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Initiation of superfluorescence in a large sphere

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We investigate the early stages of superfluorescent emission from a large spherical volume containing atoms all initially in the same excited state. The electric polarization fluctuations that characterize a fully inverted atomic system, and which trigger superfluorescence, are found to cause field amplification only in vector spherical-harmonic multipole modes of limited order. The field and polarization fluctuations obey Gaussian statistics, a result that has been used to calculate the angular correlation of intensities radiated in any two directions. Components of the electric field radiated in two arbitrary directions and circularly polarized in opposite senses are found to be uncorrelated. They thus have no intensity-intensity or higher-order correlations. Field components which are circularly polarized in the same sense, however, are shown to be correlated over an angular range that is proportional to the ratio of the wavelength to the radius of the sphere.

I. INTRODUCTION

Superfluorescence is a spontaneous emission process in which a large number of atoms, all initially in the same excited state, radiate coherently. Several theoretical papers[1–4] have studied this phenomenon in detail for the cylindrical geometry in which measurements have thus far been made.[5] Nearly all of the calculations which have been carried out analytically, and most of the numerical ones, have assumed the problem to be spatially one dimensional. They have thereby described the radiated pulse as propagating and amplifying in a single direction. The simplifying feature of cylindrical geometry is that nearly all of the superfluorescent radiation is confined to the end-fire modes. It is worth emphasizing, however, that superfluorescence is a three-dimensional phenomenon; it is capable of producing pulse fields with interesting correlation properties over a continuum of propagation directions. The simplest geometry with which to investigate these is probably the spherical one to which we shall devote the present paper.

Because the radiation rate varies as the field depletes the atomic excitation present, the equations which describe superfluorescence are intrinsically nonlinear. In the early stages of the emission process, however, before the field has become intense enough to influence the atoms appreciably, the equations are still in effect linear and can be solved analytically. Much can be learned from exact calculations which cover this linear phase since it lasts for most of the time interval prior to the pulse peak. The linear regime indeed contains all the quantum fluctuations and amplification phenomena which characterize the initially slow buildup of the superfluorescent pulse. Statistical predictions for the field strengths which hold in this appreciable interval should be quite accessible to experimental verification.

Two greatly simplified analyses of a similar collective radiation problem have been carried out by Ernst and Stehle[6] and by Rehler and Eberly.[7] Although these analyses do address the nonlinear aspects of the problem, they are based in effect upon mean-field approximations which do not accurately represent the time dependence of the process or the polarization properties of the field.

The vector nature of the electromagnetic field plays a more central role in the analysis of the spherical problem than the cylindrical one. We shall find it convenient to regard the field as an expansion in vector spherical harmonics of both the electric and magnetic types and shall calculate the amplification rates for each of these. We shall show that only spherical harmonics up to a certain limiting order undergo significant amplification and their intensities increase with time t as $t^{-1}\exp(\text{const}\times t^{1/2})$, a result rather different from the time dependences found in Refs. 6 and 7. Those of higher order never become appreciably excited.

The linearity of the problem we discuss makes it possible to analyze some statistical aspects of the emission process. The atomic polarization fluctuations in the initial, fully inverted state of the atoms will be shown to have a Gaussian distribution. The electromagnetic fields radiated at early times may be expressed as time-dependent linear combinations of the initial polarization variables. The field amplitudes therefore tend also to have a Gaussian quasiprobability distribution. Components of the radiation field which are circularly polarized in opposite senses will be shown to be uncorrelated and therefore to have no intensity-intensity or higher-order correlations. Components which are circularly polarized in the same sense, on the other hand, show nonzero correlation. For them we calculate an expression for the angular dependence of the intensity-intensity correlation function. The angular range of such a correlation is found to be proportional to the ratio of the wavelength to the radius of the radiating sphere.

Since in the linear domain the fluctuations of field amplitude have a Gaussian probability distribution, it follows that the angular range of correlation must likewise be small for the entire hierarchy of intensity correlation functions. In other words, the initial radiation from a large sphere in any single experiment tends to be concentrated along a single ray that points in a random direction.

II. THE ATOMIC MODEL AND EQUATIONS OF MOTION

The radiating system we discuss consists of N identical atoms, each with two energy levels separated by an energy $\hbar\omega_0$. We take the atoms to be uniformly distributed over a spherical volume of radius R. To secure a simple description of the initial atomic state we may for definiteness take the upper of the two energy levels to correspond to an orbital S state $|s\rangle$. Then if, for example, the lower of the two states is a P state, the radiation will proceed as an electric dipole transition. The P state must, however, be triply degenerate to preserve rotational symmetry (Fig. 1). (An alternative initial state could be the spherically symmetrically excited triply degenerate P states that lie above the nondegenerate S state to which transitions occur. Later in this section we shall show that the linear equations that describe the early stages of this alternative type of radiative process are essentially the same.) To describe transitions between the S state and the three P states we introduce the electric dipole moment operator for the jth atom

$$\boldsymbol{\mu}_j=\mu(\boldsymbol{\sigma}_j^+ +\boldsymbol{\sigma}_j^-) . \tag{1}$$

The Cartesian components of $\boldsymbol{\sigma}_j^\pm$ may be regarded as the raising and lowering operators connecting the upper state $|s\rangle$ with the three lower states $|p_a\rangle$ ($a=x,y,z$), i.e., they are defined as

$$\begin{aligned}\boldsymbol{\sigma}_j^+&=(|s\rangle_{j\,j}\langle p_x| \;,\; |s\rangle_{j\,j}\langle p_y| \;,\; |s\rangle_{j\,j}\langle p_z|)\\ &=(\boldsymbol{\sigma}_j^-)^\dagger ,\end{aligned} \tag{2}$$

so that there are three equal dipole matrix elements

$$\mu={}_j\langle s|x|p_x\rangle_j={}_j\langle s|y|p_y\rangle_j={}_j\langle s|z|p_z\rangle_j . \tag{3}$$

By appropriately fixing the phases of the states $|p_a\rangle$ we may determine μ to be real.

The polarization density $\mathbf{P}(\mathbf{r},t)$ in the sample is easily expressed in terms of $\boldsymbol{\sigma}_j^\pm$. If we write

$$\mathbf{P}(\mathbf{r},t)=\mathbf{P}^+(\mathbf{r},t)+\mathbf{P}^-(\mathbf{r},t) , \tag{4}$$

we may let

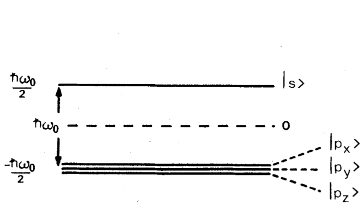

FIG. 1. Atomic-level structure. The strictly degenerate p orbitals are shown as nondegenerate for ease of visualization.

$$\mathbf{P}^+(\mathbf{r},t)=[\mathbf{P}^-(\mathbf{r},t)]^\dagger=\mu\sum_j \boldsymbol{\sigma}_j^+\delta^{(3)}(\mathbf{r}-\mathbf{r}_j) . \tag{5}$$

The Hamiltonian H of the system is given by

$$H=(H_0)_{\rm atom}+(H_0)_{\rm rad}+H_{\rm int} , \tag{6}$$

in which $(H_0)_{\rm atom}$ and $(H_0)_{\rm rad}$ are the free Hamiltonians for the atomic and the radiation systems, and $H_{\rm int}$ is the interaction Hamiltonian which is given in the dipole approximation by

$$H_{\rm int}=-\mu\sum_j[\boldsymbol{\sigma}_j^+\cdot\mathbf{E}^{(+)}(\mathbf{r}_j,t)+\boldsymbol{\sigma}_j^-\cdot\mathbf{E}^{(-)}(\mathbf{r}_j,t)] . \tag{7}$$

The operators $\mathbf{E}^{(\pm)}(\mathbf{r},t)$ are the positive- and negative-frequency parts of the electric field. In obtaining Eq. (7) we have neglected the antiresonant terms $\boldsymbol{\sigma}_j^+\cdot\mathbf{E}^{(-)}$ and $\boldsymbol{\sigma}_j^-\cdot\mathbf{E}^{(+)}$ which oscillate at frequencies close to $2\omega_0$ in the interaction picture.

For completeness, we also introduce the atomic inversion operator σ_j^3, defined as

$$\sigma_j^3=(|s\rangle_{j\,j}\langle s|-\tfrac{1}{2}) , \tag{8}$$

and the corresponding inversion density operator

$$R_3(\mathbf{r},t)=\sum_{j=1}^N \sigma_j^3\delta^{(3)}(\mathbf{r}-\mathbf{r}_j) . \tag{9}$$

The commutation rules for $\boldsymbol{\sigma}_j^\pm$ and σ_j^3 may be obtained from their definitions, (2) and (8), and are

$$[\boldsymbol{\sigma}_j^\pm,\sigma_{j'}^3]=\mp\delta_{jj'}\boldsymbol{\sigma}_j^\pm , \tag{10}$$

$$[\sigma_{ja}^+,\sigma_{j'b}^-]=\delta_{jj'}(\delta_{ab}|s\rangle_{j\,j}\langle s|-|p_b\rangle_{j\,j}\langle p_a|) \tag{11}$$

$(a,b=x,y,z)$.

With the aid of these commutators the Heisenberg equations of motion for the polarization and population inversion densities $\mathbf{P}^+(r,t)$ and $R_3(r,t)$ are readily found to be

$$\begin{aligned}\frac{\partial}{\partial t}\mathbf{P}^+(\mathbf{r},t)&=i\omega_0\mathbf{P}^+(\mathbf{r},t)+\frac{i\mu^2}{\hbar}\mathbf{E}^{(-)}(\mathbf{r},t)\left[R_3(\mathbf{r},t)+\frac{n_0}{2}\right]\\ &\quad-\frac{i\mu^2}{\hbar}\mathbf{E}^{(-)}(\mathbf{r},t)\cdot\mathbf{R}(\mathbf{r},t) ,\end{aligned} \tag{12}$$

$$\frac{\partial}{\partial t}R_3(\mathbf{r},t)=\frac{i}{\hbar}[\mathbf{P}^+(\mathbf{r},t)\cdot\mathbf{E}^{(+)}(\mathbf{r},t)-\mathbf{P}^-(\mathbf{r},t)\cdot\mathbf{E}^{(-)}(\mathbf{r},t)] . \tag{13}$$

In these equations n_0 is the number density of atoms in the sphere, and $\mathbf{R}(r,t)$ is a 2nd-rank tensor operator with components

$$R_{ab}=\sum_j |p_a\rangle_{j\,j}\langle p_b|\delta^{(3)}(\mathbf{r}-\mathbf{r}_j) . \tag{14}$$

These equations are supplemented by the field equation

$$\nabla\times\nabla\times\mathbf{E}^{(\pm)}(\mathbf{r},t)=-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}[\mathbf{E}^{(\pm)}(\mathbf{r},t)+\mathbf{P}^{\mp}(\mathbf{r},t)] \tag{15}$$

which completes the system of equations that describes the general atom-field interaction. Specification of the initial state (complete atomic inversion, photon vacuum) and the Sommerfeld radiation condition (outgoing waves at infinity) are then sufficient to determine both $\mathbf{E}^{(\pm)}(r,t)$ and $\mathbf{P}^{\pm}(r,t)$ and the expectation values of all their products uniquely.

