assumed random spatial orientation of the proton-hydrogen atom axis with respect to the projectile-beam direction. This new internuclear axis, rather than the projectile-beam axis, acts as the quantization direction for the resultant excited hydrogen atom. While largely conjectural, this hypothesis is substantiated by the measured Lyman-α-radiation polarization and also the measurement of Balmer-α-radiation polarization from this same reaction.[23] Measurements at 3- and 6-keV projectile energy yielded zero polarization for the Lyman-α radiation with rms deviation of the data consistent with that from the proton projectile reactions. We therefore conclude that no serious instrumental effect influences the polarizations reported here.

[23] D. H. Jaecks and F. J. de Heer (unpublished).

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Spontaneous Emission in the Presence of a Prescribed Classical Field*

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The effect of the application of a classical driving field on the spectrum of spontaneous emission from a simple physical system is studied. The physical system consists of an ensemble of two-level atoms interacting with a relaxation mechanism. The single Lorentzian line-shape characteristic of the power spectrum of spontaneous emission for the undriven case is split into components by the driving field. This splitting is associated with the establishment of definite phase relations between the corresponding components of the field spectrum.

INTRODUCTION

WE will investigate the spectral distribution of the spontaneous emission from a system of atoms subjected to a strong, near-resonant field. The material system is modeled by an ensemble of two-level atoms interacting with a relaxation mechanism.

The spectral distribution of the spontaneous emission will be unaffected by the driving field if the latter is not sufficiently large to significantly alter the state of the atomic system in a relaxation time.[1] If the driving field is sufficiently large we expect the spectral distribution to be changed from the undriven case. When the frequency associated with the interaction energy (of the atomic currents with the electromagnetic field) is small compared to the resonance frequency, useful calculational techniques are based on treating the interaction Hamiltonian as a perturbation of the full Hamiltonian. This technique is not valid for the present problem. Rautian and Sobelman[2] have considered the situation that the material system is coupled to many modes of the radiation field, but that initially only one mode of the field is in a high-energy eigenstate. They obtained a solvable finite set of equations for the atom field probability amplitudes by truncating the infinite set to correspond to a small number of multiphoton processes. It appears to us that the validity of this procedure is limited to relatively small initial field energies. Bergmann[3] has investigated the problem of spontaneous emission from a two-level system with incident beams of radiation which are initially either in a coherent state or in an n-photon state. His treatment attempts to avoid a perturbation-theory approach and uses approximations which retain only diagonal elements of the field time-development operator. In addition to these approximations his equations of motion are restricted to material two-level systems which cannot develop into mixed states. We feel it is necessary to consider the effect of mixed states, and indeed the main features of the spontaneously radiated power spectrum for large fields are simply related to the time development of these states.

In our work we take the large driving field to be classical and prescribed in its time dependence. The Hamiltonian now includes separate terms which describe the interactions of the atomic current with the prescribed classical field and with the small quantum-mechanical field which causes spontaneous emission. We compute the spectral distribution of the spontaneous emission to second order in the small interaction. The general solution gives the spectral power radiated in terms of the characteristic frequencies which describe the problem, and the atomic and relaxation-mechanism parameters. This solution has been used as a basis for determining the fundamental noise properties

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[1] I. R. Senitzky, Phys. Rev. **119**, 1807 (1960).
[2] S. G. Rautian and I. I. Sobelman, Zh. Eksperim. i Teor. Fiz. 41, 456 (1961) [English transl.: Soviet Phys.—JETP 14, 328 (1962)].
[3] S. M. Bergmann, J. Math. Phys. 8, 159 (1967).

of nonlinear quantum amplifiers of the type discussed by Senitzky *et al.*[4] The effects described in this paper have analogs in a dynamical theory of laser oscillators developed by Barone.[5] In both of these applications significant effects are associated not only with the alteration of the power spectrum from the undriven case, but also with the development of phase relations between parts of the current spectrum.

FORMULATION

The system to be considered consists of an ensemble of two-level atoms interacting with the quantum field, a prescribed classical field, and a relaxation mechanism. If the dimensions of the atom can be taken to be small compared to the scale of spatial variations of the electromagnetic field (dipole approximation), the Hamiltonian takes the form

$$H=H_a+H_f-\mathbf{j}\cdot\mathbf{A}/c-\mathbf{j}\cdot\mathbf{A}_c(t)/c-\mathbf{j}\cdot\mathbf{F}. \tag{1}$$

The terms H_a and H_f represent the free atom and field Hamiltonians. The term $\mathbf{j}\cdot\mathbf{A}/c$ represents the interaction between the material current operator $\mathbf{j}$ and the quantum field described by the vector potential operator $\mathbf{A}$. The term $\mathbf{j}\cdot\mathbf{A}_c(t)/c$ represents the similar form of interaction with the prescribed classical field represented by the vector-potential c number $\mathbf{A}_c(t)$. Despite the fact that the magnitude of this field is not required to be small, the quadratic term $A_c^2(t)$ (which would otherwise appear) may be dropped in the dipole approximation. In this approximation it merely adds a time-dependent term to the phase of the wave function. The last term $\mathbf{j}\cdot\mathbf{F}$ in the Hamiltonian represents the interaction between the material current operator and a relaxation mechanism coordinate $\mathbf{F}$. The atomic current operator $\mathbf{j}$ has the matrix elements

$$\mathbf{j}_{kl}=-\frac{ihe}{m}\int d\mathbf{r}\,\psi_k^*(\mathbf{r})\nabla\psi_l(\mathbf{r}) \tag{2}$$

in the representation in which the Hamiltonian of the free atom H_a is diagonal, i.e.,

$$H_a\psi_k=E_k\psi_k.$$

The Hamiltonian for the free quantum field is given by

$$H_f=\frac{1}{2}\int d\mathbf{r}[\mathbf{E}^2(\mathbf{r})+\mathbf{H}^2(\mathbf{r})], \tag{3}$$

where $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic field operators. In terms of the vector potential operator $\mathbf{A}$ we have the relations

$$\mathbf{H}=\nabla\times\mathbf{A},\quad \mathbf{E}=-\partial_t\mathbf{A}/c. \tag{4}$$

[4] B. Senitzky, G. Gould, and S. Cutler, Phys. Rev. **130**, 1460 (1963).

