Quantum macroscopic effect in resonance optics

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According to quantum electrodynamics, the reflection of resonance radiation by a medium containing excited atoms (molecules) is not determined by its refractive index. Actually, the reflection process is described by two interfering channels. The coherent channel contains elastic scattering processes and gives rise to the selective component. The incoherent channel modifies the quantum state of the medium and produces Raman scattering and stimulated emission. It contributes to both selective and diffuse components of scattered radiation. It is shown that, when the reflectance of a thermally excited medium for resonance radiation is calculated, the result obtained in the semiclassical theory is different at the macroscopic level from that found in quantum electrodynamics.

1. According to the semiclassical theory of radiation that operates with nonquantized electromagnetic fields, the evolution of the electromagnetic field in a medium is wholly determined by its permittivity. If the medium consists of two-level atoms (molecules) with ground and excited state concentrations n_μ and n_m, respectively, the permittivity of the medium for $(n_\mu - n_m)\lambda^3 \ll 1$ is found to be [1] proportional to the difference $n_\mu - n_m$. We suggested in our previous papers[2,3] that the semiclassical approach is inadequate as a way of describing the evolution of resonance radiation in inhomogeneous excited media because their permittivity does not determine the Poynting vector. This conclusion was based on a study of the reflection of resonance radiation by thermally excited media. The question is also of independent interest. The point is that the reflectance of an inverted medium for resonance radiation, calculated from the semiclassical theory of radiation, is not in agreement[4–6] with experimental data. It was pointed out in Ref. 7 that a possible reason for this discrepancy was that lateral waves were not taken into account. We now draw attention to another reason, namely, the fact that, when the quantum-mechanical properties of radiation are taken into account qualitatively at the macroscopic level, this modifies the calculated reflectance even for thermally excited media. This is obviously unexpected and requires rigorous demonstration. In this paper, we present a proof of this proposition that does not rely on perturbation theory since, as will be shown below, the direct evaluation of the reflectance of an excited medium cannot be carried out in finite-order perturbation theory because of the presence of stimulated processes.

To begin with, we must establish the reason why the necessity for field quantization in reflection problems was not noted before. It was assumed that the reflectance of a dispersive medium can be calculated by the methods of standard statistical Green's functions.[8] The field correlation functions are then unimportant, but several effects slip out of view. In systems with a dense energy spectrum, the higher-order terms in V (V is the volume of the system) in the Feynman expansion[8] are asymptotically taken into account by the decoupling of correlation functions. The reason for this is that, to derive closed equations in the Bogolyubov chain, higher-order correlation functions must be expressed in terms of lower-order functions. In systems with a sparse spectrum (e.g., one-dimensional systems), decoupling is not in general admissible.[9] Complications also arise in boundary-value problems. On the other hand, there is no problem with diffuse reflection because coherent processes are then unimportant, and reflectance evaluated in the lowest order in the coupling constant is proportional to the number of scattering targets, i.e., $R^{(n)} \sim nV$ (n is the concentration of these targets). The admissibility of decoupling of correlation functions in the case of coherent (Fresnel) reflection is not as obvious. The reflectance is then proportional to the square of the concentration of scattering targets, i.e., $R^{(c)} \propto n^2$, so that $R^{(c)}/R^{(n)} \propto V^{-1}$. In general, decoupling will not guarantee that terms of this structure will be correctly evaluated. The question then is whether higher-order correlations should be taken into account in boundary value problems. The answer is that they should because of the presence of stimulated emission processes, provided the excited atoms in the medium contribute significantly to the reflected radiation.

Nonclassical states of the electromagnetic field (antibunching of photons and squeezed states) have recently attracted close attention. These states are identified by analyzing higher-order correlations. We now draw attention to quantum-mechanical effects that can be seen under the conditions of interference. They arise from the interaction between a field and a medium if there are two or more photons in the virtual states that determine scattering. This is what happens in the case of scattering by excited atoms, which is virtually described in the resonance approximation in terms of the emission of the scattered photon, followed by the absorption of the latter. These quantum-mechanical effects are not small and their contribution to the resultant signal can reach 100%. The Poynting vector, i.e., the lowest-order correlation function, carries information about these effects.

There is an elegant technique[10] for the analysis of the evolution of higher-order correlations, based on Hall's generalization[11] of the thermodynamic variant of Wick's theorem.[8,12] The absence of correlations from the medium follows from Hall's generalization, provided they were not present in the initial state. In our case, the situation is different. There are no correlations in the initial state, there can be only one scattered photon, and the number of photons is

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multiplied up during the evolution process as a consequence of stimulated emission.

2. We propose a different modification of the standard formalism that enables us to avoid the decoupling of photon-photon correlations in any order of perturbation theory and yet obtain a closed Dyson-type equation. We shall decouple the atom-atom correlations in the usual way,[8,12] assuming that the scatterers are uncorrelated. The theory contains no other approximations and the calculations are performed exactly. This is an important point. As noted above, when the medium contains excited atoms, the Feynman series contains singular terms that can be given a meaning only by the partial summation of infinite subsequences. Different methods of summation then lead to different results. The formalism that we are proposing automatically selects the regular part $\langle\hat{\mathbf{s}}\rangle^{(c)}$, whereas the irregular part $\langle\hat{\mathbf{s}}\rangle^{(n)}$ can be shown to be positive-definite without resorting to perturbation theory. Our formalism is based on the unitary transformation of the $\hat{s}$-matrix expansion and, obviously, retains all the advantages of standard Feynman-diagram type techniques, summation rules, and Dyson equations. Under equilibrium conditions, it retains the dispersion relations, the Kallén-Lehmann representations, and so on. Moreover, the absence of photon-photon correlation decoupling means that field moments of any order can be calculated.

We shall describe the electromagnetic field in a medium in terms of the density matrix ρ of the system, in which we have evaluated the trace of the atomic variables of the medium. We are therefore dealing with a significantly more detailed description of the field, which is based on expectation values and finite-order correlations. We shall determine the evolution of the density matrix of the subsystem (electromagnetic field) when, generally speaking, it is not strongly coupled to the environment. The arguments of the matrix ρ are the occupation numbers. Under equilibrium conditions, and when the coupling between the radiation and matter is weak, this matrix determines the Gibbs distribution. We will find the equation for it. Actually, we are concerned with the derivation of the analogs of the constitutive equations and Maxwell's equations in the medium at the quantum-mechanical level. In its formal aspect, the formulation of the problem resembles Refs. 13–15 in its derivation of the controlling equation, but the formalism that we employ will enable us to retain the advantages of the method of second quantization.

Let us introduce auxiliary,[16] mutually hermitian, creation (annihilation) operators $\hat{\mathfrak{N}}_{\mathbf{N}}^{+}$ ($\hat{\mathfrak{N}}_{\mathbf{N}}$) for the ensemble of photons as a whole. The components of the vector $\mathbf{N}$ are the occupation numbers $N_{\mathbf{k}\lambda}$ of the optical modes characterized by wave vector $\mathbf{k}$ and polarization index λ. By analogy with phase volumes in statistical physics, we refer to these operators as Γ operators, in contrast with the mean operators that represent the creation and annihilation of single-particle states.

