RADIATIVE TRANSFER EQUATIONS IN BROAD-BAND, TIME-VARYING FIELDS

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ABSTRACT

A derivation of the equation of transfer is obtained by starting with Maxwell’s equations in the “slowly varying envelope” form. Particular attention is paid to characterizing the intensity that is “seen” by the atom (which is found to be related to a Wigner distribution of the electric field). The equation of transfer is found to be valid for “broad-band” slowly varying radiation fields.

Subject heading: radiative transfer

I. INTRODUCTION

The derivation of the equations of statistical equilibrium and the emission and absorption coefficients for use in the theory of radiative transfer has recently been considered by Cooper et al. (1982) (hereinafter CBBH). Their results apply specifically to steady-state situations for broad-band, multimode radiation. The equation of transfer, as derived by Landi Degl’Innocenti and Landi Degl’Innocenti (1975), was assumed to be valid. Recently this derivation for intense, broad-band, possibly time-varying fields has been questioned (W. Molander, private communication; J. Oxenius, private communication). The purpose of this note is to sketch the derivation of the equations of transfer, starting from the “slowly varying envelope” form of Maxwell’s equations (Graham and Haken 1968; Litvak 1970; Allen and Eberly 1975).

II. STATISTICAL EQUILIBRIUM EQUATIONS AND DEFINITION OF INTENSITY

The concept of the intensity spectrum of light in a time-varying situation has been discussed by Courtens and Szöke (1977) and Eberly and Wódkiewicz (1977). Specific account is taken of the bandwidth of the measuring detector in their definition of intensity. In the case considered here the more important question is: To what aspects of the radiation does the atom respond? Specifically, what “intensity” should we use in the statistical equilibrium equations?

This question may be immediately answered from the derivation of CBBH. Ignoring, for convenience, the velocity of the atom and statistical weight factors, we need to reexamine for the time-dependent situation, the absorption (and stimulated emission) term (designated here as Q) in the (angle averaged) statistical equilibrium equations, viz.:

\[
Q = \frac{B_i}{4\pi} \int d\Omega \int_{-\infty}^{\infty} d\omega' I(\omega', \hat{r'}) \phi(\omega') (n_i - n_u)
\]

(of eq. [26], from CBBH). From CBBH, equations (22) and (23), this term corresponds to

\[
Q = \text{Re} \int_{-\infty}^{t} dt' \langle \delta'(t) \hat{\delta}^*(t') \rangle_{\Lambda_v} \exp \left[ \left( i\omega_{ul} - \kappa' \right)(t - t') \right] \left[ n_r(t') - n_u(t') \right]_{\Lambda_v}
\]

where the average is over the possible variations of the electric field of the radiation. For a broad-band radiation field (when the rapidly varying part \( e^{-i\Omega t} \) associated with the mean frequency is factored out), the correlation time of \( \langle \delta'(t) \hat{\delta}^*(t') \rangle_{\Lambda_v} \) is short, corresponding to roughly \( 1/\Delta \), where \( \Delta \) is the bandwidth of the radiation. \( \Delta \) is typically at least a Doppler width. On the other hand, the populations change on a time scale of at most the absorption rate or the lifetime which is much larger. Thus the decorrelation discussed in CBBH can be used:

\[
Q = \text{Re} \int_{-\infty}^{t} dt' \langle \delta'(t) \hat{\delta}^*(t') \rangle_{\Lambda_v} \exp \left[ \left( i\omega_{ul} - \kappa' \right)(t - t') \right] \left[ n_r(t') - n_u(t') \right]_{\Lambda_v}
\]

\[
\approx \text{Re} \int_{-\infty}^{t} dt' \langle \delta'(t) \hat{\delta}^*(t') \rangle_{\Lambda_v} \exp \left[ \left( i\omega_{ul} - \kappa' \right)(t - t') \right]
\]

\[
\approx \text{Re} \int_{-\infty}^{\infty} \int_{0}^{t} ds \langle \delta'(t) \hat{\delta}^*(t - s) \rangle_{\Lambda_v} \exp \left[ \left( i\omega_{ul} - \kappa' \right)s \right]
\]

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The second line in equation (3) follows, since \( \langle n(t) - n_u(t) \rangle_{\lambda v} \) changes only slowly during the correlation time of \( \langle \delta(t)\delta^*(t') \rangle_{\lambda v} \) and further, the lower limit may be replaced by \(-\infty\) for the same reason.