[5] S. R. Barone (to be published).

The rate of change of the field Hamiltonian can be found from the Heisenberg equations of motion:

$$(d/dt)H_f=(1/ih)[H_f,H].$$

Using the commutation relations for the field operators (Ref. 6), we obtain

$$(d/dt)H_f=-\mathbf{j}\cdot\mathbf{E}=-\mathbf{j}\cdot\partial_t\mathbf{A}/c. \tag{5}$$

This relation may, of course, also be obtained from Maxwell's equations. The average power radiated by an atom at the time t, when the interactions are turned on in the distant past, $t_0=-\infty$, is given by the quantum and thermodynamic ensemble expectation value:

$$P(t)=\langle t_0=-\infty\,|\dot{H}_f(t)\,|\,t_0=-\infty\,\rangle_\theta. \tag{6}$$

The subscript θ in Eq. (6) signifies the average over the thermodynamic ensemble representing the relaxation mechanism. This averaging will be chracterized more precisely later.

SECOND-ORDER PERTURBATION

We now exploit the fact that the interaction energy H_I of the atom and the quantum field,

$$H_I=-\mathbf{j}\cdot\mathbf{A}/c, \tag{7}$$

is small compared to the unperturbed Hamiltonian H_0:

$$H_0=H-H_I. \tag{8}$$

The Hamiltonian H_0 includes the terms describing the current interactions with the classical field and the loss mechanism.

The time development of the transformation functions under the full Hamiltonian can be formally related to quantities which develop under the unperturbed Hamiltonian H_0 by the expression[7]

$$\langle t\,|\,t_0\rangle_H=\langle t|\left(\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}H_I(t')dt'\right)\right)_+|\,t_0\rangle_{H_0}.$$

The subscript $+$ on the exponential means that the integrands in the formal expansion of the exponential are positively time ordered (operators with later time arguments appear to the left of those with earlier time arguments). The subscripts H and H_0 on the transformation functions mean that the time development of each function is determined by the corresponding Hamiltonian. To first order in the interaction energy we have

$$\langle t\,|\,t_0\rangle_H=\langle t\,|\,t_0\rangle_{H_0}+\frac{i}{ch}\int_{t_0}^{t}dt'\langle t|\mathbf{j}(t')\cdot\mathbf{A}(t')\,|\,t_0\rangle_{H_0},$$

[6] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954).

[7] J. Schwinger, Lecture Notes, Brandeis University Summer Institute in Theoretical Physics, Brandeis University, 1960 (unpublished).

and the complex-conjugate expression

$$\langle t_0|t\rangle_H=\langle t_0|t\rangle_{H_0}-\frac{i}{c\hbar}\int_{t_0}^{t}dt'\,\langle t_0|\mathbf{A}(t')\cdot\mathbf{j}(t')|t\rangle_{H_0}.$$

Under the Hamiltonian H_0, $\mathbf{A}(t)$ develops as a free quantum field, and $\mathbf{j}(t)$ develops as a current coupled to a classical field and a relaxation mechanism. These expansions may be inserted into expression (6) for the thermodynamic expectation value of H_f when the states $\langle t_0|$ in the latter expression are represented by a complete set of states $\langle t|$ at the time t. We thereby obtain for the power radiated at time t, $P(t)$, to second order,

$$P(t)=\lim_{t_0\to-\infty}\frac{2}{c^2}\,\mathrm{Re}\int_{t_0}^{t}dt'\left(\frac{-i}{\hbar}\right)\langle t_0|\dot{A}_i(t)A_j(t')|t_0\rangle_{H_0}\times\langle t_0|j_i(t)j_j(t')|t_0\rangle_{H_0,\theta},\quad(9)$$

where the subscripts (i,j) label the spatial vector components, and the convention is used whereby a summation over repeated indices is understood. We have assumed that the initial state of the uncoupled field is the vacuum, hence $\langle t_0|A|t_0\rangle_{H_0}=0$.

ELECTROMAGNETIC STRUCTURE

The role of the electromagnetic structure is made explicit by recognizing that the vacuum expectation value of time-ordered field operators can be identified with the Green's function dyadic, namely,

$$\frac{-i}{\hbar}\langle\mathrm{vac},t_0|\Big(A_i(\mathbf{r},t)A_j(\mathbf{r}',t')\Big)_+|\mathrm{vac},t_0\rangle=G_{ij}(\mathbf{r},t;\mathbf{r}',t'),\quad(10)$$

where $\mathbf{G}$ satisfies the differential equation

$$\left(\nabla^2-\frac{1}{c^2}\partial_t^2\right)\mathbf{G}(\mathbf{r},t;\mathbf{r}',t')=\boldsymbol{\delta}_t(\mathbf{r},\mathbf{r}')\delta(t-t'),\quad(11)$$

with $\boldsymbol{\delta}_t$ representing the transverse delta-function dyadic. This identification may be verified by substituting the expression (10) for G in Eq. (11) and using the equal time commutation relations:

$$i[\dot{A}_i(\mathbf{r},t),A_j(\mathbf{r}',t)]=\hbar c^2(\boldsymbol{\delta}_t(\mathbf{r},\mathbf{r}'))_{ij}.$$

