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Power Spectrum of Light Scattered by Two-Level Systems

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The power spectrum of the light scattered by a two-level atom driven near resonance by a monochromatic classical electric field is evaluated. The atom is assumed to relax to equilibrium with the driving field via radiation damping, which is treated by explicitly coupling the atom to the quantized electromagnetic field modes. The power spectrum of the scattered field is directly obtainable from the two-time atomic dipole moment correlation function, which is evaluated by a method based on a Markoff-type assumption analogous to that used to evaluate the time evolution of single-time atomic expectation values.

I. INTRODUCTION

THE purpose of this paper is to evaluate the power spectrum of the radiation scattered by an atomic system driven by a strong incident field, which is assumed to oscillate harmonically near one of the atomic resonance frequencies. The atom is assumed to be isolated and fixed in position, and to come into equilibrium with the driving field through the effect of radiation damping. Other relaxation processes, notably collisional[1–3] ones, are omitted from our analysis, as is the effect of the Doppler shifts associated with thermal motion.

In the model we consider, the scattered radiation possesses, in addition to a monochromatic, elastic component, inelastic components that are the result of the alteration of the atomic states by the time-dependent driving field. In the limit of weak driving fields, the scattering is almost entirely elastic, and its intensity is given (as it must be) by the usual atomic cross-section formula. As the intensity of the driving field is increased, however, inelastic scattering begins to contribute, becoming appreciable when the frequency Ω of induced atomic transitions becomes comparable to the natural relaxation rate κ. In the limit of strong driving fields ($\Omega \gg \kappa$), inelastic scattering predominates over elastic scattering. The power spectrum of the scattered field in this limit has peaks centered at the incident field frequency ω and at the displaced frequencies $\omega \pm \Omega$, with widths proportional to the atomic relaxation rate κ.

The model we have adopted as the basis of our analysis consists of a two-level atom driven by a classical electric field. The effect of radiation damping, which has been analyzed in detail in a previous article,[4] emerges naturally as the result of the coupling of the atom to the quantized electromagnetic field modes into which the atom radiates. In Ref. 4 the time evolution of the reduced density operator for the atom, and hence of the mean values of atomic operators at a given time was found. The analysis of the present paper consists of an extension of the methods of Ref. 4 appropriate to the evaluation of the correlation function representing the product of the atomic dipole moment at two different times. The Fourier transform of this atomic correlation function is, apart from some simple factors, the power spectrum of the scattered field.

In the next section of this paper, the basic model is introduced, and the power spectrum of the scattered field is related to the atomic dipole moment correlation function. In Sec. III explicit solutions are presented for the time evolution of the reduced density operator for the atom, and the relative magnitudes of the elastic and inelastic scattering intensities are evaluated. In

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[1] J. H. Van Vleck and V. F. Weisskopf, Rev. Mod. Phys. **17**, 227 (1945).

[2] R. Karplus and J. Schwinger, Phys. Rev. **73**, 1020 (1948).

[3] M. Newstein [Phys. Rev. **167**, 89 (1968)] has evaluated the power spectrum of the scattered field in the case in which the atomic relaxation is due to the strong collisional process analyzed in Refs. 1 and 2.

[4] B. R. Mollow and M. M. Miller, Ann. Phys. (N. Y.) **52**, 464 (1969).

Sec. IV the power spectrum of the scattered field is found, and various limiting cases are discussed.

II. DRIVEN TWO-LEVEL ATOM IN RESONANT APPROXIMATION

We take as our basic model a fixed atom with two energy eigenstates $|0\rangle_a$ and $|1\rangle_a$, with energies 0 and $\hbar\omega_0$, respectively. An arbitrary atomic operator may then be expressed as a linear combination of the four basis operators

$$a=|0\rangle_a\,{}_a\langle 1|, \tag{2.1a}$$

$$a^\dagger=|1\rangle_a\,{}_a\langle 0|, \tag{2.1b}$$

$$a^\dagger a=|1\rangle_a\,{}_a\langle 1|, \tag{2.1c}$$

and

$$aa^\dagger=|0\rangle_a\,{}_a\langle 0|. \tag{2.1d}$$

The atom (assumed to lie at the origin of coordinates) is coupled to the electromagnetic field, in the dipole approximation, by the expression

$$H_I(t)=-\mathbf{d}(t)\cdot\mathbf{E}(0,t), \tag{2.2}$$

where $\mathbf{E}(\mathbf{r},t)$ is the electric field at position $\mathbf{r}$ and time t, and $\mathbf{d}(t)$ is the dipole moment operator for the atom, which may be expressed in terms of the dipole matrix element $\boldsymbol{\mu}={}_a\langle 1|\mathbf{d}|0\rangle_a$ as

$$\mathbf{d}(t)=\boldsymbol{\mu}a^\dagger(t)+\boldsymbol{\mu}^*a(t). \tag{2.3}$$

In the resonant approximation, we may express the electric field as the sum of positive- and negative-frequency parts

$$\mathbf{E}(\mathbf{r},t)=(1/\sqrt{2})[\mathbf{E}^{(+)}(\mathbf{r},t)+\mathbf{E}^{(-)}(\mathbf{r},t)], \tag{2.4}$$

and approximate the interaction Hamiltonian as

$$H_I(t)=-(1/\sqrt{2})[a^\dagger(t)\boldsymbol{\mu}\cdot\mathbf{E}^{(+)}(0,t)+a(t)\boldsymbol{\mu}^*\cdot\mathbf{E}^{(-)}(0,t)]. \tag{2.5}$$