Defining the time-dependent intensity \( I(\omega', \vec{k}', t) \) by

\[
\langle \delta(t)\delta^*(t-s) \rangle_{\lambda v} = \int \frac{d\Omega}{4\pi} \int d\omega' I(\omega', \vec{k}'; t)e^{-i\omega's},
\]

we obtain

\[
Q = \frac{B_{pe}}{4\pi} \int d\Omega \int_{-\infty}^{\infty} d\omega' I(\omega', \vec{k}'; t)\phi(\omega')\langle n(t) - n_u(t) \rangle_{\lambda v}.
\]

Thus, with this definition of intensity, the intensity and populations in the statistical equilibrium rate equations just become time dependent.

It should be noted that equation (4) is equivalent to the Page (1952) and Lampard (1954) definition of the intensity. Eberly and Wódkiewicz (1977) also show that their definition reduces to this spectrum in the limit of short correlation times, as are of interest in this note.

The above results were obtained using the method of CBBH, where, for convenience, the radiation field was treated classically. It is straightforward to show that a direct replacement of the classical electric field by quantum mechanical operators is possible. Specifically we obtain in equation (4) the normally ordered correlation function

\[
\langle E^-|(t)|E^+|(t-s) \rangle_{\lambda v}
\]

in the term describing absorption, and the anti-normally ordered correlation function

\[
\langle E^+|(t)|E^-|(t-s) \rangle_{\lambda v}
\]

in the emission term. Here \( E^+ \) and \( E^- \) denote the positive and negative frequency part of the radiative field in the Heisenberg picture (Agarwal 1974); \( \langle \cdot \rangle_{\lambda v} \) now becomes a trace over the total density operator of the system (since the electric field at any point in space implicitly depends on the entire atomic system). In particular, we note that although the operators \( E^+ \) and \( E^- \) act only on the radiation field variables, the occupation of the field states in the density operator depends on the radiation which may be due to all the atoms in the system; i.e., the implicit dependence on the atomic variables is via the mode occupation at a given position and at a given time. Note that we use \( \delta \) for classical fields.

Bringing the correlation function (6b) into normal order (by using the free field commutation relation, corresponding to a treatment of radiative decay in lowest order perturbation theory), we recover the right-hand side of equation (5) and in addition a term describing the radiative decay of the upper level (which corresponds to the \( \Gamma_n n_u \) term in CBBH).

III. EQUATION OF TRANSFER

We sketch below the derivation of the equation of transfer, starting from Maxwell’s equation for a quantized radiation field. The central result is that for a broad-band field with many uncorrelated modes, the absorption and emission coefficients can be expressed in terms of atomic dipole correlation functions, as has been assumed in CBBH.

The transfer equation deals with the time and spatial change of the spectral intensity in a given direction at a certain “point” and “time” in the medium. There would be a contradiction in defining an intensity of a mode \( (\omega, k) \) at a given time and space point \( (t, x) \), where \( k \leftrightarrow x \) and \( t \leftrightarrow \omega \) are interpreted as conjugate Fourier transform variables; instead, a meaningful definition of this spectral intensity can only be given as a macroscopic level, where the dependence on \( x \) and \( t \) can be understood in a coarse grained sense.

Suppose we divide space into cells \( V = L^3 \), where \( L \) is much larger than the (mean) wavelength of the radiation field. Then in each of these cells we can write a mode expansion of the electric field (Louisell 1973) in the Heisenberg picture:

\[
E^+|(r + x, t) = \sum_{k\lambda} \frac{e^{i\lambda k x}}{2\kappa_0 V} b_{k\lambda}(r, t)e^{ik\lambda x},
\]

where \( r \) has to be interpreted as labeling different cells in space, while \( x \) is the spatial variable inside this box. Here \( b_{k\lambda}(r, t) \) is the destruction operator and \( e_{k\lambda} \) \( (\lambda = 1, 2) \) the light polarization vector of the mode \( k\lambda \); \( b_{k\lambda}(r, t) \) acts on the photon occupation states associated with the given cell and thus the dependence on \( r \) is implicit. (Its \( t \) dependence is of course of the form \( e^{-i\omega t} \) times a slowly varying function of time, which may be associated with the changing intensity in the mode of interest.)

