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Stationary and Nonstationary Processes in Pumped Resonant Media

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The response of a system of coherently pumped atoms to weak electromagnetic perturbing fields is discussed in general terms. The pump field is prescribed, and is assumed to propagate unattenuated through the medium which the (identical) atoms comprise, exciting a particular transition through either electric or magnetic dipole coupling. The perturbing field, which is assumed to act through electric dipole coupling, is allowed to be the (resonant) field emitted by the atoms, a weak external field absorbed by the atoms under resonant conditions, or a resonant or nonresonant wave propagating through the pumped medium. All of these cases are conveniently described within the same general framework, based on an analysis of the (single-atom) two-time dipole-moment correlation function. This function contains nonstationary components as well as stationary ones, owing to the time dependence imposed by the pump field. We are accordingly led to introduce a time-dependent linear susceptibility, which is inhomogeneous and anisotropic, due, respectively, to the variation of the pump phase within the medium and the preferred direction established by the pump-field polarization. Such disparate effects as parametric frequency conversion, emission- and absorption-line splitting, and time-dependent modulation of resonant emission and absorption functions in optical-pumping experiments emerge naturally from a single unified formalism.

I. INTRODUCTION

In a wide variety of physical processes, atoms which are driven by a strong coherent "pump" field interact weakly at the same time with fields other than the pump field. These may be either the fields emitted by the atoms or fields incident upon the atoms from external sources. Processes in the latter category occur in the ac Stark effect,[1] optical-pumping and double-resonance experiments,[2] and, more generally, any experiment in which the effect of the pump field is monitored by observing the change it produces in the absorption of a weak "probe" or "signal" field. (An interesting example of the latter is the phenomenon of saturated absorption,[3] in which the pump and signal fields induce resonant transitions between the same pair of atomic states.[4]) The emission of radiation by strongly driven atoms has been discussed in a number of recent publications, both for the case of transitions between the same pair of states as are coupled by the pump field[5–7] and for the case of transitions between one of these states and an undriven state of the atom.[8] Finally, one may mention as processes in this same general category the nonlinear processes of stimulated parametric amplification and frequency conversion of light waves.[9]

The purpose of this paper is to analyze in some generality the time-dependent electromagnetic properties of a pumped medium consisting of identical stationary atoms in the case in which the pump field oscillates at a frequency near resonance for transitions between a particular pair of atomic states. The pump field is treated as a prescribed classical function, and is assumed to propagate unattenuated through the medium. The fundamental processes both of emission and absorption are described in a unified way in terms of the correlation function which represents the product of the atomic dipole moment at two different times. An important advantage of this method of formulation is that of enabling one to carry out the fundamental calculations without explicit reference to the perturbing (emitted or incident) fields, since the correlation functions are evaluated in the presence of the pump field alone.

Because the atoms are driven throughout the process in question by a time-dependent field, their electromagnetic properties are described by nonstationary functions as well as by stationary ones. The stationary functions which describe the emission[6,8] and absorption[8,10] of radiation by a single atom have been discussed previously within the general framework adopted herein. The analysis of the stationary functions is developed in this paper from a somewhat more unified point of view, and is extended to include an evaluation of the dielectric susceptibility. In addition, the nonstationary functions which describe both emission and absorption of radiation are evaluated. In certain cases, it should be emphasized, a single process requires an evaluation of both stationary and nonstationary functions for its complete description. In the frequency splitting (subharmonic generation) of light, for example, the intensity of light at either subharmonic frequency

is given by the familiar stationary cross-spectral density, while coincidence counting rates for the same process depend upon the value of the nonstationary correlation function which describes coherent photon-pair emission.

It is found that the absorption of radiation can be fully described by introducing a time-dependent linear complex susceptibility. The susceptibility, which has components oscillating at harmonics up to the second of the pump-field frequency, is both inhomogeneous (due to the variation of the pump-field phase within the medium) and anisotropic (due to the preferred direction of the pump-field polarization). Such physically different effects as the parametric frequency conversion of light waves and the time-dependent modulation of emission and absorption functions in double-resonance experiments are thus naturally described within the same general formalism.

The description of double-resonance experiments is formulated in terms of a coherent optical field, which leads to solutions of rather more complex and interesting structure than does the more commonly considered broadband field, the results for which can be obtained directly from those presented here. In contrast to the broadband case, it is found that for a coherent optical field, oscillating components in the absorption rate survive even in the limit of intense pump fields, where the pump-field-induced mean dipole moment vanishes. Similarly, while the *total* emission rate has an oscillating component only for moderate pump-field intensities, the emission process is characterized even in the limit of intense pump fields by appreciable time-dependent correlations between the fields emitted at different frequencies.

In Sec. II the atomic dipole-moment correlation function is introduced and the basic method of evaluating it is described in general terms. The evaluation is then carried out in Sec. III under the conditions imposed by a harmonic resonant driving field. Section IV is devoted to an analysis of the emission process (both stationary and nonstationary), and Sec. V to a similar analysis of the processes relating to dielectric susceptibility and absorption.

II. DIPOLE-MOMENT CORRELATION FUNCTION

The lowest-order interaction of an atom with the electromagnetic field is fully characterized, in the electric dipole approximation, by the atomic correlation function $\langle\mu(t)\mu(t')\rangle$, where μ is the atomic electric dipole-moment operator. The emission of photons, for example, is described by an electromagnetic-field correlation function[11,12] which is simply proportional to the cross-spectral function

$$g^{(1,1)}(t',t)=\langle\mu^{(-)}(t')\mu^{(+)}(t)\rangle, \tag{2.1}$$

the superscripts + and − denoting positive- and negative-frequency components according to the usual convention. The effect of a weak perturbing or signal field

$$E'(t)=E_0'e^{-i\nu t}+E_0'^{*}e^{i\nu t} \tag{2.2}$$

on the other hand, is to produce a small change in the equilibrium atomic density operator which is given to lowest order by the relation[8,13]

$$\Delta\rho(t)=(i\hbar)^{-1}\int_{-\infty}^{t}dt'\,[H'(t'),\rho], \tag{2.3}$$

where ρ is the unperturbed density operator and $H'(t)=-\mu(t)\cdot E'(t)$. By evaluating the expectation value $\mathrm{Tr}[\mu\Delta\rho(t)]$ of the dipole moment induced by the perturbing field and then multiplying the result by the number density N of (identical) atoms, one finds that the polarization at any point in the medium is

$$P'(t)=\chi(\nu,t)E_0'e^{-i\nu t}+\chi^{*}(\nu,t)E_0'^{*}e^{i\nu t}, \tag{2.4}$$

where the (second-rank tensor) complex electric susceptibility is given as[14]

$$\chi^{\alpha\beta}(\nu,t)=i(N/\hbar)\int_{-\infty}^{t}dt'\langle[\mu^{\alpha}(t),\mu^{\beta}(t')]\rangle e^{i\nu(t-t')}. \tag{2.5}$$

The superscripts in this relation are polarization indices and the statistical average is taken in the absence of the perturbing field. The susceptibility contains time-dependent and orientation-nonsymmetric components in the case to be considered, in which the atom is driven throughout the process in question by a time-varying pump field with a specified polarization. In addition, there is a (suppressed) spatial dependence, due to the variation of the pump-field phase within the medium.

The dipole-moment correlation function $\langle\mu(t)\mu(t')\rangle$ may be expressed as

$$\langle\mu(t)\mu(t')\rangle=\sum_{j,k,j',k'}\mu_{kj}\langle a_{jk}(t)a_{j'k'}^{\dagger}(t')\rangle\mu_{j'k'}, \tag{2.6}$$

where $a_{jk}=|k\rangle\langle j|$ and $a_{jk}^{\dagger}=|j\rangle\langle k|$ are the atomic-lowering and -raising operators, with expectation values

$$\langle a_{jk}\rangle=\rho_{jk},\quad \langle a_{jk}^{\dagger}\rangle=\rho_{kj} \tag{2.7a}$$

and moments

$$\langle a_{jk}^{\dagger}a_{jk}\rangle=\rho_{jj},\quad \langle a_{jk}a_{jk}^{\dagger}\rangle=\rho_{kk}. \tag{2.7b}$$

The dipole-matrix element $\mu_{jk}=\langle j|\mu|k\rangle$ is assumed to vanish for $j=k$,

$$\mu_{jj}=0. \tag{2.8}$$

Stationary processes involving resonant transi-

tions from an initial state $|j\rangle$ to a final state $|k\rangle$ are described by the function

$$\tilde{g}_{0(j\to k)}(\nu)=\mu_{jk}\mu_{kj}\int_{-\infty}^{\infty}d\tau\, e^{i\nu\tau}\langle a_{jk}^{\dagger}a_{jk}(\tau)\rangle, \quad (2.9)$$

which is appreciable for $\nu\approx(E_j-E_k)/\hbar$. For $E_j>E_k$, the function given by Eq. (2.9) is directly proportional to the emission spectrum for the transition in question, while the absorption of radiation from the signal field in Eq. (2.2) during transitions between the same pair of states is described by the linear absorption coefficient

$$\beta_{0(jk)}(\nu)=(\nu N/2\hbar c)[\tilde{g}_{0(k\to j)}(-\nu)-\tilde{g}^{T}_{0(j\to k)}(\nu)], \quad (2.10)$$

the second term on the right-hand side representing stimulated emission. [The symbol T indicates transposition of the (suppressed) vector indices.] The relation (2.10) follows directly from the expression

$$\chi_{0(jk)}(\nu)=(iN/\hbar)\mu_{kj}\mu_{jk}\int_{0}^{\infty}d\tau\, e^{i\nu\tau}\langle[a_{jk}(\tau),a_{jk}^{\dagger}]\rangle \quad (2.11)$$

for the stationary part of the complex susceptibility for the transition in question. It should be emphasized that no simple relationship exists between the functions $\tilde{g}_{0(j\to k)}(\nu)$ and $\tilde{g}_{0(k\to j)}(\nu)$ if either of the two states $|j\rangle$ or $|k\rangle$ is one of the pump-field-coupled pair of states $|0\rangle$ and $|1\rangle$, and that there is thus, in general, no simple proportionality between the emission and absorption spectra corresponding to transitions between a particular pair of atomic states.

