Atomic decay in saturated resonant light scattering

B. R. Mollow
Department of Physics, University of Massachusetts, Boston, Massachusetts 02125
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The emission spectrum is found for transitions between a pair of atomic levels which are coupled by a coherent applied field, in the case in which decay to other levels of lower energy takes place during the emission process. The problem is solved by first finding the spectral distribution of radiation emitted by an atom which is initially prepared in some combination of the two coupled states, and which then decays to states of lower energy. (The solution found to this initial-value problem is valid even when many photons are emitted before the decay takes place.) It is then shown how the solution so obtained may be used to describe the case in which the atom is continually excited to the pair of levels in question by a weak, incoherent pumping mechanism. Limiting cases are discussed and compared with those which apply when no decay out of the two-dimensional subspace of coupled states takes place. The method of solution, which is based on an evaluation of the appropriate atomic correlation function by means of the quantum fluctuation-regression theorem, is compared to the method of simply adding imaginary terms, proportional to the decay constants, to the state energies. The latter method is of far more restricted validity, in that it depends on a particular strong inequality which must be assumed to hold between the decay constants.

The radiation emitted by an atom during transitions between a pair of (nondegenerate) levels which are coupled to one another throughout the emission process by a coherent near-resonant applied field has been the subject of numerous theoretical studies.[1-8] These have been motivated, in part, by a recognition of the importance of the driven spontaneous emission process in determining quantum noise in lasers, nonlinear amplifiers, "superradiant" processes, and, more generally, in any process in which the atoms are subject to the resonant action of a large mean field.[9,10] The recent advent of high-intensity tunable cw lasers has now raised the possibility that the essential features of the driven emission process may be clearly observable in resonant scattering experiments,[11] where the high laser intensity may in certain cases lead to line splittings large enough to remain clearly visible in the presence of Doppler broadening.

Accurate solutions for the emission spectrum in the process under discussion have so far been obtained in simple closed form, only for the case in which no decay out of the two-dimensional subspace of coupled states takes place.[12] The purpose of this paper is to treat the problem as proposed in one of its earliest formulations,[1] in which one or both levels are assumed to be excited states which decay to states of lower energy during the emission process. The analysis presented here represents a substantial improvement over that of Ref. 1, which was limited in its range of validity by a strong inequality which was assumed to hold between certain decay constants.[12a]

The density operator for an atom which is driven by an applied electric field $\vec{E}(t)$, while being governed by a general relaxation process, obeys the equations

$$(d/dt + i\omega_{jk} + \kappa'_{jk})\rho_{jk}(t) = i\hbar^{-1}\vec{E}(t)\cdot[\vec{\mu}, \rho(t)]_{jk} \quad (j \neq k), \tag{1a}$$

$$(d/dt + \kappa_j)n_j(t) - \sum_k n_k(t)\kappa_{kj} = i\hbar^{-1}\vec{E}(t)\cdot[\vec{\mu}, \rho(t)]_{jj}, \tag{1b}$$

where $\omega_{jk} \equiv (E_j - E_k)/\hbar$, $n_j \equiv \rho_{jj}$, $\vec{\mu}$ is the electric-dipole operator, and $\kappa_j = \sum_k \kappa_{jk}$. Of particular, though not exclusive interest is the case of purely radiative relaxation, for which κ_{jk} is the Einstein A coefficient for transitions from the state $|j\rangle$ to the state $|k\rangle$, and $\kappa'_{jk} = \frac{1}{2}(\kappa_j + \kappa_k)$. The electric field $\vec{E}(t) = \vec{E}_0 e^{-i\omega t} + \text{c.c.}$ is assumed to couple a single pair of states $|0\rangle$ (*not* necessarily the ground state) and $|1\rangle$, i.e., $\omega \approx \omega_{10}$.

It will be assumed throughout this paper that the decay process proceeds only to states of lower energy, and that no decay sequence of the form $|1\rangle \rightarrow |j\rangle \rightarrow |0\rangle$ takes place. Also, it is convenient to begin by assuming that the atom is initially prepared in some combination of the states $|0\rangle$ and $|1\rangle$, and thus to defer a discussion of the effect of the original process of excitation until the initial-value problem has been solved.