Consider now the atom \( i \) in box \( r \) at the position \( x_i \). The intensity seen by this atom is according to equation (4) proportional to the Fourier transform of the correlation function

\[
\langle E^-|(r + x_i, t)|E^+|(r + x_i, t - s) \rangle_{\lambda v}
\]

with respect to \( s \). Inserting the mode expansion (7) into equation (8), we find, using the approximation (see below)

\[
\langle b_{k\lambda}^+(r, t)|b_{k\lambda}^-(r, t') \rangle_{\lambda v} \approx \delta_{k\lambda} \delta_{k\lambda'} N_{k\lambda}(r, t) \exp(i\omega_s),
\]

where \( \delta_{k\lambda} \) is the Kronecker delta, \( N_{k\lambda}(r, t) \) is the density operator of the system (since the electric field at any point in space implicitly depends on the entire atomic system), and \( \delta_{k\lambda} \delta_{k\lambda'} \) acts only on the radiation field variables.

Thus, the intensity and populations in the statistical equilibrium rate equations just become time dependent.

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Bringing the correlation function (6b) into normal order (by using the free field commutation relation, corresponding to a treatment of radiative decay in lowest order perturbation theory), we recover the right-hand side of equation (5) and in addition a term describing the radiative decay of the upper level (which corresponds to the \( \Gamma_n n_u \) term in CBBH).
with $s = t - t'$, the expression

$$
\langle E^{-}(r, x_{i}, t)E^{+}(r, x_{i}, t - s) \rangle _{\lambda v} = \sum_{k_{x}} \frac{\hbar \omega_{k}}{2c_{0} V} N_{k_{x}}(r, t) \exp (i\omega_{k} s),
$$

(10)

where $N_{k}(r, t)$ is the number of photons in the mode $(k, \lambda)$ at the "position" $r$ and "time" $t$ (compare eq. [4]). Its explicit time dependence, of course, comes from the change of occupancy with time of the photon states within the cell, i.e., from the variation of the radiation part of the density matrix. Approximation (9) is based on the two assumptions that (i) the different modes of the radiation field are uncorrelated (which in particular is true for a chaotic radiation field) (Louisell 1973) and (ii) the time dependence of the $N_{k_{x}}(r, t)$ (due to changes of occupancy of radiation modes in the cell) is slow on the time scale of the inverse bandwidth $1/\Delta$ of the light (a condition for the Page-Lampard definition of the spectrum to be meaningful, as discussed in § II). This follows because factoring out a high-frequency part $e^{-i\omega_{s}}$, we see that the time scale associated with equation (10) is $s \sim 1/\Delta$. Note that since we are superposing a series of plane waves, this second condition also implies that the spectral intensity varies slowly on the distance $L \gg c/\Delta$. Classically, equation (10) corresponds to

$$
\langle \delta(t) \delta^{*}(t - s) \rangle = \sum_{\mu} |\delta_{\mu}(t)|^{2} \exp (i\omega_{\mu} s)
$$

(given by CBBH, eq. [25]), again assuming the mode amplitude changes slowly in time.