Specifically, we shall be concerned with nonrelativistic gas atoms each of which contains a single valence electron. Spin effects will be neglected. We then associate a field operator $\psi(\mathbf{r}, \mathbf{R})$ with the gas atoms in the Schroedinger representation, where $\mathbf{R}$ is the position vector of the center of gravity of an individual atom and $\mathbf{r}$ the position vector of the valence electron. We associate the operator $\hat{A}_{\nu}(\mathbf{r})$ with the electromagnetic field. Assuming a quasiresonant interaction, i.e., $|k-\omega_{m\mu}| \ll k+\omega_{m\mu}$, where $\omega_{m\mu}$ is the optical transition frequency of the atoms, we adopt the following Schroedinger equation for the system ($\hbar = c = 1$):

$$i\frac{\partial\Psi}{\partial t} = \left[\hat{H}_a + \hat{H}_{ph} - \frac{e}{m}\int \hat{\psi}^{+}(\mathbf{r},\mathbf{R})\,\mathbf{p}\hat{\mathbf{A}}(\mathbf{r})\,\hat{\psi}(\mathbf{r},\mathbf{R})\,d\mathbf{r}\,d\mathbf{R}\right]\Psi \tag{1}$$

where

$$\hat{\psi}(\mathbf{r},\mathbf{R}) = \sum_{ip}\psi_i(\mathbf{r}-\mathbf{R})\,e^{i\mathbf{pr}}\frac{\hat{b}_{ip}}{V^{1/2}},$$

$$\hat{H}_a = \sum_{ip}\varepsilon_{ip}\hat{b}_{ip}^{+}\hat{b}_{ip}, \qquad \varepsilon_{ip} = \varepsilon_i + \frac{p^2}{2M},$$

in which ψ_i is the wave function describing the internal structure of the atoms with energy ε_i. We shall use $\mathbf{p}$ to represent the momentum of the atom, M the mass of the atomic residue, $V = L_x L_y L_z$ the quantization volume, and $\hat{b}_{ip}^{+}$ ($\hat{b}_{ip}$) the creation (annihilation) operators for the atom in a state $(i,\mathbf{p})$.

Next, we assume that

$$\hat{A}_{\nu}(\mathbf{r}) = \sum_{\mathbf{k}\lambda}\frac{e_{\mathbf{k}\nu}^{\lambda}}{[2\omega(\lambda)V]^{1/2}}\hat{\alpha}_{\mathbf{k}\lambda}e^{i\mathbf{kr}} + \text{h.c.},$$

$$\hat{H}_{ph} = \sum_{\mathbf{k}\lambda}k\hat{\alpha}_{\mathbf{k}\lambda}^{+}\hat{\alpha}_{\mathbf{k}\lambda},$$

where $\mathbf{e}_{\mathbf{k}}^{\lambda}$ is the linear polarization unit vector, $\hat{\alpha}_{\mathbf{k}\lambda}^{+}$ ($\hat{\alpha}_{\mathbf{k}\lambda}$) are the photon creation (annihilation) operators, and the vacuum term in $\hat{H}_{\text{ph}}$ has been discarded. In the gauge with zero scalar potential, we have

$$\omega(\lambda) = \begin{cases} (k^2+\mu^2)^{1/2}, & \lambda = 1,2 \\ \mu, & \lambda = 3, \end{cases}$$

in which μ will be allowed to tend to zero at the end of the calculation.[17] The statistical properties of the operators $\hat{b}_{ip}^{+}$ and $\hat{b}_{ip}$ are unimportant when the gas is not temperature degenerate. We assume that

$$[\hat{b}_{ip}, \hat{b}_{i'p'}^{+}] = \delta_{ii'}\delta_{pp'}.$$

To evaluate the required density matrix, we introduce the functions

$$\Phi^0(\mathbf{N}|\xi) = \prod_{\mathbf{k}\lambda}\varphi(N_{\mathbf{k}\lambda}|\xi_{\mathbf{k}\lambda}),$$

that describe, in the second-quantization representation, the free electromagnetic field of the configuration defined by the vector $\mathbf{N}$, where $\varphi(N|\zeta)$ represents the wave functions of the quantum-mechanical oscillator.

Let

$$\hat{\Phi}(\xi) = \sum_{\mathbf{N}}\hat{\mathfrak{N}}_{\mathbf{N}}\Phi^0(\mathbf{N}|\xi).$$

Assuming that $\hat{\mathfrak{N}}_{\mathbf{N}}$ and $\hat{\mathfrak{N}}_{\mathbf{N}}^{+}$ act in some new space Γ, we construct the unitary operator

$$\hat{O} = \hat{\Phi}^{+}(\xi)\rangle_{\Gamma}{}^{0},$$

which transforms the dynamic formalism from the second

quantization space to the space Γ. The vacuum basis vector in the new representation is denoted by $\rangle_\Gamma^0$, and the remaining basis vectors are constructed by applying the operators $\hat{\mathfrak{A}}_N^+$ to it. It is convenient at this point to note that the set $\hat{\mathfrak{A}}_N^+\rangle_\Gamma^0$ is sufficient basis for physical states. The repeated application of the operator $\hat{\mathfrak{A}}_N^+$ will not be encountered. It is precisely this fact that favors the use of the Γ space.

If some state of the free electromagnetic field is described in the second quantization representation $\Phi^0(\mathbf{N}^0|\zeta)$, then the following wave function corresponds to it in Γ space:

$$\hat{O}\Phi^0(\mathbf{N}^0|\zeta)=\int\sum_{\mathbf{N}}\hat{\mathfrak{A}}_{\mathbf{N}}{}^+\hat{\Phi}(\mathbf{N}|\zeta)\rangle_\Gamma{}^0\Phi^0(\mathbf{N}^0|\zeta)\,d\zeta=\hat{\mathfrak{A}}_{\mathbf{N}^0}{}^+\rangle_\Gamma{}^0,$$

where

$$d\zeta=\prod_{\mathbf{k}\lambda}d\zeta_{\mathbf{k}\lambda}.$$

The inverse transformation is also valid:

$$\hat{O}^+\hat{\mathfrak{A}}_{\mathbf{N}^0}{}^+\rangle_\Gamma{}^0=\left\langle\sum_{\mathbf{N}}\hat{\Phi}(\mathbf{N}|\zeta)\hat{\mathfrak{A}}_{\mathbf{N}^0}{}^+\right\rangle_\Gamma^0=\Phi^0(\mathbf{N}^0|\zeta).$$

There is no need to specify the commutation relations for the oscillary operators $\hat{\mathfrak{A}}_{\mathbf{N}}$ and $\hat{\mathfrak{A}}_{\mathbf{N}}^+$ when the unitary operator $\hat{O}$ is constructed. They are unrelated to the statistical properties of the fields, and can remain arbitrary. Since the application of the operator $\hat{\mathfrak{A}}_{\mathbf{N}}^+$ generates a complete state of the electromagnetic field, it follows that $\hat{\mathfrak{A}}_{\mathbf{N}}$ and $\hat{\mathfrak{A}}_{\mathbf{N}}^+$ can only appear alternately in the theory (by analogy with the description of the kinetics of a photon by $\hat{a}_{\mathbf{k}\lambda}$ and $\hat{a}_{\mathbf{k}\lambda}^+$). However, the commutation relations for both Fermi and Bose fields then lead to identical final expressions. We shall assume for simplicity that

$$[\hat{\mathfrak{A}}_{\mathbf{N}},\ \hat{\mathfrak{A}}_{\mathbf{N}'}{}^+]=\delta_{\mathbf{N}\mathbf{N}'}.$$