The superfluorescent radiation process evolves via linear equations as long as the population inversion can be assumed to be only insignificantly depleted. Specifically then, for such early times

$$R_3(\mathbf{r},t) \simeq R_3(\mathbf{r},0) = \frac{n_0}{2}\ , \quad \mathbf{R}(\mathbf{r},t) \simeq \mathbf{R}(\mathbf{r},0) = 0\ . \tag{16}$$

This approximation reduces Eq. (12) and its conjugate to the form

$$\frac{\partial}{\partial t}\mathbf{P}^{\pm}(\mathbf{r},t) = \pm i\omega_0 \mathbf{P}^{\pm}(\mathbf{r},t) \pm (i\mu^2 n_0/\hbar)\mathbf{E}^{(\mp)}(\mathbf{r},t)\ . \tag{17}$$

Another type of atomic transition, as we have noted earlier, one in which the triply degenerate P states lie above the S state, can be treated by means of the same set of equations. Formally the same linearized equation (17) is then obtained if the system is initially in the spherically symmetric condition in which an incoherent mixture of equal amounts of the three P states is present. It is only necessary in that case to make an appropriate interchange of the roles of S and P states in the definition of $\sigma^{\pm}$ and to replace μ^2 by $\mu^2/3$. This initial state is represented by the atomic density operator ρ_0 given by

$$\rho_0 = \tfrac{1}{3}(\,|p_x\rangle\langle p_x| + |p_y\rangle\langle p_y| + |p_z\rangle\langle p_z|\,)\ .$$

In this case only two-thirds of the atoms are capable of radiating in any given direction. Specifically, if the z axis is chosen to be along the direction of observation then only transitions from the $l=1$, $m=\pm 1$ states to the $l=0$, $m=0$ states may occur giving rise to equal amounts of left- and right-circularly polarized radiation.

III. THE SOLUTION PROCEDURE AND DETAILS OF SOLUTION

In view of the spherical symmetry of the problem, we can solve the coupled equations (15) and (17) by expanding all operators in spherical harmonic modes, Y_{lm}, each specified by the type (electric or magnetic multipole) and the indices (l,m). The smallest value of l that occurs in the expression for radiation fields is $l=1$, which corresponds to dipole radiation. For a sample of radius $R \gg \lambda_0 = 2\pi c/\omega_0$, as we shall see, many higher values of l are just as important.

Equation (15) does not take precisely the form of a wave equation due to the presence of $\nabla\times\nabla\times\mathbf{E}$ rather than $-\nabla^2\mathbf{E}$. The difference $\nabla\times\nabla\times\mathbf{E}-(-\nabla^2\mathbf{E}) = \nabla(\nabla\cdot\mathbf{E})$ is nonzero, for one of the Maxwell equations implies

$$\nabla(\nabla\cdot\mathbf{E}) = -\nabla(\nabla\cdot\mathbf{P})\ ,$$

the right-hand side of which does not vanish in general. A convenient method of solution proceeds by transforming away locally at every point the $\nabla(\nabla\cdot\mathbf{E})$ term, thereby reducing Eq. (15) to a simple wave equation which can then be solved by using scalar Green's-function techniques.

There are two useful and convenient linear differential operators that can accomplish this transformation, viz. $\mathbf{L}\cdot$ and $\mathbf{L}\cdot\nabla\times$ where $\mathbf{L}=(1/i)\mathbf{r}\times\nabla$ is the angular momentum operator. These operators both yield zero when applied to a gradient. Since they also commute with ∇^2, both $\mathbf{L}\cdot\mathbf{E}^{(\pm)}$ and $\mathbf{L}\cdot\nabla\times\mathbf{E}^{(\pm)}$ satisfy the same scalar wave equation:

$$\left[\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\eta^{(\pm)} = \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\phi^{\mp}\ , \tag{18}$$

where

$$\eta^{(\pm)} = \mathbf{L}\cdot\mathbf{E}^{(\pm)} \ \text{ or } \ \mathbf{L}\cdot\nabla\times\mathbf{E}^{(\pm)} \tag{19a}$$

and correspondingly

$$\phi^{\mp} = \mathbf{L}\cdot\mathbf{P}^{\mp} \ \text{ or } \ \mathbf{L}\cdot\nabla\times\mathbf{P}^{(\mp)}\ . \tag{19b}$$

In view of the Maxwell equations $\nabla\times\mathbf{E}^{(\pm)} \simeq \pm(i\omega_0/c)\mathbf{B}^{(\pm)}$, $\nabla\times\mathbf{B}^{(\pm)} \simeq \mp(i\omega_0/c)\mathbf{E}^{(\pm)}$, we note that $\mathbf{L}\cdot\mathbf{E}^{(\pm)} \sim \mathbf{r}\cdot\nabla\times\mathbf{E}^{(\pm)} \sim \mathbf{r}\cdot\mathbf{B}^{(\pm)}$. We likewise have $\mathbf{L}\cdot\nabla\times\mathbf{E}^{(\pm)} \sim \mathbf{r}\cdot\nabla\times\mathbf{B}^{(\pm)} \sim \mathbf{r}\cdot\mathbf{E}^{(\pm)}$. We are thus in fact solving for the radial components of $\mathbf{B}^{(\pm)}$ and $\mathbf{E}^{(\pm)}$. A knowledge of the radial components $\mathbf{r}\cdot\mathbf{E}$ and $\mathbf{r}\cdot\mathbf{B}$ uniquely determines the electric and magnetic multipole coefficients and therefore according to a theorem demonstrated later in this section determines the full vectors $\mathbf{E}$ and $\mathbf{B}$ in the free-space exterior to the spherical volume. We now proceed to the actual solution.

We consider first $\mathbf{L}\cdot$ or $\mathbf{L}\cdot\nabla\times$ acting on Eq. (17). We may write the result for both cases as

$$\left[\frac{\partial}{\partial t} \mp i\omega_0\right]\phi^{\pm}(\mathbf{r},t) = \pm\frac{i\mu^2 n_0}{\hbar}\eta^{(\mp)}(\mathbf{r},t)\ . \tag{20}$$

We next solve Eq. (18) with appropriate initial and boundary conditions. We may write the retarded solution as

$$\eta^{(\mp)}(\mathbf{r},t) = \eta^{(\mp)}_{\text{vac}}(\mathbf{r},t) - (4\pi c^2)^{-1}\int \frac{(\partial^2/\partial t^2)\phi^{\pm}(\mathbf{r}',t-|\mathbf{r}-\mathbf{r}'|/c)}{|\mathbf{r}-\mathbf{r}'|}d^3\mathbf{r}'\ , \tag{21}$$

in which $\eta^{(\mp)}_{\text{vac}}(\mathbf{r},t)$ is the solution of the scalar wave equation (18) in vacuum ($\phi^{\pm}=0$). We may note here that $\eta^{(\mp)}_{\text{vac}}$ (and $\eta^{(\mp)}$ in general) contain no remnants of the longitudinal part of $\mathbf{E}^{(\mp)}_{\text{vac}}$ (and $\mathbf{E}^{(\mp)}$). This is because the operator $\nabla\times$ annihilates any longitudinal fields expressible as gradients. In other words, $\eta^{(\mp)}_{\text{vac}}$ is expressible entirely in terms of the freely evolving transverse photon annihilation and creation operators.

By removing the rapidly oscillating factors $\exp(\pm i\omega_0 t)$, we can define more slowly varying amplitudes $\Pi^{\pm}$ and $\Upsilon^{(\mp)}$ for $\phi^{\pm}$ and $\eta^{(\mp)}$:

$$\phi^{\pm}(\mathbf{r},t)=\Pi^{\pm}(\mathbf{r},t)\exp(\pm i\omega_0 t)\ ,$$
$$\eta^{(\mp)}(\mathbf{r},t)=\Upsilon^{(\mp)}(\mathbf{r},t)\exp(\pm i\omega_0 t)\ . \tag{22}$$

If we then make the slowly varying envelope approximation by assuming

$$\left|\frac{\partial^2}{\partial t^2}\Pi^{\pm}\right| \ll \omega_0 \left|\frac{\partial}{\partial t}\Pi^{\pm}\right| \ll \omega_0^2 \Pi^{\pm}\ , \tag{23}$$

etc., we see that Eqs. (20) and (21) assume simpler forms when written in terms of $\Pi^{\pm}$ and $\Upsilon^{(\mp)}$. These can then be combined into the single equation for $\Pi^{\pm}$,

$$\frac{\partial}{\partial t}\Pi^{\pm}(\mathbf{r},t)=\pm\frac{i\mu_0^2 n_0}{\hbar}\Upsilon^{(\mp)}_{\rm vac}(\mathbf{r},t)\pm\frac{i\mu^2 n_0 k_0^2}{4\pi\hbar}\int \Pi^{\pm}\left[\mathbf{r}',t-\frac{|\mathbf{r}-\mathbf{r}'|}{c}\right]\exp(\mp ik_0|\mathbf{r}-\mathbf{r}'|)/|\mathbf{r}-\mathbf{r}'|\,d^3\mathbf{r}'\ . \tag{24}$$

It is sufficient to solve Eq. (24) for the lower sign only; the solution for the upper sign is just the Hermitian conjugate. We begin by resolving Π^- and $\Upsilon^{(+)}$ into the normalized spherical-harmonic modes $Y_{lm}(\theta,\phi)$:

$$\Pi^-(\mathbf{r},t)=\sum_{l,m} c_{lm}^-(r,t)Y_{lm}(\theta,\phi)\ ,$$
$$\Upsilon^{(+)}(\mathbf{r},t)=\sum_{l,m} e_{lm}^{(+)}(r,t)Y_{lm}(\theta,\phi)\ , \tag{25}$$

where $\sum_{l,m}$ stands for $\sum_{l=0}^{\infty}\sum_{m=-l}^{l}$ and $r=|\mathbf{r}|$. Substitution of these expansions in Eq. (24) and use of the orthogonality property of the Y_{lm} functions immediately yield

$$\frac{\partial}{\partial t}c_{lm}^-(r,t)+i\Lambda_0 e_{lm}^{(0)}(r,t)=-\frac{ik_0^2\Lambda_0}{4\pi}\int_0^R dr' r'^2\sum_{l',m'}\int d^2\Omega\int d^2\Omega'\frac{\exp(ik_0|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|}c_{l'm'}^-\left[r',t-\frac{|\mathbf{r}-\mathbf{r}'|}{c}\right]$$
$$\times Y_{lm}^*(\theta,\phi)Y_{l'm'}(\theta',\phi')\ , \tag{26}$$

where

$$\Lambda_0=\frac{n_0\mu^2}{\hbar}\ , \tag{27}$$

$d^2\Omega$, $d^2\Omega'$ are elements of the solid angle, and $e_{lm}^{(0)}(r,t)$ are the coefficients of $\zeta_{\rm vac}^{(+)}(\mathbf{r},t)$ in the sense of Eq. (25).