We are interested in finding the radiated field in the case in which the atom is driven by a prescribed classical electric field $\mathbf{E}_{\rm cl}(\mathbf{r},t)$, with positive- and negative-frequency parts $\mathcal{E}(\mathbf{r},t)$ and $\mathcal{E}^*(\mathbf{r},t)$ and polarization specified by the unit vector $\hat{e}_0$:

$$\mathbf{E}_{\rm cl}(\mathbf{r},t)=(1/\sqrt{2})[\mathcal{E}(\mathbf{r},t)+\mathcal{E}^*(\mathbf{r},t)]\hat{e}_0. \tag{2.6}$$

We therefore express the total electric field as the sum of this classical expression and a quantum-mechanical field expanded in a region of volume V. Denoting by ω_k, $\hat{e}_k$, and b_k the frequency, polarization vector, and annihilation operator, respectively, for the field mode specified by the index k, we have

$$\mathbf{E}^{(+)}(\mathbf{r},t)=\mathcal{E}(\mathbf{r},t)\hat{e}_0+i(\hbar/V)^{1/2}\sum_k \omega_k^{1/2}\hat{e}_k b_k(t)e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{2.7}$$

The interaction Hamiltonian thus takes the form

$$H_I(t)=-\hbar a^\dagger(t)[\lambda\mathcal{E}(0,t)+i\sum_k g_k b_k(t)]+\text{H.c.}, \tag{2.8}$$

where

$$\lambda\equiv(\boldsymbol{\mu}\cdot\hat{e}_0)/\hbar\sqrt{2} \tag{2.9}$$

and

$$g_k\equiv(\boldsymbol{\mu}\cdot\hat{e}_k)(\omega_k/2\hbar V)^{1/2}. \tag{2.10}$$

The Heisenberg field operator $\mathbf{E}(\mathbf{r},t)$ may be solved for in terms of its initial value (at a time before the driving field is turned on) and the time-dependent atomic dipole moment operator by formally integrating the equations of motion for the operators $b_k(t)$ and and $b_k^\dagger(t)$ and substituting the resulting expressions into Eq. (2.7). The result is, of course, the familiar retarded field generated by a point dipole[5] plus a freely propagating component $\mathbf{E}_f(\mathbf{r},t)$. If we make use of the fact that the time dependence of the atomic lowering operator $a(t)$ is specified by a rapid oscillation near the resonant frequency ω_0 times a relatively slowly varying function of time, we find that the positive-frequency part of the field in the scattering region (where the incident field vanishes) may be approximated by the expression

$$\mathbf{E}^{(+)}(\mathbf{r},t)=\boldsymbol{\varphi}(\mathbf{r})a(t-r/c)+\mathbf{E}_f^{(+)}(\mathbf{r},t), \tag{2.11}$$

where

$$\boldsymbol{\varphi}(\mathbf{r})=(-\omega_0^2\sqrt{2}/4\pi c^2 r^3)(\boldsymbol{\mu}\times\mathbf{r})\times\mathbf{r}, \tag{2.12}$$

and the freely propagating field operator $\mathbf{E}_f^{(+)}(\mathbf{r},t)$ consists of a sum of (initial) field annihilation operators b_k times harmonically time-varying factors. Since we are assuming the field to be unexcited initially, it follows that the freely propagating term in Eq. (2.11) will not contribute to the *normally ordered* field correlation functions. The mean value of the scattered field is thus

$$\langle\mathbf{E}^{(+)}(\mathbf{r},t)\rangle=\boldsymbol{\varphi}(\mathbf{r})\langle a(t-r/c)\rangle, \tag{2.13}$$

and the first-order field correlation function[6] is

$$G_{jk}^{(1)}(\mathbf{r}',t';\mathbf{r},t)\equiv\langle E_j^{(-)}(\mathbf{r}',t')E_k^{(+)}(\mathbf{r},t)\rangle \tag{2.14}$$

$$=\varphi_j(\mathbf{r}')\varphi_k(\mathbf{r})\times\langle a^\dagger(t'-r'/c)a(t-r/c)\rangle. \tag{2.15}$$

We will be interested in cases in which the atomic system is in equilibrium with the driving field, so that the atomic correlation function $\langle a^\dagger(t')a(t)\rangle$ depends only on the time difference $t-t'$,

$$\langle a^\dagger(t')a(t)\rangle=g(t-t'). \tag{2.16}$$

In this case, the function $G^{(1)}$ also depends only on $t-t'$. At $\mathbf{r}'=\mathbf{r}$, for example, we have

$$G_{jk}^{(1)}(\mathbf{r},t';\mathbf{r},t)=\varphi_j(\mathbf{r})\varphi_k(\mathbf{r})g(t-t'). \tag{2.17}$$

The power spectrum of the scattered field

$$I(\nu;\mathbf{r})\equiv\int_{-\infty}^{\infty}d\tau\,e^{i\nu\tau}\sum_j G_{jj}^{(1)}(\mathbf{r},0;\mathbf{r},\tau) \tag{2.18}$$

[5] See, e.g., L. Landau and E. Lifshitz, *The Classical Theory of Fields* (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1951), Eq. (9-20).

[6] R. J. Glauber, Phys. Rev. **130**, 2529 (1963).

at any point in space is thus equal to a simple factor times the Fourier transform of the atomic correlation function $g(\tau)$:

$$I(\nu;\mathbf{r})=|\boldsymbol{\varphi}(\mathbf{r})|^2\tilde{g}(\nu), \tag{2.19}$$

$$\tilde{g}(\nu)\equiv\int_{-\infty}^{\infty}d\tau\, e^{i\nu\tau}g(\tau). \tag{2.20}$$

The mean intensity of the scattered field is

$$\begin{aligned}\langle\mathbf{E}^{(-)}(\mathbf{r},t)\cdot\mathbf{E}^{(+)}(\mathbf{r},t)\rangle&=\frac{1}{2\pi}\int d\nu\, I(\nu;\mathbf{r})\\&=|\boldsymbol{\varphi}(\mathbf{r})|^2g(0)\\&=|\boldsymbol{\varphi}(\mathbf{r})|^2\bar{n}_\infty,\end{aligned} \tag{2.21}$$

where

$$\bar{n}_\infty\equiv\lim_{t\to\infty}\langle a^\dagger(t)a(t)\rangle$$

is the equilibrium probability of finding the atom in its excited state.