Under the stated conditions, the density-matrix elements referring to the pair of coupled states obey the equations, in the resonant approximation,

$$(d/dt - i\delta)\alpha'(t) = i\hbar^{-1}\vec{E}_0\cdot\vec{\mu}_{10}[n_0(t) - n_1(t)],$$
$$(d/dt + i\delta^*)\alpha'^*(t) = -i\hbar^{-1}\vec{E}_0^*\cdot\vec{\mu}_{10}^*[n_0(t) - n_1(t)],$$

$$(d/dt+\kappa_1)n_1(t) = -i\hbar^{-1}\vec{E}_0^*\cdot\vec{\mu}_{10}^*\,\alpha'(t) + i\hbar^{-1}\vec{E}_0\cdot\vec{\mu}_{10}\alpha'^*(t)$$
$$(d/dt+\kappa_0)n_0(t) - n_1(t)\kappa_{10} = i\hbar^{-1}\vec{E}_0^*\cdot\vec{\mu}_{10}^*\,\alpha'(t) - i\hbar^{-1}\vec{E}_0\cdot\vec{\mu}_{10}\alpha'^*(t)\,, \tag{2}$$

where $\rho_{10}(t)\equiv\alpha'(t)e^{-i\omega t}$, $\delta\equiv\Delta\omega+i\kappa'_{10}$, and $\Delta\omega\equiv\omega-\omega_{10}$.

The emission of photons during atomic transitions may be analyzed by introducing an appropriate expansion of the quantized electromagnetic field into oscillating modes, and then integrating the equations of motion for the Heisenberg photon-annihilation and -creation operators in the dipole approximation. One finds in this way that the mean number of photons radiated into a field mode of angular frequency ν_k and unit polarization vector $\hat{e}_k$ during transitions from $|1\rangle$ to $|0\rangle$, within the time interval T is

$$\bar{N}_k = A_k\int_0^T dt\int_0^t dt'\,e^{i\nu_k(t-t')}\langle a^\dagger(t')a(t)\rangle + \text{c.c.}, \tag{3}$$

where $\nu_k\approx\omega_{10}$, $A_k\equiv\nu_k|\vec{\mu}_{10}\cdot\hat{e}_k|^2/2\hbar V$, V is the quantization volume, and the lowering and raising operators $a\equiv|0\rangle\langle1|$ and $a^\dagger\equiv|1\rangle\langle0|$ on the right-hand side are evaluated in the Heisenberg picture. The quantity $\bar{N}_k$ becomes small compared to unity as $V\to\infty$, and in this limit represents the probability that a single photon has been radiated into the mode in question, irrespective of whether or not photons have been radiated into other modes. It should be emphasized here that although in the limit $V\to\infty$ the probability that more than one photon is radiated during transitions from $|1\rangle$ to $|0\rangle$ *into the same mode* is small enough to be neglected, the probability that more than one photon is radiated *at all* during transitions between the same pair of states need not be small. Indeed, for sufficiently intense driving fields, the mean total number of photons which are radiated before the atom decays to a state other than $|0\rangle$ or $|1\rangle$ will become quite appreciable if $\kappa_{10}\gg\kappa_0$, $\kappa_1-\kappa_{10}$, and will remain small compared to unity only if $\kappa_{10}\ll\kappa_1+\kappa_0$.

The atomic correlation function in Eq. (3) is easily evaluated by means of the quantum *fluctuation-regression theorem*.[13,14] One finds the relation[3]

$$\langle a^\dagger(t')a(t)\rangle = [n_1(t')\mathfrak{u}'_{\alpha\alpha}(t-t') + \alpha'^*(t')\mathfrak{u}'_{\alpha n_0}(t-t')]\times e^{-i\omega(t-t')}, \qquad t>t' \tag{4}$$

in which the functions $\mathfrak{u}'(t)$ are the time-dependent coefficients which appear in the general solution to Eqs. (2) for $\alpha'(t)$,

$$\alpha'(t) = \mathfrak{u}'_{\alpha\alpha}(t)\alpha'(0) + \mathfrak{u}'_{\alpha n_0}(t)n_0(0) + \mathfrak{u}'_{\alpha n_1}(t)n_1(0) + \mathfrak{u}'_{\alpha\alpha^*}(t)\alpha'^*(0). \tag{5}$$

By substituting Eq. (4) into Eq. (3) and taking the limit $T\to\infty$, one finds

$$\bar{N}_k = A_k\,2\,\mathrm{Re}\{\hat{n}_1(0)\hat{\mathfrak{u}}'_{\alpha\alpha}(\nu_k-\omega) + \hat{\alpha}'^*(0)\hat{\mathfrak{u}}'_{\alpha n_0}(\nu_k-\omega)\}, \tag{6}$$

in which the caret denotes the Laplace transform with imaginary argument

$$\hat{f}(\nu)\equiv\int_0^\infty dt\,e^{i\nu t}f(t) \tag{7}$$

of the time-dependent functions $n_1(t)$, $\alpha'^*(t)$, and $\mathfrak{u}'(t)$.