The spatial boundary conditions on the Green's function are the same as those on the vector potential. The temporal boundary conditions are determined, according to Eq. (10), by the state of the uncoupled field at time t_0; in this case, the vacuum. This implies that $G(t,t')$ is causal, that is, the spectral distribution consists of only positive frequencies for $t>t'$, and of only negative frequencies for $t<t'$, i.e.,

$$\mathbf{G}(\mathbf{r},t;\mathbf{r}',t')=i\int_0^\infty d\omega\,\mathbf{B}(\mathbf{r},\mathbf{r}';\omega)e^{-i\omega|t-t'|},\quad(12)$$

where the spectral density $\mathbf{B}$ is a real dyadic which characterizes the Green's function. The spectral density may be related to the modal properties of the electromagnetic structure. Thus, if we label the proper modes of the structure by the resonant frequency ω and the degeneracy parameter α, viz., $\mathbf{A}(\omega,\alpha,\mathbf{r})$, we have, by the requirements of completeness and orthonormality,

$$\int_0^\infty d\omega\int d\alpha\,\rho_\alpha(\omega)\mathbf{A}(\omega,\alpha,\mathbf{r})\mathbf{A}(\omega,\alpha,\mathbf{r}')=\boldsymbol{\delta}_t(\mathbf{r}-\mathbf{r}'),\quad(13)$$

where ρ is the mode density. The expansion of the Green's function in terms of these proper modes leads to the relation between the spectral density and the mode density:

$$B_{ij}(\mathbf{r},\mathbf{r}';\omega)=\frac{-c^2}{2\omega}\int d\alpha\,\rho_\alpha(\omega)A_i(\omega,\alpha,\mathbf{r})A_j(\omega,\alpha,r').\quad(14)$$

In terms of the electromagnetic Green's function the power radiated is given to second order by

$$P(t)=\lim_{t_0\to-\infty}\frac{2}{c^2}\,\mathrm{Re}\int_{t_0}^{t}dt'\dot{G}_{ij}(\mathbf{r},t;\mathbf{r},t')\times\langle t_0|j_i(t)j_j(t')|t_0\rangle_{H_0,\theta},\quad(15)$$

where $\mathbf{r}$ is the location of the radiating atom. Using the spectral representation of the Green's function, we obtain the spectral resolution of the power,

$$P(\omega,t)=\frac{2\omega}{c^2}B_{ij}(\mathbf{r}=\mathbf{r}',\omega)\,\mathrm{Re}\lim_{t_0\to-\infty}\int_{t_0}^{t}dt'\,e^{-i\omega(t-t')}\times\langle t_0|j_i(t)j_j(t')|t_0\rangle_{H_0,\theta},\quad(16)$$

where

$$P(t)=\int_0^\infty d\omega\,P(\omega,t).\quad(17)$$

DRIVEN MATERIAL SYSTEM

We will now be more specific regarding the nature of the driven material system and discuss a particular situation which is of interest for the determination of the noise properties of a nonlinear quantum amplifier.[4] Here a gas of molecules, subjected to relaxing collisions, is contained in an externally coupled cavity. The cavity is excited by an applied field whose spatial distribution is determined by a monochromatic excitation of the cavity near a resonance. Two levels of the molecule are separated by a near-resonant energy spacing and are connected by a $\Delta m=0$ electric-dipole transition. Within the frequency range of interest the spatial behavior of the spectral density $\mathbf{B}$ can be taken to be the same as the excitation field. The classical field is monochromatic and plane polarized at the location of each molecule:

$$\mathbf{A}_c(t)=\boldsymbol{\varepsilon}A_0\cos\omega_0 t.\quad(18)$$

Only the current component parallel to the local

polarization axis radiates in the $\Delta m=0$ transition. If we understand by the scalar operator j, the component of the current vector operator parallel to the local polarization axis, we have

$$P(\omega,t)=\frac{2\omega}{c^2}\boldsymbol{\varepsilon}\cdot\mathbf{B}\cdot\boldsymbol{\varepsilon}\,\mathrm{Re}\lim_{t_0\to-\infty}\int_{-\infty}^{t}dt'e^{-i\omega(t-t')}\times\langle t_0|\,j(t)j(t')\,|t_0\rangle_{H_0,\theta}. \quad (19)$$

For the purpose of this calculation, the important property of the relaxation mechanism is as follows: In the absence of the coupling to the fields it causes the population of the energy levels of the uncoupled material system to relax, in the time τ, to a thermal distribution corresponding to a temperature T. We model this mechanism as follows: Each molecule suffers collisions of instantaneous duration, the times between collisions being distributed randomly according to a Poisson distribution with the mean τ. The effect of a collision is to throw the molecule into a superposition of energy eigenstates such that an ensemble average leads to a thermal mixture. This is accomplished by the unitary time-development (under H_0, including $\mathbf{j}\cdot\mathbf{F}$) operator $U_F(t,t_0)$ which acts on the initial state of the system $|\alpha,t_0\rangle$, to give

$$U_F(t,t_0)\,|\alpha,t_0\rangle=U(t,t_c)\sum_k(\sqrt{P_k})e^{i\phi_k(\alpha,t_0,t_c)}\,|E_k,t_c\rangle,$$

where t_c is the time of the last collision previous to t, P_k is the probability distribution corresponding to the temperature T,

$$P_k=e^{-E_k/kT},$$

and $U(t,t_c)$ is the time-development operator in the absence of coupling to the relaxation mechanism. The phase $\phi_k(\alpha,t_0,t_c)$ is a function of the initial state of the system, and the time of the last collision. The following ensemble-average relation holds:

$$\langle e^{i\phi_k(\alpha,t_0,t_c)}e^{-i\phi_{k'}(\alpha',t_0',t_c')}\rangle=\delta_{k,k'}\delta_{\alpha,\alpha'}\delta_{t_0,t_0'}\delta_{t_c,t_c'}.$$