The expectation value on the right-hand side of Eq. (2.6) is easily evaluated in the Markoff approximation by means of the quantum *fluctuation-regression theorem*,[15] whenever the time-dependent solutions to the equations of motion for the atomic density matrix are known. These take the general form

$$\rho_{jk}(t)=\sum_{n,m}\mathfrak{U}_{jk;nm}(t,t')\rho_{nm}(t') \quad (t>t'), \quad (2.12)$$

where the functions $\mathfrak{U}(t,t')$ satisfy the Hermiticity relation

$$\mathfrak{U}^{*}_{jk;nm}(t,t')=\mathfrak{U}_{kj;mn}(t,t') \quad (2.13)$$

and the initial condition

$$\mathfrak{U}_{jk;nm}(t',t')=\delta_{jn}\delta_{km}. \quad (2.14)$$

The expectation value in Eq. (2.6) may be expressed in terms of the functions $\mathfrak{U}(t,t')$ and the equilibrium density-matrix elements $\rho_{jk}(t')$ as

$$\langle a_{jk}(t)a_{j'k'}^{\dagger}(t')\rangle=\sum_{n,m}\mathfrak{U}_{jk;nm}(t,t')\langle a_{nm}(t')a_{j'k'}^{\dagger}(t')\rangle$$
$$=\sum_{m}\mathfrak{U}_{jk;j'm}(t,t')\rho_{k'm}(t') \quad (t>t'), \quad (2.15)$$

the latter relation following from Eq. (2.7a) and the identity $a_{nm}a_{j'k'}^{\dagger}=\delta_{nj'}a_{k'm}$.

III. RESONANT PUMP FIELD

Let us now suppose that the atom is driven by a strong classical pump field

$$F(t)=F_0e^{-i\omega t}+F_0^{*}e^{i\omega t}, \quad (3.1)$$

which for the sake of generality will be allowed to be electric or magnetic, with $\lambda\hbar$ denoting the relevant (electric or magnetic) dipole-moment operator. It is convenient to introduce off-diagonal decay rates κ'_{jk}, transition rates[16] κ_{jk} from the state $|j\rangle$ to the state $|k\rangle$, and the diagonal decay rate $\kappa_j=\sum_k\kappa_{jk}$. The equations of motion for the density matrix are then

$$\left(\frac{d}{dt}+i\omega_{jk}+\kappa'_{jk}\right)\rho_{jk}(t)-\delta_{jk}\sum_{m}\rho_{mm}(t)\kappa_{mj}$$
$$=iF(t)\cdot[\lambda,\rho(t)]_{jk}, \quad (3.2)$$

where $\kappa'_{jj}\equiv\kappa_j$ and

$$\omega_{jk}\equiv(E_j-E_k)/\hbar. \quad (3.3)$$

(The state labels do not necessarily indicate energy ordering.)

It will be assumed that the pump field induces resonant transitions between a single pair of states $|0\rangle$ and $|1\rangle$ (not necessarily the ground state and first-excited state), so that $\omega_{10}\approx\omega$. Then the only off-diagonal matrix elements which are nonvanishing in equilibrium are $\rho_{10}(t)=\bar{\alpha}_{10}e^{-i\omega t}$ and $\rho_{01}(t)=\bar{\alpha}_{10}^{*}e^{i\omega t}$, and it follows directly from Eqs. (2.6) and (2.15) that the dipole-moment correlation function is given for $t>t'$ by the relation

$$\langle\mu(t)\mu(t')\rangle=\sum_{j,k,j',k'=0}^{\infty}\mu_{kj}\mu_{j'k'}\bar{n}_{k'}\mathfrak{U}_{jk;j'k'}(t,t')$$
$$+\bar{\alpha}_{10}e^{-i\omega t'}\sum_{j,k,j'=0}^{\infty}\mu_{kj}\mu_{j'1}\mathfrak{U}_{jk;j'0}(t,t')$$
$$+\bar{\alpha}_{10}^{*}e^{i\omega t'}\sum_{j,k,j'=0}^{\infty}\mu_{kj}\mu_{j'0}\mathfrak{U}_{jk;j'1}(t,t')$$
$$(t>t'), \quad (3.4)$$

where $\bar{n}_j\equiv\rho_{jj}$ is the (constant) equilibrium occupation number for the state $|j\rangle$. For $t<t'$, the function $\langle\mu(t)\mu(t')\rangle$ may be found from the Hermiticity relation

$$\langle\mu(t)\mu(t')\rangle=\langle\mu(t')\mu(t)\rangle^{\dagger}, \quad (3.5)$$

where $\dagger$ means complex conjugation and transposition of the suppressed vector indices.

The equilibrium matrix elements which refer to the pair of strongly coupled states may be expressed, if relaxation mechanisms connecting these states to other states of the atom can be ignored, as

$$\bar{\alpha}_{10} = iF_0 \cdot \lambda_{10}(\bar{n}_0 - \bar{n}_1)/z^*, \qquad (3.6)$$
$$\bar{n}_0 - \bar{n}_1 = (\bar{n}_0^{(0)} - \bar{n}_1^{(0)})\,\kappa|z|^2/(\kappa'_{10}\Omega^2 + \kappa|z|^2),$$

where $\bar{n}_j^{(0)}$ is the equilibrium occupation number for the state $|j\rangle$ in the absence of the pump field, and

$$z \equiv \kappa'_{10} + i\Delta\omega, \qquad (3.7a)$$
$$\Delta\omega \equiv \omega - \omega_{10}, \qquad (3.7b)$$
$$\kappa \equiv \kappa_{10} + \kappa_{01}, \qquad (3.7c)$$
$$\Omega \equiv 2|F_0 \cdot \lambda_{10}|, \qquad (3.7d)$$
$$\Omega' \equiv [\Omega^2 + (\Delta\omega)^2]^{1/2}. \qquad (3.7e)$$

It is assumed that the parameter Ω is small compared to $\omega \approx \omega_{10}$, and hence that harmonic production and other nonresonant phenomena associated with the action of the pump field by itself can be ignored. (The same assumption enables one to ignore refinements on damping theory such as those proposed by Lehmberg.[17])

The general form of the time-dependent functions $\mathfrak{U}_{jk;nm}(t,t')$ in the presence of the pump field (3.1) is conveniently expressed in terms of parameters $\bar{\omega}_{jk} \equiv -\bar{\omega}_{kj}$, defined by relations similar to Eq. (3.3), but with $E_1 \to E_1 + \frac{1}{2}\hbar\Delta\omega$ and $E_0 \to E_0 - \frac{1}{2}\hbar\Delta\omega$:

$$\bar{\omega}_{jk} \equiv \omega_{jk} \qquad (j,k \geq 2), \qquad (3.8a)$$
$$\bar{\omega}_{j1} \equiv \omega_{j1} - \tfrac{1}{2}\Delta\omega \quad (j \geq 2), \qquad (3.8b)$$
$$\bar{\omega}_{j0} \equiv \omega_{j0} + \tfrac{1}{2}\Delta\omega \quad (j \geq 2), \qquad (3.8c)$$
$$\bar{\omega}_{10} \equiv \omega. \qquad (3.8d)$$

One finds the general time dependence

$$\mathfrak{U}_{jk;nm}(t,t') = e^{i(\bar{\omega}_{nm} - \bar{\omega}_{jk})t'}\mathfrak{U}_{jk;nm}(t-t') = e^{-i\bar{\omega}_{jk}t + i\bar{\omega}_{nm}t'}\mathfrak{U}'_{jk;nm}(t-t'), \qquad (3.9)$$

where the functions $\mathfrak{U}(\tau) \equiv \mathfrak{U}(\tau, 0)$ are given in terms of the slowly varying functions $\mathfrak{U}'(\tau)$ by the relation

$$\mathfrak{U}_{jk;nm}(\tau) = e^{-i\bar{\omega}_{jk}\tau}\mathfrak{U}'_{jk;nm}(\tau). \qquad (3.10)$$

In the general equation (3.2) for the time-dependent density matrix, the elements $\rho_{jk}(t)$ are coupled to one another, in the resonant approximation, only within the following three categories: (a) the category consisting of the off-diagonal elements for which $j \geq 2$ and $k \geq 2$; (b) the category consisting of the elements $\rho_{j0}(t), \rho_{j1}(t)$, and their complex conjugates, for all $j \geq 2$; and (c) the category consisting of all of the diagonal elements and of $\rho_{10}(t)$ and $\rho_{01}(t)$. We shall consider these categories in turn.