We can rewrite Eq. (26) in terms of the Laplace transforms

$$\tilde{c}_{lm}^-(r,s)=\int_0^\infty c_{lm}^-(r,t)\exp(-st)dt\ ,$$
$$\tilde{e}_{lm}^{(0)}(r,s)=\int_0^\infty e_{lm}^{(0)}(r,t)\exp(-st)dt\ . \tag{28}$$

Since $c_{lm}^-(r',t')$ vanishes for $t'<0$, the Laplace transform of

$$c_{lm}^-\left[r',t-\frac{|\mathbf{r}-\mathbf{r}'|}{c}\right]$$

is

$$\tilde{c}_{lm}^-(r',s)\exp(-s\,|\mathbf{r}-\mathbf{r}'|/c)\ .$$

In terms of Laplace transforms, Eq. (26) simplifies considerably, for we may now carry out the Ω,Ω' integrations explicitly. This we do by using the addition theorem[8]

$$\exp(i\beta_s|\mathbf{r}-\mathbf{r}'|)/|\mathbf{r}-\mathbf{r}'|$$
$$=4\pi i\beta_s\sum_{l,m} j_l(\beta_s r^<)h_l^{(1)}(\beta_s r^>)Y_{lm}(\theta,\phi)Y_{lm}^*(\theta',\phi')\ , \tag{29}$$

where $r^{<,>}$ = smaller (greater) of r,r', and $j_l(x),h_l^{(1)}(x)$ are the spherical Bessel functions[8] of order l. The equation for the transforms of the coefficients is then

$$s\tilde{c}_{lm}^-(r,s)-c_{lm}^-(r,0)+i\Lambda_0\tilde{e}_{lm}^{(0)}(r,s)$$
$$=\Lambda_0 k_0^2\beta_s\int_0^R dr' r'^2\tilde{c}_{lm}^-(r',s)j_l(\beta_s r^<)h_l^{(1)}(\beta_s r^>)\ , \tag{30}$$

in which we have written

$$\beta_s=k_0+is/c\ . \tag{31}$$

We note that in the passage from Eq. (26) to Eq. (30) the different angular momentum (l,m) modes have become decoupled. The spherical symmetry of the problem ensures that the different spherical-harmonic modes evolve independently.

We can solve Eq. (30) by using the Green's-function relation

$$[\nabla^2+\beta_s^2-l(l+1)/r^2][j_l(\beta_s r^<)h_l^{(1)}(\beta_s r^>)]$$
$$=i\delta(r-r')/\beta_s r'^2\ , \tag{32}$$

which is easily derived from the relation

$$(\nabla^2+\beta_s^2)[\exp(i\beta_s|\mathbf{r}-\mathbf{r}'|)/|\mathbf{r}-\mathbf{r}'|]=-4\pi\delta^{(3)}(\mathbf{r}-\mathbf{r}')$$

together with Eq. (29). We now apply the operator $\nabla^2+\beta_s^2-l(l+1)/r^2$ to Eq. (30) and thereby obtain

$$[\nabla^2+\gamma_s^2-l(l+1)/r^2] \times[s\tilde{c}_{lm}^{-}(r,s)-c_{lm}^{-}(r,0)+i\Lambda_0 e_{lm}^{(0)}(r,s)] = i\Lambda_0 k_0^2[c_{lm}^{-}(r,0)-i\Lambda_0\tilde{e}_{lm}^{(0)}(r,s)]/s\ , \quad (33)$$

in which we have written

$$\gamma_s^2=\beta_s^2-i\Lambda_0 k_0^2/s\ . \quad (34)$$

We may regard Eq. (33) as an explicit equation for the unknown $\tilde{c}_{lm}^{-}(r,s)$. If β_s in the definition (32) of the Green's function is replaced by γ_s, we may use that definition to construct for Eq. (33) the solution

$$s\tilde{c}_{lm}^{-}(r,s)-c_{lm}^{-}(r,0)+i\Lambda_0 e_{lm}^{(0)}(r,s) = \Lambda_0 k_0^2\int_0^R [c_{lm}^{-}(r',0)-i\Lambda_0\tilde{e}_{lm}^{(0)}(r',s)] \times\tilde{L}_l(r^<,r^>;s)r'^2dr'\ , \quad (35)$$

where the Green's function is written as

$$\tilde{L}_l(r^<,r^>;s)=\gamma_s j_l(\gamma_s r^<)h_l^{(1)}(\gamma_s r^>)/s\ . \quad (36)$$

The Laplace inversion of Eq. (35) is formally straightforward. It leads to

$$\frac{\partial}{\partial t}c_{lm}^{-}(r,t)=-i\Lambda_0 e_{lm}^{(0)}(r,t)+\Lambda_0 k_0^2\left[\int_0^R c_{lm}^{-}(r',0)L_l(r^<,r^>;t)r'^2dr'-i\Lambda_0\int_0^R dr'r'^2\int_0^t dt' e_{lm}^{(0)}(r',t')L_l(r^<,r^>;t-t')\right], \quad (37)$$

in which

$$L_l(r^<,r^>;t)=\mathscr{L}^{-1}(\tilde{L}_l(r^<,r^>;s))\ , \quad (38)$$

where $\mathscr{L}^{-1}$ denotes the inverse Laplace transform. Equation (37) is the formal solution for the time derivative of the polarization coefficient operator $\dot{c}_{lm}^{-}(r,t)$ for $r<R$. But since from Eqs. (20), (22), and (25)

$$e_{lm}^{(+)}(r,t)=i\Lambda_0^{-1}(\partial/\partial t)c_{lm}^{-}(r,t)\ , \quad (39)$$

we obtain an explicit expression for the electric field variable $e_{lm}^{(+)}(r,t)$ inside the sphere $(r<R)$. It is given by

$$e_{lm}^{(+)}(r,t)=e_{lm}^{(0)}(r,t)+ik_0^2\left[\int_0^R c_{lm}^{-}(r',0)L_l(r^<,r^>;t)r'^2dr'-i\Lambda_0\int_0^R dr'r'^2\int_0^t dt' e_{lm}^{(0)}(r',t')L_l(r^<,r^>;t-t')\right]. \quad (40)$$

Equations (37) and (40) embody the physical mechanism of the linear initiation process. The fluctuating initial polarization and vacuum field specified by the coefficients $c_{lm}^{-}(r',0)$ and $e_{lm}^{(0)}(r',t')$, respectively, act to induce the cooperative quantum decay of the atomic excitation. They are both needed to describe correctly the dynamics of the operators $c_{lm}^{-}(r',t)$ and $e_{lm}^{(+)}(r',t)$. In expectation values of normally ordered products of the coefficient operators, however, the vacuum field operators $e_{lm}^{(0)}(r',t')$ make no contribution, for the initial state has no photons present. We shall employ the normal-ordering scheme, since it is the one most naturally adapted to the description of the usual forms of photon detection by absorption.[9] Inasmuch as the vacuum field operators do not contribute to the expectation values which interest us, we shall take the liberty of dropping them from Eqs. (37) and (40) and the others that follow. Their omission entails no error apart from the consideration of vacuum fluctuations. With this understanding we obtain

$$e_{lm}^{(+)}(r,t)=i\Lambda_0^{-1}(\partial/\partial t)c_{lm}^{-}(r,t)=ik_0^2\int_0^R c_{lm}^{-}(r',0)L_l(r^<,r^>;t)r'^2dr'\ . \quad (41)$$

From Eqs. (21)—(23) we can also calculate $e_{lm}^{(+)}(r,t)$ outside the sphere $(r>R)$. Its Laplace transform is given by

$$\tilde{e}_{lm}^{(+)}(r,s)=ik_0^2\beta_s\left[\int_0^R dr'r'^2\tilde{c}_{lm}^{-}(r',s)j_l(\beta_s r')\right]h_l^{(1)}(\beta_s r)\ , \quad (42)$$

where we have once again used the identity (29). With the aid of Eq. (35), Eq. (42) may be expressed as

$$\tilde{e}_{lm}^{(+)}(r,s)=\frac{ik_0^2\beta_s}{s}\left[\int_0^R dr'r'^2\left[c_{lm}^{-}(r',0)+\Lambda_0 k_0^2\int_0^R c_{lm}^{-}(r'',0)\tilde{L}_l(r^{\ll},r^{\gg};s)r''^2dr''\right]j_l(\beta_s r')\right]h_l^{(1)}(\beta_s r)\ , \quad (43)$$

where $r^{\ll,\gg}$ = smaller (larger) of r',r''.

The first term in the large parentheses in Eq. (43) represents the independent-atom spontaneous emission (intensity proportional to N) whereas the second term represents the evolution of the cooperative radiation process (intensity proportional to N^2). We expect the intensity contributed by the cooperative emission to dominate the independent-atom radiation at all save the earliest times. In studying the asymptotic behavior of intensities at large times in the linear regime we may therefore drop the first term of Eq. (43). We assume further that detection takes place in the far field $(r\gg R)$, for which we have

$$h_l^{(1)}(\beta_s r)\simeq(-i)^{l+1}\exp(i\beta_s r)/\beta_s r=(-i)^{l+1}\exp(ik_0 r)\exp(-sr/c)/\beta_s r\ .$$

The Laplace inversion of Eq. (43) is now easily carried out for $r\gg R$ and we obtain

$$e_{lm}^{(+)}(r,t)\simeq\frac{(-i)^l k_0^4\Lambda_0}{r}\exp(ik_0 r)\int_0^R dr' r'^2\int_0^R dr'' r''^2 c_{lm}^-(r'',0)L_l'(r^{\ll},r^{\gg};t-r/c)\,, \tag{44}$$

where $L_l'(r^{\ll},r^{\gg};t)$ is the Laplace inverse of $s^{-1}\tilde{L}_l(r^{\ll},r^{\gg};s)j_l(\beta_s r')$, i.e.,

$$L_l'(r^{\ll},r^{\gg};t)=(2\pi i)^{-1}\int_{\epsilon-i\infty}^{\epsilon+i\infty}s^{-1}\exp(st)\tilde{L}_l(r^{\ll},r^{\gg};s)j_l(\beta_s r')ds\,. \tag{45}$$

We must now evaluate the evolution kernels L_l and L_l' in order to obtain quantitative predictions. Their transforms $\tilde{L}_l$ and $\tilde{L}_l'$ have a rather complicated structure [Eqs. (36), (34), and (45)] which precludes exact inversion. We note, however, that simplification occurs if $\beta_s=k_0+is/c$ is replaced by k_0 in Eqs. (34) and (45). Such an approximation amounts to the neglect of retardation manifest in Eqs. (24) and (26) and is justified if the time scale of change of $L_l(r^{\ll},r^{\gg};t)$ and $L_l'(r^{\ll},r^{\gg};t)$ is much larger than the maximum retardation $2R/c$. It may help in considering such retardation effects to recall that we have assumed all the atoms to be in the same excited state at the initial moment. Any practical scheme to excite the atoms would be likely to involve retardation effects which are implicitly ignored in specifying the initial state. We shall thus assume that the sphere is small enough that retardation effects can safely be neglected. With this simplification we have

$$L_l(r^<,r^>;t)=k_0(2\pi i)^{-1}\times\int_{\epsilon-i\infty}^{\epsilon+i\infty}s^{-1}j_l(\gamma_s r^<)\times h_l^{(1)}(\gamma_s r^>)\exp(st)ds\,, \tag{46}$$

and

$$L_l'(r^{\ll},r^{\gg};t)=k_0 j_l(k_0 r')(2\pi i)^{-1}\times\int_{\epsilon-i\infty}^{\epsilon+i\infty}s^{-2}j_l(\gamma_s r^<)\times h_l^{(1)}(\gamma_s r^>)\exp(st)ds\,, \tag{47}$$

i.e.,

$$L_l'(r^{\ll},r^{\gg};t)=j_l(k_0 r')\int_0^t L_l(r^{\ll},r^{\gg};t')dt' \tag{48}$$

with

$$\gamma_s=k_0(1-i\Lambda_0/s)^{1/2}\,. \tag{49}$$

It is now straightforward to evaluate the integrals in Eqs. (46) and (47) for $k_0R\gg 1$ by asymptotic techniques. We shall assume for the moment that in the long-time limit the major contributions to the integrals in Eqs. (41) and (44) come from the $r',r''\approx R$ regions and then demonstrate that to be true. With this assumption the Bessel functions $j_l(\gamma_s r^<)$, $h_l^{(1)}(\gamma_s r^>)$, etc., can be expanded asymptotically into exponentials of the Debye form. The details of this evaluation are presented in Appendix A. Here we just give the results which can be classified according to the value of l.

