1}{2}c$, $z = +\frac{1}{2}c$. The contribution from the surface $|z| \leqslant \frac{1}{2}c$, $x^2+y^2 = R^2$ is neglected as a diffraction pattern correction to the coherent processes (compare Bullough 1962, appendix 1 in particular).

The surface integral proves to be (Bullough 1970a, equation (2.10))

$$-2\pi \exp\{-\tfrac{1}{2}\mathrm{i}(k-k_s)c\} \exp \mathrm{i}k_s z(k_s+k)k_s^{-1}$$
$$-2\pi \exp\{+\tfrac{1}{2}\mathrm{i}(k+k_s)c\} \exp -\mathrm{i}k_s z(k_s-k)k_s^{-1}. \tag{A.5}$$

Note that, although (A.1) (at least) has the single wavevector $\boldsymbol{k}$ up the positive z axis, (A.5) has wavevectors $\pm\boldsymbol{k}_s$ both up and down the z axis: there is a 'reflected' wave.

Since, by assumption, $\hat{\boldsymbol{u}}$ is orthogonal to the z axis, the term in $\boldsymbol{kk}\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{x})$ in (A.4) does not contribute to $I(\boldsymbol{k}, k_s)$. Such a contribution is in any case possible only from longitudinal rather than transverse waves. The result (A.4) now allows us to perform the integration over $\boldsymbol{x}$ and, with the Lorentz field term discarded, the result is

$$I(\boldsymbol{k}, k_s) = 2e^2x_{0s}^2\hbar^{-1}V^{-1}\left\{\frac{4\pi k_s^2}{k^2-k_s^2}+\frac{2\pi\mathrm{i}k_s}{c}\left(\frac{1-\exp\{-\mathrm{i}c(k-k_s)\}}{(k-k_s)^2}\right)\right.$$
$$\left.+\frac{2\pi\mathrm{i}k_s}{c}\left(\frac{1-\exp\{+\mathrm{i}c(k+k_s)\}}{(k+k_s)^2}\right)\right\}. \tag{A.6}$$

From this we find the imaginary and real parts

$$\mathrm{Im}(I(\boldsymbol{k}, k_s)) = 4e^2x_{0s}^2\hbar^{-1}V^{-1}\left(\frac{\sin^2\{\frac{1}{2}(k-k_s)c\}}{\frac{1}{2}c(k-k_s)^2}+\frac{\sin^2\{\frac{1}{2}(k+k_s)c\}}{\frac{1}{2}c(k+k_s)^2}\right) \tag{A.7a}$$

$$\mathrm{Re}\,(I(\boldsymbol{k}, k_s)) = 2e^2x_{0s}^2\hbar^{-1}V^{-1}\left\{\frac{4\pi k_s^2}{k^2-k_s^2}-\frac{2\pi k_s}{c}\left(\frac{\sin c(k-k_s)}{(k-k_s)^2}-\frac{\sin c(k+k_s)}{(k+k_s)^2}\right)\right\}. \tag{A.7b}$$

It will be noticed that the results (A.6) and (A.7) are all invariant under $k \to -k$ in agreement with the analysis from (A.1) to (A.2).

In the case when $k = 0$ we now have

$$\mathrm{Im}(I(\boldsymbol{0}, k_s)) = 8e^2x_{0s}^2\hbar^{-1}V^{-1}\left(\frac{\sin^2\frac{1}{2}k_sc}{\frac{1}{2}k_sc}\right) \tag{A.8a}$$

$$\mathrm{Re}(I(\boldsymbol{0}, k_s)) = 8\pi e^2x_{0s}^2\hbar^{-1}V^{-1}\left(-1+\frac{\sin k_sc}{k_sc}\right). \tag{A.8b}$$

The first of these is identically $\mathscr{D}$ for the same slab exhibited in (I.17a); the second is the shifting term which yields the coherent shift for the slab in a simple Dicke state $|r, m\rangle$ exhibited in equation (12) of this paper II. (The shift $\frac{1}{2}(4\pi n)e^2x_{0s}^2$ is related to a longitudinal shift depending on the square of the 'plasma frequency' ω_p^2 (Bullough 1969).)

In the case when $k = k_s$ (resonance case) we have

$$\mathrm{Im}(I(\boldsymbol{k}_s, k_s)) = 4e^2x_{0s}^2k_s\hbar^{-1}V^{-1}\left(\tfrac{1}{2}c+\frac{\sin^2 k_sc}{2k_s^2c}\right) \tag{A.9a}$$

$$\mathrm{Re}(I(\boldsymbol{k}_s, k_s)) = 2\pi e^2x_{0s}^2\hbar^{-1}V^{-1}k_sc^{-1}\sin 2k_sc. \tag{A.9b}$$

The expression (A.7b) is not defined when $k = k_s$ and we have defined it as the limit $k \to k_s$; since (A.7a) is invariant under $k \to -k$ this is also the limit $k \to -k_s$.

The result (A.9*a*) is not the result which leads to (I.25). It leads in fact to

$$\Gamma_{\text{coh}} = \frac{(\frac{1}{2}N+m)(\frac{1}{2}N-m+1)}{N} 6\pi\Gamma_0 n k_s^{-3}\left(\frac{1}{4}k_s c + \frac{1}{4}\frac{\sin^2 k_s c}{k_s c}\right). \tag{A.10}$$

The result (A.9*b*) is not the result evaluated from equation (II.17) when $k = k_s$.