It follows directly from Eqs. (2) and (5) that the functions $\hat{\mathfrak{u}}'$ in Eq. (6) have the values

$$\hat{\mathfrak{u}}'_{\alpha\alpha}(\nu) = [i/F(\nu)]\{(\nu-\delta^*)(\nu+i\kappa_1)(\nu+i\kappa_0) - \tfrac{1}{2}\Omega^2(\nu+i\kappa_-)\},$$
$$\hat{\mathfrak{u}}'_{\alpha n_0}(\nu) = -i\hbar^{-1}\vec{E}_0\cdot\vec{\mu}_{10}(\nu-\delta^*)(\nu+i\kappa_1)/F(\nu)\,, \tag{8}$$

where

$$\kappa_- = \tfrac{1}{2}(\kappa_1-\kappa_{10}+\kappa_0)\,, \tag{9}$$

Ω is the power-broadening parameter $2|\vec{\mu}_{10}\cdot\vec{E}_0|/\hbar$, and $F(\nu)$ is the fourth-degree polynomial

$$F(\nu) = (\nu+\delta)(\nu-\delta^*)(\nu+i\kappa_1)(\nu+i\kappa_0) - \Omega^2(\nu+i\kappa_-)(\nu+i\kappa'_{10}). \tag{10}$$

The coefficients $\hat{n}_1(0)$ and $\hat{\alpha}'^*(0)$ in Eq. (6) depend on the initial density-matrix elements $n_0(0)\equiv N_0$ and $n_1(0)\equiv N_1$ (the initial off-diagonal elements are assumed to vanish) and may be found from the relations

$$\hat{\alpha}'(0) = [\hat{n}_1(0)-\hat{n}_0(0)]\,\vec{\mu}_{10}\cdot\vec{E}_0/\hbar\delta\,,$$
$$\kappa_1\hat{n}_1(0) - N_1 = -\kappa_0\hat{n}_0(0) + \hat{n}_1(0)\kappa_{10} + N_0 = [\hat{n}_0(0)-\hat{n}_1(0)]\,\tfrac{1}{2}\Omega^2\kappa'_{10}/|\delta|^2\,. \tag{11}$$

In many experimental situations in which decay processes are observed, the system is not initially prepared in an excited state at a single known instant of time, but rather is continually reexcited at random times after decays have taken place. Such an incoherent excitation process may be represented by introducing appropriate transition rates into Eqs. (1). Its effect is to produce nonzero *equilibrium* density matrix elements $\bar{n}_j$ and $\rho_{10}(t)=\rho_{01}^*(t)=\bar{\alpha}e^{-i\omega t}$ (where $\bar{n}_j$ and $\bar{\alpha}$ are constants) and thus an equilibrium value for the atomic cor-

relation function in Eq. (4) which depends only on the time difference $t-t'$. The expectation value in Eq. (3) then grows linearly with time for large times (rather than assuming a finite asymptotic value) and has the value $\bar{N}_k = A_k \tilde{g}(\nu_k)T$, where

$$\tilde{g}(\nu) = 2\,\mathrm{Re}\,[\bar{n}_1 \hat{\mathfrak{U}}'_{\alpha\alpha}(\nu-\omega) + \bar{\alpha}^* \hat{\mathfrak{U}}'_{\alpha n_0}(\nu-\omega)]\,. \tag{12}$$

One may draw a connection between the initial-value problem solved above and the stationary process presently under discussion simply by expressing the equilibrium excitation rates to the states $|0\rangle$ and $|1\rangle$ in the present case as

$$\sum_{j(j\neq 0,1)} \bar{n}_j \kappa_{j0} \equiv RN_0\,, \qquad \sum_{j(j\neq 0,1)} \bar{n}_j \kappa_{j1} \equiv RN_1\,, \tag{13}$$

where $N_0 + N_1 \equiv 1$ and R is the total excitation rate into the $|0\rangle - |1\rangle$ subspace. It is then a simple matter to show that the constant parameters in Eq. (12) are just

$$\bar{n}_1 = R\hat{n}_1(0), \quad \bar{\alpha}^* = R\hat{\alpha}'^*(0)\,, \tag{14}$$

where $\hat{n}_1(0)$ and $\hat{\alpha}'^*(0)$ are the quantities evaluated in Eqs. (11) in the absence of the excitation mechanism, i.e., for $R=0$.