Case (a). $(l+\frac{1}{2})<k_0r^<<k_0r^>\le k_0R$:

$$L_l(r^<,r^>;t)\approx -i(8\pi r^> r^<)^{-1/2}\{2\beta(\alpha_>^2-1)(\alpha_<^2-1)[(\alpha_>^2-1)^{1/2}+(\alpha_<^2-1)^{1/2}]\}^{-1/4}\times\exp(i\nu A_0^+ +\{2\Lambda_0 t\nu[(\alpha_>^2-1)^{1/2}+(\alpha_<^2-1)^{1/2}]\}^{1/2})\,, \tag{50}$$

$$L_l'(r^<,r^>;t)\simeq\Lambda_0^{-1}(2\beta)^{1/2}[(\alpha_>^2-1)^{1/2}+(\alpha_<^2-1)^{1/2}]^{-1/2}j_l(k_0r')L_l(r^<,r^>;t)\,. \tag{51}$$

In these equations A_0^+, $\alpha_>$, $\alpha_<$, ν, and β are given by

$$A_0^+=(\alpha_>^2-1)^{1/2}+(\alpha_<^2-1)^{1/2}-\arccos(\alpha_>^{-1})-\arccos(\alpha_<^{-1})\,, \tag{52}$$

$$\nu=l+\tfrac{1}{2}\,,\quad \alpha_>=k_0r^>/\nu\,,\qquad \alpha_<=k_0r^</\nu\,,\quad \beta=\Lambda_0t/\nu\,. \tag{53}$$

Case (b). $l\gg(k_0R/2)^2$:

$$L_l(r^<,r^>;t)\approx-\frac{i}{(2l+1)r^>}(r^</r^>)^l\times\exp(i\Lambda_0t/2)J_0(\Lambda_0t/2)\,, \tag{54}$$

$$L_l'(r^<,r^>;t)\approx-i(2l+1)^{-1}(r^</r^>)^l(r^>)^{-1}\times\int_0^t\exp(i\Lambda_0t'/2)J_0(\Lambda_0t'/2)dt'=-i(2l+1)^{-1}(r^</r^>)^l(r^>)^{-1}\times j_l(k_0r')tM(\tfrac{1}{2};2;i\Lambda_0t)\,. \tag{55}$$

In the latter equation $M(a;b;z)$ is the single-valued Kummer function.[10] In the limit $\Lambda_0t\to\infty$,

$$tM(\tfrac{1}{2};2;i\Lambda_0t)\sim t^{1/2}\,,$$

and

$$J_0(\Lambda_0t/2)\sim t^{-1/2}\cos(\Lambda_0t/2-\pi/4)\,.$$

Thus, L_l has oscillatory decay whereas L_l' shows only a slow $t^{1/2}$ growth in time.

For $l\sim k_0r^<,k_0r^>$ the behavior of L_l as a function of time is given by the function $E_{1,7/6}(i\Lambda_0t)$, defined[11] by

$$E_{\alpha,\beta}(z)=\sum_{k=0}^{\infty}z^k/\Gamma(\alpha k+\beta)\,.$$

The modulus of this function $|E_{1,7/6}(i\Lambda_0t)|$ shows no monotonic growth with t; in fact, for large Λ_0t ($\Lambda_0t\gg1$) it decreases as $(\Lambda_0t)^{-1/6}$.

From the above considerations we conclude that the modes (l,m) for which $l>k_0R$ do not amplify significantly in the time during which modes (l,m) with $l<k_0R$ do. We will henceforth call the (l,m) modes with $l<k_0R$ the *amplifying modes* and the remaining (l,m) modes the *nonamplifying modes.*

For the purposes of later calculations we shall simplify Eq. (50) by assuming that $\alpha_>,\alpha_< \gg 1$. This approximation corresponds to extending the simplification which takes place for $l \ll k_0R$ to those amplifying modes for $l \sim O(k_0R)$ which do not in fact amplify quite as rapidly. Equation (50) then reduces to

$$L_l(r^<,r^>;t) \approx -i(8\pi)^{-1/2}(k_0rr')^{-1}[\Lambda_0 t k_0(r+r')]^{-1/4} \times \exp[ik_0(r+r')] \times \exp[2\Lambda_0 t k_0(r+r')]^{1/2}\,, \tag{56}$$

which is independent of l for $l<k_0R$. The corresponding reduction for Eq. (51) is

$$L_l'(r^{\ll},r^{\gg};t) \approx -ij_l(k_0r')(4\pi\Lambda_0 k_0 r' r'')^{-1}\{16\pi^2(\Lambda_0 t)/[k_0(r'+r'')]^3\}^{-1/4}\exp[ik_0(r'+r'')]\exp[2\Lambda_0 t k_0(r'+r'')]^{1/2}\,. \tag{57}$$

With the aid of expression (57) for L_l', we can carry out the r' integration in Eq. (44) approximately in the long-time limit, $(2\Lambda_0 k_0 R t)^{1/2} \gg 1$. The result for $r \gg R \gg k_0^{-1}$ is

$$e_{lm}^{(+)}(r,t) \approx (8\pi)^{-1/2}k_0 \exp(ik_0r)r^{-1}\int_0^R \exp\{ik_0r''+[2\Lambda_0(t-r/c)k_0(R+r'')]^{1/2}\} \times [k_0\Lambda_0(t-r/c)(R+r'')]^{-1/4}c_{lm}^-(r'',0)r''dr''\,. \tag{58}$$

This can be rewritten in terms of $L_l(r^<,r^>;t)$, given asymptotically by Eq. (56) as

$$e_{lm}^{(+)}(r,t) \approx ik_0^2(R/r)\exp[ik_0(r-R)]\int_0^R dr'\,r'^2 L_l(r',R;t-r/c)c_{lm}^-(r',0)\,. \tag{59}$$

It has not been necessary thus far to distinguish between $\mathbf{L}\cdot\mathbf{P}^\pm(\mathbf{r},t)$ and $\mathbf{L}\cdot\nabla\times\mathbf{P}^\pm(\mathbf{r},t)$, or correspondingly, between $\mathbf{L}\cdot\mathbf{E}^{(\mp)}(\mathbf{r},t)$ and $\mathbf{L}\cdot\nabla\times\mathbf{E}^{(\mp)}(\mathbf{r},t)$. But now, in order to discuss separately the electric and the magnetic multipoles (related to $\mathbf{L}\cdot\nabla\times\mathbf{P}^\pm$ and $\mathbf{L}\cdot\mathbf{P}^\pm$, respectively) we have to make the distinction.

We let $c_{lm}^-(r,t)$ and $d_{lm}^-(r,t)$ denote the coefficient operators in the expansion of the slowly varying amplitudes of $\mathbf{L}\cdot\mathbf{P}^-$ and $\mathbf{L}\cdot\nabla\times\mathbf{P}^-$, respectively:

$$\mathbf{L}\cdot\mathbf{P}^-(\mathbf{r},t)=\exp(-i\omega_0 t)\sum_{l,m}c_{lm}^-(r,t)Y_{lm}(\theta,\phi)\,, \tag{60}$$

$$\mathbf{L}\cdot\nabla\times\mathbf{P}^-(\mathbf{r},t)=\exp(-i\omega_0 t)\sum_{l,m}d_{lm}^-(r,t)Y_{lm}(\theta,\phi)\,. \tag{61}$$

Then from Eqs. (25) and (59), for $r \gg R$ we obtain in the long-time limit

$$\left.\begin{matrix}\mathbf{L}\cdot\mathbf{E}^{(+)}(\mathbf{r},t)\\ \mathbf{L}\cdot\nabla\times\mathbf{E}^{(+)}(\mathbf{r},t)\end{matrix}\right\} \approx ik_0^2(R/r)\exp[ik_0(r-R)-i\omega_0 t]\sum_{l,m}{}'\int_0^R dr' r'^2 L_l(r',R;t-r/c)\times\left\{\begin{matrix}c_{lm}^-(r',0)\\ d_{lm}^-(r',0)\end{matrix}\right\}\times Y_{lm}(\theta,\phi)\,. \tag{62}$$

The prime on $\sum$ in this and the subsequent equations indicates a sum over only the amplifying (l,m) modes, i.e., those for which $l<ak_0R$ $(a\approx 1)$. The Hermitian adjoints of Eqs. (62) give the corresponding relations for the negative-frequency field variables. The foregoing equations, as we have noted earlier, in fact determine the radial components $\mathbf{r}\cdot\mathbf{B}^{(+)}$ and $\mathbf{r}\cdot\mathbf{E}^{(+)}$ as expansions in terms of the various spherical harmonics. Each term in the first sum and the corresponding term in the second sum of Eqs. (62) together specify the two Debye potentials[12] in terms of which the full free-space vector field $\mathbf{E}^{(+)}(\mathbf{r},t)$ may be written as

$$\begin{aligned}\mathbf{E}^{(+)}(\mathbf{r},t) \simeq -R\exp(-ik_0R-i\omega_0 t)\Bigg[&\sum_{l,m}{}'[l(l+1)]^{-1}(\nabla\times\{\nabla\times[\mathbf{r}\exp(ik_0r)Y_{lm}(\theta,\phi)/r]\})\\ &\times\int_0^R dr'\,r'^2 L_l(r',R;t-r/c)d_{lm}^-(r',0)\\ &+k_0^2\sum_{l,m}{}'[l(l+1)]^{-1}\{\nabla\times[\mathbf{r}\exp(ik_0r)Y_{lm}(\theta,\phi)/r]\}\\ &\times\int_0^R dr'\,r'^2 L_l(r',R;t-r/c)c_{lm}^-(r',0)\Bigg]\,.\end{aligned} \tag{63}$$

The magnetic field $\mathbf{B}^{(+)}$ given by

$$\mathbf{B}^{(+)}(\mathbf{r},t)=(ik_0)^{-1}\nabla\times\mathbf{E}^{(+)}(\mathbf{r},t)$$

assumes the same form as Eq. (63) provided the replacements, $d_{lm}^{-}(r',0)\rightarrow c_{lm}^{-}(r',0)$ and $c_{lm}^{-}(r',0)\rightarrow -d_{lm}^{-}(r',0)$, are made. With the aid of the identity[12]

$$\mathbf{r}\cdot(\nabla\times\{\nabla\times[\mathbf{r}f(r)Y_{lm}(\theta,\phi)]\})=l(l+1)f(r)Y_{lm}(\theta,\phi) \tag{64}$$

and the result

$$\nabla^2[f(r)Y_{lm}(\theta,\phi)]\simeq -k_0^2 f(r)Y_{lm}(\theta,\phi)$$

which holds approximately if the dominant dependence of $f(r)$ is of the form $\exp(ik_0 r)/r$, one may easily verify that $\mathbf{E}^{(+)}$ and $\mathbf{B}^{(+)}$ as determined above do indeed satisfy the free-space Maxwell equations as well as Eqs. (62). In expression (63) the first sum represents the electric multiple contribution whereas the second sum represents the magnetic multipole contribution.