An alternative equivalent expression for the spectral intensity can be given in terms of a Wigner transform of the electric field correlation function. Consider the quantity

$$
f(k, x, t) = \int \frac{d^{3}x}{(2\pi)^{3}} e^{ik_{x}x} \langle E^{-}(x - \frac{1}{2} \xi, t) \cdot E^{+}(x + \frac{1}{2} \xi, t) \rangle _{\lambda v},
$$

(11)

defined as the Fourier transform of the spatial normally ordered electric field correlation function. (A more general definition would consider $\langle E_{\mu}^{-}(t)E_{\nu}^{+}(t) \rangle _{\lambda v}$, with $\mu, \nu$ the light polarization indices, which can be used to define a Stokes matrix.) From equation (11) it follows that

$$
\int d^{3}k f(k, x, t) = \langle E^{-}(x, t)E^{+}(x, t) \rangle _{\lambda v},
$$

(12)

is proportional to the total light intensity at $(x, t)$, while

$$
\int d^{3}k \zeta f(k, x, t) = \sum_{k_{x}} \frac{\hbar \omega_{k}}{2c_{0}} \langle b_{k_{x}}^{+}(t)b_{k_{x}}(t) \rangle _{\lambda v}.
$$

(13)

is proportional to the number of photons in the mode $k$, where here $b$ and $b^{+}$ are the creation and destruction operators corresponding to a mode decomposition in whole space; i.e., there is now no explicit dependence on the cell position $r$, so that

$$
E^{+}(x, t) = i \sum_{k_{x}} \int d^{3}k \left[ \frac{\hbar \omega_{k}}{2c_{0}(2\pi)^{3}} \right]^{1/2} \epsilon_{k_{x}} e^{i\omega_{k}t}b_{k_{x}}(t).
$$

The Wigner function $f(k, x, t)$ cannot be interpreted as a joint probability distribution of finding photons in the mode $k$ at position $x$ and time $t$, as $f(k, x, t)$ is not necessarily positive definite. (Notice, similar problems occur with the Page-Lampard definition of the spectrum [Eberly and Wódkiewicz 1977; Wolf 1978].) We can, however, define a macroscopically averaged spectral intensity as

$$
I(k, r, t) = \frac{1}{V} \int_{V} d^{3}x \frac{1}{(\Delta k)^{3}} \int d^{3}k \zeta f(k, k', r + x, t),
$$

(14)

with $V$ denoting a macroscopic volume with $L \gg c/\Delta$ and $r$ a coarse grained spatial variable. In addition, we have averaged over the (small) wave vector interval $\Delta k_{x} = 2\pi/L$, etc., with $L = V$. Inserting the mode decomposition (7) into equations (11) and (14), we find

$$
I(k, r, t) = \frac{\hbar \omega_{k}}{2c_{0}(2\pi)^{3}} \sum_{x} N_{k_{x}}(r, t),
$$

(15)

which, therefore, can be identified as being proportional to the number of photons in the mode $k$ in the cell $r$. This agrees, according to equations (9) and (10), with the Page-Lampard definition of the spectrum. Again we stress that equations (15) and (10) correspond to the classical definition of equation (4).

We now turn to the derivation of a transfer equation for the spectral intensity (14). The total Hamiltonian of our system of two-level atoms is

$$
H = H_{A} + H_{F} - \sum_{i} \mu_{i} E^{-}(x_{i}, t) + \text{h.c.},
$$

(16)

with $H_{A} = \sum \hbar \omega_{i}^{(0)} a_{i} a_{i}^{+}$ and $H_{F}$ the Hamiltonian operator of the atoms (which can, if necessary, include interaction with perturbers) and radiation field, respectively. Here $a_{i}$ is the destruction operator of the $i$th atoms (corresponding to the raising

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part of the dipole operator $d^{(+)}$ in CBBH; $\mu$ is the dipole matrix element. The electric field operator $E(x, t)$ in the Heisenberg picture obeys the Maxwell equation

$$\left( \frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \nabla_x^2 \right) E(x, t) = -\mu_0 \frac{\partial^2}{\partial t^2} P(x, t)$$

(17)

with $P(x, t)$ the polarization operator of the medium (see below). Since we are interested in a frequency band small compared to a mean frequency $\bar{\omega} \approx \omega_{10}$ of the light, we make (consistent with the rotating wave approximation [RWA]) a slowly varying amplitude approximation (Graham and Haken 1968)

$$E^\pm(x, t) = \bar{E}^\pm(x, t)e^{i\omega t},$$

(18)

where the time variation of $\bar{E}^\pm(x, t)$ is slow on the optical time scale $1/\bar{\omega}$. The wave equation (17) then reduces to

$$\left( \frac{1}{c^2} \frac{\partial^2}{\partial t^2} + \frac{\omega^2}{c^2} + \nabla_x^2 \right) \bar{E}^\pm(x, t) = -\mu_0 \bar{\omega}^2 \bar{P}^\pm(x, t)$$