(a) In the first category, the density-matrix elements are unaffected by the pump field, and the time-dependent coefficients in Eq. (2.12) are just

$$\mathfrak{U}_{jk;j'k'}(t,t') = \delta_{jj'}\delta_{kk'}e^{-(i\omega_{jk} + \kappa'_{jk})(t-t')}, \quad t > t', \quad j \neq k \quad (j,k \geq 2.) \qquad (3.11)$$

(b) In the second category, the matrix elements are coupled in pairs, with the two elements $\rho_{j1}(t)$ and $\rho_{j0}(t)$ coupled to each other for every value of $j \geq 2$.[8] The functions $\mathfrak{U}(t,t')$, which are nonvanishing in this category, are thus the four functions $\mathfrak{U}_{j1;j1}(t,t')$, $\mathfrak{U}_{j0;j0}(t,t')$, $\mathfrak{U}_{j1;j0}(t,t')$ and $\mathfrak{U}_{j0;j1}(t,t')$, and the functions which are related to these through Eq. (2.13). The Laplace-transform functions

$$\hat{\mathfrak{U}}'(s) \equiv \int_0^\infty d\tau\, e^{-s\tau}\mathfrak{U}'(\tau) \qquad (3.12)$$

in this category are given by the relations[8]

$$\begin{aligned}
\hat{\mathfrak{U}}'_{j1;j1}(s) &= (s + \kappa'_{j0} - \tfrac{1}{2}i\Delta\omega)/f_j(s),\\
\hat{\mathfrak{U}}'_{j0;j0}(s) &= (s + \kappa'_{j1} + \tfrac{1}{2}i\Delta\omega)/f_j(s),\\
\hat{\mathfrak{U}}'_{j1;j0}(s) &= -iF_0^* \cdot \lambda_{10}^*/f_j(s),\\
\hat{\mathfrak{U}}'_{j0;j1}(s) &= -iF_0 \cdot \lambda_{10}/f_j(s), \qquad (j \geq 2)
\end{aligned} \qquad (3.13)$$

in which the polynomial $f_j(s)$ is defined as

$$f_j(s) \equiv (s + \kappa'_{j1} + \tfrac{1}{2}i\Delta\omega)(s + \kappa'_{j0} - \tfrac{1}{2}i\Delta\omega) + \tfrac{1}{4}\Omega^2 \qquad (j \geq 2). \qquad (3.14)$$

(c) Under conditions in which relaxation couplings between either of the states $|0\rangle$ and $|1\rangle$ and other states of the atom can be ignored, this category is effectively reduced to the four matrix elements $\rho_{11}(t), \rho_{00}(t), \rho_{10}(t)$, and $\rho_{01}(t)$. The time-dependent coefficients $\mathfrak{U}'(\tau)$ in this category which enter into Eq. (3.4) have the Laplace transforms[10]

$$\begin{aligned}
\hat{\mathfrak{U}}'_{10;10}(s) &= \frac{(s+z)(s+\kappa) + \frac{1}{2}\Omega^2}{f(s)},\\
\hat{\mathfrak{U}}'_{10;11}(s) &= \frac{iF_0 \cdot \lambda_{10}(s+z)[-s + \kappa(\bar{n}_0^{(0)} - \bar{n}_1^{(0)})]}{sf(s)},\\
\hat{\mathfrak{U}}'_{10;00}(s) &= \frac{iF_0 \cdot \lambda_{10}(s+z)[s + \kappa(\bar{n}_0^{(0)} - \bar{n}_1^{(0)})]}{sf(s)},\\
\hat{\mathfrak{U}}'_{10;01}(s) &= 2(F_0 \cdot \lambda_{10})^2/f(s),
\end{aligned} \qquad (3.15)$$

where the polynomial $f(s)$ is defined in terms of the parameters defined by Eqs. (3.7) as

$$f(s) = (s+\kappa)(s+z)(s+z^*) + \Omega^2(s + \kappa'_{10}). \qquad (3.16)$$

IV. EMISSION

A. Time-Dependent Atomic Correlation Function

The dipole-moment correlation function as given by Eqs. (3.4) and (3.9) has the general form

$$\langle \mu(t)\mu(t')\rangle = g_0(t-t') + \sum_{n=\pm1,\pm2} e^{-in\omega t'}g_{n\omega}(t-t'), \qquad (4.1)$$

in which the functions $g(\tau)$, by virtue of Eq. (3.5), satisfy the Hermiticity relations

$$g_0(\tau)=g_0^{\dagger}(-\tau) \tag{4.2a}$$

and

$$g_{-n\omega}(\tau)=e^{in\omega\tau}g_{n\omega}^{\dagger}(-\tau) \tag{4.2b}$$

for $-\infty\leq\tau\leq\infty$.

By making use of Eqs. (2.13), (2.8), and (3.9) in Eq. (3.4), one finds that the functions $g(\tau)$ are given for $\tau\geq 0$ by the relations

$$g_0(\tau)\underset{(\tau\geq 0)}{=}\sum_{j,k=0}^{\infty}\mu_{kj}\mu_{jk}\bar{n}_k\mathfrak{u}_{jk;jk}(\tau)+\sum_{j=0}^{\infty}[\bar{\alpha}_{10}\mu_{1j}\mu_{j1}\mathfrak{u}_{j1;j0}(\tau)+\bar{\alpha}_{10}^{*}\mu_{0j}\mu_{j0}\mathfrak{u}_{j0;j1}(\tau)], \tag{4.3}$$

$$g_{\omega}(\tau)\underset{(\tau\geq 0)}{=}\sum_{j=2}^{\infty}\{\mu_{0j}\mu_{j1}[\bar{n}_1\mathfrak{u}_{j0;j1}(\tau)+\bar{\alpha}_{10}\mathfrak{u}_{j0;j0}(\tau)]+\mu_{j1}\mu_{0j}\bar{n}_j\mathfrak{u}_{j1;j0}^{*}(\tau)\}, \tag{4.4a}$$

$$g_{-\omega}(\tau)\underset{(\tau\geq 0)}{=}\sum_{j=2}^{\infty}\{\mu_{1j}\mu_{j0}[\bar{n}_0\mathfrak{u}_{j1;j0}(\tau)+\bar{\alpha}_{10}^{*}\mathfrak{u}_{j1;j1}(\tau)]+\mu_{j0}\mu_{1j}\bar{n}_j\mathfrak{u}_{j0;j1}^{*}(\tau)\}, \tag{4.4b}$$

$$g_{2\omega}(\tau)\underset{(\tau\geq 0)}{=}\mu_{01}\mu_{01}e^{-i\omega\tau}[\bar{n}_1\mathfrak{u}'_{10;01}(\tau)+\bar{\alpha}_{10}\mathfrak{u}'_{10;00}(\tau)], \tag{4.5a}$$

$$g_{-2\omega}(\tau)\underset{(\tau\geq 0)}{=}\mu_{10}\mu_{10}e^{i\omega\tau}[\bar{n}_0\mathfrak{u}'^{*}_{10;01}(\tau)+\bar{\alpha}_{10}^{*}\mathfrak{u}'^{*}_{10;11}(\tau)]. \tag{4.5b}$$

B. Emission Cross-Spectral Density; Resonant Multiphoton Processes

The function $g_0(\tau)$ in Eq. (4.1) determines the stationary part of the process under consideration, the remaining functions characterizing time-dependent or nonstationary processes. The (average) spectral density of the emitted radiation, for example, is proportional to the Fourier transform

$$\tilde{g}_0(\nu)=\int_{-\infty}^{\infty}d\tau\,e^{-i\nu\tau}g_0(\tau). \tag{4.6}$$

By making use of Eqs. (4.3) and (4.2a) in this relation one finds, with the aid of Eqs. (2.13), (3.10), and (3.12),

$$\begin{aligned}\tilde{g}_0(\nu)=&\sum_{j,k=0}^{\infty}\mu_{jk}\mu_{kj}\bar{n}_j\,2\,\mathrm{Re}\,\hat{\mathfrak{u}}'_{jk;jk}(-i(\nu-\bar{\omega}_{jk}))\\&+\sum_{j=0}^{\infty}\{\mu_{1j}\mu_{j1}\,2\,\mathrm{Re}\,[\bar{\alpha}_{10}\hat{\mathfrak{u}}'_{j1;j0}(i(\nu-\bar{\omega}_{1j}))]\\&\quad+\mu_{0j}\mu_{j0}\,2\,\mathrm{Re}\,[\bar{\alpha}_{10}^{*}\hat{\mathfrak{u}}'_{j0;j1}(i(\nu-\bar{\omega}_{0j}))]\}.\end{aligned} \tag{4.7}$$

Inasmuch as the functions $\hat{\mathfrak{u}}'(s)$ are appreciable only near $s=0$, it is a simple matter to pick out the terms in Eq. (4.7) corresponding to transitions between any particular pair of states. Transitions from $|j\rangle$ to $|1\rangle$ (where $j\geq 2$ and $E_j>E_1$), for example, are accompanied by the emission of radiation with spectral density[8]

$$\tilde{g}_{0(j\to 1)}(\nu)=\mu_{j1}\mu_{1j}\bar{n}_j\,2\,\mathrm{Re}\,\hat{\mathfrak{u}}'_{j1;j1}(-i(\nu-\bar{\omega}_{j1})), \tag{4.8}$$

while if $E_j<E_1$, the spectral density corresponding to the transition $|1\rangle\to|j\rangle$ is

$$\begin{aligned}\tilde{g}_{0(1\to j)}(\nu)=\mu_{1j}\mu_{j1}\,2\,\mathrm{Re}\,&[\bar{n}_1\hat{\mathfrak{u}}'_{j1;j1}(i(\nu-\bar{\omega}_{1j}))\\&+\bar{\alpha}_{10}\hat{\mathfrak{u}}'_{j1;j0}(i(\nu-\bar{\omega}_{1j}))].\end{aligned} \tag{4.9}$$

Similar expressions, with the state indices 1 and 0 interchanged, hold for the case of transitions from $|j\rangle$ to $|0\rangle$ and $|0\rangle$ to $|j\rangle$, respectively. Finally, the spectral density corresponding to the transition from the upper driven state $|1\rangle$ to the lower state $|0\rangle$ may be found simply by setting $j=0$ in Eq. (4.9).[6]

It should be noted that although these solutions are described as representing direct transitions between specified pairs of states, in fact rather complicated processes, involving multiple atomic transitions and the emission and absorption of arbitrarily high numbers of pump-field and emission-field quanta, are implicitly present, and are correctly described by the same formalism. The only restriction is to processes in which one resonant photon is emitted or absorbed during each atomic transition.