The excitation mechanism which induces transitions to the states $|0\rangle$ and $|1\rangle$ from states of lower energy alters the general time dependence of the density operator, and hence alters the functions $\mathfrak{U}'(t)$ in Eq. (5). It is clear, however, that for sufficiently small excitation-transition rates κ_{j0} and κ_{j1} (roughly speaking, for sufficiently small R) the effect of the excitation mechanism on the functions $\mathfrak{U}'(t)$ may be neglected. This approximation requires that R be vanishingly small compared to the other relaxation rates, and is valid, in particular, only when the equilibrium excitation probability is small ($\bar{n}_0 + \bar{n}_1 \ll 1$). For sufficiently small R then, the functions $\hat{\mathfrak{U}}'$ in Eq. (12) are well approximated by the values found for them in Eqs. (8), so that Eqs. (12), (8), (11), and (14) represent a complete solution to the proposed problem whenever the equilibrium reexcitation rates are known. (The solution in question specifies $\bar{N}_k$ as RT times the asymptotic value found for it in the absence of reexcitation.)

That the assumption $R \to 0$ is really necessary for the validity of the approximation being made here perhaps becomes most readily apparent when one notes that the *coherent part* of the spectral density $\tilde{g}(\nu)$, which represents the elastic scattering of the incident light, is not present in Eqs. (12) and (8). This part has the value

$$\tilde{g}_{\mathrm{coh}}(\nu) = 2\pi |\bar{\alpha}|^2 \delta(\nu-\omega)\,, \tag{15}$$

and is thus proportional to R^2 (since $\bar{\alpha}$ is proportional to R). It is thus vanishingly small in integrated value for $R \to 0$, compared to the incoherent part as given by Eqs. (12) and (8), which is proportional to R. The failure of the coherent part of $\tilde{g}(\nu)$ to appear directly in Eqs. (12) and (8) is due to the omission of terms proportional to R in the functions $\mathfrak{U}'$, an omission which affects as well the validity of the expressions found for the *incoherent* part of $\tilde{g}(\nu)$, except in the limit $R \to 0$. [The problem with an appreciable reexcitation rate can of course still be solved by the general method based on the fluctuation-regression theorem, but only by solving the general Eqs. (1), which involve the occupation numbers of all states which are part of the decay sequence or reexcitation sequence. The answer cannot in general be expressed in terms of the equilibrium reexcitation rate R and the decay constants which refer to the states $|0\rangle$ and $|1\rangle$ alone. (One of the effects of appreciable R is a broadening of the spectral lines.)]

In the stated approximation, the limiting forms of the function $\tilde{g}(\nu)$ are readily found. For weak incident fields ($\Omega \to 0$) and for $|\Delta\omega| \equiv |\omega - \omega_{10}| \gg \kappa_{jk}, \kappa'_{jk}$, the function is sharply peaked at the three frequencies ω_{10}, ω, and $\omega + \Delta\omega$. It is well approximated by the relation[14a]

$$\tilde{g}(\nu) = \frac{2\bar{n}_1 \kappa'_{10}}{(\nu-\omega_{10})^2 + \kappa'^2_{10}} + \frac{\frac{1}{2}\Omega^2}{(\Delta\omega)^2}\left(\frac{B_0\kappa_0}{(\nu-\omega)^2+\kappa_0^2} + \frac{B_1\kappa_1}{(\nu-\omega)^2+\kappa_1^2}\right) + \frac{\frac{1}{8}\Omega^4}{(\Delta\omega)^4}\,\frac{\bar{n}_0\kappa'_{10}}{(\nu-\omega-\Delta\omega)^2+\kappa'^2_{10}}\,, \tag{16}$$

where

$$B_0 = \bar{n}_0 - \bar{n}_1\kappa_{10}/(\kappa_1-\kappa_0)\,, \qquad B_1 = \bar{n}_1[1+\kappa_{10}/(\kappa_1-\kappa_0)]\,. \tag{17}$$

The first term in Eq. (16) (which is given accurately only when $N_1 \neq 0$) represents the familiar spontaneous emission field at the resonant frequency ω_{10}.