The reason is that because $I(\boldsymbol{k}, k_s)$ is invariant under $\boldsymbol{k} \to -\boldsymbol{k}$ it is evaluated with an integrand which contains both a 'transmitted' and a 'reflected' wave. Physically this makes complete sense since the *total* radiation rate from V cannot depend on whether waves with wavevectors $\boldsymbol{k}$ or $-\boldsymbol{k}$ transmit. However the actual *direction* of this emission does depend on the choice of $\boldsymbol{k}$ or $-\boldsymbol{k}$. In evaluating (I.25) by the route sketched in the paper I we have to evaluate

$$e^2 x_{0s}^2 \hbar^{-1} k_s \int \mathrm{d}\boldsymbol{k}(\mathbf{U} - \hat{\boldsymbol{k}}\hat{\boldsymbol{k}}) : \hat{\boldsymbol{u}}\hat{\boldsymbol{u}}\delta(k_x)\delta(k_y)\delta(k-k_s)\left(\frac{\sin\{\frac{1}{2}(k_0-k)c\}}{\frac{1}{2}c(k_0-k)}\right)^2 \tag{A.11}$$

where k_0 is the magnitude of the incident wavevector and k refers to outgoing photons.

There is an ambiguity about the product $\delta(k_x)\delta(k_y)\delta(k-k_s)$ since this is

$$\delta(k_x)\delta(k_y)\delta(k_z \pm k_s).$$

But if the integral is to be invariant under $\boldsymbol{k} \to -\boldsymbol{k}$ we need to interpret this as

$$\delta(k_x)\delta(k_y)\delta(k-k_s) = \delta(k_x)\delta(k_y)\tfrac{1}{2}\{\delta(k_z-k_s)+\delta(k_z+k_s)\} \tag{A.12}$$

precisely, and then instead of (I.25) we get (A.10). This now means that photons leave the slab in the two directions normal to the slab with, however, different probabilities. It is still true that this coherent emission is in the direction normal to the slab only which is then parallel or antiparallel to the direction of the wavevector $\boldsymbol{k}_0$.

In the case of a resonant pulse moving with wavevector $\boldsymbol{k}_0$ of magnitude k_s in the sample it might however be argued that the radiation rate is associated with the coherent transmission of energy by the pulse. In this case we need the result that radiation rates are in the direction of $\boldsymbol{k}_0$. We achieve this result on resonance by deliberately choosing the expression

$$\delta(k_x)\delta(k_y)\delta(k-k_s) = \delta(k_x)\delta(k_y)\delta(k_z-k_s) \tag{A.13}$$

instead of (A.12).

In the one case where the problem of the excitation of a phased Dicke state by an incident pulse is soluble, namely the excitation of $|\frac{1}{2}N, -\frac{1}{2}N+1; \boldsymbol{k}\rangle$ in refractive index theory there are no difficulties. Refractive index theory (pseudoboson theory) is an all order perturbation theory: it selects an infinite (but incomplete) set of virtual processes from the perturbation theory. It shows that an excitation with wavevector $\boldsymbol{k}_0$ propagates with that vector: the propagator $\mathbf{F}$ carries the mode function $\exp(i\boldsymbol{k}_0 \cdot \boldsymbol{x}')$ to $\exp(i\boldsymbol{k}_0 \cdot \boldsymbol{x})$. Associated with this is a propagation of intensity in the medium with wavevector $\boldsymbol{k}_0$ also. However, the radiation out of the region into a region without matter occurs from *both* surfaces of the slab if this is the choice for V, and more generally it occurs from all parts of the surface of V (although in this case the matter excitation cannot be labelled by a single wavevector $\boldsymbol{k}_0$). The failure of conservation of energy this surface emission implies is averted by the fact that it is necessary to excite the excitation by a free field incident from outside.

The perturbation theory of these two papers is not capable of this degree of sophistication. For simplicity we have therefore imposed the condition that coherent resonant

radiation occurs solely in the direction of the incident resonant wavevector $\boldsymbol{k}_0$ by, for example, choosing (A.13) rather than (A.12) for the slab. It should however be realized that *if* a system can be placed in a phased Dicke state $|r, m; \boldsymbol{k}_0\rangle$, and the excitation mechanism immediately switched off, the state $|r, m; \boldsymbol{k}_0\rangle$ radiates from (in the case of the slab) both surfaces and will then evolve accordingly. The method of excitation used by Brewer and Shoemaker (1971) seems closely to approach this desideratum.

The argument presented in the texts of both papers therefore uses the following:

$$\mathrm{Im}(I(\boldsymbol{k}, k_s)) = 8e^2x_{0s}^2k_s\hbar^{-1}V^{-1}\frac{\sin^2\frac{1}{2}(k-k_s)c}{\frac{1}{2}c(k-k_s)^2} \tag{A.14a}$$

$$\mathrm{Re}(I(\boldsymbol{k}, k_s)) = 4e^2x_{0s}^2\hbar^{-1}V^{-1}\left(\frac{2\pi k_s}{k-k_s} - \frac{2\pi k_s}{c}\frac{\sin c(k-k_s)}{(k-k_s)^2}\right). \tag{A.14b}$$

On resonance these reduce to $8e^2x_{0s}^2\hbar^{-1}V^{-1}(\frac{1}{2}k_sc)$ in agreement with (I.25) and to zero (in agreement with (II.17)) respectively. The results for $\boldsymbol{k} = 0$ are unchanged and are just (A.8*a*) and (A.8*b*) respectively. The reader should have no difficulty in recalculating the initial rates and shifts appropriate to a slab placed initially in the phased Dicke state $|r, m; \boldsymbol{k}_0\rangle$ if he requires them. The arbitrary choices (A.14) rather than (A.7) in no way affect the conclusions of the two papers and simplify their presentation.

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