(19)

with the slowly-varying polarization amplitude

$$\bar{P}^\pm(x, t) = \sum_i \sum_\gamma \frac{d^3k}{(2\pi)^3} e^{ik(x-x_i)} \epsilon_{k\gamma} (\epsilon_{k\gamma} \cdot \mu) \tilde{a}_i(t) \approx \sum_i \delta_T(x-x_i) \cdot \mu \tilde{a}_i(t),$$

(20)

where $\delta_T$ denotes the transverse $\delta$-function (Louisell 1973). Note that the expectation value

$$\langle P(x, t) \rangle_{Av} = \text{tr} \{ p(t) \bar{P}^\pm(x, t) \} e^{-i\omega t} + \text{c.c.}$$

(21)

[with $\rho(t)$ the density operator of the total system] is the polarization vector of the medium (Graham and Haken 1968). The classical analog to equation (19) in the slowly varying envelope approximation is obvious (see Allen and Eberly 1975).

We are now in a position to derive the equation of motion for the Wigner function (11) and hence the intensity; differentiating equation (11) with respect to time and using equation (20), we obtain

$$\left( \frac{1}{c^2} \frac{\partial}{\partial t} + \frac{k}{|k|} \cdot \nabla_x \right) f(k, x, t) = -i \left[ \frac{\omega}{2 \epsilon_0 c} \sum_\gamma \frac{d^3k}{(2\pi)^3} e^{-i k \cdot x} \epsilon_{k\gamma} \langle E^\gamma \rangle_{Av} \right]_x \nabla_x \cdot \tilde{a}_i(t)

+ i \left[ \frac{\omega}{2 \epsilon_0 c} \sum_\gamma \frac{d^3k}{(2\pi)^3} e^{-i k \cdot x} \epsilon_{k\gamma} \langle E^\gamma \rangle_{Av} \right] \nabla_x \cdot \mu \tilde{a}_i(t),$$

(22)

This equation describes the time and spatial change of the Wigner function due to the power delivered by the atoms to the field. If the quantum averages on the right-hand side of equation (22) are decorrelated, we see that the change of the spectral intensity of the light is proportional to the product of the electric field and the polarization of the medium (again, as we expected classically). Averaging equation (22) over the volume $V$ according to equation (14) we find

$$\left( \frac{1}{c^2} \frac{\partial}{\partial t} + \frac{k}{|k|} \cdot \nabla_x \right) I(k, r, t) = -i \left[ \frac{\hbar \omega}{2 \epsilon_0 c (2\pi)^3} \sum_\gamma \frac{d^3k}{(2\pi)^3} e^{-i k \cdot x} \epsilon_{k\gamma} \langle E^\gamma \rangle_{Av} \right]_x \nabla_x \cdot \tilde{a}_i(t)

+ i \left[ \frac{\hbar \omega}{2 \epsilon_0 c} \sum_\gamma \frac{d^3k}{(2\pi)^3} e^{-i k \cdot x} \epsilon_{k\gamma} \langle E^\gamma \rangle_{Av} \right] \nabla_x \cdot \mu \tilde{a}_i(t),$$

(23)

where essentially only atoms $i$ inside the volume $V$ contribute. Simplifying equation (23) we obtain

$$\left( \frac{1}{c^2} \frac{\partial}{\partial t} + \frac{k}{|k|} \cdot \nabla_x \right) I(k, r, t) = -i \left[ \frac{\hbar \omega}{2 \epsilon_0 c (2\pi)^3} \sum_{r \in V} \sum_{x \in V} \frac{\hbar \omega}{2 \epsilon_0 V} V \right]^{1/2} e^{i k \cdot (x-r)} \epsilon_{k\gamma} \langle \tilde{a}_i(t) | b_{k\gamma}(r, t) \rangle_{Av} + \text{c.c.},$$