III. SINGLE-TIME EXPECTATION VALUES

In Ref. 4 it was shown how the time-dependent reduced density operator for the atom, and thus the mean values of atomic operators at a given time, may be evaluated. The method is based on a Markoff or factorization assumption, which represents the full Schrödinger density operator $\rho(t)$ for the joint system of field modes and atom as the product of the vacuum state $|0\rangle_F$ for the field, which is defined by the condition

$$b_k|0\rangle_F=0, \tag{3.1}$$

times the reduced density operator for the atom, which is defined as

$$\rho_a(t)\equiv\mathrm{tr}_F\rho(t), \tag{3.2}$$

where tr_F means trace with respect to the fixed states of the field. The density operator for the system is thus approximated by the expression

$$\rho(t)=|0\rangle_F\,{}_F\langle 0|\rho_a(t). \tag{3.3}$$

The unitary time-evolution operator $U(t,t')$ for the system is defined by the relations

$$i\hbar\frac{d}{dt}U(t,t')=H_S(t)U(t,t') \tag{3.4}$$

and

$$U(t',t')=1, \tag{3.5}$$

where $H_S(t)$ is the Hamiltonian $H_0+H_I(t)$ evaluated in the Schrödinger picture. The density operator for the system at time t is

$$\rho(t)=U(t,t')\rho(t')U^{-1}(t,t') \tag{3.6}$$

$$=U(t,t')|0\rangle_F\,{}_F\langle 0|\rho_a(t')U^{-1}(t,t'), \tag{3.7}$$

and the reduced density operator for the atom at time t is thus given in terms of its value at t' by the relation

$$\rho_a(t)=\mathrm{tr}_F\{U(t,t')|0\rangle_F\,{}_F\langle 0|\rho_a(t')U^{-1}(t,t')\}. \tag{3.8}$$

It was shown in Ref. 4 that the solution to Eq. (3.8) obeys the differential equation

$$\begin{aligned}\frac{d}{dt}\rho_a(t)&=\kappa a\rho_a(t)a^\dagger-\tfrac{1}{2}\kappa[a^\dagger a\rho_a(t)+\rho_a(t)a^\dagger a]\\&\quad+i[\{-\omega_0a^\dagger a+a^\dagger\lambda\mathcal{E}(0,t)+a\lambda^*\mathcal{E}^*(0,t)\},\rho_a(t)],\end{aligned} \tag{3.9}$$

where κ is the natural decay rate of the atom,

$$\kappa=|\mu|^2\omega_0^3/3\pi\hbar c^3. \tag{3.10}$$

The density operator $\rho_a(t)$ may be expressed in terms of the basis operators defined by Eqs. (2.1) as

$$\rho_a(t)=\bar{n}(t)a^\dagger a+\alpha(t)a^\dagger+\alpha^*(t)a+\bar{m}(t)aa^\dagger, \tag{3.11}$$

where

$$\bar{n}(t)={}_a\langle 1|\rho_a(t)|1\rangle_a=\mathrm{tr}\{\rho(t)a^\dagger a\}, \tag{3.12a}$$

$$\alpha(t)={}_a\langle 1|\rho_a(t)|0\rangle_a=\mathrm{tr}\{\rho(t)a\}, \tag{3.12b}$$

$$\alpha^*(t)={}_a\langle 0|\rho_a(t)|1\rangle_a=\mathrm{tr}\{\rho(t)a^\dagger\}, \tag{3.12c}$$

and

$$\bar{m}(t)={}_a\langle 0|\rho_a(t)|0\rangle_a=\mathrm{tr}\{\rho(t)aa^\dagger\}. \tag{3.12d}$$

[The relation $\mathrm{tr}_a\rho_a(t)=1$ implies $\bar{m}(t)=1-\bar{n}(t)$. It will prove convenient for the purpose of later analysis, however, if we do not make explicit use of this relation, nor of the fact that $\alpha^*(t)$ is the complex conjugate of $\alpha(t)$, but develop our analysis as if $\rho_a(t)$ were a general operator in the state space of the atom.]

By substituting Eq. (3.11) into Eq. (3.9) and making use of the definitions (2.1), we find that the parameters given by Eqs. (3.12) obey the differential equations

$$\frac{d}{dt}\bar{n}(t)=-\kappa\bar{n}(t)+i\lambda\mathcal{E}(0,t)\alpha^*(t)-i\lambda^*\mathcal{E}^*(0,t)\alpha(t), \tag{3.13a}$$

$$\frac{d}{dt}\alpha(t)=-(\tfrac{1}{2}\kappa+i\omega_0)\alpha(t)-i\lambda\mathcal{E}(0,t)[\bar{n}(t)-\bar{m}(t)], \tag{3.13b}$$

$$\begin{aligned}\frac{d}{dt}\alpha^*(t)&=(-\tfrac{1}{2}\kappa+i\omega_0)\alpha^*(t)\\&\quad+i\lambda^*\mathcal{E}^*(0,t)[\bar{n}(t)-\bar{m}(t)],\end{aligned} \tag{3.13c}$$

$$\frac{d}{dt}\bar{m}(t)=-\frac{d}{dt}\bar{n}(t). \tag{3.13d}$$

We note that by virtue of Eq. (3.13d) the trace of $\rho_a(t)$ is preserved:

$$\mathrm{tr}\rho_a(t)=\bar{n}(t)+\bar{m}(t)=\mathrm{const}. \tag{3.14}$$

The general form of the solution to the first-order linear coupled equations (3.13) for each of the functions $\bar{n}$, α, α^*, and $\bar{m}$ at an arbitrary time $t'+\tau$ (where $\tau>0$), is a linear combination of all four functions at

time t'. The coefficients in these solutions depend, in the general case, both on the time difference τ and on the initial time t', since the driving field is allowed to be an arbitrary function of time. The solution for $\alpha(t'+\tau)$, for example, takes the form

$$\alpha(t'+\tau)=\mathfrak{U}_{\alpha n}(\tau;t')\bar{n}(t')+\mathfrak{U}_{\alpha\alpha}(\tau;t')\alpha(t') +\mathfrak{U}_{\alpha\alpha}{}^{*}(\tau;t')\alpha^{*}(t')+\mathfrak{U}_{\alpha m}(\tau;t')\bar{m}(t'), \quad (3.15)$$

where the functions $\mathfrak{U}(\tau;t')$ depend upon the incident or driving field between t' and $t'+\tau$.

We shall be interested in the case in which the driving field oscillates harmonically at a frequency ω which is assumed to lie near the atomic resonance frequency ω_0, so that

$$\mathcal{E}(0,t)=\mathcal{E}_0 e^{-i\omega t}, \quad (3.16)$$

where $\mathcal{E}_0$ is a complex constant. In that case the coupled equations (3.13) can be solved directly by the method of Laplace transforms. Let us introduce the parameters

$$\Omega\equiv 2|\lambda|\,|\mathcal{E}_0|, \quad (3.17a)$$

$$\Delta\omega\equiv\omega-\omega_0, \quad (3.17b)$$

and

$$z\equiv\tfrac{1}{2}\kappa+i\Delta\omega. \quad (3.17c)$$

We find that the functions $\bar{n}(t)$ and $\alpha(t)$ approach equilibrium values that are independent of initial conditions as $t\to\infty$:

$$\bar{n}_\infty\equiv\bar{n}(t\to\infty)=\frac{\tfrac{1}{4}\Omega^2}{\tfrac{1}{2}\Omega^2+(\Delta\omega)^2+\tfrac{1}{4}\kappa^2}, \quad (3.18)$$

$$\alpha_\infty(t)\equiv\alpha(t\to\infty)=e^{-i\omega t}\frac{i\lambda\mathcal{E}_0 z}{\tfrac{1}{2}\Omega^2+(\Delta\omega)^2+\tfrac{1}{4}\kappa^2}. \quad (3.19)$$

The only time-dependent solutions we shall need to consider are those for the functions $\mathfrak{U}_{\alpha\alpha}(\tau;t')$ and $\mathfrak{U}_{\alpha m}(\tau;t')$, the former of which is independent of the initial time t' and the latter of which depends on t' only through the harmonic factor $e^{-i\omega t'}$:

$$\mathfrak{U}_{\alpha m}(\tau;t')=\mathfrak{U}_{\alpha m}{}'(\tau)e^{-i\omega t'}. \quad (3.20)$$

We find that the Laplace transform functions of $\mathfrak{U}_{\alpha\alpha}$ and $\mathfrak{U}_{\alpha m}{}'$ are given by

$$\hat{\mathfrak{U}}_{\alpha\alpha}(s)\equiv\int_0^\infty d\tau\, e^{-s\tau}\mathfrak{U}_{\alpha\alpha}(\tau) =\frac{(s+i\omega+\kappa)(s+i\omega+z)+\tfrac{1}{2}\Omega^2}{f(s+i\omega)}, \quad (3.21)$$

$$\hat{\mathfrak{U}}_{\alpha m}{}'(s)\equiv\int_0^\infty d\tau\, e^{-s\tau}\mathfrak{U}_{\alpha m}{}'(\tau) =i\lambda\mathcal{E}_0\left[\frac{(s+i\omega+\kappa)(s+i\omega+z)}{(s+i\omega)f(s+i\omega)}\right], \quad (3.22)$$

in which the function $f(s)$ is the third-degree polynomial

$$f(s)\equiv(s+\kappa)(s+z)(s+z^*)+\Omega^2(s+\tfrac{1}{2}\kappa) \quad (3.23a)$$

$$=s^3+2\kappa s^2+[\Omega^2+(\Delta\omega)^2+(5/4)\kappa^2]s +\kappa[\tfrac{1}{2}\Omega^2+(\Delta\omega)^2+\tfrac{1}{4}\kappa^2]. \quad (3.23b)$$

It is useful to compare the mean intensity of the scattered field as given by Eq. (2.21) with the square of the mean field as given by Eq. (2.13), which is just the coherent monochromatic field obtained by replacing the atomic dipole moment operator by its mean value. The ratio of the intensity of the coherently scattered light to the total scattering intensity, when the atom is in equilibrium, is, by virtue of Eqs. (3.18) and (3.19),

$$\frac{I_{\rm coh}}{I_{\rm tot}}=\frac{\langle\mathbf{E}^{(-)}\rangle\cdot\langle\mathbf{E}^{(+)}\rangle}{\langle\mathbf{E}^{(-)}\cdot\mathbf{E}^{(+)}\rangle}=\frac{\langle a^\dagger\rangle\langle a\rangle}{\langle a^\dagger a\rangle} =\frac{|\alpha_\infty|^2}{\bar{n}_\infty}=\frac{(\Delta\omega)^2+\tfrac{1}{4}\kappa^2}{\tfrac{1}{2}\Omega^2+(\Delta\omega)^2+\tfrac{1}{4}\kappa^2}, \quad (3.24)$$

where κ, Ω, and $\Delta\omega$ are given by Eqs. (3.10), (3.17a), and (3.17b), respectively. In the limit of weak incident fields ($\Omega\ll|z|$), the scattered field is almost completely coherent, and its intensity is given by the usual atomic cross-section formula, evaluated in the dipole approximation. For very strong incident fields, on the other hand, the coherent part of the scattered field is only a very small part of the total scattered field. The remaining radiation is incoherent, in the sense that its contribution to the total scattering intensity must be added, if many atoms are present, to similar expressions for each driven atom. The intensity of the incoherently scattered light becomes appreciable when saturation effects becomes important, i.e., when the incident field is strong enough to keep the atom in its excited state an appreciable fraction of the time.