Under these conditions we have, for the thermal expectation value of the current correlation function,

$$\langle t_0|\,j(t)j(t')\,|t_0\rangle_{H_0,\theta}=\int_{-\infty}^{t'}\frac{dt_c}{\tau}e^{-(t-t_c)/\tau}\times\langle t_c|\,j(t)j(t')\,|t_c\rangle_{\theta,F=0}, \quad (20)$$

where the ensemble average θ leads to a thermal mixture at the time t_c. The expression for the spectral power becomes

$$P(\omega,t)=\frac{2\omega}{c^2}\boldsymbol{\varepsilon}\cdot B\cdot\boldsymbol{\varepsilon}\,\mathrm{Re}\int_{-\infty}^{t}dt'\,e^{-[\tau^{-1}+i\omega](t-t')}\times\int_{-\infty}^{t'}\frac{dt_c}{\tau}\frac{e^{-(t'-t_c)/\tau}}{\tau}\langle t_c|\,j(t)j(t')\,|t_c\rangle_{\theta,F=0}, \quad (21)$$

where we have explicitly accounted for the relaxation mechanism. The remaining implicit time development is for a two-level system coupled only to a classical field.

We are now required to evaluate the expectation value of the current correlation function which develops dynamically under the Hamiltonian

$$H_0(F=0)=H_a-\mathbf{j}\cdot\mathbf{A}_c(t)/c. \quad (22)$$

In the representation in which the Hamiltonian H_a of the two-level system is diagonal, we have

$$\langle t|\,H_a(t)\,|t\rangle=\tfrac{1}{2}\hbar\omega_a\begin{pmatrix}+1 & 0\\ 0 & -1\end{pmatrix}, \quad (23)$$

$$\langle t|\,j(t)\,|t\rangle=|\,j_{12}|\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}, \quad (24)$$

where ω_a is the resonant frequency of the two-level system, and j_{12} is the matrix element of the current element given in Eq. (2).

The expressions (23) and (24) follow directly from our description of the two-level system when we choose the phase of the wave functions such that the matrix element j_{12} is real. It is convenient to express the pertinent dynamical variables in terms of the Pauli spin operators σ_x, σ_y, and σ_z which satisfy the commutation relations

$$\boldsymbol{\sigma}\times\boldsymbol{\sigma}=2i\boldsymbol{\sigma}. \quad (25)$$

In terms of the spin vector $\boldsymbol{\sigma}$, the Hamiltonian $H_0(F=0)$ may be written

$$H_0(F=0)=\frac{\hbar\omega_a}{2}\sigma_z-\frac{|\,j_{12}|A(t)}{c}\sigma_x. \quad (26)$$

The equation of motion is

$$i\hbar d\boldsymbol{\sigma}/dt=[\boldsymbol{\sigma},H_0(F=0)], \quad (27)$$

which has the formal solution

$$\sigma_i(t)=U_{ij}(t,t')\sigma_j(t'). \quad (28)$$

Using the commutation relations (25) we obtain for the expectation value of the current correlation function

$$\langle t_c|\,j(t)j(t')\,|t_c\rangle_{\theta,F=0}=|\,j|^2\{U_{xx}(t,t')+i[U_{xz}(t,t')U_{yz}(t',t_c)-U_{xy}(t,t')U_{zz}(t',t_c)]\times\langle t_c|\,\sigma_z(t_c)\,|t_c\rangle_\theta\}. \quad (29)$$

The thermal expectation value $\langle t_c|\,\sigma_z(t_c)\,|t_c\rangle_\theta$ is $(\frac{1}{2}\hbar\omega_a)^{-1}$ times the average energy of the two-level system immediately after a collision; thus

$$\langle t_c|\,\sigma_z(t_c)\,|t_c\rangle_{|\theta|}=\tanh\tfrac{1}{2}\frac{\hbar\omega_a}{kT}. \quad (30)$$

The frequency K, given by

$$K=|\,j_{12}|A_0/\hbar c, \quad (31)$$

measures the additional broadening, due to saturation by the applied field, $\varepsilon A_0 \cos\omega_0 t$, of the response of the material system to this field. In Appendix A we evaluate the elements of the time-development matrix $U(t,t')$ to first order in the small quantities K/ω_a and $(\omega-\omega_0)/\omega$. The results of this evaluation are, for the pertinent elements of the matrix U,