(24)

where we used

$$\frac{1}{V} \int d^3x e^{-i k \cdot x} \epsilon_{k\gamma} | E^\gamma(r+x, t) \cdot \epsilon_{k\gamma} | = i \left( \frac{\hbar \omega}{2 \epsilon_0 V} V \right)^{1/2} \epsilon_{k\gamma} b_{k\gamma}(r, t).$$

(25)

The physical interpretation of equation (24) becomes clear when we consider the time rate of change of the photon numbers $N_k(r, t)$ in a specific cell (as derived from the Heisenberg equations).

$$\frac{dN_k(r, t)}{dt} = \sum_{k=1}^{\infty} \sum_{x \in V} \left[ \frac{\hbar \omega}{2 \epsilon_0 V} V \right]^{1/2} e^{i k \cdot (x-r)} \langle \tilde{a}_i^\dagger(r, t) | b_{k\gamma}(r, t) \rangle_{Av} \mu \cdot \epsilon_{k\gamma} + \text{c.c.}.$$  
(26)
and compare it with the transport equation (24). The right-hand side of equation (26) has, when we factorize the quantum average, the form of an electric field (via eq. [25]) times the atomic polarization ($\langle a(t) \rangle$) in the RWA and thus describes the power delivered by the atom to the particular field mode $k$. According to equation (24) the time derivative in equation (26) has to be interpreted as a total derivative.

In the following we express, in the source terms in equations (24) and (26), the absorption and emission coefficients in the form of dipole autocorrelation functions, thereby extending Mollow's (1973) treatment of absorption and emission profiles to our problem. Mollow expresses the polarization of the medium induced by a weak classical field of a given frequency in terms of atomic dipole correlation functions, which are calculated in the absence of the probe field. We adapt below Mollow's (1975, p. 1929) arguments by separating from the total electric field a single mode $k$ and calculating the emission and absorption profile of this particular mode in the presence of the remaining modes of the field. This gives us again expressions for the absorption and emission coefficients in terms of dipole correlation functions. For a (broad-band) multimode field (large $V$) with many uncorrelated modes, the exclusion of the mode $k$, when calculating the correlation functions, is negligible as argued by Mollow, and our results reduce to the ones which were the starting points in CBBH. Note that although the field intensity of a particular mode may be small, the total high intensity of all modes (which enters the atomic statistical equilibrium equations) may be strong enough to cause power broadening. These arguments are, of course, not valid for single mode (laser) light.

To evaluate the source term in equation (24) we go to the interaction representation with respect to the mode $k$. We write the total Hamiltonian now for a specific cell characterized by $r$, as

$$H = H_0 + H_1$$

with

$$H_1 = -\mu \sum_{j \neq k} a_j(t) \left( \frac{\hbar \omega}{2\varepsilon_0 V} \right)^{1/2} \epsilon_{kl} b_{kl}(r, t) e^{i(k \cdot x - \omega t)} + \text{h.c.}$$

where $H_1$ [classically of form $-d \cdot \mathbf{E}(t)$] describes the interaction of the atom under consideration with the mode $k$. $H_0$ of course includes all the other modes, and the implicit dependence on position of cell is through $b_{kl}(r, t)$. In the interaction picture the density matrix (of the total system)

$$\rho'(t) = e^{iH_0 t} \rho(t) e^{-iH_0 t}$$

obeys

$$\rho'(t) = \rho'(0) - i \frac{\hbar}{\epsilon_0} \int_0^t dt' [H_1 \rho'(t'), \rho'(t')] .$$

A similar equation was used by Degl'Innocenti and Degl'Innocenti (1975), but here we retain $\rho'(t')$ rather than the perturbation limit $\rho'(0)$. Substituting this result into equation (24), we are left with expressions of the form

$$\text{tr} \left[ b_{kl} a_k(t) \rho(t) \right] = -i \frac{\hbar}{\epsilon_0} \int_0^t dt' \text{tr} \left\{ b_{kl} a_k(t') \rho(t') \right\} ,$$

where we assumed the density matrix to factorize at $t = 0$, giving a zero average of the electric field (corresponding to random phases). The operator $b_{kl}(r, t)$ commutes apart from the free field Hamiltonian $H_F$ with $H_0$ since it does not explicitly contain the mode $k$.