We now apply the unitary transformation to (1) via the operators $\hat{O}$ and $\hat{O}^+$:

$$i\frac{\partial\Psi_\Gamma}{\partial t}=\Big[\hat{H}_a+\sum_{\mathbf{N}}\varepsilon(\mathbf{N})\hat{\mathfrak{N}}_{\mathbf{N}}{}^+\hat{\mathfrak{A}}_{\mathbf{N}}$$
$$-\frac{e}{m}\int\hat{\Phi}^+\hat{\psi}^+\hat{\mathbf{p}}\hat{\mathbf{A}}\hat{\psi}\hat{\Phi}d\zeta\,d\mathbf{r}\,d\mathbf{R}\Big]\Psi_\Gamma,\quad\Psi_\Gamma=\hat{O}\Psi,\qquad(2)$$

where $\varepsilon(\mathbf{N})$ is the energy of the free electromagnetic field with occupation vector $\mathbf{N}$. The expectation value of any electromagnetic field operator can now be found from the formula

$$\langle\hat{K}\rangle=\int\langle\hat{\Phi}^+(\zeta)\hat{K}\hat{\Phi}(\zeta)\rangle_\Gamma\,d\zeta,$$

where $\rangle_\Gamma=\Psi_\Gamma$. Therefore, the construct

$$\rho(\zeta,\zeta'')=\langle\tilde{\Phi}^+(\zeta')\tilde{\Phi}(\zeta)\rangle_\Gamma\qquad(3)$$

plays the part of the density matrix of the electromagnetic field in the medium.

To evaluate (3), it is convenient to use in Γ space the formalism of quantum-mechanical Green's functions in the form proposed by Keldysh[18]

$$\mathscr{D}_{ll'}(\zeta,t,\zeta',t')=-i\langle\hat{T}_c\tilde{\Phi}_l(\zeta,t)\tilde{\Phi}_{l'}{}^+(\zeta',t')\hat{S}\rangle_\Gamma,\qquad(4)$$

where the field operators are taken in the interaction representation and the subscript l describes the time contour that starts from $(l'=1)$ $t\to-\infty$, extends to $t\to\infty$ and returns again $(l=2)$ to $t\to-\infty$, where T_c is the chronological operator on this contour. The operator $\hat{S}$ in (4) has the form

$$\hat{S}=\hat{T}_c\exp\Big\{\sum_l(-1)^{l+1}\frac{ie}{m}\int\tilde{\Phi}_l{}^+\tilde{\psi}_l{}^+\hat{\mathbf{p}}\hat{\mathbf{A}}\tilde{\psi}_l\tilde{\Phi}_l d\zeta\,d\mathbf{r}\,d\mathbf{R}\,dt\Big\},$$

$$\tilde{\psi}=\sum_{ip}\psi_i(\mathbf{r}-\mathbf{R})\frac{\hat{b}_{ip}}{V^{1/2}}\exp(i\mathbf{pR}-i\varepsilon_{ip}t),$$

$$\tilde{\Phi}=\sum_{\mathbf{N}}\hat{\mathfrak{A}}_{\mathbf{N}}\Phi^0(\mathbf{N}|\zeta)\exp[-i\varepsilon(\mathbf{N})t].$$

The average in (4) is evaluated over the initial state before the interaction between the field and the medium is turned on. In the subsequent analysis, we shall assume in (4) that quantum-mechanical averaging and statistical averaging over the ensemble of systems are employed. The interaction between the gas atoms and the reservoir can be explicitly included in (4). It can also be taken into account in the initial state. The influence of the reservoir via the mass operators is seen in the broadening of the atomic energy levels. The sign of the imaginary part of the mass operator is determined by the causality principle.

We shall assume a Gaussian distribution for the atomic ensemble prior to the interaction with radiation. As noted above, this will enable us to use the thermodynamic variant[8,12] of Wick's theorem to simplify the average product of operators $\tilde{\psi}$. The product of operators $\tilde{\Phi}$ can be evaluated exactly by using the algebraic Wick theorem[19] and the obvious fact that $(\tilde{\Phi})^i\rangle_\Gamma\equiv0$ for $i>1$. This identity follows from the completeness of the basis $\hat{\mathfrak{A}}_{\mathbf{N}}^+$ for physical states and is responsible for the linear dependence of the Feyman terms on the normal product of operators $\hat{\mathfrak{A}}_{\mathbf{N}}$ and $\hat{\mathfrak{A}}_{\mathbf{N}}^+$. We emphasize once again the completeness of the basis $\hat{\mathfrak{A}}_{\mathbf{N}}^+\rangle_\Gamma^0$, which excludes N-products containing more than one annihilation operator $\hat{\mathfrak{A}}_{\mathbf{N}}$. It is precisely this point in the derivation of closed equations that enables us to avoid the decoupling of photon-photon correlations.

The Feynman graphs thus split into the sum of two series:

$$\mathscr{D}_{ll'}=\Delta_{ll'}-i\rho_{ll'}$$

The first of these does not contain the normal products of operators $\tilde{\Phi}$; they necessarily appear in each term of the second series, but only with the single operator $\hat{\mathfrak{A}}_{\mathbf{N}}$ and $\hat{\mathfrak{A}}_{\mathbf{N}}^+$. In the standard technique, the Dyson equation for the vacuum averages is[19]

$$\mathscr{D}=\mathscr{D}^0+\mathscr{D}^0\hat{\mathscr{P}}\mathscr{D}.$$

In our formalism, this equation is complicated by the addition to each of the cofactors of terms containing the average of the normal product of operators $\hat{\mathfrak{A}}_{\mathbf{N}}$ and $\hat{\mathfrak{A}}_{\mathbf{N}}^+$, but in such a way that each new term depends linearly on this average.