$$\begin{aligned}
U_{xx}(t,t') &= \tfrac{1}{2}A_{++}(t-t')e^{i\omega_0(t-t')} \\
&\quad +\tfrac{1}{2}A_{+-}(t-t')e^{i\omega_0(t+t')}+\text{c.c.}, \\
U_{xy}(t,t') &= \tfrac{1}{2}iA_{++}(t,t')e^{i\omega_0(t,t')} \\
&\quad -\tfrac{1}{2}iA_{+-}(t-t')e^{i\omega_0(t+t')}+\text{c.c.}, \\
U_{xz}(t,t') &= A_{+z}(t-t')e^{i\omega_0 t}+\text{c.c.}, \\
U_{yz}(t',t_c) &= -iA_{+z}(t'-t_c)e^{i\omega_0 t'}+\text{c.c.}, \\
U_{zz}(t',t_c) &= A_{zz}(t'-t_c),
\end{aligned} \tag{32}$$

where the A coefficients are given as inverse Laplace transforms:

$$A(t-t')=\frac{1}{2\pi i}\int_c dp\, a(p)e^{p(t-t')}, \tag{33}$$

and the pertinent a coefficients are given by

$$\begin{aligned}
a_{++} &= (p^2+i\Delta p+\tfrac{1}{2}K^2)/D, \\
a_{+-} &= (\tfrac{1}{2}K^2)/D, \\
a_{\pm z} &= (\pm\tfrac{1}{2}iK)(p\pm i\Delta)/D, \\
a_{zz} &= (p^2+\Delta^2)/D,
\end{aligned} \tag{34}$$

with

$$D=p(p^2+K^2+\Delta^2) \tag{35}$$

and

$$\Delta=(\omega_a-\omega_0). \tag{36}$$

It will be noted that the time dependence of the A coefficients in Eq. (32) is slow compared to that of the exponentials in the same expression. In deriving this form we have taken $\omega_0>0$.

POWER SPECTRUM

From Eqs. (21) and (29) we have, as the expression for the spectral power distribution,

$$\begin{aligned}
P(\omega,t) &= \frac{2\omega}{c^2}|j_{12}|^2\boldsymbol{\varepsilon}\cdot\mathbf{B}\cdot\boldsymbol{\varepsilon}\,\mathrm{Re}\int_{-\infty}^{t}dt'e^{-(\tau^{-1}+i\omega)(t-t')} \\
&\quad\times\int_{-\infty}^{t'}\frac{dt_c}{\tau}e^{-(t'-t_c)/\tau}\{U_{xx}(t,t')+i\langle t_c|\sigma_z(t_c)|t_c\rangle_\theta \\
&\quad\times[U_{xz}(t,t')U_{yx}(t',t_c)-U_{xy}(t,t')U_{zz}(t',t_c)]\}.
\end{aligned} \tag{37}$$

From the form of the elements of the matrix U given by Eq. (32), we observe that $P(\omega,t)$ is of the form

$$P(\omega,t)=\mathrm{Re}[P_0(\omega)+P_1(\omega)e^{2i\omega_0 t}]. \tag{38}$$

The constant part, $\mathrm{Re}P_0(\omega)$, is the measurable power spectrum. The time-dependent part gives information on phase relations between components of the field

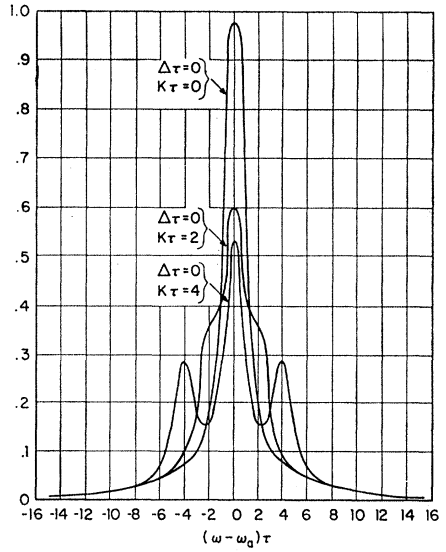

FIG. 1. Spectral distribution of spontaneous emission applied field on resonance.

spectrum. The resonant contributions to $P_0(\omega)$ come from those terms in the current correlation function which have the factor $e^{i\omega_0(t-t')}$ in their time dependence. Substituting these terms from Eq. (32) into (37) we obtain (see Appendix B)

$$\begin{aligned}
P_0(\omega) &= \frac{2\omega}{c^2}|j|^2\boldsymbol{\varepsilon}\cdot\mathbf{B}\cdot\boldsymbol{\varepsilon}\,\mathrm{Re}\{\tfrac{1}{2}a_{++}(p_1)(1+\tfrac{1}{2}\langle\sigma\rangle a_{zz}(p_2)) \\
&\quad -\langle\sigma\rangle a_{+z}(p_1)a_{-z}(p_2)\},
\end{aligned} \tag{39}$$

where the a coefficients are given in Eq. (34) and

$$p_1=[\tau^{-1}+i(\omega-\omega_0)], \tag{40}$$

$$p_2=\tau^{-1}. \tag{41}$$

The expectation value $\langle\sigma\rangle$ is

$$\langle\sigma\rangle\equiv\langle t_c|\sigma_z(t_c)|t_c\rangle_\theta=\tanh\tfrac{1}{2}\frac{\hbar\omega_a}{kT},$$

by Eq. (34).