The terms proportional to Ω^2 in Eq. (16) represent scattering and stimulated emission processes, due to the direct action of the incident field. It is instructive to consider the case $\bar{n}_1 \ll \bar{n}_0$, $B_1 \approx 0$, $B_0 \approx \bar{n}_0$, for which the terms in question reduce to a Lorentzian function of width κ_0 and integrated intensity $2\pi\bar{n}_0\frac{1}{4}\Omega^2/(\Delta\omega)^2$. If $|0\rangle$ were the ground

state of the atom, then by taking the limit $\kappa_0 \to 0$ one could recover the familiar result for the coherent elastic scattering intensity. Here the interpretation of the terms in question is somewhat different. The nonzero spectral width κ_0 of the emission field is due to the damping of the driven oscillating atomic current which is caused by the decay of the atom from the state $|0\rangle$ to states of lower energy. Though inelastic, the process would nevertheless still have a coherent character if all of the atoms were initially prepared in the state $|0\rangle$ and the transient emission process were observed in the absence of any reexcitation mechanism. When a weak reexcitation mechanism is present, however, the process becomes almost completely *incoherent* by the time the steady state is reached, simply because two different atoms are then unlikely to be excited to within the $|0\rangle$-$|1\rangle$ subspace at the same time. The coherent part of the scattering intensity is then given by Eq. (15), and, as noted above, is vanishingly small compared to the incoherent part in the limit $R \to 0$.

The term proportional to Ω^4 in Eq. (16) represents a parametric process in which two photons are absorbed and two emitted.[6,8,15] One of the emitted photons [the one represented in Eq. (16)] has frequency $\omega+\Delta\omega$, while the other, with frequency $\omega-\Delta\omega=\omega_{10}$, is masked by the much larger spontaneous emission field. Here again the fact that an atom is excited only a small fraction of the time implies that the process, which would have a coherent character if $|0\rangle$ were the ground state (with the two-photon amplitudes due to different atoms adding coherently in the forward direction) is largely incoherent in the case under discussion.

In the limit of intense incident fields, the spectral intensity is sharply peaked at the frequencies ω, $\omega+\Omega$, and $\omega-\Omega$, and is well approximated by the relation

$$\bar{g}(\nu)=\bar{n}_1\left(\frac{\kappa'_{10}}{(\nu-\omega)^2+\kappa'^2_{10}}+\frac{\frac{1}{2}\Gamma}{(\nu-\omega-\Omega)^2+\Gamma^2}+\frac{\frac{1}{2}\Gamma}{(\nu-\omega+\Omega)^2+\Gamma^2}\right), \tag{18}$$

in which

$$\Gamma=\tfrac{1}{2}\kappa'_{10}+\tfrac{1}{4}(\kappa_1+\kappa_0+\kappa_{10}) \tag{19a}$$

$$=\tfrac{1}{2}(\kappa_1+\kappa_0)+\tfrac{1}{4}\kappa_{10}\,, \tag{19b}$$

with Eq. (19b) holding in the case of purely radiative decay, where $\kappa'_{10}=\frac{1}{2}(\kappa_1+\kappa_0)$. The radiative decay of the atom out of the $|0\rangle$-$|1\rangle$ subspace thus broadens the emission lines equally at the sideband frequencies and at the central frequency ω, and leads to equal widths for all three lines in the limit $(\kappa_1+\kappa_0)/\kappa_{10}\to\infty$.

In the analysis of Rautian *et al.*,[1] closed solutions were obtained only for the limiting case $\kappa_{10}\ll\kappa_1+\kappa_0$, i.e., only for the case of vanishingly small probability that more than one photon is emitted before the atom decays to a state other than $|0\rangle$ or $|1\rangle$. When κ_{10} is eliminated from Eqs. (2), the resulting equations are the same as those obtained by simply introducing imaginary parts $-\frac{1}{2}i\kappa_0$ and $-\frac{1}{2}i\kappa_1$ into the energies of the states $|0\rangle$ and $|1\rangle$, respectively. It is not difficult to show that the general relation (4) for the atomic correlation function then reduces to the relation[1]

$$\langle a^\dagger(t')a(t)\rangle=\langle V^\dagger(t')a^\dagger V^{\dagger-1}(t')V^\dagger(t)aV(t)\rangle\,, \qquad t\geq t' \tag{20}$$

where $V(t)$ is the 2×2 time-development matrix for a pure state of the system with complex energies, under the influence of an applied field. The method adopted in this paper, by contrast to that of Ref. 1, depends on no assumption about the relative magnitudes of any of the decay constants, and thus retains its validity even when the method of adding imaginary terms to the state energies ceases to be applicable.[16]