As a means of illustrating this point, it is convenient to examine the limiting form of the spectral density given by Eq. (4.9) when κ'_{j0} and κ'_{j1} are small compared to Ω'. One finds the function to be sharply peaked at the frequencies $\omega_{1j}+\frac{1}{2}\Delta\omega\pm\frac{1}{2}\Omega'$, and to be well approximated by the expression

$$\begin{aligned}\tilde{g}_{0(1\to j)}(\nu)=\mu_{1j}\mu_{j1}\Bigg(&\frac{\kappa'_{j+}[\bar{n}_1(1-\Delta\omega/\Omega')+\mathfrak{W}\Delta\omega/\kappa'_{10}\Omega']}{(\nu-\omega_{1j}-\frac{1}{2}\Delta\omega-\frac{1}{2}\Omega')^2+\kappa'^2_{j+}}\\&+\frac{\kappa'_{j-}[\bar{n}_1(1+\Delta\omega/\Omega')-\mathfrak{W}\Delta\omega/\kappa'_{10}\Omega']}{(\nu-\omega_{1j}-\frac{1}{2}\Delta\omega+\frac{1}{2}\Omega')^2+\kappa'^2_{j-}}\Bigg)\end{aligned}$$

$$(\Omega'\gg\kappa'_{j1},\kappa'_{j0}), \tag{4.10}$$

in which the widths are defined as

$$\kappa'_{j\pm}=\tfrac{1}{2}(\kappa'_{j0}+\kappa'_{j1})\pm\tfrac{1}{2}(\kappa'_{j0}-\kappa'_{j1})\Delta\omega/\Omega', \tag{4.11}$$

and $\mathfrak{W}$, the rate at which quanta are absorbed from the pump field, is given by the relation

$$\mathfrak{W}=\tfrac{1}{2}\Omega^2\kappa'_{10}(\bar{n}_0-\bar{n}_1)/|z|^2\approx\tfrac{1}{2}\Omega^2\kappa'_{10}(\bar{n}_0-\bar{n}_1)/(\Delta\omega)^2, \tag{4.12}$$

the latter approximation holding for $\kappa'_{10} \ll |\Delta\omega|$.

In the limit of strong pump fields, the peaks in the spectral density in Eq. (4.10) become equal in width and in integrated strength. The function is well approximated in this limit as

$$\tilde{g}_{0(1\to j)}(\nu) = \mu_{1j}\mu_{j1}\bar{n}_1\kappa'_j\{[(\nu-\omega_{1j}-\tfrac{1}{2}\Omega)^2+\kappa'^2_j]^{-1} + [(\nu-\omega_{1j}+\tfrac{1}{2}\Omega)^2+\kappa'^2_j]^{-1}\} \quad (\Omega \gg |\Delta\omega|, \kappa'_{jk}), \quad (4.13)$$

where

$$\kappa'_j = \tfrac{1}{2}(\kappa'_{j0}+\kappa'_{j1}). \quad (4.14)$$

In the limit of weak pump fields, on the other hand, the spectral density for the same transition is, to order Ω^2,

$$\tilde{g}_{0(1\to j)}(\nu) = \mu_{1j}\mu_{j1}\left(\frac{\tfrac{1}{2}\kappa'_{j0}\bar{n}_0\Omega^2/(\Delta\omega)^2}{(\nu-\omega+\omega_{j0})^2+\kappa'^2_{j0}} + \frac{\kappa'_{j1}[2\bar{n}_1-\tfrac{1}{2}\bar{n}_0\Omega^2/(\Delta\omega)^2]}{(\nu-\omega_{1j})^2+\kappa'^2_{j1}}\right) \quad (\Omega, \kappa'_{jk} \ll |\Delta\omega|). \quad (4.15)$$

If the state $|1\rangle$ is populated by transitions (e.g., collisional) other than those induced from the state $|0\rangle$ by the pump field, then in the limit $\Omega \to 0$, the second term on the right-hand side of Eq. (4.15) is predominant, and in fact reduces to the familiar spontaneous emission field for the $|1\rangle \to |j\rangle$ transition. The (smaller) first term, on the other hand, represents a Raman-type transition from the state $|0\rangle$ to the state $|j\rangle$, induced by the absorption of a pump-field photon of frequency ω followed by the emission of a photon of frequency ν. The state $|1\rangle$ is an intermediate state in this process, and does not enter into the determination of the width κ'_{j0} or the emission resonance frequency $\omega - \omega_{j0}$.[18]

It should be emphasized that the equilibrium occupation numbers in all of these relations are to be determined in the presence of the pump field, and may even be wholly due to the action of the pump field. An example of the latter possibility is the case in which $|0\rangle$ is the ground state of the atom, and the atomic relaxation is purely radiative [$\kappa'_{jk} = \tfrac{1}{2}(\kappa_j+\kappa_k)$, where κ_j and κ_k are the natural widths of the states $|j\rangle$ and $|k\rangle$]. It is found in this case that the relative magnitudes of the two terms in Eq. (4.15) are reversed, with the second now the smaller. [It is given as zero in this case by Eq. (4.15) and must be evaluated by returning to Eq. (4.10).] One finds in this case that

$$\tilde{g}_{0(1\to j)}(\nu) = \mu_{1j}\mu_{j1}\left(\frac{\tfrac{1}{4}\kappa_j\Omega^2/(\Delta\omega)^2}{(\nu-\omega+\omega_{j0})^2+\tfrac{1}{4}\kappa_j^2} + \frac{\tfrac{1}{16}(\kappa_1+\kappa_j)\Omega^4/(\Delta\omega)^4}{(\nu-\omega_{1j})^2+\tfrac{1}{4}(\kappa_1+\kappa_j)^2}\right). \quad (4.16)$$

The second term in this relation, which resembles in resonance frequency and in width a spontaneous-emission transition from $|1\rangle$ to $|j\rangle$, may be shown to represent the photon emitted during the last transition in the sequence $|0\rangle \to |1\rangle \to |0\rangle \to |1\rangle \to |j\rangle$, where the transitions $|0\rangle \to |1\rangle$ in each case are induced by the absorption of a pump-field photon, while the transition $|1\rangle \to |0\rangle$ is accompanied by the emission of a photon of frequency near the pump-field resonance frequency ω_{10}.

It is instructive here to examine the transition $|j\rangle \to |0\rangle$, for a state $|j\rangle$ with energy higher than the state $|0\rangle$. One finds in this case the spectral density

$$\tilde{g}_{0(j\to 0)}(\nu) = \mu_{j0}\mu_{0j}\bar{n}_j\left(\frac{2\kappa'_{j0}}{(\nu-\omega_{j0})^2+\kappa'^2_{j0}} + \frac{\tfrac{1}{2}\kappa'_{j1}\Omega^2/(\Delta\omega)^2}{(\nu-\omega+\omega_{1j})^2+\kappa'^2_{j1}}\right) \quad \text{for } \Omega, \kappa'_{jk} \ll |\Delta\omega|. \quad (4.17)$$

The first term in this relation corresponds to the spontaneous-emission field. The (smaller) second term is due to the composite process $|j\rangle \to |0\rangle \to |1\rangle$, with a photon of frequency ν emitted during the first stage and a pump-field photon of frequency υ absorbed during the second: The width κ'_{j1} and the resonance frequency $\omega - \omega_{1j}$ for the emission are determined solely by the initial and final states $|j\rangle$ and $|1\rangle$ of the composite process. The state $|j\rangle$ may here be populated by collisions or by radiative transitions from higher states, e.g., as part of a cascade process.

A further possibility, which arises when inversion asymmetry is present, is that a state $|j\rangle$ intermediate in energy between the states $|1\rangle$ and $|0\rangle$ and coupled to both may be populated wholly by the transitions from the state $|1\rangle$ represented in Eq. (4.16). [This implies $\bar{n}_j = \tfrac{1}{4}\kappa_{10}\Omega^2/\kappa_j(\Delta\omega)^2$.] The first terms on the right-hand sides of Eqs. (4.16) and (4.17) then can be shown to correspond to a frequency-splitting process, in which a pump-field photon, absorbed during a transition from $|0\rangle$ to $|1\rangle$, divides its energy between two (nearly resonant) photons of smaller energy, emitted successively during the transitions $|1\rangle \to |j\rangle$ and $|j\rangle \to |0\rangle$. [Exact energy conservation in this process is not evident from an examination of the stationary spectral functions considered so far, but requires an analysis (presented below) of the relevant nonstationary functions.] The small second terms on the right-hand sides of Eqs. (4.16) and (4.17) in this case are due, in addition to the process discussed above, following Eq. (4.16) (which now must be completed with a

$|j\rangle \rightarrow |0\rangle$ transition and is thus a three-photon emission process), to two additional processes, both beginning with the elementary-transition sequence $|0\rangle \rightarrow |1\rangle \rightarrow |j\rangle \rightarrow |0\rangle$, one ending with a repetition of this same sequence, the other with the sequence $|0\rangle \rightarrow |1\rangle \rightarrow |0\rangle$. The former process involves the emission of four photons whose energies sum to 2ω. These processes are rather difficult to analyze by the usual methods of scattering theory, even in the weak-pump-field limit presently under discussion. By far the better method is to deal directly with the correlation functions, which are the quantities of direct physical significance, and which can be evaluated by an entirely systematic method which automatically includes the effects of whatever processes are important.