A particularly simple special case occurs when the relaxation process tends to equalize the populations of the two levels, viz., $\langle\sigma\rangle=0$, and the applied field is tuned to the central atomic frequency, $\omega_a=\omega_0$. For this case we have

$$P_0(\omega)=\frac{\omega}{c^2}|j|^2\boldsymbol{\varepsilon}\cdot\mathbf{B}\cdot\boldsymbol{\varepsilon}\,\mathrm{Re}a_{++}, \tag{42}$$

where

$$\begin{aligned}
a_{++} &= \frac{\tfrac{1}{2}}{\tau^{-1}+i(\omega-\omega_0)}+\frac{\tfrac{1}{4}}{\tau^{-1}+i[(\omega-\omega_0)+K]} \\
&\quad +\frac{\tfrac{1}{4}}{\tau^{-1}+i[(\omega-\omega_0)-K]}.
\end{aligned} \tag{43}$$

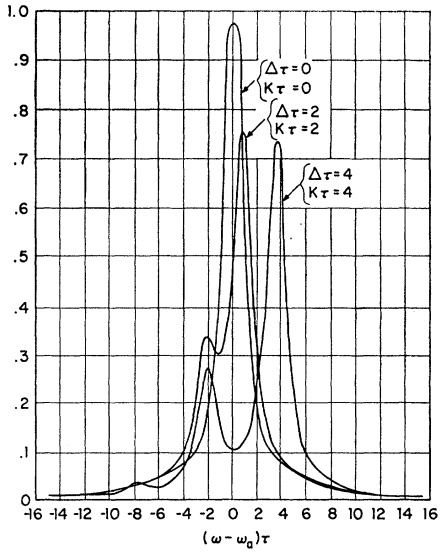

FIG. 2. Spectral distribution of spontaneous emission applied field off resonance.

Thus, the effect of the large classical field is to split the single Lorentzian line-shape characteristic of ordinary spontaneous emission into a structure consisting of three Lorentzian components. One component is centered at the central atomic frequency and the other two, of half the amplitude, are displaced symmetrically on either side by the amount of the saturation parameter K. Each Lorentzian has the width τ^{-1} characteristic of spontaneous emission of the undriven system. This situation is illustrated in Fig. 1 where the spectral power is given for several values of the dimensionless saturation parameter $K\tau$ as a function of the dimensionless frequency $(\omega-\omega_a)\tau$, where ω is the frequency of the radiated photon and ω_a is the central atomic frequency. Since the driving frequency ω_0 is tuned to the atomic resonance, we have $\Delta=(\omega_a-\omega_0)=0$.

More complicated spectral distributions occur when the driving field is not tuned to the atomic resonance. These are illustrated in Fig. 2, for several values of the dimensionless parameters $\Delta\tau$ and $K\tau$.

CURRENT PHASE RELATIONS

The existence of a double frequency term in the expression (38) for the power spectrum is indicative of phase relations between spectral components of the spontaneously radiating current. This term arises from the fact that due to the existence of the driving field, the spontaneous current fluctuations $\langle j(t)j(t')\rangle$ are not merely a function of the difference $t-t'$, but rather of the individual times t and t'. The fact that the quantum-mechanical ensemble average does not factor is essential to the proper description of spontaneous emission. Indeed, the expectation value of $j(t)$, which is the current driven by the classical field, $A_0\cos\omega_0 t$, is monochromatic according to

$$\langle j(t)\rangle=\int_{-\infty}^{t}\frac{dt_c}{\tau}e^{-(t-t_c)/\tau}\langle t_c|\,j(t)\,|t_c\rangle$$
$$=|j|\langle\sigma\rangle\int_{-\infty}^{t}\frac{dt_c}{\tau}e^{-(t-t_c)/\tau}U_{xz}(t,t_c)$$
$$=\frac{|j|\langle\sigma\rangle}{\tau^{-1}(\tau^{-2}+K^2+\Delta^2)}(\Delta K\cos\omega_0 t+\tau^{-1}K\sin\omega_0 t). \tag{44}$$

Nevertheless, in order to obtain a simple picture of the phase relations, it is convenient to introduce a fictitious classical stochastic current $j_c(t)$ whose stochastic-ensemble-averaged correlation function equals the quantum-mechanical-ensemble-averaged correlation function:

$$\langle j_c(t)j_c(t')\rangle_s\equiv\langle j(t)j(t')\rangle. \tag{45}$$

Useful pictorial information can be obtained from the derived form of $j_c(t)$. The fictitious current has a spectrum centered near ω_0 and hence may be written

$$j_c(t)=j_a(t)\sin\omega_0 t+j_f(t)\cos\omega_0 t, \tag{46}$$

where j_a and j_f are slowly varying stochastic functions of t. For the case $\Delta=0$, from Eq. (44), $j_a(t)\sin\omega_0 t$ represents a current component amplitude modulated relative to the driven current $\langle j(t)\rangle$, and $j_f(t)\cos\omega_0 t$ represents a frequency-modulated current component. For the previously discussed particular case, $\langle\sigma\rangle=0$, the right-hand side of Eq. (45) is

$$\langle j(t)j(t')\rangle=|j_{12}|^2U_{xx}(t-t')e^{-(t-t')/\tau}$$
$$=\tfrac{1}{2}|j|^2[(1+\cos K(t-t'))\cos\omega_0(t-t')$$
$$+(1-\cos K(t-t'))\cos\omega_0(t+t')]e^{-(t-t')/\tau}. \tag{47}$$

Substituting Eq. (46) into the left-hand side of Eq. (45), we obtain the identities

$$\langle j_a(t)j_a(t')\rangle_s=\cos K(t-t')e^{-(t-t')/\tau},$$
$$\langle j_f(t)j_f(t')\rangle_s=e^{-(t-t')/\tau}, \tag{48}$$
$$\langle j_a(t)j_f(t')\rangle_s=\langle j_f(t)j_a(t')\rangle_s=0.$$

We may now identify the three components of the spectral power distribution given in Eqs. (42) and (43) when we consider it to have been generated by the equivalent classical stochastic current. The central component of the power spectrum is generated by the current component phased for frequency modulation relative to the driven current, and the two side bands are generated by the current component phased for amplitude modulation. The equivalent stochastic classical fields generated by the currents obey the corresponding phase relations relative to the driving field.