The structure of the Feynman series is found to be as follows:

$$\Delta_{ll'}=\Delta_{ll'}^0+\sum_{l_1l_2}\Delta_{ll_1}^0(-1)^{l_1+1}\hat{\mathscr{P}}_{l_1l_2}\Delta_{l_2l'},\qquad(5)$$

$$\rho_{ll'}=\rho_{ll'}^{0}+\sum_{l_1l_2}\rho_{ll_1}^{0}(-1)^{l_1+1}\hat{\mathscr{P}}_{l_1l_2}\Delta_{l_2l'}$$
$$+\sum_{l_1l_2}\Delta_{ll_1}^{0}(-1)^{l_1+1}\hat{\mathscr{P}}_{l_1l_2}^{(n)}\Delta_{l_2l'}+\sum_{l_1l_2}\Delta_{ll_1}^{0}(-1)^{l_1+1}\hat{\mathscr{P}}_{l_1l_2}\rho_{l_2l'}. \quad (6)$$

The operator $\hat{\mathscr{P}}_{l_1l_2}$ does not contain normal products of the operators $\tilde{\Phi}$. However, these products are present in each term of the operator $\hat{\mathscr{P}}_{l_1l_2}^{(n)}$. We note that the condition

$$i\Delta_{ll'}^{0}=\langle \bar{T}_c\tilde{\Phi}_l\tilde{\Phi}_{l'}^{+}\rangle_{\Gamma}^{0}$$

leads to

$$\Delta_{12}^{0}=\Delta_{12}=\hat{\mathscr{P}}_{12}=0. \quad (7)$$

To construct the required density matrix ρ, we need only know ρ_{12}, since it is clear from (3), (4), and (7) that $\rho=i\mathscr{D}_{12}$ for $t=t'$. From (6) we find that

$$(1-\Delta_r\hat{\mathscr{P}}_r)\rho_{12}=\rho_{12}^{0}(1+\hat{\mathscr{P}}_a\Delta_a)-\Delta_r^{0}\mathscr{P}_{12}^{(n)}\Delta_a, \quad (8)$$

where, in accordance with Ref. 18 and (7), we have

$$\Delta_r=\Delta_{11},\ \Delta_a=-\Delta_{22}=\Delta_r^{+},\ \hat{\mathscr{P}}_r=\hat{\mathscr{P}}_{11},\ \hat{\mathscr{P}}_a=\hat{\mathscr{P}}_{22}=\hat{\mathscr{P}}_r^{+}.$$

We now apply the operator $1+\Delta_r\hat{\mathscr{P}}_r$, to the left-hand side of (8), and write (5)–(8) in the symmetric form

$$\rho_{12}=\rho_{12}^{(c)}+\rho_{12}^{(n)}, \quad (9)$$

$$\rho_{12}^{(c)}=(1+\Delta_r\hat{\mathscr{P}}_r)\rho_{12}^{0}(1+\hat{\mathscr{P}}_a\Delta_a), \quad (10)$$

$$\rho_{12}^{(n)}=-\Delta_r\mathscr{P}_{12}^{(n)}\Delta_a,\quad \Delta_r=\Delta_r^{0}+\Delta_r^{0}\hat{\mathscr{P}}_r\Delta_r. \quad (11)$$

According to (9), the density matrix of the photon subsystem splits into the sum of two components. The coherent component $\rho_{12}^{(c)}$ describes elastic scattering processes in which atoms of the medium return to their initial (including translational) quantum-mechanical state. The incoherent component $\rho_{12}^{(n)}$ describes processes in which the atoms change their quantum-mechanical state. The latter includes spontaneous emission, Raman scattering, and stimulated emission. In extended media and time-dependent situations, the matrix $\rho_{12}^{(c)}$ describes the initial stage of relaxation, and $\rho_{12}^{(n)}$ the kinetic stage.

The structure of (9) can be elucidated by the following elementary considerations. Let Φ_i represent the wave functions of the different states of the medium, and let us suppose that, prior to its interaction with radiation, the medium was in the state Φ_0. When the interaction is turned on, the wave function of the system can be written in the form

$$\Psi=f_0\varphi_0+\sum_{i\neq 0}f_i\varphi_i.$$

The parameters of the electromagnetic field appear only in the functions f_i. The density matrix of the electromagnetic field that appears as a result of the quantum-mechanical averaging over the parameters of the medium is given by

$$\rho=\mathrm{Sp}'\,\Psi\Psi^{*}=\rho^{(c)}+\rho^{(n)},$$
$$\rho^{(c)}=f_0f_0^{*},\quad \rho^{(n)}=\sum_{i\neq 0}f_if_i^{*}.$$

Expression (9) shows that this structure of the matrix ρ is preserved when the extended medium is considered not in the pure but in the mixed state. If $\hat{s}$ is the operator form of the Poynting vector, then according to (9)

$$\langle\hat{\mathbf{s}}\rangle=\mathrm{Sp}\,\hat{\mathbf{s}}\rho_{12}=\langle\hat{\mathbf{s}}\rangle^{(c)}+\langle\hat{\mathbf{s}}\rangle^{(n)}. \quad (12)$$

Consequently, the coherent and incoherent channels do not interfere.[20] This absence of interference is a consequence of the orthogonality of the wave functions of the medium, the net result of which is that coherent and incoherent scattering occur in different final quantum states. The assignment of elastic scattering and stimulated emission to different reaction channels, leads to a number of differences between the results obtained in quantum theory and in semiclassical theory operating with unquantized electromagnetic fields. We note that the decoupling of photon-photon operators, i.e., the use of the standard Keldysh technique,[18] would automatically place stimulated emission in the coherent channel, and would thus distort the final results.

Let $\hat{A}_\nu$ represent the field amplitude operator. From (9) we then have

$$\langle\hat{A}_\nu\rangle=\mathrm{Sp}\,\hat{A}_\nu\rho_{12}=\langle\hat{A}_\nu\rangle^{(c)}+\langle\hat{A}_\nu\rangle^{(n)}. \quad (13)$$

From the additivity of the amplitudes of different channels we cannot as yet conclude that interference occurs between them. First, the initial state may be a Fock state, in which case $\langle\hat{A}_\nu\rangle=0$. If, prior to scattering, the radiation was in a quantum-mechanically coherent[21] state $|\alpha\rangle$, and the condition

$$|\langle\hat{\mathbf{s}}\rangle|\propto\langle\hat{\mathbf{A}}\rangle^{2}$$

was satisfied for it, then after scattering this proportionality will be violated because (12) and (13) cannot then be simultaneously satisfied.

We therefore conclude that, as a result of scattering, the quasiclassical state $|\alpha\rangle$ ceases to be quasiclassical, provided both coherent and incoherent channels participate in the scattering process. This condition is always satisfied if the medium contains excited atoms that participate in stimulated emission.

3. Let us now consider reflection from a plane-parallel excited medium in greater detail. We confine our attention to the single-loop approximation for the polarization operators:

$$\hat{\mathscr{P}}_r=-\left(\frac{e}{m}\right)^{2}$$
$$\times\int\sum_{\nu_1\nu_2}\hat{p}^{\nu_1}\hat{A}_{\nu_1}G_r(X_1,X_2)\Delta_{11}^{0}\hat{p}^{\nu_2}\hat{A}_{\nu_2}G_{12}(X_2,X_1)\,d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{R}_1\,d\mathbf{R}_2,$$

$$\hat{\mathscr{P}}_{12}^{(n)}=\left(\frac{e}{m}\right)^{2}$$

$$\times\int\sum_{\nu_1\nu_2}\hat{p}^{\nu_1}\hat{A}_{\nu_1}G_{12}(X_1,X_2)\rho_{12}\hat{p}^{\nu_2}\hat{A}_{\nu_2}G_{21}(X_2,X_1)\,d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{R}_1\,d\mathbf{R}_2,$$

which will suffice for $n\ll k_3$. We take n to be the total concentration of scattering atoms, X the set of variables $(\mathbf{r},\mathbf{R},t)$ and $G_{u'}$ the atomic Green's functions $G_r=G_{11}-G_{12}$. Scattering of radiation is, in general, both diffuse and directed (selective) in character.[22]