Rearranging terms, we find in the RWA that equation (30) is proportional to

$$\frac{i}{\hbar} \int_0^t dt' \text{tr} \left\{ [b_{kl} a_k(t), b_{kl}(r, t') a_k(t')] \rho(t') \right\} ,$$

keeping only the $i = j$ terms (see below). To the extent that the intensity of the single mode $k \lambda$ is weak (which does not imply that the total intensity is small), we can factorize (decorrelate) in the above average, the electric field ($b_{kl}$ terms) from the atomic variables since $b_{kl}(r, t)$ is only for mode $k$ and the other variables are assumed to be independent of this single mode. In other words, that part of the density operator $\rho'(t')$ which describes the occupancy of radiation mode $k$ is assumed to be independent of all the other modes and to be constant in time (i.e., intensity in cell is "slowly varying" on time scales of interest), and conversely its exclusion has negligible effect on the evolution of the total density operator of the system. In first order perturbation theory this would be exact.

In the same approximation we can drop the superscript $I$ referring to the interaction representation. Quantities like $a_k(t)$, etc., are identified with the Heisenberg operator ignoring the interaction with the mode $k \lambda$, and thus, since the contribution of the mode $k$ is small, we are able to replace $a_k(t)$ by $a(t)$, etc., in equation (31), where $a(t)$ now evolves via the full Hamiltonian $H$ of equation (27) (i.e., the field due to the single mode $k$ from a large number of modes has a negligible effect on the overall evolution). Note that these approximations do not apply to single mode (laser) radiation. At the same time we can use equations (9) and (15) to rewrite equation (31) as a product of dipole correlation functions and the spectral intensity $I(k, r, t)$. (In general, when polarization effects are not ignored, we would get a sum over the different Stokes components of the intensity.)
Equation (31) splits into two contributions. The first one describes stimulated emission and absorption and is proportional to

$$2 \text{Re} \left[ \int_0^{t'} dt' e^{-i\omega(t-t')} \langle a(t), a(t') \rangle \right] I(k, r, t) \rightarrow \kappa(\omega_0, t) I(k, r, t),$$

(32)

where the stimulated emission and absorption coefficient is the Laplace transform of the dipole correlation function in agreement with CBBH. In addition, the antinormal ordering of the field operators in equation (31) for the emission term gives a spontaneous emission coefficient

$$2 \text{Re} \left[ \int_0^{t'} dt' e^{-i\omega(t-t')} \langle a(t) a(t') \rangle \right]$$

(compare § II, eqs. [6]) describing the spontaneous radiative decay into the mode $k$. Thus we obtain the equation of transfer

$$\left( C^+ + \frac{k}{|k|} \cdot \nabla_r \right) I(k, r, t) = -\kappa(\omega, t) I(k, r, t) + j(\omega, t).$$

(34)

In order to obtain these results we had to put the time arguments $t = t'$ equal in the $b_{k'}(r, t)$ terms in equation (31) (after factoring out $\exp [-i\omega(t-t')]$ terms, as we did in conjunction with eq. [9]). In addition, we had to replace the upper limit on $s = t - t'$ in the dipole correlation functions by infinity. The typical value of $\Delta \omega = \omega_k - \omega_0$ corresponds to at least a Doppler width $\Delta$, so the times of interest in the transform of the correlation function $1/\Delta$ are short. Populations (and hence, by the regression theorem, the slowly varying parts of the correlation functions) and related intensities change on time scales corresponding to radiative decay times or induced transition lifetimes (absorption times), and these times are long compared to $1/\Delta$; thus the approximations obtaining equations (32) and (33) from equation (31) are valid for broad-band fields of astrophysical interest.