DISCUSSION

We have studied how the spectrum of spontaneous emission from a simple physical system is affected by the application of a classical driving field. The single Lorentzian line shape, characteristic of emission from a two-level system coupled to a relaxation mechanism, is split into components by the driving field. Furthermore, the splitting of the components of the power spectrum is associated with the establishment of definite phase relations between the corresponding components of the field spectrum. These effects become significant when the strength of the driving field is sufficient to appreciably alter the state of the material system in a relaxation time. The splitting of the power spectrum can be associated with the sinusoidal modulation of the population of the upper level of the material system, between relaxation collisions, due to the coupling to the driving field. The establishment of phase relations between the components of the field spectrum can be associated with response characteristics of the driven material system. Senitzky *et al.*[4] have shown that the linear response to an additional small signal depends on the phase of the small signal relative to the driving field. The susceptibility of the medium to a small signal phased for frequency modulation relative to a resonant driving field has a Lorentzian line shape centered at the central atomic frequency. The susceptibility to a small signal phased for amplitude modulation consists of the sum of two Lorentzians, symmetrically displaced relative to the central frequency. These three peaks in the susceptibility and the associated phase relations correspond to the same features of the spontaneous emission spectrum.

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APPENDIX A: TIME DEVELOPMENT MATRIX FOR TWO-LEVEL SYSTEM IN A CLASSICAL FIELD

Starting with the equations of motion (26) and the specified field $A(t)=A_0\cos\omega_0 t$, we have

$$\begin{aligned} d\sigma_x/dt &= -\omega_a\sigma_y\,,\\ d\sigma_y/dt &= \omega_a\sigma_x+2K\sigma_z\cos\omega_0 t\,,\\ d\sigma_z/dt &= -2K\sigma_y\cos\omega_0 t\,, \end{aligned} \tag{A1}$$

where

$$K=|j|A_0/\hbar c.$$

It is convenient to transform to a rotating coordinated system by the transformations

$$\begin{aligned} \sigma_+ &= \tfrac{1}{2}(\sigma_x+i\sigma_y)e^{-i\omega_0 t}\,,\\ \sigma_- &= \tfrac{1}{2}(\sigma_x-i\sigma_y)e^{+i\omega_0 t}. \end{aligned} \tag{A2}$$

The quantities σ_+, σ_-, and σ_z are slowly varying compared to the time dependence associated with the frequency ω_0. The transformed spin components obey the equations

$$\begin{aligned} &(d/dt-i\Delta)\sigma_+ = i(\tfrac{1}{2}K)\sigma_z(1+e^{-2i\omega_0 t})\,,\\ &(d/dt+i\Delta)\sigma_- = -i(\tfrac{1}{2}K)\sigma_z(1+e^{+2i\omega_0 t})\,,\\ &(d/dt\sigma_z) = iK(\sigma_+-\sigma_-)+iK(\sigma_+e^{2i\omega_0 t}-\sigma_-e^{-2i\omega_0 t})\,, \end{aligned} \tag{A3}$$

where the frequency difference Δ is given by

$$\Delta=(\omega_a-\omega_0)\,.$$

We may obtain σ correctly to order (K/ω_0) by neglecting the terms involving $e^{\pm 2i\omega_0 t}$ in Eqs. (A3). Dropping the double-frequency terms, we get for the equations of motion

$$\begin{aligned} &(d/dt-i\Delta)\sigma_+ = i(\tfrac{1}{2}K)\sigma_z\,,\\ &(d/dt+i\Delta)\sigma_- = -i(\tfrac{1}{2}K)\sigma_z\,,\\ &(d/dt)\sigma_z = iK(\sigma_+-\sigma_-) \quad \text{for } \omega_0>0. \end{aligned} \tag{A4}$$

We introduce the Laplace transforms

$$\sigma(p)=\int_{t'}^{\infty} dt\, e^{-pt}\sigma(t)\,, \tag{A5}$$

with $\sigma(t)$ given by

$$\sigma(t)=\frac{1}{2\pi i}\int_c e^{pt}\sigma(p)dp\,, \tag{A6}$$

where the contour c is to the right of the poles of $\sigma(p)$ in the complex p plane. We may solve the resultant linear equations for $\sigma(p)$ to obtain

$$\begin{aligned} \sigma_+(p) &= \frac{e^{-pt'}}{p(p^2+K^2+\Delta^2}\{(p^2+i\Delta p+\tfrac{1}{2}K^2)\sigma_+(t')\\ &\quad +\tfrac{1}{2}K^2 2\sigma_-(t')+\tfrac{1}{2}iK(p+i\Delta)\sigma_z(t')\}\,,\\ \sigma_-(p) &= \sigma_+^*(p^*)\,,\\ \sigma_z(p) &= \frac{e^{-pt'}}{p(p^2+K^2+\Delta^2)}\{iK(p+i\Delta)\sigma_+(t')\\ &\quad -iK(p-i\Delta)\sigma_-(t')+(p^2+\Delta^2)\sigma_z(t')\}\,. \end{aligned} \tag{A7}$$