Despite the fact that, for $n\ll K_3$, the selective component is energetically small,[23] we now proceed to investigate

it. On the one hand, it can be resolved experimentally because it is directional. On the other hand, by studying it, we can demonstrate a number of qualitative properties that also appear when light is scattered by denser objects. Under homogeneous time-independent conditions,

$$G_{ll'}=\sum_{jj'}\psi_j(\mathbf{r}-\mathbf{R})\psi_{j'}^{*}(\mathbf{r}'-\mathbf{R}')\int G_{ll'}^{jj'}(\mathbf{p},E)$$

$$\times\exp[i\mathbf{p}(\mathbf{R}-\mathbf{R}')-iE(t-t')]\frac{dE}{2\pi V},$$

$$\Delta_{11}^{0}=\int\frac{\exp[-iE(t-t')]}{E-\hat{H}_{ph}+i0}\frac{dE}{2\pi}.$$

If the scattering atoms in the medium experience the effects of a reservoir (collisions with electrons, impurities, and so on), then if we take the mass operator $\hat{M}_r$ into account, we obtain

$$G_r^{jj'}(\mathbf{p},E)=\delta_{jj'}(E-\varepsilon_{j\mathbf{p}}+i\gamma_j/2)^{-1}.$$

The real part of the mass operator is omitted and the sign of the imaginary part is uniquely determined by the causality principle. In the absence of reservoir particles, $i\gamma$ is replaced with $i0$. The Keldysh technique[18] then leads to

$$G_{12}=-G_r\hat{M}_{12}G_a.$$

Suppose that the width of the energy spectrum of the reservoir particles is significantly greater than γ. The dependence of M_{12} on E can then be neglected, and we have

$$G_{12}^{jj'}(\mathbf{p},E)=-2\pi i\delta_{\gamma_j}(E-\varepsilon_{j\mathbf{p}})N_{jj'}(\mathbf{p}),$$

$$\delta_\gamma(E)=-[(E+i\gamma/2)^{-1}-(E-i\gamma/2)^{-1}]/2\pi i,$$

where $N_{jj'}(\mathbf{p})$ are the occupation numbers of the atomic states. Thus, if space is homogeneous and the scattering process is time-independent, we obtain the following expression for $\hat{\mathscr{P}}_r$ in the energy representation:

$$\hat{\mathscr{P}}_r(E)$$

$$=\sum_{\mathbf{k}_1\lambda_1\mathbf{k}_2\lambda_2}[\hat{\alpha}_{\mathbf{k}_1\lambda_1}A_r^{\mathbf{k}_1\lambda_1\mathbf{k}_2\lambda_2}(E-\hat{H}_{ph})\hat{\alpha}_{\mathbf{k}_2\lambda_2}^{+}+\hat{\alpha}_{\mathbf{k}_1\lambda_1}^{+}C_r^{\mathbf{k}_1\lambda_1\mathbf{k}_2\lambda_2}(E-\hat{H}_{ph})\hat{\alpha}_{\mathbf{k}_2\lambda_2}],$$

$$A_r^{\mathbf{k}_1\lambda_1\mathbf{k}_2\lambda_2}(E)=\sum_{mm'\mu\mathbf{p}}\frac{p_{m'\mu}^{\lambda_1}(\mathbf{k}_1)p_{m\mu}^{\lambda_2*}(\mathbf{k}_2)}{2V[\omega(\lambda_1)\omega(\lambda_2)]^{1/2}}$$

$$\times\frac{N_{mm'}(\mathbf{p})\delta_{\mathbf{k}_1\mathbf{k}_2}}{E+\omega_{m\mu}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{p}/2M+i\gamma/2},\qquad(14)$$

$$C_r^{\mathbf{k}_1\lambda_1\mathbf{k}_2\lambda_2}(E)=\sum_{m\mu\mu'\mathbf{p}}\frac{p_{m\mu'}^{\lambda_1*}(\mathbf{k}_1)p_{m\mu}^{\lambda_2}(\mathbf{k}_2)}{2V[\omega(\lambda_1)\omega(\lambda_2)]^{1/2}}$$

$$\times\frac{N_{\mu\mu'}(\mathbf{p})\delta_{\mathbf{k}_1\mathbf{k}_2}}{E-\omega_{m\mu}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{p}/2M+i\gamma/2}.\qquad(15)$$

In the two-level approximation, the Zeeman sublevels of the atomic excited states are assigned the index m in these expressions, whereas the unexcited states are assigned the index μ; γ is the energy width of the excited state. Radiative broadening is neglected. In the dipole approximation

$$p_{m\mu}^{\lambda}(\mathbf{k})=\frac{e}{m}\int\psi_m^{*}\mathbf{e}_{\mathbf{k}}^{\lambda}\hat{\mathbf{p}}\psi_\mu\,d\rho,\qquad\rho=\mathbf{r}-\mathbf{R}.$$

The detailed expression for $\hat{\mathscr{P}}_{12}^{(n)}$ will not be reproduced here, and we shall confine ourselves to proving only some of the general properties that will lead us to conclude that the coherent scattering channel provides the lower bound for the Poynting vector in the reflected flux: $|\langle\hat{\mathbf{s}}\rangle|\geqslant|\langle\hat{\mathbf{s}}\rangle^{(c)}|$. The Doppler effect is not taken into account here, especially since it appears in a nontrival manner.[24] The situation is thus assumed to be dominated by collision broadening. If the scattering medium takes the form of a plane-parallel layer of thickness L, then in the Wigner approximation we must make the following replacement[16] in (14)

$$\delta_{\mathbf{k}_1\mathbf{k}_2}\to\delta(k_{1x},k_{2x})\delta(k_{1y},k_{2y})\vartheta_L(k_{2z}-k_{1z}),$$

whereas in (15)

$$\delta_{\mathbf{k}_1\mathbf{k}_2}\to\delta(k_{1x},k_{2x})\delta(k_{1y},k_{2y})\vartheta_L(k_{1z}-k_{2z}),$$

where

$$\vartheta_L(q)=\int_0^L e^{-iqz}\frac{dz}{L_z}$$

It will be assumed below that the scattering medium occupies the half-space $z>0$ and that $2L=L_z$. The Poynting vector of the radiation reflected by the medium is shown by (12) to consist of two components. The coherent component is responsible for the directionally reflected beam. In lowest-order perturbation theory in the concentration of the scattering particles, this component is associated with the expression[16]

$$\langle\mathbf{s}\rangle^{(c)}=\sum_{\lambda=1,2}\mathbf{q}\frac{\sin^2(k_{0z}L)}{4k_{0z}^2V}|c^{\mathbf{q}\lambda\mathbf{k}_0\lambda_0}(k_0,k_0)|^2,$$

$$\mathbf{q}=\{k_{0x},k_{0y},-k_{0z}\}.$$

We assume that the mode $(\mathbf{k}_0,\lambda_0)$ of the radiation incident on the medium contains only one photon, and

$$\rho_{12}^{0}(E)=2\pi\delta(E-k_0)|\mathbf{k}_0\lambda_0\rangle\langle\mathbf{k}_0\lambda_0|.$$