C. Nonstationary Emission Functions; Cross-Spectral Terms

The nonstationary terms in Eq. (4.1) corresponding to the transitions of interest may also be evaluated in a straightforward manner. The contribution of the oscillating terms proportional to g_ω and $g_{-\omega}$ in the cross-spectral correlation function defined by Eq. (2.1), for example, is

$$g_\omega^{(1,1)}(t', t) = e^{-i\omega t} g_\omega^{(-)}(t' - t) + [t \rightarrow t']^\dagger, \quad (4.18)$$

where the superscript on the right-hand side is a frequency signature. By making use of Eqs. (4.4) and (3.10), one finds that appreciable contributions to this function occur for the case of transitions involving any state $|j\rangle$ for which either $E_j > E_1$ or $E_j < E_0$. In the first case, one finds, with the aid of Eqs. (4.4) and (4.2b), and after inverting the Laplace transforms in Eqs. (3.13) and (3.14) under the special assumption $\kappa'_{j1} = \kappa'_{j0} \equiv \kappa'_j$, the contribution

$$g_{\omega(j)}^{(1,1)}(t', t) = (-2i\bar{n}_j/\Omega')e^{-\kappa'_j|t-t'|}\sin\tfrac{1}{2}\Omega'(t - t') \times [F_0 \cdot \lambda_{10}\mu_{j1}\mu_{0j}e^{i\bar{\omega}_{j1}t' - i\bar{\omega}_{j0}t} + F_0^* \cdot \lambda_{10}^*\mu_{j0}\mu_{1j}e^{i\bar{\omega}_{j0}t' - i\bar{\omega}_{j1}t}] \quad (E_j > E_1), \quad (4.19)$$

where Ω' is defined by Eq. (3.7e). For $E_j < E_0$, on the other hand, one finds the contribution

$$g_{\omega(j)}^{(1,1)}(t', t) \underset{(t \geq t')}{=} \mu_{0j}\mu_{j1}e^{i\bar{\omega}_{0j}t' - i\bar{\omega}_{1j}t} \times [\bar{n}_0 \mathfrak{u}'^*_{1;j0}(t - t') + \bar{\alpha}_{10}\mathfrak{u}'^*_{1;j1}(t - t')] + (1 \rightarrow 0) \quad (E_j < E_0), \quad (4.20)$$

in which the interchange of the state indices 1 and 0 must be understood to imply the interchange $\bar{\alpha}_{10} \rightarrow \bar{\alpha}_{10}^*$.

The expressions given by Eqs. (4.19) and (4.20) both represent interference, the first between the fields emitted during the two transitions $|j\rangle \rightarrow |1\rangle$ and $|j\rangle \rightarrow |0\rangle$, and the second between the fields emitted during the two transitions $|1\rangle \rightarrow |j\rangle$ and $|0\rangle \rightarrow |j\rangle$. The interference is of course produced by the action of the pump field in coupling the states $|0\rangle$ and $|1\rangle$, and vanishes for $F_0 = 0$.

In the second case ($E_j < E_0$), the interference terms lead to an oscillatory time dependence in the *total* emission rate $g^{(1,1)}(t, t)$. By evaluating Eq. (4.20) at $t' = t$ with the aid of Eq. (2.14) one finds, after adding the total stationary emission rate [$= \int \tilde{g}_0(\nu)d\nu/2\pi$], the relation

$$g_j^{(1,1)}(t, t) = \mu_{0j}\mu_{j0}\bar{n}_0 + \mu_{1j}\mu_{j1}\bar{n}_1 + \mu_{0j}\mu_{j1}\bar{\alpha}_{10}e^{-i\omega t} + \mu_{1j}\mu_{j0}\bar{\alpha}_{10}^* e^{i\omega t}. \quad (4.21)$$

An oscillatory component in the total emission rate is thus present in this case whenever the off-diagonal matrix element $\bar{\alpha}_{10}$ referring to the pair of pump-field coupled states is nonvanishing. The expression given by Eq. (4.19), on the other hand, vanishes identically at $t' = t$, and hence no oscillatory component in the total emission rate is present for transitions from an upper state $|j\rangle$, where $E_j > E_1$, to the coupled states $|0\rangle$ and $|1\rangle$.

The nonstationary components in the Eqs. (4.19)–(4.21) are simplest to interpret when the oscillations at the pump-field frequency are slow compared to the oscillations of the emitted radiation, i.e., when $\omega \approx \omega_{10} \ll \omega_{j1} \approx \omega_{j0}$. This condition is well satisfied in double-resonance or optical-pumping experiments,[2] where the pump field oscillates at rf or at microwave frequencies, coupling pairs of states within a Zeeman or hyperfine multiplicity, while the process is monitored by observing its effect on optical transitions. The nonstationary functions in Eqs. (4.19) and (4.20) in such cases appear in squared form in the expression for the intensity autocorrelation function[11,12] of the emitted field, leading to oscillations in this function both at the pump-field frequency ω and at the (lower) Rabi frequency Ω'.[19] Unlike the oscillations in the total emission rate, these oscillations survive even the limit of intense pump fields, where $\bar{\alpha}_{10} \rightarrow 0$.

It should be emphasized that the phase of the oscillations in the nonstationary components depends on the phase of the pump field, and hence in general varies within the medium. The contributions from different atoms to the emission field will in general tend to cancel one another except at observation points very nearly in the direction of propagation of the pump field, unless the pump-field wavelength is large compared to the sample size.

D. Double-Positive Frequency Nonstationary Functions

The emission process has been described so far only in terms of the cross-spectral correlation function for the radiated field, which is proportional to the cross-spectral atomic correlation function defined by Eq. (2.1). The field correlation function which represents the mean value of the product of two field-annihilation operators is also important, however, especially in cases in which photons are emitted in pairs, where its value is needed to determine the field-intensity autocorrelation function[20,21] and hence the coincidence counting rates. The function in question may be shown to be proportional to the double-positive frequency, time-ordered atomic correlation function[21]

$$g^{(0,2)}(t_2, t_1) \equiv \langle (\mu^{(+)}(t_2)\mu^{(+)}(t_1))_T \rangle . \tag{4.22}$$

The Fourier transform function

$$\bar{g}^{(0,2)}(\nu_2, \nu_1) = (2\pi)^{-1} \iint_{-\infty}^{\infty} dt_2\, dt_1 \times e^{i\nu_2 t_2 + i\nu_1 t_1} g^{(0,2)}(t_2, t_1) \tag{4.23}$$

is proportional, in lowest order, to the wave amplitude for finding two photons coherently radiated (i.e., with definite phase relationship) at the frequencies ν_1 and ν_2.

Appreciable resonant contributions to this function occur, according to Eqs. (4.1), (4.4a), and (3.10), whenever a state $|j\rangle$ is intermediate in energy between the states $|0\rangle$ and $|1\rangle$ and is coupled to both of them. The frequencies of the emitted photons add up exactly to ω, showing that the process so described is one of frequency splitting of the pump field. To lowest order in the pump-field intensity, under the assumption that the atomic states in question have nonvanishing populations even for zero pump-field intensity, one finds

$$\bar{g}^{(0,2)}(\nu_2, \nu_1) = \delta(\nu_1 + \nu_2 - \omega)\mu_{0j}\mu_{j1} i F_0 \cdot \lambda_{10} \times \left(\frac{\bar{n}_0}{\zeta_1 \Delta\omega} - \frac{\bar{n}_1}{\zeta_1' \Delta\omega} - \frac{\bar{n}_j}{\zeta_1^* \zeta_1'^*} \right) + (\nu_1 \rightleftharpoons \nu_2)^T \quad (\Omega, \kappa_{jk}' \ll |\Delta\omega|), \tag{4.24}$$

where $\zeta_1 \equiv \nu_1 - (\omega - \omega_{j0}) - i\kappa_{j0}'$ and $\zeta_1' \equiv \nu_1 - \omega_{1j} - i\kappa_{j1}'$.

Each of the three terms exhibited in Eq. (4.24) represents a process in which a pump-field photon is absorbed during the transition $|0\rangle \rightarrow |1\rangle$, and photons of frequency ν_1 and ν_2 are emitted during the transitions $|1\rangle \rightarrow |j\rangle$ and $|j\rangle \rightarrow |0\rangle$, respectively. The three processes differ from one another only in the order in which these events occur. In the term proportional to $\bar{n}_0$ the absorption occurs first, while in the term proportional to $\bar{n}_1$, both emissions occur before the absorption. The term proportional to $\bar{n}_j$, finally, represents a process in which the emission at frequency ν_2 occurs first, followed by the absorption and then by the emission at frequency ν_1. In the processes represented by the terms proportional to $\bar{n}_1$ and $\bar{n}_j$, one must think of the states $|1\rangle$ and $|j\rangle$ as initial states in a perturbation-theory sense, having been populated, e.g., by collisional processes.