Transforming back to the laboratory frame, we have

$$\begin{aligned} \sigma_x(t) &= \frac{1}{2\pi i}\int dp[\sigma_+(p)e^{(p+i\omega_0)t}+\sigma_-(p)e^{(p-i\omega_0)t}]\,,\\ \sigma_y(t) &= \frac{1}{2\pi i}\int dp[-i\sigma_+(p)e^{(p+i\omega_0)t}+i\sigma_-(p)e^{(p-i\omega_0)t}]\,,\\ \sigma_z(t) &= \frac{1}{2\pi i}\int dp\, e^{pt}\sigma_z(p)\,. \end{aligned} \tag{A8}$$

We relate $\sigma(p)$ to $\sigma(t')$ by Eqs. (A5) and (A6) to obtain

$$\sigma_i(t)=\sum_i U_{ij}(t,t')\sigma_j(t'), \quad i=x,y,z, \quad j=x,y,z \tag{A9}$$

where

$$\begin{aligned}
&U_{xx}(t,t')=\tfrac{1}{2}A_{++}e^{i\omega_0(t-t')}+\tfrac{1}{2}A_{+-}e^{i\omega_0(t+t')}+\text{c.c.},\\
&U_{xy}(t,t')=\tfrac{1}{2}iA_{++}e^{i\omega_0(t-t')}-\tfrac{1}{2}iA_{+-}e^{i\omega_0(t+t')}+\text{c.c.},\\
&U_{xz}(t,t')=A_{+z}e^{i\omega_0 t}+\text{c.c.},\\
&U_{yy}(t,t')=\tfrac{1}{2}A_{++}e^{i\omega_0(t-t')}-\tfrac{1}{2}A_{+-}e^{i\omega_0(t+t')}+\text{c.c.},\\
&U_{yx}(t,t')=-\tfrac{1}{2}iA_{++}e^{i\omega_0(t-t')}-\tfrac{1}{2}iA_{+-}e^{i\omega_0(t+t')}+\text{c.c.},\\
&U_{yz}(t,t')=(-i)A_{+z}e^{i\omega_0 t}+\text{c.c.},\\
&U_{zx}(t,t')=\tfrac{1}{2}A_{z+}e^{-i\omega_0 t'}+\text{c.c.},\\
&U_{zy}(t,t')=\tfrac{1}{2}iA_{z+}e^{-i\omega_0 t'}+\text{c.c.},\\
&U_{zz}(t,t')=A_{zz}.
\end{aligned} \tag{A10}$$

The A coefficients are functions of the time difference $(t-t')$ given by

$$A(t,t')=\frac{1}{2\pi i}\int dp\, e^{p(t-t')}a(p). \tag{A11}$$

The a coefficients are given by

$$\begin{aligned}
a_{++}&=(p^2+i\Delta p+\tfrac{1}{2}K^2)/D,\\
a_{+-}&=(\tfrac{1}{2}K^2)/D,\\
a_{z+}&=a_{+z}=(\tfrac{1}{2}iK)(p+i\Delta)/D,\\
a_{zz}&=(p^2+\Delta^2)/D,
\end{aligned} \tag{A12}$$

where

$$D=p(p^2+K^2+\Delta^2). \tag{A13}$$

APPENDIX B: EVALUATION OF POWER SPECTRUM

The integration to be performed in Eq. (37) is

$$I=\int_{-\infty}^{t} dt' \exp[-(\tau^{-1}+i\omega)(t-t')]$$

$$\times\int_{-\infty}^{t'}\frac{dt_c}{\tau}\exp[-(t'-t_c)/\tau]\{U_{xx}(t,t')+i\langle\sigma\rangle \times[U_{xz}(t,t')U_{yz}(t',t_c)-U_{xy}(t,t')U_{zz}(t',t_c)]\}.$$

For ω, $\omega_0>0$ the resonant contribution to the constant component comes from the terms

$$\begin{aligned}
&U_{xx}(t,t')\rightarrow\tfrac{1}{2}A_{++}(t-t')e^{i\omega_0(t-t')},\\
&U_{xz}(t,t')U_{yz}(t',t_c)\rightarrow iA_{+z}(t-t')A_{-z}(t'-t_c)e^{i\omega_0(t-t')},\\
&U_{xy}(t,t')U_{zz}(t',t_c)\rightarrow\tfrac{1}{2}iA_{++}(t-t')A_{zz}(t'-t_c)e^{i\omega_0(t-t')}.
\end{aligned}$$

Changing to the new variables of integration,

$$\tau'=t-t', \quad \tau''=t'-t_c,$$

we obtain

$$I=\frac{1}{\tau}\int_0^\infty d\tau' \exp\{-[\tau^{-1}+i(\omega-\omega_0)]\tau'\}\int_0^\infty d\tau'' \times\exp[-(\tau''/\tau)]\{\tfrac{1}{2}A_{++}(\tau')[1+\tfrac{1}{2}\langle\sigma\rangle A_{zz}(\tau'')]-\langle\sigma\rangle A_{+z}(\tau')A_{-z}(\tau'')\}.$$

Substituting

$$A(t)=\frac{1}{2\pi i}\int_c dp\, e^{pt}a(p),$$

and using the Laplace transform relations

$$\int_0^\infty dt\, e^{-p't}\frac{1}{2\pi i}\int_c dp\, e^{pt}a(p)=a(p'),$$

we obtain

$$I=\tfrac{1}{2}a_{++}(p_1)[1+\tfrac{1}{2}\langle\sigma\rangle a_{zz}(p_2)]-\langle\sigma\rangle a_{+z}(p_1)a_{-z}(p_2),$$

where

$$\begin{aligned}
p_1&=[\tau^{-1}+i(\omega-\omega_0)],\\
p_2&=\tau^{-1}.
\end{aligned}$$