IV. POWER SPECTRUM OF SCATTERED FIELD

In order to describe more precisely the characteristics of the scattered field, it is necessary to evaluate the first-order field correlation function (2.14), which, we have seen, is determined by the two-time atomic correlation function $\langle a^\dagger(t')a(t)\rangle$. It is not difficult to show that this function may be expressed in terms of the Schrödinger density operator at time t' and the time-independent Schrödinger operators a and $a^\dagger$ as

$$\langle a^\dagger(t')a(t)\rangle=\mathrm{tr}\{\rho(t')a^\dagger U^{-1}(t,t')aU(t,t')\} =\mathrm{tr}\{|0\rangle_{F\,F}\langle 0|\rho_a(t')a^\dagger U^{-1}(t,t')aU(t,t')\}, \quad (4.1)$$

where $U(t,t')$ is the time-development operator defined by Eqs. (3.4) and (3.5), and in reaching the final step we have made use of the Markoff approximation (3.3).

The right-hand side of Eq. (4.1) may be evaluated by an entirely straightforward method, which is analogous to that used to obtain the mean values of atomic operators at a given time. Let us recall that the expecta-

tion value

$$\alpha(t)=\mathrm{tr}[\rho(t)a] \tag{4.2}$$

is given, according to Eq. (3.7), by the relation

$$\alpha(t)=\mathrm{tr}\{|0\rangle_F\,{}_F\langle 0|\rho_a(t')U^{-1}(t,t')aU(t,t')\}. \tag{4.3}$$

Comparison of the right-hand sides of Eqs. (4.1) and (4.3) shows that the former may be obtained from the latter simply by making the substitution

$$\rho_a(t')\rightarrow\rho_a(t')a^\dagger. \tag{4.4}$$

The evaluation of the two-time correlation function $\langle a^\dagger(t')a(t)\rangle$ is thus formally identical to the evaluation of the single-time mean value $\alpha(t)=\langle a(t)\rangle$, the only difference being that the non-Hermitian operator $\rho_a(t')a^\dagger$ must be used in place of the density operator $\rho_a(t')$. This means that we must make the substitutions, on the right-hand side of Eq. (3.15),

$$\bar{n}(t')={}_a\langle 1|\rho_a(t')|1\rangle_a\rightarrow{}_a\langle 1|\rho_a(t')a^\dagger|1\rangle_a=0, \tag{4.5a}$$

$$\alpha(t')={}_a\langle 1|\rho_a(t')|0\rangle_a\rightarrow{}_a\langle 1|\rho_a(t')a^\dagger|0\rangle_a=\bar{n}(t'), \tag{4.5b}$$

$$\alpha^*(t')={}_a\langle 0|\rho_a(t')|1\rangle_a\rightarrow{}_a\langle 0|\rho_a(t')a^\dagger|1\rangle_a=0, \tag{4.5c}$$

$$\bar{m}(t')={}_a\langle 0|\rho_a(t')|0\rangle_a\rightarrow{}_a\langle 0|\rho_a(t')a^\dagger|0\rangle_a=\alpha^*(t'). \tag{4.5d}$$

We have then

$$\begin{aligned}g(\tau;t')&\equiv\langle a^\dagger(t')a(t'+\tau)\rangle\\&=\mathfrak{U}_{\alpha\alpha}(\tau;t')\bar{n}(t')+\mathfrak{U}_{\alpha m}(\tau;t')\alpha^*(t'),\end{aligned} \tag{4.6}$$

where the quantities $\mathfrak{U}(\tau;t')$ are the same functions[7] that appear in the solution for the atomic mean value $\alpha(t)$.

Let us now consider the case in which the driving field is the harmonic function given by Eq. (3.16), so that $\mathfrak{U}_{\alpha\alpha}(\tau;t')$ is independent of t', and $\mathfrak{U}_{\alpha m}(\tau;t')$ has the form given by Eq. (3.20). Let us also assume that the atom is in equilibrium with the field, so that $\bar{n}(t')$ and $\alpha(t')$ are given by the asymptotic expressions (3.18) and (3.19), with the latter evaluated at time t'. The atomic correlation function in Eq. (4.6) is then independent of the initial time t', and is given by the relation

$$g(\tau)=\mathfrak{U}_{\alpha\alpha}(\tau)\bar{n}_\infty+\mathfrak{U}_{\alpha m}'(\tau)\left[\frac{-i\lambda^*\mathcal{E}_0^*z^*}{\frac{1}{2}\Omega^2+(\Delta\omega)^2+\frac{1}{4}\kappa^2}\right]. \tag{4.7}$$

The Laplace transform of $g(\tau)$

$$\hat{g}(s)\equiv\int_0^\infty d\tau\, e^{-s\tau}g(\tau) \tag{4.8}$$

thus satisfies the relation

$$\hat{g}(s)=\hat{\mathfrak{U}}_{\alpha\alpha}(s)\bar{n}_\infty+\hat{\mathfrak{U}}_{\alpha m}'(s)\left[\frac{-i\lambda^*\mathcal{E}_0^*z^*}{\frac{1}{2}\Omega^2+(\Delta\omega)^2+\frac{1}{4}\kappa^2}\right], \tag{4.9}$$

where the Laplace transform functions on the right-hand side are given by Eqs. (3.21) and (3.22).