IV. STATISTICAL ASPECTS: AVERAGE INTENSITIES AND THEIR ANGULAR CORRELATIONS

Since during the initial stages of amplification the electromagnetic field depends linearly on the atomic polarization variables $c_{lm}^{\pm}(r',0)$ and $d_{lm}^{\pm}(r',0)$, the average field intensity and the intensity-intensity correlation are averages of bilinear and quadrilinear forms, respectively, of these operators. We shall address here the more general problem of determining the statistics of the fluctuations of $c_{lm}^{\pm}(r',0), d_{lm}^{\pm}(r',0)$ by calculating the expectation values of all of their products.

A. Properties of the fully inverted initial state

By projecting out the spherical harmonic coefficients in Eqs. (60) and (61) we find

$$\left.\begin{matrix} c_{lm}^{-}(r,0) \\ d_{lm}^{-}(r,0)\end{matrix}\right\} = \int d^2\Omega\, Y_{lm}^{*}(\theta,\phi)\left\{\begin{matrix}\mathbf{L}\cdot\mathbf{P}^{-}(\mathbf{r},0) \\ \mathbf{L}\cdot\nabla\times\mathbf{P}^{-}(\mathbf{r},0)\end{matrix}\right. . \tag{65}$$

It is more convenient to consider integrals over the full spherical volume. This we do by using the Hankel transforms $c_{lmk}^{-}(0), d_{lmk}^{-}(0)$ defined as

$$\left.\begin{matrix} c_{lm}^{-}(r,0) \\ d_{lm}^{-}(r,0)\end{matrix}\right\} = \int_0^{\infty}\left\{\begin{matrix} c_{lmk}^{-}(0) \\ d_{lmk}^{-}(0)\end{matrix}\right\}\times j_l(kr)k^2 dk \tag{66}$$

with the inverse relations

$$\left.\begin{matrix} c_{lmk}^{-}(0) \\ d_{lmk}^{-}(0)\end{matrix}\right\} = \frac{2}{\pi}\int_0^{R}\left\{\begin{matrix} c_{lm}(r',0) \\ d_{lm}(r',0)\end{matrix}\right\}\times j_l(kr')r'^2 dr' , \tag{67}$$

in which we have used the fact that $c_{lm}^{-}(r',0)$ and $d_{lm}^{-}(r',0)$ vanish for $r'>R$. From Eq. (65) we then find

$$\left.\begin{matrix} c_{lmk}^{-}(0) \\ d_{lmk}^{-}(0)\end{matrix}\right\} = \frac{2}{\pi}\int_V d^3r\, \Pi_{lmk}^{*}(\mathbf{r})\times\left\{\begin{matrix}\mathbf{L}\cdot\mathbf{P}^{-}(\mathbf{r},0) \\ \mathbf{L}\cdot\nabla\times\mathbf{P}^{-}(\mathbf{r},0)\end{matrix}\right. , \tag{68}$$

in which we have written

$$\Pi_{lmk}(\mathbf{r})=j_l(kr)Y_{lm}(\theta,\phi) . \tag{69}$$

On using some vector identities and then integrating by parts we may show that

$$\left.\begin{matrix} c_{lmk}^{-}(0) \\ d_{lmk}^{-}(0)\end{matrix}\right\} = \frac{2}{i\pi}\int_V d^3r\, \mathbf{P}^{-}(\mathbf{r},0)\cdot\left\{\begin{matrix}\nabla\times(\mathbf{r}\Pi_{lmk}^{*}) \\ \nabla\times[\nabla\times(\mathbf{r}\Pi_{lmk}^{*})]\end{matrix}\right. . \tag{70}$$

Then using the definition (5) of the initial polarization field and the relations $\langle\sigma_{ja}^{+}\sigma_{jb}^{-}\rangle=\delta_{jj'}\delta_{ab}$ $(a,b=x,y,z)$ followed by some algebraic manipulations, including several integrations by parts, one obtains the results

$$\langle c_{lm}^{+}(r,0)c_{l'm'}^{-}(r',0)\rangle = n_0\mu^2\delta_{ll'}\delta_{mm'}l(l+1)\delta(r-r')/r^2 , \tag{71}$$

$$\langle c_{lm}^{+}(r,0)d_{l'm'}^{-}(r',0)\rangle = 0 , \tag{72}$$

and

$$\langle d_{lm}^{+}(r,0)d_{l'm'}^{-}(r',0)\rangle = n_0\mu^2 l(l+1)\delta_{ll'}\delta_{mm'} \times\left[\frac{\partial^2}{\partial r\partial r'}\frac{\delta(r-r')}{r^2}+\frac{l+2}{r'}\frac{\partial}{\partial r}\frac{\delta(r-r')}{r^2}+\frac{l+2}{r}\frac{\partial}{\partial r'}\frac{\delta(r-r')}{r'^2}+\frac{(l+2)^2}{rr'}\frac{\delta(r-r')}{r^2} +\frac{1}{2}\frac{d}{dR}[\delta(r-R)\delta(r'-R)/R^2]\right] . \tag{73}$$

Equation (72) represents the mutual independence of electric and magnetic multipole evolution. The vanishing of Eqs. (71) and (73) for $l=0$ indicates an absence of source fluctuations for $l=0$. Thus as we have noted earlier, the lowest radiating multipole is a dipole ($l=1$).

In the limit of large particle number N, the averages of the higher-order products are reducible to forms involving only the 2nd-order products. Clearly, products involving an odd number of $c_{lm}^{\pm}$ or $d_{lm}^{\pm}$ have zero expectation values in the initial state. On the other hand, the $2p$-order averages ($p=1,2,\ldots$) can be reduced to a sum of products of 2nd-order averages as follows:

$$\langle c_{l_1m_1}^{+}(r_1)\cdots c_{l_pm_p}^{+}(r_p)c_{l_1'm_1'}^{-}(r_1')\cdots c_{l_p'm_p'}^{-}(r_p')\rangle \simeq \sum_P \langle c_{l_1m_1}^{+}(r_1)c_{l_1'm_1'}^{-}(r_1')\rangle\cdots\langle c_{l_pm_p}^{+}(r_p)c_{l_p'm_p'}^{-}(r_p')\rangle . \tag{74}$$

The sum in this equation runs over all $p!$ permutations of the primed coordinates. The time coordinate which is 0 has been dropped to simplify the notation. Equation (74) is accurate only for $p \ll N$. A formally identical relation is also obtained for the d_{lm}'s. From these composition rules it is evident that the fluctuations of the operators $c_{lm}(r,0)$ and $d_{lm}(r,0)$ in the initial state follow Gaussian statistics for N sufficiently large. More precisely, the coherent-state amplitude[9] corresponding to each of the initial polarization coefficient operators $c_{lm}(r,0)$ and $d_{lm}(r,0)$ assumes random values with a Gaussian quasiprobability distribution. Since the electromagnetic field can be written as a linear combination of $c_{lm}(r,0)$ and $d_{lm}(r,0)$, the fluctuations of the electromagnetic field also obey the Gaussian statistics in the linear regime.

B. Average intensity of (l,m) multiple radiation; average delay times

We now discuss in detail the two lowest-order averages of the electromagnetic field in the linear regime. For $l<k_0R$, in the limit of $k_0R \gg 1$ and long times, both magnetic and electric multipoles of a given order (l,m) radiate the same average intensity,

$$\begin{aligned} I_{lmE}(\mathbf{r},t) &\simeq I_{lmM}(\mathbf{r},t) \\ &\cong cR^2k_0^2n_0\mu^2[l(l+1)]^{-1} \\ &\quad\times |\nabla\times[\mathbf{r}\exp(ik_0r)Y_{lm}(\theta,\phi)/r]|^2 \\ &\quad\times \int_0^R dr'r'^2\,|L_l(r',R;t-r/c)|^2 . \end{aligned} \tag{75}$$

The details of the derivation are presented in Appendix B.

For large times $\tau=t-r/c$, the integral in Eq. (75) is straightforwardly evaluated, for then only r' close to R contribute significantly when expression (56) for L_l is introduced. The resulting intensity is

$$\begin{aligned} I_{lmE}(\mathbf{r},t) &\simeq \{\hbar\omega_0/[16\pi l(l+1)\tau]\} \\ &\quad\times \exp[4(\Lambda_0k_0R\tau)^{1/2}]\,|\nabla Y_{lm}(\theta,\phi)|^2 , \end{aligned} \tag{76}$$

an expression which is valid for

$$(2\Lambda_0k_0R\tau)^{1/2} \gg 1 . \tag{77}$$

Thus the time dependence of the intensity is asymptotically of the same form, $\tau^{-1}\exp(\alpha\sqrt{\tau})$, as in the one-dimensional model of superfluorescence.[4] This formal correspondence is a hint of a one-dimensional character for the emission from a large sphere. In any single experiment, as we shall see, the radiation does tend to be confined close to just one dominant radial direction. The time dependence expressed by Eq. (76) is qualitatively different from the purely exponential time dependence found by Ernst and Stehle[6] and by Rehler and Eberly.[7]

The total energy flow rate $W(\tau)$ from the sample in all such (l,m) modes is given by integrating $2I_{lmE}(\mathbf{r},t)$ over all solid angles and summing over l from 1 to $l_0=ak_0R$. For long times τ we may use Eq. (76) to obtain the result

$$W(\tau) \simeq [\hbar\omega_0(ak_0R)^2/8\pi\tau]\exp[4(\Lambda_0k_0R\tau)^{1/2}] . \tag{78}$$

Its short-time behavior is perhaps best seen from Eqs. (44) and (47), which show that $W(\tau)$ is zero at $\tau=0$ and rises at first quadratically with τ. The number of photons $N(\tau)$ emitted in time interval τ is given by the integral

$$N(\tau) = \frac{1}{\hbar\omega_0}\int_0^\tau W(\tau')d\tau' .$$

Since $W(\tau')$ is small for small τ', $N(\tau)$ may in the long-time limit (77) be estimated by means of Eq. (78). The result is

$$N(\tau) \simeq \frac{(ak_0R)^2}{4\pi}\,\frac{\exp[4(\Lambda_0k_0R\tau)^{1/2}]}{4(\Lambda_0k_0R\tau)^{1/2}} . \tag{79}$$

Even though the linear regime is confined to the initial stages of radiation during which $N(\tau) \ll N/2$, we see from Eq. (79) that when $N \gg (k_0R)^2$ there should nevertheless exist a finite interval of time in which the solution follows the asymptotic expressions (76) and (78) accurately.