If $|0\rangle$ is the ground state and the relaxation is purely radiative [the situation described in Eqs. (4.16) and (4.17)], however, one must think of $|0\rangle$ as the initial state for any process described in elementary perturbation-theory terms. In this case [which Eq. (4.24) is insufficiently accurate to describe] one finds that the function defined by Eq. (4.23) is well approximated in the limit of weak pump fields by the expression

$$\bar{g}^{(0,2)}(\nu_2, \nu_1) = \delta(\nu_1 + \nu_2 - \omega)\mu_{0j}\mu_{j1} \frac{i F_0 \cdot \lambda_{10}}{\Delta\omega} \times \left(\frac{1}{\zeta_1} - \frac{\bar{n}_j}{\zeta_1'^*} \right) + (\nu_1 \rightleftharpoons \nu_2)^T . \tag{4.25}$$

While the first term still represents the elementary frequency-splitting process during the transition sequence $|0\rangle \rightarrow |1\rangle \rightarrow |j\rangle \rightarrow |0\rangle$, the term proportional to $\bar{n}_j$ may be shown to be due to phase coherence between the wave function for the photon pair thus emitted and the wave function which describes the four-photon emission process already mentioned as being responsible, in part, for the terms centered at $\nu_1 = \omega_{1j}$ and $\nu_2 = \omega - \omega_{1j}$ in the spectral densities given by Eqs. (4.16) and (4.17).

Another contribution of interest to the function $g^{(0,2)}(t_2, t_1)$ is made by the terms proportional to $g_{2\omega}$ in Eq. (4.1). These are due to the action of the pump field on the states $|0\rangle$ and $|1\rangle$ themselves, and involve no other state of the atom.[22] The contribution in question has the form $e^{i\omega(t_2+t_1)}$ times a function of $t_2 - t_1$, thus leading to the factor $\delta(\nu_1 + \nu_2 - 2\omega)$ in the spectral function defined by Eq. (4.23). In the weak-pump-field limit, the latter function is found to be sharply peaked at the frequencies $\omega - \Delta\omega$ and $\omega + \Delta\omega$, thus representing the coherent emission of pairs of photons at these frequencies. The basic process in operation here consists of the absorption of two pump-field photons followed by the emission of two photons at nearby frequencies ν_1 and ν_2, the relation $\nu_1 + \nu_2 = 2\omega$ representing energy conservation. (The atomic transition sequence is $|0\rangle \rightarrow |1\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |0\rangle$, with the absorptions and emissions occurring during the transitions $|0\rangle \rightarrow |1\rangle$ and $|1\rangle \rightarrow |0\rangle$,

respectively.) What is being described here is in fact the spontaneous part of the process which has been called light-by-light scattering[23] when it is stimulated by the initial presence of a photon at either of the frequencies ν_1 or ν_2.

The (spontaneous) two-photon emission process under discussion here must of course contribute to the ordinary cross-spectral density defined by Eq. (4.6), since $\int |g^{(0,2)}(\nu_2, \nu_1)|^2 d\nu_2$ is proportional to the probability of finding one of the pair of emitted photons with frequency ν_1. The contribution one finds in this way in the weak-pump-field limit (where processes of higher order may be neglected) is indeed in agreement with the result which has previously been found for the cross-spectral density in the same limit.[6] Proper interpretation of the effect as one of coherent two-photon emission, in addition to implying an increased coincidence counting rate, implies that the emission is sharply peaked in the direction of the pump beam, where the contributions from different atoms to $g^{(0,2)}$ add coherently. These facts cannot be deduced from an examination of the stationary cross-spectral density alone.

V. TIME-DEPENDENT LINEAR SUSCEPTIBILITY AND ABSORPTION

A. General Relations

The time-dependent complex susceptibility as given by Eqs. (2.5) and (4.1) has the general form

$$\chi(\nu, t) = \chi_0(\nu) + \sum_{n=\pm1,\pm2} e^{-in\omega t} \chi_{n\omega}(\nu). \tag{5.1}$$

Although ν must of course be understood as positive in physical applications, Eq. (2.5) defines a perfectly well-behaved function for all positive and negative values of ν. The function so defined satisfies the relation

$$\chi(\nu, t) = \chi^*(-\nu, t), \tag{5.2}$$

which is simply a reflection of the invariance of the polarization (2.4) under the interchange $E_0' e^{-i\nu t} \rightarrow E_0'^* e^{i\nu t}$ [which leaves the signal field (2.2) unchanged]. It follows from Eq. (5.2) that the coefficients in Eq. (5.1) obey the identities

$$\chi_{-n\omega}(\nu) = \chi^*_{n\omega}(-\nu). \tag{5.3}$$

By substituting Eqs. (4.3)–(4.5) and (4.2) into Eqs. (4.1) and (2.5), we find, with the aid of Eqs. (2.13) and (3.10), that the coefficients $\chi_0(\nu)$, $\chi_\omega(\nu)$, and $\chi_{2\omega}(\nu)$ in Eq. (5.1) are given by the relations[23(a)]

$$\chi_0(\nu) = i(N/\hbar) \sum_{j,k=0}^{\infty} \mu_{kj}\mu_{jk}(\bar{n}_k - \bar{n}_j)\hat{u}'_{jk;jk}(-i(\nu - \bar{\omega}_{jk}))$$

$$+ (iN/\hbar) \sum_{j=0}^{\infty} \{[\bar{\alpha}_{10}\mu_{1j}\mu_{j1}\hat{u}'_{j1;j0}(-i(\nu - \bar{\omega}_{j1})) + \bar{\alpha}^*_{10}\mu_{0j}\mu_{j0}\hat{u}'_{j0;j1}(-i(\nu - \bar{\omega}_{j0}))] - [\nu \rightarrow -\nu]^*\}, \tag{5.4a}$$

$$\chi_\omega(\nu) = i(N/\hbar)\left\{\sum_{j=2}^{\infty} \mu_{0j}\mu_{j1}[(\bar{n}_1 - \bar{n}_j)\hat{u}'_{j0;j1}(-i(\nu - \bar{\omega}_{j1})) + \bar{\alpha}_{10}\hat{u}'_{j0;j0}(-i(\nu - \bar{\omega}_{j1}))]\right\} - i\frac{N}{\hbar}\begin{Bmatrix}\nu \rightarrow -\nu \\ 0 \rightarrow 1\end{Bmatrix}^*, \tag{5.4b}$$

and

$$\chi_{2\omega}(\nu) = i(N/\hbar)\mu_{01}\mu_{01}\{(\bar{n}_1 - \bar{n}_0)\hat{u}'_{10;01}(-i(\nu + \omega)) + \bar{\alpha}_{10}[\hat{u}'_{10;00}(-i(\nu + \omega)) - \hat{u}'_{10;11}(-i(\nu + \omega))]\}. \tag{5.4c}$$

B. Stationary Components

For values of the signal-field frequency ν far from any atomic resonance, the functions $\hat{u}'(s)$ are not appreciably affected by the pump field, and are thus given by the approximate relation

$$\hat{u}'_{jk;nm}(i\Delta\nu) = \delta_{jn}\delta_{km}(i\Delta\nu)^{-1} \quad (|\Delta\nu| \gg \Omega', \kappa). \tag{5.5}$$

The stationary part of the susceptibility as expressed by Eq. (5.4a) is thus equal in this limit to the expression

$$\chi_0(\nu) = N \sum_{j,k=0}^{\infty} \frac{\mu_{kj}\mu_{jk}(\bar{n}_k - \bar{n}_j)}{\hbar(\omega_{jk} - \nu)}. \tag{5.6}$$

This relation makes no explicit reference to the pump field, and in fact is the standard perturbation-theory result for the real off-resonance susceptibility for stationary systems. The pump field nevertheless plays an important implicit role, in determining the occupation numbers $\bar{n}_j$. It should be emphasized in this connection that the susceptibility $\chi_0(\nu)$ as given by Eq. (5.6) is in general anisotropic, a preferred direction being defined by the polarization of the pump field.