In order to evaluate the spectral correlation function $\tilde{g}(\nu)$ defined by Eq. (2.20), it is first necessary to find the asymptotic form of $g(\tau)$ in the limit $\tau\rightarrow\infty$, which originates from the poles of $\hat{g}(s)$ on the imaginary axis of the s plane. Since it is readily shown that the real parts of the roots of the polynomial $f(s)$ defined by Eq. (3.23) are all negative (in fact, they all lie between $-\kappa$ and $-\frac{1}{2}\kappa$), it is clear that the only contribution to $g(\tau\rightarrow\infty)$ comes from the pole at $s=-i\omega$ on the right-hand side of Eq. (3.22). The residue of the pole is, by virtue of Eqs. (4.9), (3.22), (3.17), (3.19), and (3.23),

$$\begin{aligned}\lim_{s\rightarrow i\omega}[(s+i\omega)\hat{g}(s)]&=\frac{\frac{1}{4}\Omega^2z^*}{\frac{1}{2}\Omega^2+(\Delta\omega)^2+\frac{1}{4}\kappa^2}\left(\frac{\kappa z}{f(0)}\right)\\&=\tfrac{1}{4}\Omega^2|z|^2/(\tfrac{1}{2}\Omega^2+|z|^2)^2=|\alpha_\infty|^2,\end{aligned} \tag{4.10}$$

where $|\alpha_\infty|$ is the modulus of the right-hand side of Eq. (3.19). The contribution of this term to the correlation function $g(\tau)$ is the coherent, harmonically varying expression

$$g_{\mathrm{coh}}(\tau)=|\alpha_\infty|^2e^{-i\omega\tau} \tag{4.11}$$

$$=\alpha_\infty^*(t')\alpha_\infty(t'+\tau), \tag{4.12}$$

which may thus be obtained by replacing the atomic operators $a^\dagger(t')$ and $a(t'+\tau)$ by their equilibrium expectation values. The Laplace transform of the remaining, incoherent part of the atomic correlation function is

$$\hat{g}_{\mathrm{inc}}(s)\equiv\hat{g}(s)-|\alpha_\infty|^2/(s+i\omega). \tag{4.13}$$

By making use of Eqs. (3.19), (3.21)–(3.23), and (4.9), after a lengthy reduction we find that $\hat{g}_{\mathrm{inc}}(s)$ may be expressed in the form

$$\begin{aligned}\hat{g}_{\mathrm{inc}}(s)=&\frac{\frac{1}{2}\bar{n}_\infty\Omega^2}{\frac{1}{2}\Omega^2+|z|^2}\\&\times\left(\frac{(s+i\omega)^2+2\kappa(s+i\omega)+(\frac{1}{2}\Omega^2+\kappa^2)}{f(s+i\omega)}\right).\end{aligned} \tag{4.14}$$

The spectral correlation function $\tilde{g}(\nu)$ defined by Eq. (2.20) may be obtained from the Laplace transform $\hat{g}(s)$ defined by Eq. (4.8) simply by noting that $g(-\tau)=g^*(\tau)$, and hence $\tilde{g}(\nu)=2\,\mathrm{Re}[\hat{g}(-i\nu)]$. Applying this relation to the incoherent part of g, and adding the result to the Fourier transform of the coherent expression (4.11), after some work we find, with the aid of Eq. (3.23b),

$$\begin{aligned}\tilde{g}(\nu)=&2\pi|\alpha_\infty|^2\delta(\nu-\omega)\\&+\bar{n}_\infty\kappa\Omega^2\left(\frac{(\nu-\omega)^2+(\frac{1}{2}\Omega^2+\kappa^2)}{|f(i(\nu-\omega))|^2}\right).\end{aligned} \tag{4.15}$$

The spectral atomic correlation function $\tilde{g}(\nu)$, and hence, by virtue of Eq. (2.19), the power spectrum of

[7] This result is a special case of a *quantum regression thoerem* derived by M. Lax. See, e.g., M. Lax, Phys. Rev. **129**, 2342 (1963), and related references.

the scattered field, is thus expressed in terms of the parameters defined by Eqs. (3.10) and (3.17)–(3.19) and the function $f(s)$ defined by Eq. (3.23).

Before discussing the limiting forms of the function $\tilde{g}(\nu)$, it is convenient to express it in terms of the three roots of the polynomial $f(s)$. If the parameter

$$\zeta \equiv [\Omega^2+(\Delta\omega)^2-\tfrac{1}{12}\kappa^2]^3+\tfrac{3}{4}\kappa^2[\tfrac{1}{2}\Omega^2-(\Delta\omega)^2-\kappa^2/36]^2 \quad (4.16)$$

is positive, then there will be one real root s_0 and two complex-conjugate roots s_+ and $s_-=s_+^*$, while if $\zeta<0$, all three roots s_0, s_+, and s_- will be real. In either case we have

$$|f(i\nu)|^2=(\nu^2+s_0^2)(\nu^2+s_+^2)(\nu^2+s_-^2), \quad (4.17)$$

and the function $\tilde{g}(\nu)$ may be expressed in the form

$$\tilde{g}(\nu)=2\pi|\alpha_\infty|^2\delta(\nu-\omega)+\bar{n}_\infty\kappa\Omega^2 \times\left(\frac{(\nu-\omega)^2+(\tfrac{1}{2}\Omega^2+\kappa^2)}{[(\nu-\omega)^2+s_0^2][(\nu-\omega)^2+s_+^2][(\nu-\omega)^2+s_-^2]}\right) \quad (4.18)$$