With the aid of Eq. (79) we obtain an order-of-magnitude estimate of the average delay time $\langle t_D\rangle$ for the evolution of the amplifying modes. It may roughly be defined to be the time needed for the emission of $N/2$ photons. We then have the approximate result

$$\langle t_D\rangle \approx (16\Lambda_0k_0R)^{-1}\{\ln[2\pi N/(ak_0R)^2]\}^2 . \tag{80}$$

Since Λ_0 is proportional to N, $\langle t_D\rangle$ also takes the form characteristic of the one-dimensional theory, $(1/N)(\ln N)^2$. The delay time t_D itself will show fluctuations about the average $\langle t_D\rangle$ which can be calculated in the present model by following the procedure of Ref. 13.

A natural physical interpretation can be given to the result in Eq. (80). Since $16\Lambda_0k_0R \sim NT_1^{-1}[1/(k_0R)]^2$, where $T_1^{-1}=\mu^2k_0^3/(\pi\hbar)$ is the spontaneous emission rate of a single excited atom, it is evident that not all atoms but only a fraction of order $(k_0R)^{-2}$ participate in the cooperative radiation process in a single pulse. But $4\pi(k_0R)^{-2}$ is the solid angle subtended by the characteristic diffraction pattern of a sphere of radius R. That only $N(k_0R)^{-2}$ atoms cooperate results from the fact that photons emitted only in a solid angle $4\pi(k_0R)^{-2}$ initiate the cooperative decay of atomic excitation. [In the terminology of Rehler and Eberly[7] $(k_0R)^{-2}$ is the shape factor μ of the sample.] In fact, all intensity-intensity and higher-order correlations may be expected, therefore, only to extend over a maximum solid angle of order $4\pi(k_0R)^{-2}$. The emission process, in other words, may tend to be highly directional for each pulse.

C. Angular intensity-intensity correlation function

The angular distribution of radiation intensity which emerges from the sphere in any single pulse is extremely random. The ensemble average of the intensity taken over many pulses is isotropic and has no angular dependence whatever. It may nevertheless be true that the individual pulses are quite anisotropic in their structure. If that is so it will become evident by consideration of the angular correlation function for the radiated intensities.

We discuss here the angular correlation of intensities detected along two rays which enclose an angle ψ (Fig. 2). Along each ray is placed a detector sensitive to either left

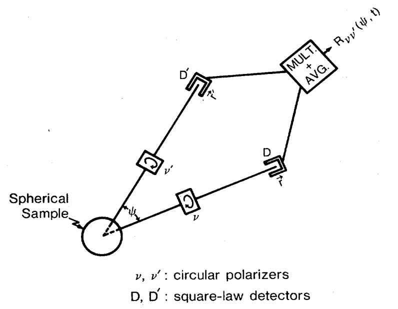

FIG. 2. Intensity-intensity correlation.

or right circular polarizations. The output photocurrents are multiplied and the product averaged over many pulses to give a correlation function which is proportional to the joint photon-counting rate in the two beams.[9] We assume that the detectors are placed the same distance $r=|\mathbf{r}|=|\mathbf{r}'|$ away from the center of the radiating sphere.

For each direction $\mathbf{r}$ we introduce a pair of complex circular polarization vectors $\hat{e}_\nu(\hat{r})$ corresponding for $\nu=\pm 1$ to the right and left circular polarization, respectively. The circularly polarized components of the field vector are then defined as

$$\mathscr{E}_\nu^{(+)}=\hat{e}_\nu^*(\hat{r})\cdot\mathbf{E}^{(+)} .$$

We introduce the normalized intensity correlation function $R_{\nu\nu'}(\psi,t)$ by writing the joint counting rate as

$$\begin{aligned}\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(-)}(\mathbf{r}',t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\mathscr{E}_\nu^{(+)}(\mathbf{r},t)\rangle &= \langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r},t)\rangle\langle \mathscr{E}_{\nu'}^{(-)}(\mathbf{r}',t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle \\ &\quad\times[1+R_{\nu\nu'}(\psi,t)] . \end{aligned} \tag{81}$$

In view of the Gaussian statistics of the electric field and the consequent reduction of the fourth-order field correlation function we can write

$$R_{\nu\nu'}(\psi,t)=\frac{|\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle|^2}{\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r},t)\rangle\langle \mathscr{E}_{\nu'}^{(-)}(\mathbf{r}',t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle} . \tag{82}$$

The computation of the second-order averages is analogous to that of the average intensity in Sec. IV B (see Appendix B for details). In the long-time limit one obtains

$$\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle_{\mathrm{El}}\approx\nu\nu'\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle_{\mathrm{Mag}} , \tag{83}$$

in which the subscripts "El" and "Mag" stand for the electric and magnetic multipole contributions to the correlation function. The factor $\nu\nu'$ comes from the difference in the angular dependence of the electric and magnetic multipole radiation patterns.

An important consequence of Eq. (83) is that field components of opposite circular polarizations are completely uncorrelated along any two rays, for, if $\nu\nu'=-1$, then

$$\begin{aligned}\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(r',t)\rangle &= \langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle_{\mathrm{Mag}} \\ &\quad+\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_{\nu'}^{(+)}(\mathbf{r}',t)\rangle_{\mathrm{El}} \\ &=0 .\end{aligned}$$

The only correlated emission, therefore, is of photons circularly polarized in the same sense, for which we have

$$R_{\nu\nu}(\psi,t)=\frac{|\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r}',t)\rangle|^2}{|\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r},t)\rangle|^2} . \tag{84}$$

To evaluate this correlation function we note that according to Eqs. (63) and (71)

$$\begin{aligned}\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r}',t)\rangle &\simeq 2\langle \mathscr{E}_\nu^{(-)}(\mathbf{r},t)\mathscr{E}_\nu^{(+)}(\mathbf{r}',t)\rangle_{\mathrm{Mag}} \\ &\simeq 2R_0^2k_0^4n_0\mu^2{\sum_{l,m}}' \,[l(l+1)]^{-1}(\hat{e}_\nu(\hat{r})\cdot\{\nabla\times[\mathbf{r}\exp(-ik_0r)Y_{lm}^*(\theta,\phi)/r]\}) \\ &\qquad\times(\hat{e}_\nu^*(\hat{r}')\cdot\{\nabla'\times[\mathbf{r}'\exp(ik_0r')Y_{lm}(\theta',\phi')/r']\})\int_0^R d\rho\,\rho^2\,|L_l(\rho,R;t-r/c)|^2 .\end{aligned} \tag{85}$$

By noting the fact that $\int_0^R d\rho\,|L_l|^2\rho^2$ is approximately independent of l for the amplifying modes, we may cast Eq. (84) in the form

$$R_{\nu\nu}(\psi,t)\simeq\frac{\left|{\sum_{l,m}}'[l(l+1)]^{-1}[\hat{e}_\nu(\hat{r})\cdot\nabla Y_{lm}^*(\theta,\phi)][\hat{e}_\nu^*(\hat{r}')\cdot\nabla' Y_{lm}(\theta',\phi')]\right|^2}{\left|{\sum_{l,m}}'[l(l+1)]^{-1}[\hat{e}_\nu(\hat{r})\cdot\nabla Y_{lm}^*(\theta,\phi)][\hat{e}_\nu^*(\hat{r})\cdot\nabla Y_{lm}(\theta,\phi)]\right|^2} . \tag{86}$$

The summations in Eq. (86) may be carried out straightforwardly. The details are presented in Appendix C.

It is convenient to write

$$R_{\nu\nu}(\psi,t)=|S(\psi)/S(0)|^2 .$$

The result of carrying out the summations of Eq. (86) can then be expressed by writing

$$\begin{aligned}S(\psi)=\sum_{l=1}^{l_0}[(2l+1)/l(l+1)] \\ \times[P_l''(\cos\psi)(-\sin^2\psi)+P_l'(\cos\psi)(1+\cos\psi)] ,\end{aligned} \tag{87}$$

where $P_l(z)$ is the Legendre polynomial of order l, $P_l'(z)$ and $P_l''(z)$ are its first two derivatives, and $l_0=ak_0R$ is the upper cutoff on the sum.

For $\psi=0$, since $P_l'(1)=\frac{1}{2}l(l+1)$, we have

$$S(0)=l_0(l_0+2)\ . \tag{88}$$

Now, for $l_0\rightarrow\infty$ (i.e., $k_0R\rightarrow\infty$), since[14]

$$\begin{aligned}\sum_{l=1}^{\infty}[(2l+1)/l(l+1)]P_l(\cos\psi)&=\sum_{l=1}^{\infty}[P_l(\cos\psi)/l)+P_l(\cos\psi)/(l+1)]\\&=-1-\ln[(1-\cos\psi)/2]\ ,\end{aligned}$$

we have (for $\psi\neq0$)

$$\begin{aligned}S(\psi)\underset{k_0R,l_0\rightarrow\infty}{\longrightarrow}&\{-[\sin^2\psi/(1-\cos\psi)^2]\\&+(1+\cos\psi)/(1-\cos\psi)\}\\&=0\ .\end{aligned} \tag{89}$$

Thus, for an infinitely large sphere there is no angular correlation at all of fields for nonvanishing ψ.

For values of k_0R that are large but not infinite, it is convenient to write $S(\psi)$ in a somewhat different form. On subtracting Eq. (89) from Eq. (87), we obtain

$$\begin{aligned}S(\psi)=\sum_{l=l_0+1}^{\infty}&[(2l+1)/l(l+1)]\\&\times[\sin^2\psi P_l''(\cos\psi)-(1+\cos\psi)P_l'(\cos\psi)]\ .\end{aligned} \tag{90}$$

Since for large l and ψ not too close to π we may write $P_l(\cos\psi)$ as[15]

$$P_l(\cos\psi)\simeq J_0[(l_0+\tfrac{1}{2})\psi](\psi/\sin\psi)^{1/2}\ , \tag{91}$$

we can evaluate the sum in Eq. (90) asymptotically. The result we find for $R_{\nu\nu}(\psi)$ in Appendix C is

$$\begin{aligned}R_{\nu\nu}(\psi)&=|S(\psi)/S(0)|^2\\&\simeq4J_1^2[(l_0+\tfrac{3}{2})\psi]/[(l_0+\tfrac{3}{2})\psi]^2\ .\end{aligned} \tag{92}$$

The first zero of the Bessel function J_1 occurs at $\psi\approx3.83/(l_0+\frac{3}{2})$, giving $R_{\nu\nu}(\psi)$ a width $\Delta\psi$ of the order of $1/(l_0+\frac{3}{2})$, or since $l_0=ak_0R$ ($a\approx1$),

$$\Delta\psi\sim(k_0R)^{-1}\ . \tag{93}$$

Since expression (86) [as well as (92)] is manifestly independent of the time variable, we may expect it to hold for times longer than the linear initiation regime over which it is rigorously valid.