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[11] In a recent experiment by F. Schuda, C. R. Stroud, Jr., and M. Hercher, J. Phys. B 7, L198 (1974), the sidebands in the saturated resonant emission spectrum were clearly resolved for a transition within the sodium D_2 line. A decay-modified scattering process similar to the one discussed in this paper may be observed by tuning the laser to the resonant frequency of the $F=2\to2$ transition within the sodium D_2 line, 60 MHz lower than the resonant frequency of the $F=2\to3$ transition investigated by Schuda *et al.* A three-peaked structure has in fact been observed recently

for laser frequencies near the $F=2 \rightarrow 2$ transition by S. Ezekiel, R. E. Grove, and F. Y. Wu (private communication).

[12]This restriction is unnecessary, however, in the strong-collision model of R. Karplus and J. Schwinger, Phys. Rev. 73, 1020 (1948). The general answer in that case may be found by putting $1-2\bar{n}_T \rightarrow \bar{m}_T - \bar{n}_T$ in Eqs. (3.12) and (2.15) of Ref. 4. The restriction in question is also unnecessary when the emission transition involves only *one* of the states coupled by the applied field.

[12a]*Note added in proof.* Since this manuscript was submitted, a similar analysis of the same problem (though apparently restricted to the case of radiative damping) has been made by A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 66, 1229 (1974) [Sov. Phys.—JETP 39, 601 (1974)]. Kazantsev's treatment of atomic relaxation is not clear, however, and his results certainly disagree with those found for the radiative case in this paper. In the strong-field limit, for example, Kazantsev's Eqs. (33) and (27) imply sideband widths twice as great as those of the undisplaced spectral line when there is no decay out of the two-dimensional subspace of coupled states. This is in contrast to the width ratio 3:2 implied by Eqs. (18) and (19b) of this paper in the same case ($\kappa_0 = 0$, $\kappa_1 = \kappa_{10}$).

[13]M. Lax, Phys. Rev. 172, 350 (1968), and references cited therein.

[14]In Ref. 8 it is rigorously shown that the validity of the fluctuation-regression theorem as applied to resonant light scattering is limited only by the assumption of the smallness of the saturated linewidth compared to the optical atomic resonance frequency.

[14a]*Note added in proof.* A nonzero initial occupation number N_1 (or pumping rate RN_1) is assumed in Eq. (16). If the atom is prepared in the lower level $|0\rangle$ (so that $N_1 = 0$), on the other hand, then one finds by returning to Eqs. (12), (8), and (10) that in the weak-field limit, the terms directly proportional to the incident field intensity in $\tilde{g}(\nu)$ have the value

$$2\,\mathrm{Re}[i\bar{n}_1(\nu+\delta)^{-1} - \tfrac{1}{2}i\Omega\,\bar{\alpha}^*(\nu+\delta)^{-1}(\nu+i\kappa_0)^{-1}],$$

independent of the ratio between detuning and damping parameters, and with $\bar{n}_1$ and $\bar{\alpha}$ obtainable from Eqs. (14) and (11). This result agrees exactly with the one obtained by A. Omont, E. W. Smith, and J. Cooper [Astrophys. J. 175, 185 (1972), Eq. (53)]. In the case of purely radiative relaxation, our result accordingly agrees exactly with Eq. (57) of the same reference, and hence equally with the scattering spectrum found originally by V. F. Weisskopf [Observatory 56, 291 (1933)]. As Omont *et al.* note, the correct expression under discussion here is rather more complicated in form than the (erroneous) one found by W. Heitler [*Quantum Theory of Radiation* (Clarendon, Oxford, 1954), p. 198].

[15]R. I. Sokolovskii, Zh. Eksp. Teor. Fiz. 59, 799 (1970) [Sov. Phys.—JETP 32, 438 (1971)].

[16]In Ref. 8 it is shown, however, that radiative atomic decay as determined by general decay constants *can* be described by the addition of imaginary terms to the state energies, provided that the radiation field modes are retained and an appropriate multiphoton calculation is carried out. The result found in this way for the atomic cross-spectral correlation function is shown to be identical to the one obtained by the fluctuation-regression theorem, which in turn is shown to be expandable in a series in which the right side of Eq. (20) is the leading term.