or

$$\tilde{g}(\nu)=2\pi|\alpha_\infty|^2\delta(\nu-\omega)+\frac{D_0}{(\nu-\omega)^2+s_0^2} +\frac{D_+}{(\nu-\omega)^2+s_+^2}+\frac{D_-}{(\nu-\omega)^2+s_-^2}, \quad (4.19)$$

in which the parameters D_0, D_+, and D_- are defined as

$$D_0=\bar{n}_\infty\kappa\Omega^2\left(\frac{\tfrac{1}{2}\Omega^2+\kappa^2-s_0^2}{(s_0^2-s_+^2)(s_0^2-s_-^2)}\right), \quad (4.20a)$$

$$D_+=\bar{n}_\infty\kappa\Omega^2\left(\frac{\tfrac{1}{2}\Omega^2+\kappa^2-s_+^2}{(s_+^2-s_0^2)(s_+^2-s_-^2)}\right), \quad (4.20b)$$

$$D_-=\bar{n}_\infty\kappa\Omega^2\left(\frac{\tfrac{1}{2}\Omega^2+\kappa^2-s_-^2}{(s_-^2-s_0^2)(s_-^2-s_+^2)}\right). \quad (4.20c)$$

We may note that the time-dependent atomic correlation function $g(\tau)$, which is the Fourier transform of $\tilde{g}(\nu)$, is given by

$$g(\tau)=|\alpha_\infty|^2e^{-i\omega\tau}-(D_0/2s_0)e^{-i\omega\tau+s_0|\tau|} -(D_+/2s_+)e^{-i\omega\tau+s_+|\tau|}-(D_-/2s_-)e^{-i\omega\tau+s_-|\tau|}. \quad (4.21)$$

When the parameter ζ defined by Eq. (4.16) is negative, Eq. (4.19) expresses the incoherent part of $\tilde{g}(\nu)$ as the sum of three Lorentzian functions centered at $\nu=\omega$, with widths $-s_0$, $-s_+$, and $-s_-$, respectively. It is important to note, however, that one of the parameters D_0, D_+, or D_- (the one associated with the root of intermediate magnitude) is negative, and hence the Lorentzian in which it appears has negative weight. It is an interesting feature of the model we are considering that, in the limit $|\nu-\omega|\to\infty$, the spectral density falls to zero as $(\nu-\omega)^{-4}$, rather than as $(\nu-\omega)^{-2}$, as it would for positive Lorentzian functions.

When the parameter ζ defined by Eq. (4.16) is positive, the roots s_+ and s_- of $f(s)$ are the complex-conjugate quantities

$$s_\pm=\sigma\pm i\Omega', \quad (4.22)$$

where σ and Ω' are real. In this case it is convenient to express the function $\tilde{g}(\nu)$ in the form

$$\tilde{g}(\nu)=2\pi|\alpha_\infty|^2\delta(\nu-\omega)+\bar{n}_\infty\kappa\Omega^2 \times\left(\frac{(\nu-\omega)^2+(\tfrac{1}{2}\Omega^2+\kappa^2)}{[(\nu-\omega)^2+s_0^2][(\nu-\omega-\Omega')^2+\sigma^2][(\nu-\omega+\Omega')^2+\sigma^2]}\right) \quad (4.23)$$

or

$$\tilde{g}(\nu)=2\pi|\alpha_\infty|^2\delta(\nu-\omega)+\frac{D_0}{(\nu-\omega)^2+s_0^2} +\frac{M-(\nu-\omega-\Omega')N}{(\nu-\omega-\Omega')^2+\sigma^2}+\frac{M+(\nu-\omega+\Omega')N}{(\nu-\omega+\Omega')^2+\sigma^2}, \quad (4.24)$$

in which the real quantities M and N are defined as

$$M=\tfrac{1}{2}\sigma(D_+/s_++D_-/s_-), \quad (4.25a)$$

$$N=-\tfrac{1}{2}i(D_+/s_+-D_-/s_-). \quad (4.25b)$$

As an illustration of these results, let us consider the case in which the incident field is exactly on resonance ($\Delta\omega=0$). The roots of the polynomial $f(s)$ are then[4]

$$s_0=-\tfrac{1}{2}\kappa, \quad (4.26a)$$

$$s_+=-\tfrac{3}{4}\kappa+(\tfrac{1}{16}\kappa^2-\Omega^2)^{1/2}, \quad (4.26b)$$

$$s_-=-\tfrac{3}{4}\kappa-(\tfrac{1}{16}\kappa^2-\Omega^2)^{1/2}. \quad (4.26c)$$

These roots are all real for $\Omega<\frac{1}{4}\kappa$, while for $\Omega>\frac{1}{4}\kappa$ the roots s_+ and s_- have the form given by Eq. (4.22), with

$$\sigma=-\tfrac{3}{4}\kappa, \quad (4.27a)$$

$$\Omega'=(\Omega^2-\tfrac{1}{16}\kappa^2)^{1/2}. \quad (4.27b)$$

The parameters D_0, M, and N in Eq. (4.24) are then given by

$$D_0=\tfrac{1}{2}\kappa\bar{n}_\infty, \quad (4.28)$$

$$M=\tfrac{3}{8}\kappa\bar{n}_\infty\left(\frac{\Omega^2-\tfrac{1}{2}\kappa^2}{\Omega^2+\tfrac{1}{2}\kappa^2}\right), \quad (4.29a)$$

$$N=\frac{\tfrac{1}{8}\kappa\bar{n}_\infty}{\Omega'}\left(\frac{5\Omega^2-\tfrac{1}{2}\kappa^2}{\Omega^2+\tfrac{1}{2}\kappa^2}\right). \quad (4.29b)$$

We have plotted the function $\tilde{g}(\nu)$ for representative values of Ω (and $\Delta\omega=0$) in Fig. 1.