When the signal-field frequency is near an atomic-resonance frequency, the stationary part of the complex susceptibility as given by Eq. (5.4a) is easily evaluated with the aid of Eqs. (3.13) and (3.15). One finds,[24] for $E_j > E_1$ and $\nu \approx \omega_{j1} > 0$,

$$\chi_{0(j1)}(\nu)=i\frac{N}{\hbar}\mu_{1j}\mu_{j1}\left(\frac{(\bar{n}_1-\bar{n}_j)[-i(\nu-\omega_{j1}+\Delta\omega)+\kappa'_{j0}]-iF_0^*\cdot\lambda_{10}^*\bar{\alpha}_{10}}{f_j(-i(\nu-\omega_{j1}+\frac{1}{2}\Delta\omega))}\right)\quad(E_j>E_1),\tag{5.7a}$$

while for $E_j<E_1$ and $\nu\approx\omega_{1j}>0$, one finds

$$\chi_{0(1j)}(\nu)=i\frac{N}{\hbar}\mu_{j1}\mu_{1j}\left(\frac{(\bar{n}_j-\bar{n}_1)[i(\nu-\omega_{1j}-\Delta\omega)+\kappa'_{j0}]+iF_0^*\cdot\lambda_{10}^*\bar{\alpha}_{10}}{f_j(i(\nu-\omega_{1j}-\frac{1}{2}\Delta\omega))}\right)^*\quad(E_j<E_1),\tag{5.7b}$$

where ω_{jk} and $\Delta\omega$ are defined by Eqs. (3.3) and (3.7b), respectively, and $f_j(s)$ is defined by Eq. (3.14). The two cases $\nu\approx\omega_{j0}$ (where $E_j>E_0$) and $\nu\approx\omega_{0j}$ (where $E_j<E_0$) can be treated by interchanging the state indices 1 and 0 in Eqs. (5.7a) and (5.7b), respectively, and making the substitutions $\Delta\omega\to-\Delta\omega$ and $F_0^*\cdot\lambda_{10}^*\bar{\alpha}_{10}\to F_0\cdot\lambda_{10}\bar{\alpha}_{10}^*$. Finally, if the signal field induces resonant transitions between the same pair of states $|0\rangle$ and $|1\rangle$ as are coupled by the pump field,[10] i.e., if $\nu\approx\omega$, the stationary part of the susceptibility is

$$\chi_{0(10)}(\nu)=i\frac{N}{\hbar}\mu_{01}\mu_{10}(\bar{n}_0-\bar{n}_1)\times\left(\frac{(-i\Delta\nu+\kappa)(-i\Delta\nu+z)+\frac{1}{2}i\Omega^2\Delta\nu/z}{f(-i\Delta\nu)}\right),\tag{5.8}$$

where $\Delta\nu\equiv\nu-\omega$, $f(s)$ is defined by Eq. (3.16), and the remaining parameters are defined by Eqs. (3.7).

The limiting forms of these functions are readily found. The function defined by Eq. (5.7a), for example, in the limit $\kappa'_{j1},\ \kappa'_{j0}\ll\Omega'$, behaves much as if the state $|1\rangle$ had split up into two states with energies $E_1+\frac{1}{2}\hbar(\Delta\omega\pm\Omega')$ and widths $\kappa'_{j\pm}$ [with the latter defined by Eq. (4.11)]. The function is well approximated in this limit as

$$\chi_{0(j1)}(\nu)=\frac{1}{2}\frac{N}{\hbar}\mu_{1j}\mu_{j1}\left(\frac{(\bar{n}_1-\bar{n}_j)(1+\Delta\omega/\Omega')+i\mathcal{W}z/\kappa'_{10}\Omega'}{\omega_{j1}-\frac{1}{2}\Delta\omega+\frac{1}{2}\Omega'-\nu-i\kappa'_{j-}}+(\Omega'\to-\Omega')\right)\quad(\Omega'\gg\kappa'_{j1},\kappa'_{j0};E_j>E_1),\tag{5.9}$$

where $\mathcal{W}$ is defined by Eq. (4.12) and the substitution $\Omega'\to-\Omega'$ implies $\kappa'_{j-}\to\kappa'_{j+}$. For weak pump fields, one finds that

$$\chi_{0(j1)}(\nu)=\frac{N}{\hbar}\mu_{1j}\mu_{j1}\left(\frac{\bar{n}_1-\bar{n}_j}{\omega_{j1}-\nu-i\kappa'_{j1}}+\frac{(\bar{n}_0-\bar{n}_j)\frac{1}{4}\Omega^2/(\Delta\omega)^2}{\omega_{j0}-\omega-\nu-i\kappa'_{j0}}\right)\quad(\Omega,\kappa'_{jk}\ll|\Delta\omega|;\ E_j>E_1).\tag{5.10}$$

The first term in this relation is the familiar expression for the complex susceptibility near a homogeneously broadened (undriven) resonance line. The smaller pump-field-dependent second term represents a process in which the atom, initially in the state $|0\rangle$, reaches the state $|j\rangle$ after absorbing two photons, a pump-field photon of frequency ω and a signal-field photon of frequency ν. The state $|1\rangle$ is an intermediate state in this process, and consequently does not enter into the evaluation of the resonance frequency or width.

In the limit of strong pump fields, the complex susceptibility is split into two terms of equal magnitude, each identical in form to that for an undriven line. It is well approximated in this limit by the relation

$$\chi_{0(j1)}(\nu)=\tfrac{1}{2}(N/\hbar)\mu_{1j}\mu_{j1}(\bar{n}_1-\bar{n}_j)[(\omega_{j1}+\tfrac{1}{2}\Omega-\nu-i\kappa'_j)^{-1}+(\omega_{j1}-\tfrac{1}{2}\Omega-\nu-i\kappa'_j)^{-1}],\tag{5.11}$$

where $\kappa'_j=\frac{1}{2}(\kappa'_{j1}+\kappa'_{j0})$.

The mean rate at which energy per unit volume is absorbed from the signal field may in all cases be expressed as

$$W'_0=2cE'_0{}^*\beta_0(\nu)E'_0,\tag{5.12}$$

where β_0, the (second-rank tensor) linear absorption coefficient is defined as

$$\beta_0(\nu)=\nu[\chi_0(\nu)-\chi_0^\dagger(\nu)]/2ic,\tag{5.13}$$

and may be found from the solutions for the emission spectral densities by using Eq. (2.10).

C. Nonstationary Components; Off-Resonance Functions

The meaning of the time-dependent terms in the complex susceptibility given by Eq. (5.1) is shown by substituting them into Eq. (2.4). The term proportional to $\chi_\omega(\nu)$, for example, induces the polarization component

$$P'_1(t)=\chi_\omega(\nu)E'_0e^{-i(\nu+\omega)t}+\text{c.c.},\tag{5.14}$$

while the term proportional to $\chi_{-\omega}(\nu)$ induces the component

$$P'_{-1}(t)=\chi_{-\omega}(\nu)E'_0e^{-i(\nu-\omega)t}+\text{c.c.}\quad\text{for }\omega<\nu\tag{5.15a}$$

$$=\chi_{-\omega}^*(\nu)E'_0{}^*e^{-i(\omega-\nu)t}+\text{c.c.}\quad\text{for }\omega>\nu,\tag{5.15b}$$

the positive-frequency part being exhibited in each

case. The electric fields generated by the polarization components in Eqs. (5.14) and (5.15a) are examples of frequency up and down conversion, respectively, and correspond to the absorption of a signal-field photon followed by its re-emission at a different frequency, the energy difference being made up by the emission or absorption of a pump-field photon. The process described in Eq. (5.15b), on the other hand, is stimulated subharmonic generation (or frequency splitting), and consists of the absorption of a pump-field photon followed by the emission of two photons, one at the frequency $\omega - \nu$ and the other at the signal-field frequency ν. (The spontaneous part of this process was discussed in Sec. IV.)

Both of these processes are susceptible of a straightforward Hamiltonian description when ν is far from any atomic resonance. The quantities $\chi_\omega(\nu)$ and $\chi_{-\omega}(\nu)$, according to Eqs. (5.4b), (5.3), and (5.5), are then given as

$$\chi_\omega(\nu) = \frac{N}{\hbar}\bar{\alpha}_{10}\sum_{j=2}^{\infty}\left(\frac{\mu_{0j}\mu_{j1}}{\omega_{j1}-\nu}+\frac{\mu_{j1}\mu_{0j}}{\omega_{j0}+\nu}\right),$$
$$\chi_{-\omega}(\nu) = \frac{N}{\hbar}\bar{\alpha}^{*}_{10}\sum_{j=2}^{\infty}\left(\frac{\mu_{1j}\mu_{j0}}{\omega_{j0}-\nu}+\frac{\mu_{j0}\mu_{1j}}{\omega_{j1}+\nu}\right), \tag{5.16}$$

and consequently obey the identities

$$\chi^{\dagger}_{\omega}(\nu) = \chi_{-\omega}(\nu+\omega), \quad \chi^{*\dagger}_{-\omega}(\nu) = \chi^{\dagger}_{-\omega}(\omega-\nu). \tag{5.17}$$

As was indicated previously, if the phase of the pump field varies within the medium, the time-dependent susceptibilities are spatially inhomogeneous as well as anisotropic, having the spatial dependence for pump fields $F_0(z) = F_0 e^{ikz}$,

$$\chi_{n\omega}(\nu, z) = e^{inkz}\chi_{n\omega}(\nu, 0), \tag{5.18}$$

where $n = 0, \pm 1, \pm 2$. It follows directly from this relation that in a dilute medium ($\chi \ll 1$) only pairs of waves traveling parallel to the pump wave will be appreciably coupled.

It is interesting to observe that a single initial nonresonant light wave of frequency ν traveling parallel to a pump wave oscillating, say, at a microwave frequency ω, would thus soon acquire frequency components at $\nu \pm n\omega$, where $n = 1, 2, 3, \ldots$, owing to the successive operation of the processes described in Eqs. (5.14) and (5.15a).

For optimal pump intensity, $\bar{\alpha}_{10}$ can be made comparable to unity, and the susceptibilities evaluated in Eq. (5.16) become comparable to the stationary susceptibility given by Eq. (5.6). The effect could thus, in principle, be made appreciable over distances comparable to that required for the phase change due to the ordinary (real) dielectric susceptibility.[24(a)]

In lowest order, the effect we are speaking of appears as a time-dependent modulation of the signal-field absorption rate. Indeed, by directly evaluating the absorption rate $E' \cdot \dot{P}'$, with P' given by Eq. (2.4), we find that the contribution of the first three terms in Eq. (5.1) is $2\nu E'^{*}_0 \beta(\nu, t) E'_0$, where the time-dependent linear absorption coefficient $\beta(\nu, t)$ is

$$\beta(\nu, t) = \beta_0(\nu) + \beta_\omega(\nu)e^{-i\omega t} + \beta^{\dagger}_{\omega}(\nu)e^{i\omega t}, \tag{5.19}$$

$\beta_0(\nu)$ being given by Eq. (5.13), and $\beta_\omega(\nu)$ by the relation

$$\beta_\omega(\nu) = \{\nu[\chi_\omega(\nu) - \chi^{\dagger}_{-\omega}(\nu)] + \omega[\chi_\omega(\nu) + \chi^{\dagger}_{-\omega}(\nu)]\}/2ic. \tag{5.20}$$

It should be noted that the full expression (5.20) for $\beta_\omega(\nu)$ must be used in the off-resonance case, since it follows from Eqs. (5.16) that the term proportional to ω on the right-hand side of Eq. (5.20) is comparable to the term proportional to ν, even though $\omega \ll \nu$.