The results presented in this section have so far referred to measurements at equal retarded times, $\tau=(t-r/c)=(t'-r'/c)=\tau'$, for the two directions. For $\tau\neq\tau'$, if we denote the time-dependent angular correlation function by $R_{\nu\nu'}(\psi;\tau,\tau')$ we easily see that $R_{\nu\nu'}=0$ if $\nu\nu'=-1$ corresponding to opposite circular polarizations. But, for $\nu\nu'=+1$ corresponding to circular polarizations of the same sense we find that $R_{\nu\nu}(\psi;\tau,\tau')$ factorizes in the limit of long τ and τ', i.e.,

$$R_{\nu\nu}(\psi;\tau,\tau')=R_{\nu\nu}(\psi)X(\tau,\tau')\ , \tag{94}$$

where $R_{\nu\nu}$ is given by Eq. (86) or (92).

One sees from Eqs. (56) and (63) that the purely temporal factor $X(\tau,\tau')$ assumes the form

$$X(\tau,\tau')=\frac{\left|\int_0^R dr\,r^2L(r,R;\tau)L^*(r,R;\tau')\right|^2}{\int_0^R dr\,r^2|L(r,R;\tau)|^2\int_0^R dr\,r^2|L(r,R;\tau')|^2} \tag{95}$$

which in the large-τ,τ' limit reduces to the expression

$$X(\tau,\tau')\simeq\frac{4\sqrt{\tau\tau'}}{(\sqrt{\tau}+\sqrt{\tau'})^2}\ . \tag{96}$$

Many of the results obtained in the present paper should continue to hold with good accuracy when the shape of the radiating volume is no longer spherical. This is because we may expect the emission to occur along well-directed rays in a one-dimensional manner. Thus if L is a measure of the maximum linear dimension of an arbitrary sample, many of the results may be carried over from the spherical case simply by substituting L for $2R$. The analysis of the present paper is, of course, confined to the linear regime which prevails in the early stages of radiation. One effect of the nonlinearity is to couple all of the radiation field modes to one another. Such coupling may be expected to alter significantly the result for later times.

After the completion of the present work, a paper by Mostowski and Sobolewska[16] dealing with the same problem appeared. Their results, although generally parallel to ours, differ in several ways. Their analysis is based on a scalar rather than the correct vector wave equation. They are thus unable to discuss those aspects of the problem that depend intrinsically on the nature of photon polarization. An important result of this kind, as we have seen, is the complete absence of angular correlation between any two photons which are circularly polarized in opposite senses. For photons circularly polarized in the same sense, however, we find that in the long-time limit the intensity-intensity correlation extends over an angular range of order $1/k_0R$. Because the Debye multiple-reflection expansion of geometrical optics, which their analysis makes use of, implicitly assumes $k_0R=\infty$, they, on the other hand, predict zero angular range for correlation in the same, long-time limit. The average intensity in their work amplifies in time as $\tau^{-3/2}\exp(\alpha\tau^{1/2})$, whereas we find it to have the same form, $\tau^{-1}\exp(\alpha\tau^{1/2})$, as occurs in the one-dimensional problem.

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APPENDIX A: EVOLUTION KERNELS $L_l(r^<,r^>;t)$ AND $L_l'(r^<,r^>;t)$: LAPLACE INVERSION DETAILS

For $t>0$ the inversion contour can be replaced by a circle of radius $>\Lambda_0$ centered at the origin in the complex s plane. Then the transformation $i\Lambda_0/s=\omega$ gives us

$$L_l(r^<,r^>;t)=k_0(2\pi i)^{-1}\oint_{c'}d\omega\,\omega^{-1}j_l(k_0r^>\sqrt{1-\omega})\times h_l^{(1)}(k_0r^>\sqrt{1-\omega})\times\exp(i\Lambda_0t/\omega) \quad \text{(A1a)}$$

and

$$L_l'(r^<,r^>;t)=-k_0(2\pi\Lambda_0)^{-1}j_l(k_0r')\times\oint_{c'}d\omega\,j_l(k_0r^<\sqrt{1-\omega})\times h_l^{(1)}(k_0r^>\sqrt{1-\omega})\times\exp(i\Lambda_0t/\omega)\,, \quad \text{(A1b)}$$

where c' is a circle of radius smaller than 1. The branch cut for the integrands lies outside c' in the ω plane. Since the integrands in Eqs. (A1a) and (A1b) differ by a simple factor of ω, we need give details of evaluation only for one of the two integrals. The value of the other integral in the stationary phase approximation, which we use since $k_0R\gg1$, is obtained simply by appending to the result an extra factor of ω_s, the relevant stationary point in the ω plane. We choose to evaluate L_l here.

We study the following domains of l values.

Case (a). $(l+\frac{1}{2})<k_0r^<<k_0r^><k_0R$: We take the radius of c' to be very small so that $|k_0R(1-\omega)^{1/2}|>(l+\frac{1}{2})$ on it. Then for $h_l^{(1)}$ we have Debye's asymptotic formula given by[17]

$$h_l^{(1)}(u)\approx u^{-1/2}[u^2-(l+\tfrac{1}{2})^2]^{-1/4}\times\exp\{i[u^2-(l+\tfrac{1}{2})^2]^{1/2}-i(l+\tfrac{1}{2})\arccos[(l+\tfrac{1}{2})/u]-i\pi/4\} \quad \text{(A2)}$$

in which

$$[u^2-(l+\tfrac{1}{2})^2]\gg1\,.$$

Now, by writing $j_l=(h_l^{(1)}+h_l^{(2)})/2$ and using Eq. (A2) in Eq. (A1a) we obtain

$$L_l=L_l^{(1)}+L_l^{(2)}\,,$$

where $L_l^{(1)}$ and $L_l^{(2)}$ are expressed as

$$\left.\begin{matrix}L_l^{(1)}\\ L_l^{(2)}\end{matrix}\right\}\simeq k_0(4\pi i)^{-1}\oint d\omega\,\omega^{-1}\exp(i\Lambda_0t/\omega)\times h_l^{(1)}[k_0r^>(1-\omega)^{1/2}]\times\begin{cases}h_l^{(1)}[k_0r^<(1-\omega)^{1/2}]\\ h_l^{(2)}[k_0r^<(1-\omega)^{1/2}]\,.\end{cases} \quad \text{(A3)}$$

We need consider either $L_l^{(1)}$ or $L_l^{(2)}$ in detail since the calculations for both are exactly analogous. For $L_l^{(1)}$ we have the expression

$$L_l^{(1)}\simeq-(k_0/4\pi)\oint g(\omega)\exp[i\nu f(\omega)]d\omega\,, \quad \text{(A4)}$$

in which

$$g(\omega)=\omega^{-1}[x_>x_<(1-\omega)]^{-1/2}\times\{[x_>^2(1-\omega)-\nu^2][x_<^2(1-\omega)-\nu^2]\}^{-1/4}\,,$$

$$f(\omega)=[\alpha_>^2(1-\omega)-1]^{1/2}+[\alpha_<^2(1-\omega)-1]^{1/2}-\arccos[\alpha^>(1-\omega)^{1/2}]^{-1}-\arccos[\alpha^<(1-\omega)^{1/2}]^{-1}+\beta/\omega\,, \quad \text{(A5)}$$

$$\nu=l+\tfrac{1}{2},\quad x_<=k_0r^<,\quad x_>=k_0r^>\,,$$

$$\alpha_<=x_</\nu,\quad \alpha_>=x_>/\nu,\quad \beta=\Lambda_0t/\nu\,.$$

We first assume $\nu\gg1$ but the final expressions hold even for $\nu\sim O(1)$. For the function $f(\omega)$ the stationary points ω_s lying close to the origin are given as

$$\omega_s=\omega^\pm\simeq\pm i(2\beta)^{1/2}[(\alpha_>^2-1)^{1/2}+(\alpha_<^2-1)^{1/2}]^{-1/2}\,, \quad \text{(A6)}$$

provided $\beta\ll1$ as is the case for small times $t\ll\nu/\Lambda_0$.

We now deform the closed contour c' so that it passes through the stationary points $\omega^\pm$ and assumes the slopes $\theta^\pm$ of the stationary paths near $\omega^\pm$ given by

$$2\theta^\pm+\arg f''(\omega^\pm)=0,\pi\,. \quad \text{(A7)}$$

In the limit $\beta\ll1$, $\theta^\pm$ are found as

$$\theta^\pm\simeq\begin{cases}-\pi/4,\ \ \pi/4\\ \pi/4,\ \ 3\pi/4\,.\end{cases} \quad \text{(A8)}$$

By using

$$\int_0^\infty\exp(\pm iaz^2)dz=(\pi/a)^{1/2}\exp(\pm i\pi/4)/2\quad(a>0)\,, \quad \text{(A9)}$$

it is now easy to demonstrate that the two stationary segments of the contour at ω^+ contribute exactly equal amounts, whereas contributions from similar segments at ω^- cancel each other exactly. One may carry out a similar procedure for $L_l^{(2)}$. The final expressions for $L_l^{(1)}$ and $L_l^{(2)}$ are written as

$$\left.\begin{matrix}L_l^{(1)}\\ L_l^{(2)}\end{matrix}\right\}\simeq\mp i(8\pi r^>r^<)^{-1/2}[(\alpha_>^2-1)(\alpha_<^2-1)]^{-1/4}\{2\beta[(\alpha_>^2-1)^{1/2}\pm(\alpha_<^2-1)^{1/2}]\}^{-1/4}\times\exp(i\nu A_0^\pm+\{2\Lambda_0t\nu[(\alpha_>^2-1)^{1/2}\pm(\alpha_<^2-1)^{1/2}]\}^{1/2})\,, \quad \text{(A10)}$$

where

$$A_0^{\pm}=(\alpha_>^2-1)^{1/2}\pm(\alpha_<^2-1)^{1/2}-\arccos(\alpha_>^{-1})\mp\arccos(\alpha_<^{-1}) . \tag{A11}$$

It is clear from Eq. (A10) that the maximum rate of evolution of L_l in the long-time limit is obtained for $r^< \sim O(r^>)$ for which $L_l^{(2)}$ is negligible.

Case (b). $k_0r^< < (l+\frac{1}{2}) < k_0r^> \le k_0R$: This case is not important for our purposes since, as we have just noted, the major contribution to the r' integral in Eq. (43) comes from the vicinity of $r^< = 0(r^>)$ [in the limit $(2\Lambda_0tk_0R)^{1/2} \gg 1$] for which $k_0r^< > (l+\frac{1}{2})$. The calculations are similar so that we will not present them here.

Case (c). $l \gg (k_0R/2)^2$: In this range of l, both j_l and $h_l^{(1)}$ have just power-law dependences on their arguments.[8] It is then easy to show that

$$\begin{aligned} L_l(r^<,r^>;t) &\approx -[2\pi(2l+1)r^>]^{-1}(r^</r^>)^l \oint_{c'}(d\omega/\omega)\exp(i\Lambda_0t/\omega)(1-\omega)^{-1/2} \\ &= -[2\pi(2l+1)r^>]^{-1}(r^</r^>)^l \oint_c ds\,\exp(i\Lambda_0ts)[s(s-1)]^{-1/2} , \end{aligned} \tag{A12}$$

where c encloses the branch cut from 0 to 1.