For linear scattering, to which we confine our attention here, this assumption is quite natural. When the distribution of atoms over the Zeeman sublevels is uniform (this is always assumed) and the occupation numbers are $N_m(\mathbf{p})=Vn_m(\mathbf{p})$ and $N_\mu(\mathbf{p})=Vn_\mu(\mathbf{p})$, we have

$$c^{\mathbf{k}\lambda\mathbf{k}_0\lambda_0}(k_1,k_2)=\sum_{m\mu}p_{m\mu}^{\lambda*}(\mathbf{k})p_{m\mu}^{\lambda_0}(\mathbf{k}_0)$$

$$\times\int\left[\frac{n_\mu(\mathbf{p})}{k_1-\omega_{m\mu}+i\gamma/2}+\frac{n_m(\mathbf{p})}{\omega_{m\mu}-k_2+i\gamma/2}\right]\frac{d\mathbf{p}}{(2\pi)^3}.$$

For resonance ($k_0=\omega_{m\mu}$) radiation, we have $|\langle\hat{\mathbf{s}}\rangle^{(c)}|\propto(n_\mu+n_m)^2$, and, if we take the inequality $|\langle\hat{\mathbf{s}}\rangle|\geqslant|\langle\hat{\mathbf{s}}\rangle^{(c)}|$ into account, we find that this is in conflict with the semiclassical theory of radiation which predicts, in particular, that $s=0$ for $n_\mu=n_m$. When $n_m=0$, quantum electrodynamics and semiclassical theory lead to the same result.

It is noted in Ref. 3 that the sum $n_\mu + n_m$ arises quite naturally in the analysis of the decay of a one-photon (Fock) state of the electromagnetic field in a uniform medium. The point is that both the absorption of the photon (determined by n_μ) and the stimulated emission produced by it (i.e., transition to the two-photon state, determined by n_m) lead to the disappearance of the one-photon state. Since both absorption and reflection of light are determined by the same polarization operator, the relation $|\langle\hat{\mathbf{s}}\rangle^{(c)}| \propto (n_\mu + n_m)^2$ becomes understandable.

4. We must now prove that the vector $\langle\hat{\mathbf{s}}\rangle^{(n)}$ is positive-definite. We first rewrite the equation for the propagator Δ_r in the form

$$\Delta_r^{-1}=(\Delta_r^0)^{-1}-\hat{\mathscr{P}}_r. \tag{16}$$

Applying the operators Δ_r^{-1} and Δ_a^{-1} to the left and right hand side of (11), respectively, and recalling that the matrix ρ_{12}^0 describes the system in the absence of interaction, we find, using (16) and (10), that

$$\Delta_r^{-1}\rho_{12}^{(c)}=(\Delta_r^0)^{-1}\rho_{12}^0(1+\hat{\mathscr{P}}_a\Delta_a)=0$$

and

$$\Delta_r^{-1}\rho_{12}\Delta_a^{-1}=-\hat{\mathscr{P}}_{12}^{(n)}, \tag{17}$$
$$\rho_{12}(t,t')=i\mathscr{D}_{12}=\langle\check{\Phi}^+(\xi',t')\check{\Phi}(\xi,t)\rangle_\Gamma,$$

where the operators $\check{\Phi}$ and $\check{\Phi}^+$ are taken in the Heisenberg representation. Hence

$$-\mathscr{P}_{12}^{(n)}=\langle\Delta_r^{-1}\check{\Phi}^+(\xi',t')\check{\Phi}(\xi,t)\Delta_a^{-1}\rangle_\Gamma. \tag{18}$$

The operators $\check{\Phi}$ and Δ_r^{-1} commute, since they act on different arguments. From $(\Delta_r^{-1})^+=\Delta_a^{-1}$ and (18) it follows that the diagonal elements of the operator $\hat{\mathscr{P}}_{12}^{(n)}$ at $t=t'$ are positive-definite. Applying the operators Δ_r and Δ_a to the left and right hand sides of (17), respectively, and recalling (11), we obtain

$$\rho_{12}^{(n)}=\Delta_r\Delta_r^{-1}\rho\Delta_a^{-1}\Delta_a.$$

Arguments similar to those leading to (18) show that the diagonal elements of the matrix $\rho_{12}^{(n)}$ for $t=t'$ are also positive-definite. Hence, according to (9), it follows that, for the diagonal elements,

$$\rho(t,t)\geqslant\rho^{(c)}(t,t).$$

The fact that the vector $\langle\hat{\mathbf{s}}\rangle^{(n)}$, which appears as a result of scattering by the spatially localized system, is positive-definite is proved by an essentially similar argument. Suppose that the medium occupies a finite volume, and reflects the flux of radiation incident upon it. We shall find the concentration of scattered photons at a large distance $\mathbf{r}$ from the system. We shall be interested only in the incoherent channel. We shall assume that the photon-number operator at the point r is

$$\hat{n}_\nu(\mathbf{r})=\frac{1}{V}\sum_{\mathbf{k}\mathbf{k}'\lambda\lambda'}e_{\mathbf{k}'\nu}^{\lambda'}\hat{\alpha}_{\mathbf{k}'\lambda'}e_{\mathbf{k}\nu}^{\lambda}\alpha_{\mathbf{k}\lambda}\exp[i(\mathbf{k}-\mathbf{k}')\mathbf{r}]. \tag{19}$$

Integration of this expression over all space gives the particle-number operator. If we introduce the factor $2V(kk')^{-1/2}$ under the summation sign, we obtain the operator for the excitation of a test atom at the point $\mathbf{r}$.

For a quasihomogeneous configuration of the electromagnetic field in vacuum, the operator (19) readily allows us to evaluate the Poynting vector. It is clear that

$$\langle\hat{n}_\nu(\mathbf{r},t)\rangle^{(n)}=\mathrm{Sp}\,\hat{n}_\nu(\mathbf{r})\rho_{12}^{(n)}(t,t)=-\frac{1}{V}\mathrm{Sp}\sum_{\mathbf{k}\mathbf{k}'\lambda\lambda'}e_{\mathbf{k}\nu}^{\lambda}\hat{\alpha}_{\mathbf{k}\lambda}$$
$$\times\exp(i\mathbf{k}\mathbf{r})\int\exp[-iE'(t-t')]\Delta_r(E')\frac{dE'}{2\pi}\hat{\mathscr{P}}_{12}^{(n)}(t',t'')$$
$$\times\exp[-iE''(t''-t)]\Delta_a(E'')\frac{dE''}{2\pi}e_{\mathbf{k}'\nu}^{\prime}\hat{\alpha}_{\mathbf{k}'\lambda'}^{+}\exp(-i\mathbf{k}'\mathbf{r})\,dt'\,dt''. \tag{20}$$