For $\Delta\omega\neq0$, the function $\tilde{g}(\nu)$ takes on a relatively simple form in two limiting cases of interest: that of

very low and that of very high incident field intensity. If the incident field is weak enough so that the parameter Ω defined by Eq. (3.17a) is very small compared to the natural decay rate κ, the three roots of $f(s)$ are (approximately) $-\kappa$, z, and z^*. By making use of expressions (3.18) and (3.19) for $\bar{n}_\infty$ and $|\alpha_\infty|$, we see that in this limit relation (4.23) for $\tilde{g}(\nu)$ reduces to

$$\tilde{g}(\nu)=\frac{\frac{1}{4}\Omega^2}{(\Delta\omega)^2+\frac{1}{4}\kappa^2}\left(2\pi\delta(\nu-\omega)+\frac{\kappa\Omega^2}{[(\nu-\omega-\Delta\omega)^2+\frac{1}{4}\kappa^2][(\nu-\omega+\Delta\omega)^2+\frac{1}{4}\kappa^2]}\right)$$

$$\text{for } \Omega\ll\kappa. \quad (4.30)$$

The integrated spectral intensity of the incoherent part of the scattered field in this limit is only a very small fraction $[\frac{1}{4}\Omega^2/(\frac{1}{4}\kappa^2+(\Delta\omega)^2)]$ of the intensity of the coherently scattered field. For $|\Delta\omega|\gg\kappa$, the incoherent part of the power spectrum is sharply peaked at the two displaced frequencies $\omega-\Delta\omega=\omega_0$ and $\omega+\Delta\omega=\omega_0+2\Delta\omega$.

If the incident field is intense enough so that Ω is much greater than κ, we find that the roots of the polynomial $f(s)$ are well approximated by the expressions

$$s_0=-\tfrac{1}{2}\kappa\left[\frac{\Omega^2+2(\Delta\omega)^2}{\Omega^2+(\Delta\omega)^2}\right] \quad (\Omega\gg\kappa) \quad (4.31a)$$

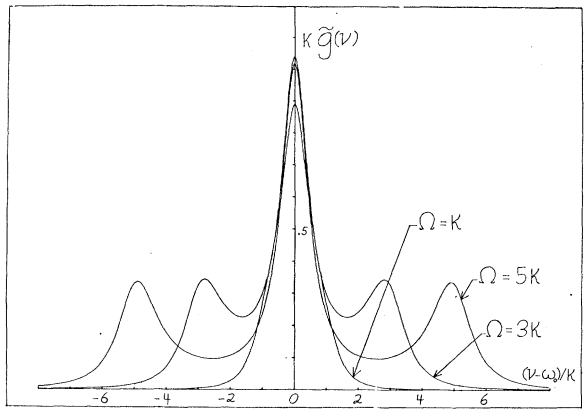

FIG. 1. Spectral density $\tilde{g}(\nu)$ for a two-level atom driven exactly on resonance.

and $s_\pm=\sigma\pm i\Omega'$, where

$$\sigma=-\kappa\left[\frac{\frac{3}{4}\Omega^2+\frac{1}{2}(\Delta\omega)^2}{\Omega^2+(\Delta\omega)^2}\right] \quad (\Omega\gg\kappa) \quad (4.31b)$$

and Ω' is the Rabi[8] frequency of population inversion in the absence of damping,

$$\Omega'=[\Omega^2+(\Delta\omega)^2]^{1/2} \quad (\Omega\gg\kappa). \quad (4.31c)$$

Since $\Omega'\gg\sigma$, s_0 in this limit, the function $\tilde{g}(\nu)$ as given by Eq. (4.23) may be approximated in the domain in which it is appreciable by a superposition of Lorentzian functions at each of its maxima at $\nu=\omega$, $\nu=\omega+\Omega'$, and $\nu=\omega-\Omega'$:

$$\tilde{g}(\nu)=2\pi|\alpha_\infty|^2\delta(\nu-\omega)+\frac{\bar{n}_\infty\kappa\Omega^2}{\Omega'^4}\left\{\frac{\frac{1}{2}\Omega^2}{(\nu-\omega)^2+s_0^2}+[\tfrac{3}{8}\Omega^2+\tfrac{1}{4}(\Delta\omega)^2]\left[\frac{1}{(\nu-\omega-\Omega')^2+\sigma^2}+\frac{1}{(\nu-\omega+\Omega')^2+\sigma^2}\right]\right\} \quad \text{for } \Omega\gg\kappa. \quad (4.32)$$

In the limit of very intense incident fields, Ω is much greater than both $|\Delta\omega|$ and κ, and we find, with the aid of the relations (3.18) and (3.19), that Eqs. (4.32) and (4.31) reduce to the relation

$$\tilde{g}(\nu)=2\pi\left(\frac{(\Delta\omega)^2+\frac{1}{4}\kappa^2}{\Omega^2}\right)\delta(\nu-\omega)+\frac{\frac{1}{4}\kappa}{(\nu-\omega)^2+\frac{1}{4}\kappa^2}+\frac{\frac{3}{16}\kappa}{(\nu-\omega-\Omega)^2+\frac{9}{16}\kappa^2}+\frac{\frac{3}{16}\kappa}{(\nu-\omega+\Omega)^2+\frac{9}{16}\kappa^2}$$

$$\text{for } \Omega\gg\kappa, |\Delta\omega|. \quad (4.33)$$

The maxima at $\nu=\omega\pm\Omega$ in this limit are one-third of the maximum at $\nu=\omega$, and the integrated spectral intensity at each of the displaced frequencies is one-half of that at the central frequency. The intensity of the coherent, elastically scattered light in this limit is inversely proportional to the incident field intensity, and is only a very small fraction of the total scattering intensity.

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[8] I. I. Rabi, Phys. Rev. 51, 652 (1937).