To evaluate the integral we deform c so that it just circumscribes the branch cut. The integral may the be written as the sum of two integrals of the same integrand which are performed over the line segments from 0 to 1 running just above and just below the real axis in the complex s plane. The sum of the two latter integrals is easily expressed as

$$\begin{aligned} L_l(r^<,r^>;t) = &-[2\pi(2l+1)r^>]^{-1} \\ &\times 2i\int_0^1 dr[r(1-r)]^{-1/2} \\ &\times\exp(i\Lambda_0tr)(r^</r^>)^l . \end{aligned}$$

On substituting $r=\sin^2\theta$ in this expression and using the formula[8]

$$J_0(x)=\frac{1}{\pi}\int_0^\pi d\theta\exp(-ix\cos\theta) ,$$

we finally obtain

$$\begin{aligned} L_l(r^<,r^>;t) \approx &-i[(2l+1)r^>]^{-1}(r^</r^>)^l \\ &\times\exp(i\Lambda_0t/2)J_0(\Lambda_0t/2) . \end{aligned} \tag{A13}$$

The high (l,m) modes thus do not evolve; they just oscillate and damp out in a time of order Λ_0^{-1} which is independent of the sample size R. This independence from R is because of the fact that to the high-l modes the sample appears to be a detail-less point emitter.

Case (d). $l \sim k_0r^< \sim k_0r^>$: For this intermediate case, too, we once again have power-law dependences. With the appropriate expressions for j_l and $h_l^{(1)}$ (Ref. 18) substituted in Eq. (A1a) we obtain

$$L_l(r^<,r^>;t) \approx K\oint_{c'} d\omega\,\omega^{-1}\exp(i\Lambda_0t/\omega)(1-\omega)^{-5/6} , \tag{A14}$$

in which

$$\begin{aligned} K = &\,ik_0\exp(i2\pi/3)(\sin^2\pi/3)\Gamma^2(\tfrac{1}{3}) \\ &\times 6^{2/3}(k_0^2r^<r^>)^{-5/6}/(18\pi^2) . \end{aligned} \tag{A15}$$

By substituting $s=\omega^{-1}$ we transform Eq. (A14) to the form

$$L_l(r^<,r^>;t)=K\oint_c ds\,s^{-1/6}(s-1)^{-5/6}\exp(i\Lambda_0ts) ,$$

where on c, $|s|=s_0>1$. By expanding $(s-1)^{-5/6}$ in a Taylor series we may show that

$$L_l=2\pi iK\left[1+\sum_{m=1}^{\infty}\frac{\frac{5}{6}(\frac{5}{6}+1)\cdots(\frac{5}{6}+m-1)}{m!}\times\frac{(i\Lambda_0t)^m}{m!}\right] . \tag{A16}$$

On writing $\frac{5}{6}+k-1=k-\frac{1}{6}$ it is easy to show that

$$\begin{aligned} &\ln[\tfrac{5}{6}(\tfrac{5}{6}+1)\cdots(\tfrac{5}{6}+m-1)] \\ &\quad\cong\ln(m!m^{-1/6}e^{-\gamma/6}) \\ &\quad\simeq\ln[(m!)^2e^{-\gamma/6}/\Gamma(m+\tfrac{7}{6})] , \end{aligned}$$

where $\gamma \simeq 0.577\cdots$ is the Euler-Mascheroni constant. With the aid of this result, Eq. (A16) reduces to the approximate form

$$L_l(r^<,r^>;t) \simeq 2\pi iK\exp(-\gamma/6)E_{1,7/6}(i\Lambda_0t) \tag{A17}$$

in which $E_{1,7/6}(i\Lambda_0t)$ is a special function defined by the relation[11]

$$E_{\alpha,\beta}(z)=\sum_{k=0}^{\infty}z^k/\Gamma(\alpha k+\beta) .$$

APPENDIX B: EQUALITY OF AVERAGED INTENSITIES FOR ELECTRIC AND MAGNETIC MULTIPOLES

From Eq. (63), on taking the (l,m) electric multipole contribution and using Eq. (79) we obtain for the corresponding averaged intensity

$$I_{lmE}(\mathbf{r},t)\approx W_{lm}(\mathbf{r})\left[\int_0^R\left|\frac{\partial}{\partial r'}[r'^2L_l^*(r',R;t-r/c)]\right|^2 dr'/r'^2 -(l+2)\int_0^R(\partial/\partial r')[r'^2L_l^*(r',R;t-r/c)]L_l(r',R;t-r/c)dr'/r' -(l+2)\int_0^R(\partial/\partial r')[r'^2L_l(r',R;t-r/c)]L_l^*(r',R;t-r/c)dr'/r' -(l+2)^2\int_0^R dr'\,|L_l(r',R;t-r/c)|^2\right], \tag{B1}$$

where W_{lm} is given by

$$W_{lm}(\mathbf{r})=cR^2n_0\mu^2[l(l+1)]^{-1}\times|\nabla\times\{\nabla\times[\mathbf{r}\exp(ik_0r)Y_{lm}(\theta,\phi)/r]\}|^2 . \tag{B2}$$

We can recombine the terms in Eq. (B1) to a more transparent form by noting that

$$(\partial/\partial r')(r'^2L_l)=r'[(\partial/\partial r')(r'L_l)+L_l] . \tag{B3}$$

Equation (B1) reduces to the form

$$I_{lmE}(r,t)\approx W_{lm}(r)\left[\int_0^R|(\partial/\partial r')(r'L_l)+L_l|^2dr' -(l+2)\int_0^R(\partial/\partial r')\times(r'^2|L_l|^2)dr'/r' +l(l+2)\int_0^R|L_l|^2dr'\right] . \tag{B4}$$

We can neglect the second term above because it involves $\partial/\partial r'$ derivatives of $r'^2|L_l|^2$, which has no fast r' variation. Furthermore, from the exact expression for L_l in Eq. (A10) (on neglecting $L_l^{(2)}$ for large times) it follows that

$$(\partial/\partial r')(r'L_l)\approx i\nu(\partial A_0^+/\partial r')(r'L_l) =ik_0[1-\nu^2/(k_0r')^2]^{1/2}(r'L_l) .$$

With the aid of this relation, Eq. (B4) may be expressed as

$$I_{lmE}(\mathbf{r},t)\approx W_{lm}(\mathbf{r})\left[k_0^2\int_0^R r'^2|L_l|^2dr' +(l+\tfrac{3}{4})\int_0^R|L_l|^2dr'\right] . \tag{B5}$$

But since in the long-time limit the major contribution comes from $r'\sim O(R)$ the first term is of order $(k_0R)^2\int_0^R|L_l|^2dr'$, whereas the second term is always less than a quantity of order $(k_0R)\int_0^R|L_l|^2dr'$ for amplifying modes ($l<k_0R$). Hence, for $k_0R\gg1$ we can ignore the second term in Eq. (B5) altogether, so that we have

$$I_{lmE}(\mathbf{r},t)\approx W_{lm}(\mathbf{r})k_0^2\int_0^R r'^2|L_l(r',R;t-r/c)|^2dr' . \tag{B6}$$

However, in the limit $k_0r\gg1$ we have the simplification

$$\nabla\times\{\nabla\times[\mathbf{r}\exp(ik_0r)Y_{lm}(\theta,\phi)/r]\} \to ik_0\exp(ik_0r)\nabla Y_{lm}(\theta,\phi) ,$$

so that Eq. (B6) may be written as

$$I_{lmE}(\mathbf{r},t)\approx cR^2n_0\mu^2k_0^4[l(l+1)]^{-1}|\nabla Y_{lm}(\theta,\phi)|^2 \times\int_0^R r'^2|L_l(r',R;t-r/c)|^2dr' , \tag{B7}$$

which is the same as the averaged intensity $I_{lmM}(\mathbf{r},t)$ of the corresponding magnetic-multipole radiation obtained easily from Eqs. (63) and (71).

APPENDIX C: DETAILS OF EVALUATION OF SUM $S(\psi)$ AND ITS ASYMPTOTIC FORM

We may write $S(\psi)$ as

$$S(\psi)=4\pi r^2\sum_{l,m}{}'[l(l+1)]^{-1}[\hat{e}_\nu(\hat{r})\cdot\nabla Y_{lm}^*(\theta,\phi)][\hat{e}_\nu^*(\hat{r}')\cdot\nabla' Y_{lm}(\theta',\phi')] =4\pi[(\partial^2/\partial\theta\,\partial\theta')-(i\nu/\sin\theta)(\partial^2/\partial\phi\,\partial\theta')+(i\nu/\sin\theta')(\partial^2/\partial\theta\,\partial\phi') +(\sin\theta\sin\theta')^{-1}(\partial^2/\partial\phi\,\partial\phi')]\sum_{l,m}{}'[l(l+1)]^{-1}Y_{lm}^*(\theta,\phi)Y_{lm}(\theta',\phi') . \tag{C1}$$

With the aid of the relation

$$\sum_{m=-l}^{l}Y_{lm}^*(\theta,\phi)Y_{lm}(\theta',\phi')=(2l+1)P_l(\cos\psi)/4\pi , \tag{C2}$$

in which the angle ψ between directions (θ,ϕ) and (θ',ϕ') is given by

$$\cos\psi=\cos\theta\cos\theta'+\sin\theta\sin\theta'\cos(\phi-\phi') , \tag{C3}$$

we may express $S(\psi)$ as

$$S(\psi)=\sum_{l=1}^{l_0}(2l+1)[l(l+1)]^{-1}[(\partial^2/\partial\theta\,\partial\theta')-(i\nu/\sin\theta)(\partial^2/\partial\phi\,\partial\theta') \\ +(i\nu/\sin\theta')(\partial^2/\partial\theta\,\partial\phi')+(\sin\theta\sin\theta')^{-1}(\partial^2/\partial\phi\,\partial\phi')]P_l(\cos\psi) . \tag{C4}$$

Since our final answer can only depend on the angle ψ (there are no other angles in the problem) we shall conveniently take $\theta=\phi=0$ after carrying out the derivatives. This straightforwardly gives us the desired expression (87) which simplifies in the limit of large k_0R.

We consider first the sum related to that in Eq. (90):

$$Z(\psi)=\sum_{l=l_0+1}^{\infty}(2l+1)[l(l+1)]^{-1}P_l(\cos\psi)$$

$$\simeq\sum_{l=l_0+1}^{\infty}2(l+\tfrac{1}{2})^{-1}P_l(\cos\psi),\quad\text{since } l\gg1 .$$

With the aid of Eq. (91) $Z(\psi)$ may be expressed as

$$Z(\psi)\simeq\sum_{l=l_0+1}^{\infty}2(l+\tfrac{1}{2})^{-1}J_0[(l+\tfrac{1}{2})\psi](\psi/\sin\psi)^{1/2} .$$

Since we shall only be interested in $\psi\ll1$, we may replace the sum by the integral

$$Z(\psi)\simeq2\int_{l_0+1}^{\infty}\{J_0[(l+\tfrac{1}{2})\psi]/(l+\tfrac{1}{2})\}dl$$
$$\simeq2\int_{(l_0+3/2)\psi}^{\infty}J_0(u)du/u .$$

From this expression $Z'(\psi)$ and $Z''(\psi)$ are easy to calculate by simple differentiation. On rewriting $R_{\nu\nu}(\psi)$ as

$$R_{\nu\nu}(\psi)\simeq[l_0(l_0+2)]^{-2}[-\sin^2\psi Z''(\psi) \\ +(1+\cos\psi)Z'(\psi)]^2$$

and using the calculated values of $Z'(\psi)$ and $Z''(\psi)$ we obtain Eq. (92).

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