D. Nonstationary Components; Resonance Functions

When the signal-field frequency is near to an atomic-resonance frequency, on the other hand, it becomes possible to approximate Eq. (5.20) as

$$\beta_\omega(\nu) = \nu[\chi_\omega(\nu) - \chi^{\dagger}_{-\omega}(\nu)]/2ic, \tag{5.21}$$

which is ν/c times the first-harmonic component in the imaginary (skew-Hermitian) part of the time-dependent susceptibility. By making use of Eqs. (5.4b) and (5.3) in Eq. (5.21), one finds that for $E_j > E_1$, the harmonic amplitude $\beta_\omega(\nu)$ in the absorption of radiation during transitions from the pair of coupled states $|0\rangle$ and $|1\rangle$ to the state $|j\rangle$ is

$$\beta_{\omega(j)}(\nu) = (N\nu/2\hbar c)\mu_{0j}\mu_{j1}\{[(\bar{n}_1 - \bar{n}_j)\hat{u}'_{j0;j1}(-i(\nu-\bar{\omega}_{j1})) + \bar{\alpha}_{1j}\hat{u}'_{j0;j0}(-i(\nu-\bar{\omega}_{j1}))] + [1 \to 0]^{*}\}$$
$$(\omega_{j1} \gg \omega_{10}), \tag{5.22a}$$

while for $E_j < E_0$, the absorption is from $|j\rangle$ to $|0\rangle$ and $|1\rangle$, and the harmonic component is

$$\beta_{\omega(j)}(\nu) = (N\nu/2\hbar c)\mu_{j1}\mu_{0j}\{[(\bar{n}_j - \bar{n}_1)\hat{u}'_{j0;j1}(i(\nu-\bar{\omega}_{1j})) - \bar{\alpha}_{10}\hat{u}'_{j0;j0}(i(\nu-\bar{\omega}_{1j}))] + [1 \to 0]^{*}\}$$
$$(\omega_{1j} \gg \omega_{10}). \tag{5.22b}$$

The harmonically varying part of the absorption function, like the time-independent part, is a sharply peaked function near the resonance maxima, and shows the effect of the pump field through splittings comparable to the Rabi frequency Ω', defined by Eq. (3.7e). This detailed structure is, however, not seen in typical double-resonance or optical-pumping experiments, where the (optical) signal field has a large frequency bandwidth. The absorption rate for a signal field with constant spectral density I is $I\gamma(t)$, where $\gamma(t)=\int\beta(\nu,t)d\nu/2\pi$. By substituting Eq. (5.19) into this relation and making use of Eq. (2.14) one finds that for the two cases described by Eqs. (5.22a) and (5.22b), the infinite-bandwidth absorption coefficient is

$$\gamma(t)=(N\nu/2\hbar c)[\mu_{1j}\mu_{j1}(\bar{n}_1-\bar{n}_j)+\mu_{0j}\mu_{j0}(\bar{n}_0-\bar{n}_j)+\mu_{0j}\mu_{j1}\bar{\alpha}_{10}e^{-i\omega t}+\mu_{1j}\mu_{j0}\bar{\alpha}^*_{10}e^{i\omega t}] \quad (\omega_{j1}\gg\omega_{10}) \quad (5.23a)$$

and

$$\gamma(t)=(N\nu/2\hbar c)[\mu_{j1}\mu_{1j}(\bar{n}_j-\bar{n}_1)+\mu_{j0}\mu_{0j}(\bar{n}_j-\bar{n}_0)-\mu_{j1}\mu_{0j}\bar{\alpha}_{10}e^{-i\omega t}-\mu_{j0}\mu_{1j}\bar{\alpha}^*_{10}e^{i\omega t}] \quad (\omega_{1j}\gg\omega_{10}). \quad (5.23b)$$

The total absorption rate from an infinite-bandwidth signal field thus contains oscillating components only if the off-diagonal matrix element $\bar{\alpha}_{10}$ is nonvanishing. The absorption rate from a coherent signal field, by contrast, has appreciable oscillating components even in the limit of very intense pump fields, where $\bar{\alpha}_{10}\to 0$. This is in direct analogy to the case of emission, where the oscillating components in Eq. (4.21) for the *total* emission rate vanish for $\bar{\alpha}_{10}\to 0$, while the nonstationary functions given by Eqs. (4.19) and (4.20) remain appreciable in the same limit.

The effects of inhomogeneous broadening of the atomic-resonance frequencies ω_{j1} and ω_{j0}, it should be noted, are much the same as those of broadening of the signal-field frequency spectrum. In the limit of infinite inhomogeneous width, in particular, the oscillatory components in the absorption rate vanish unless $\bar{\alpha}_{10}\neq 0$. It is interesting here to consider the case in which the inhomogeneous width is finite, though large compared to Ω, while Ω is in turn large compared to the homogeneous widths and to $|\Delta\omega|$, thus implying $\bar{\alpha}_{10}\to 0$. One finds in this limit for the case described by Eq. (5.22a) that the harmonic amplitude in the absorption function is well approximated by the expression

$$\beta_{\omega(j)}(\nu)=-F_0\cdot\lambda_{10}\mu_{0j}\mu_{j1}(\nu/2\hbar c)\{\{(\bar{n}_1-\bar{n}_j)[\pi N_1'(\nu)-iP\int d\omega_{j1}N_1'(\omega_{j1})/(\omega_{j1}-\nu)]\}+\{1\to 0\}^*), \quad (5.24)$$

where $N_1(\omega_{j1})d\omega_{j1}$ is the number of atoms per unit volume with resonance frequencies between ω_{j1} and $\omega_{j1}+d\omega_{j1}$, $N_1'(\omega_{j1})\equiv dN_1(\omega_{j1})/d\omega_{j1}$, and P means principal value. The oscillatory absorption component is of order (Ω/inhomogeneous width) times the stationary component $\beta_0(\nu)$ for the same transition, and, as noted above, vanishes for $N(\nu)$ = constant. [A similar analysis can be carried out for the case of large though finite spectral width of the signal field, essentially by replacing the number density $N(\nu)$ by the spectral density $I(\nu)$.]

The effect of the terms proportional to $\chi_{\pm 2\omega}(\nu)$ in $\chi(\nu,t)$ is to induce the polarization components

$$P_2'(t)=\chi_{2\omega}(\nu)E_0'e^{-i(\nu+2\omega)t}+\text{c.c.}, \quad (5.25a)$$

$$P_{-2}'(t)=\chi^*_{-2\omega}(\nu)E_0'^*e^{-i(2\omega-\nu)t}+\text{c.c.} \quad (5.25b)$$

The function $\chi_{2\omega}(\nu)$ as given by Eq. (5.4c) is a comparatively small quantity for all values of $\nu>0$. The function $\chi_{-2\omega}(\nu)$, as given by Eqs. (5.3) and (5.4c), on the other hand, is sharply peaked near $\nu=\omega$. One finds, with the aid of Eqs. (3.15) and (3.6), the relation

$$\chi_{-2\omega}(\nu)=2iN\hbar^{-1}\mu_{10}\mu_{10}(\bar{n}_0-\bar{n}_1)(F_0\cdot\lambda_{10})^{*2}\times(-i\Delta\nu+2\kappa'_{10})/zf(-i\Delta\nu). \quad (5.26)$$

where $\Delta\nu\equiv\nu-\omega$ and $f(s)$ is defined by Eq. (3.16). For small values of $\Delta\nu$, i.e., for signal-field frequencies nearly equal to the pump-field frequency, the frequency $2\omega-\nu=\omega-\Delta\nu$ of the polarization component given by Eq. (5.25b) is nearly equal to both ν and ω. By evaluating directly the rate $E'\cdot\dot{P}'$ at which energy per unit volume is absorbed from the signal field, one finds, after adding the stationary contributions described previously,

$$W'(t)=2cE_0'^*\beta_0(\nu)E_0'+[i\omega E_0'\chi_{-2\omega}(\nu)E_0'e^{-2i\Delta\nu t}+\text{c.c.}], \quad (5.27)$$

where $\beta_0(\nu)$ is given by Eqs. (5.13) and (5.8). The absorption rate thus contains a slowly varying component which oscillates at the frequency $2\Delta\nu=2(\nu-\omega)$ and is comparable in magnitude to the stationary component. This is in fact the result of interference between the signal field and a field oscillating at the frequency $\omega-\Delta\nu$ which is generated by the polarization component given by Eq. (5.25b), and which becomes parametrically coupled

to the signal field when both fields travel in the same direction as the pump field.[25] The process in operation here is "light-by-light scattering," the associated spontaneous part of which is represented by the nonstationary terms in the emission process discussed at the end of Sec. IV.

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