An approximation implicit in the decorrelation of the atom-field variables in equation (31) is the neglect of possible atom-atom coherences; similar assumptions are implicit in semiclassical “one-atom” theories (Kazantsev, Smirnov, and Yakovlev 1982). It requires the average distance between the atoms to be much larger than the wavelength of the radiation field (low atomic densities).

IV. DISCUSSION

Thus, the $\kappa(\omega, t)$ and $j(\omega, t)$ in equation (34) are directly related to the dipole autocorrelation functions for emission and absorption given by CBBH (or Mollow 1973) with $p(t)$ determining the initial conditions.

To obtain this result we excluded the mode corresponding to the direction $k$ from the total Hamiltonian. As argued earlier, for a broad-band multimode field (such as found in astrophysical situations) this is unlikely to be important; i.e., the Rabi frequency corresponding to the field in a single directional mode is likely to be small. Thus, the results of CBBH may be used directly. It should be stressed that although the field in a given direction can be considered as “weak” so that the simple equation of transfer is obtained (eq. [34]) (see also Mollow 1973), this approximation does not imply that power broadening is negligible. Power broadening (and the equations governing the level populations) depends on the response of the atom to the total field. Thus, although $B_{k'} I(k', x; t)$ for a given direction may be negligible, the power broadening which depends on $(B_{k'}/4\pi) \int d\Omega I(k, x, t)$ may not necessarily be small. (Note: this implies that the effects of the field enter the equations of statistical equilibrium and the equation of transfer in different ways.)

In a laser situation, where the intensity per mode can be very high, the decorrelation is not necessarily valid. (Other methods must then be used—see, e.g., Zoller 1979.)

The correlation functions (and their transforms) can now be calculated as CBBH. Writing more explicitly the initial time as $t_0$, the equation corresponding to equation (57) of CBBH becomes (using semiclassical expression for convenient comparison)

$$\langle \delta^*(t) C(t) \rangle_{\lambda\nu} = d_{\lambda}^3 \left\{ \langle \delta^*(t) \exp \left[ -\Gamma(t - t_0) \right] \int_0^{t_0} dt' \delta'(t') \rangle \right\} \exp \left[ (i\omega_0 - \kappa')(t_0 - t) \right] \langle n(t') \rangle_{\lambda\nu}$$

$$= d_{\lambda}^3 \left\{ \langle \delta^*(t) \exp \left[ -\Gamma(t - t_0) \right] \int_{-\infty}^{t_0} dt' \delta'(t') \rangle \right\} \exp \left[ (i\omega_0 - \kappa')(t_0 - t) \right] \langle n(t) \rangle_{\lambda\nu}$$

$$= d_{\lambda}^3 \left[ \frac{d\Omega}{4\pi} \int d\omega' I(k', x; t_0) \exp \left[ (i\omega' - \Gamma)(t - t_0) \right] \langle n(t) \rangle_{\lambda\nu} \langle n(t_0) - n(t_0) \rangle_{\lambda\nu} \right]\langle n(t_0) \rangle_{\lambda\nu} \frac{\langle n(t_0) \rangle_{\lambda\nu}}{i(\omega' - \omega_0 - \kappa')}.$$ (35)

The second line in equation (35) follows since populations are slowly varying during the correlation time for the field (and similarly the lower limit may be replaced by $-\infty$). In the last line, $I(k', x; t)$ is replaced by $I(k, x; t_0)$ since again $t$ changes little in a correlation time. The net result is to retain the form of the emission coefficient $j(\omega; t)$ (see CBBH, eq. [58] or more specifically eq. [61]; but not eq. [60], which uses steady state relationships) but with $I(k, x; t)$ and $n(t)$, etc., now being time dependent.
V. CONCLUSIONS

We have shown that for a broad-band, multimode radiation field the correlation time is sufficiently short that the equations of transfer and the equations of statistical equilibrium (as developed by CBBH) may be used with a parametric dependence of intensity \( I(k, x, t) \) and populations \([n_\alpha(t), \text{etc.}\] on time, specifically in CBBH equations (32), (61) [but not (60)], and (69). The explicit validity requires that the bandwidth of interest be large compared with the radiative decay or absorption (power broadening) rates (which determine the time scale for variation of intensity, etc.).

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