Using the representation given by (18), we can rewrite (20) in the form

$$\langle\hat{n}_\nu(\mathbf{r},t)\rangle^{(n)}=\langle\check{Y}_\nu(t)\check{Y}_\nu^+(t)\rangle_\Gamma\geqslant 0, \tag{21}$$

where

$$\check{Y}_\nu(t)=\mathrm{Sp}\sum_{\mathbf{k}\lambda}e_{\mathbf{k}\nu}^{\lambda}\hat{\alpha}_{\mathbf{k}\lambda}\exp(i\mathbf{k}\mathbf{r})$$
$$\times\int\exp[-iE'(t-t')]\Delta_r(E')\frac{dE'}{2\pi V^{1/2}}\Delta_r^{-1}\check{\Phi}^+(t')\,dt'.$$

This proves that $\langle\hat{n}_\nu\rangle$ is positive-definite. However, (21) tells us more. Its structure prevents us from using the identity given by (A.3) and, as $r\to\infty$, the following cofactors appear in (21):

$$\frac{1}{r^2}\delta\left(\frac{\mathbf{k}}{|\mathbf{k}|}-\frac{\mathbf{r}}{|\mathbf{r}|}\right)\delta\left(\frac{\mathbf{k}'}{|\mathbf{k}'|}-\frac{\mathbf{r}}{|\mathbf{r}|}\right),$$

where

$$\int\delta\left(\frac{\mathbf{k}}{|\mathbf{k}|}-\frac{\mathbf{r}}{|\mathbf{r}|}\right)d\Omega_{\mathbf{k}}=1.$$

Thus, the concentration of scattered photons decreases asymptotically as r^{-2}, and involves only photons that propagate in the direction of the vector $\mathbf{r}/|\mathbf{r}|$. Thus, while remaining positive-definite at all points, this concentration produces a radiation flux pointing away from the scattering target. The neglect of this flux leads to an underestimate of the total scattered flux, so that the inequality $|\langle\hat{\mathbf{s}}\rangle|\geqslant|\langle\hat{\mathbf{s}}\rangle^{(c)}|$ is valid under all time-dependent conditions. Of course, it would have been sufficient for our purposes to confine ourselves to the proof that the expectation value of the excitation operator in the incoherent channel is positive-definite.

5. As an aid to our understanding of the difference between the predictions of semiclassical and quantum-mechanical theories, which is particularly clear-cut for $n_\mu = n_m$, let us find the quantum-mechanical average of the vector potential in the reflected flux. We shall again assume that the scattering medium consists of thermally excited atoms occupying the half-space $z>0$. Let us suppose that the beam to be scattered is in the coherent quantum state[21]

$$|\alpha\rangle=\exp\left(-\frac{|\alpha|^2}{2}\right)\sum_n\frac{\alpha}{(n!)^{1/2}}|n\rangle$$

so that $|\langle\hat{\mathbf{s}}\rangle^0|\propto\langle\hat{\mathbf{A}}\rangle^2$, where $\langle\hat{\mathbf{s}}\rangle^0$ is the Poynting vector of the incident flux. We shall confine our attention to the case of an infinitesimal field amplitude, and assume that the density matrix for the incident radiation is

$$\rho_{12}^{0}(t_1,t_2)=[1+\alpha_{\mathbf{k}_0\lambda_0}\alpha_{\mathbf{k}_0\lambda_0}\exp(-ik_0t_1)]\,|0\rangle$$
$$\times\langle 0|\,[1+\alpha^{\cdot}_{\mathbf{k}_0\lambda_0}\hat{\alpha}_{\mathbf{k}_0\lambda_0}\exp(ik_0t_2)].$$

Passing now to the classical description, we can say that the incident radiation is characterized by the vector potential

$$\langle\hat{\mathbf{A}}(\mathbf{r},t)\rangle=\mathrm{Sp}\,\rho_{12}^{0}\hat{\mathbf{A}}(\mathbf{r})=\mathbf{e}_{\mathbf{k}_0}^{\lambda_0}(2k_0V)^{-1/2}[\alpha_{\mathbf{k}_0\lambda_0}\exp(i\mathbf{k}_0\mathbf{r}-ik_0t)$$
$$+\alpha^{\cdot}_{\mathbf{k}_0\lambda_0}\exp(-i\mathbf{k}_0\mathbf{r}+ik_0t)],$$
$$\alpha_{\mathbf{k}_0\lambda_0}=\mathrm{Sp}\,\rho_{12}^{0}\hat{\alpha}_{\mathbf{k}_0\lambda_0},\quad t_1=t_2=t.$$

For linear scattering of radiation, for which $\alpha_{\mathbf{k}_0\lambda_0}$, we can confine our attention to the single-photon approximation. To find the vector potential in the reflected flux, i.e., the part of the radiation that is characterized by classical parameters, we write the potential in the form of the sum (13), in which each of the terms is found by analogy with the discussion in Sec. 3. In second-order perturbation theory,

$$\langle\hat{A}_\nu(\mathbf{r},t)\rangle^{(c)}=\frac{1}{4k_{0z}^{2}}\sum_{\lambda m\mu}\frac{e_{\mathbf{q}\nu}^{\lambda}}{(2k_0V)^{1/2}}\exp[i(\mathbf{qr}-k_0t)]$$
$$\times p_{m\mu}^{\lambda*}(\mathbf{q})\,p_{m\mu}^{\lambda_0}(\mathbf{k}_0)$$
$$\times\left(\frac{n_\mu}{k_0-\omega_{m\mu}+i\gamma/2}+\frac{n_m}{\omega_{m\mu}-k_0+i\gamma/2}\right)+\text{h.c.} \tag{22}$$

$$\langle\hat{A}_\nu(\mathbf{r},t)\rangle^{(n)}=\frac{1}{4k_{0z}^{2}}\sum_{\lambda m\mu}\frac{e_{\mathbf{q}\nu}^{\lambda}}{(2k_0V)^{1/2}}\exp[i(\mathbf{qr}-k_0t)]$$
$$\times p_{m\mu}^{\lambda*}(\mathbf{q})\,p_{m\mu}^{\lambda_0}(\mathbf{k}_0)\,n_m$$
$$\times\left(\frac{1}{k_0-\omega_{m\mu}-i\gamma/2}-\frac{1}{k_0-\omega_{m\mu}+i\gamma/2}\right)+\text{h.c.} \tag{23}$$

As before, in the coherent channel for resonance radiation, the reflected field $\langle\hat{A}_\nu\rangle^{(c)}$ is determined by the sum $n_\mu+n_m$ and, in contrast to $\langle\hat{A}_\nu\rangle^{(n)}$, the component $\langle\hat{\mathbf{s}}\rangle^{(n)}$ is not positive-definite. For this reason, it follows from (22) and (23) that

$$\langle\hat{A}_\nu\rangle^{(c)}+\langle\hat{A}_\nu\rangle^{(n)}=\langle\hat{A}_\nu\rangle\propto n_\mu-n_m.$$

We have thus arrived at the result predicted by the semiclassical theory. In particular, when $n_\mu=n_m$, we have $\langle\hat{A}_\nu\rangle=0$ in the reflected flux. Of course, as before, we have $|\langle\hat{\mathbf{s}}|\geqslant|\langle\hat{\mathbf{s}}\rangle^{(c)}|\propto(n_\mu+n_m)^2$, so that the proportionality $|\langle\hat{\mathbf{s}}\rangle|\propto\langle\hat{\mathbf{A}}\rangle^2$ breaks down, which suggests that the quantum state of reflected light for $n_m\neq0$ is not coherent, and the classical description of the reflection process is not admissible.

6. Finally, let us determine why the Poynting vector $\langle\hat{\mathbf{s}}\rangle^{(n)}$ generated by the incoherent channel cannot be calculated by perturbation theory. The point is that one of the processes responsible for producing the reflected light is the coupling of two elementary processes, namely, the elastic scattering of the photon by one of the ground-state atoms of the medium and the subsequent stimulated emission by another (excited) atom. However, unexpectedly, this coupling does not allow an analysis by quantum-electrodynamic perturbation theory.

Let us consider this coupling for two isolated atoms 1 and 2. The second atom is excited and there are no energy widths. Suppose, for simplicity, that the atoms are different and that $\varepsilon_{1m}-\varepsilon_{1\mu}>\varepsilon_{2m}-\varepsilon_{2\mu}=\omega=\omega_0$, where ω_0 and ω are the frequencies of the incident and scattered photons. The quantum-electrodynamic amplitude for the process in the lowest-order perturbation theory is

$$\langle\hat{S}\rangle\sim e^3\frac{\delta(\varepsilon_{2m}-\varepsilon_{2\mu}-\omega)\,\delta(\omega-\omega_0)}{\varepsilon_{1\mu}-\varepsilon_{1m}+\omega+i0}$$

i.e., it is proportional to the product of two δ-functions.

This type of structure is not uncommon in perturbation theory, and usually suggests that two simultaneous, but independent, processes are taking place. In our case, the subdivision into two independent processes cannot be carried out because the second process is due to the first. However, the probability of the process is then proportional to the square of the product of the δ-functions and, in any case, the cross section (the Poynting vector) contains δ^2, i.e., it is mathematically meaningless. The expression can be given a meaning if the δ-function is "smeared out," which requires the summation of an infinite sequence of Feynman diagrams and leads to an expression that is not analytic in the charge.

We have thus shown for the incoherent channel that the reflection coefficient of an excited medium is difficult to evaluate in sixth-order perturbation theory. The situation is not saved by the presence of energy widths. This is confirmed by the direct calculations reported in Ref. 25.

Our proof that $\langle\hat{\mathbf{s}}\rangle^{(n)}$ is positive-definite has enabled us to confine our attention to $\langle\hat{\mathbf{s}}\rangle^{(c)}$. The evaluation of $\langle\hat{\mathbf{s}}\rangle^{(n)}$ is a separate problem.

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APPENDIX

Consider the integral

$$J_\nu(E)=\int\frac{d\mathbf{k}}{(2\pi)^3k}\frac{f(\mathbf{k})\exp(i\mathbf{kr})}{E-k+i0}\sum_{\lambda=1,2}p_{m\mu}^{\lambda}(\mathbf{k})\,e_{\mathbf{k}\nu}^{\lambda}, \tag{A1}$$

where $f(k)$ is an arbitrary but smooth function that does not prevent the existence of the integral. We note, first, that

$$\sum_{\lambda=1,2}p_{m\mu}^{\lambda}(\mathbf{k})\,e_{\mathbf{k}\nu}^{\lambda}=\sum_{\nu'}p_{m\mu}^{\nu'}\left(\delta_{\nu\nu'}-\frac{k_\nu k_{\nu'}}{k^2}\right),$$
$$p_{m\mu}^{\nu}=\frac{e}{m}\int\psi_m^{*}\hat{p}^{\nu}\psi_\mu\,d\rho.$$

Hence

$$J_\nu(E)=\int f(k)\sum_{\nu'}\frac{p_{m\mu}^{\nu'}}{E-k+i0}\left(\delta_{\nu\nu'}+\frac{1}{k^2}\frac{\partial}{\partial r_\nu}\frac{\partial}{\partial r_{\nu'}}\right)e^{i\mathbf{kr}}\frac{k\,dk\,d\Omega_{\mathbf{k}}}{(2\pi)^3}$$
$$=\int_0^\infty\frac{dk}{2\pi^2}\frac{f(k)}{E-k+i0}\sum_{\nu'}p_{m\mu}^{\nu'}\left(\delta_{\nu\nu'}+\frac{1}{k^2}\frac{\partial}{\partial r_\nu}\frac{\partial}{\partial r_{\nu'}}\right)\frac{\sin kr}{r}.$$

We are interested in the limiting case $r\to\infty$. The main contribution to the integral is provided by the region $k\approx E$. For this reason, at the point $k=E$, smoothly varying functions can be taken outside the integral sign. When the re-

maining singular equation is integrated, the limits can be replaced with $(-\infty, \infty)$. The theory of residues then shows that

$$J_\nu(E)=\frac{1}{2\pi r}\sum_{\nu'} f(E)\,p_{21}{}^{\nu'}\left(\delta_{\nu\nu'}+E^{-2}\frac{\partial}{\partial r_\nu}\frac{\partial}{\partial r_{\nu'}}\right)e^{i\mathbf{k}\mathbf{r}}.$$

Differentiating with respect to r and summing over a ν', we obtain the following expression:

$$J_\nu(E)=-\frac{1}{2\pi r}\sum_{\lambda=1,2} f(E)\,p_{m\mu}{}^{\lambda}(\mathbf{n})\,e_{n\nu}{}^{\lambda}e^{i\mathbf{k}\mathbf{r}},\qquad \mathbf{n}=\frac{\mathbf{r}}{|\mathbf{r}|}. \tag{A2}$$

Comparison of (A1) with (A2) enables us to write the following symbolic identity:

$$\frac{e^{i\mathbf{k}\mathbf{r}}}{E-k+i0}\underset{r\to\infty}{\longrightarrow}-\frac{4\pi^2}{kr}e^{i\mathbf{k}\mathbf{r}}\delta(E-k)\,\delta\left(\frac{\mathbf{k}}{|\mathbf{k}|}-\frac{\mathbf{r}}{|\mathbf{r}|}\right).$$

If the state $(\mathbf{k},\lambda)$ contains $N_{\mathbf{k}\lambda}$ photons, and the other states $(\mathbf{k}',\lambda')$ are also occupied, the matrix elements of the operator Δ_r^0 have the form

$$\left(E-kN_{\mathbf{k}\lambda}-\sum_{\mathbf{k}',\lambda'\neq\mathbf{k},\lambda}k'N_{\mathbf{k}'\lambda'}+i0\right)^{-1}$$

and (A2) is replaced with

$$\Delta_r{}^0e^{ikr}\underset{r\to\infty}{\longrightarrow}-\frac{4\pi^2}{kr}\delta\left(\frac{\mathbf{k}}{|\mathbf{k}|}-\frac{\mathbf{r}}{|\mathbf{r}|}\right)\times e^{i\mathbf{k}\mathbf{r}}\delta\left(k-\frac{E}{N_{\mathbf{k}\lambda}}+\frac{1}{N_{\mathbf{k}\lambda}}\sum k'N_{\mathbf{k}'\lambda'}\right). \tag{A3}$$

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