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Transient Effects in Resonance Fluorescence and Spontaneous Emission by a System of Identical Atoms

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A simple theoretical model is formulated which describes dynamical features of the interaction of two-level atoms with a continuum of radiation oscillators. In particular, this paper is concerned with resonance scattering by single atoms in the presence of an external classical field, and spontaneous emission by a system of identical atoms. In the case of resonance scattering, the response of an atom to rapid changes of the amplitude of the external field is studied. The model for spontaneous emission by a system of atoms is based on the assumptions that all atoms have the same resonance frequency, and that the atoms are contained in a volume whose extension is small compared with the wavelength of the emitted radiation. The complete evolution of a system of m atoms is determined by a set of coupled first-order linear differential equations, and explicit solutions are presented for the values $m=1, 2, 3, 4$, and 8. Pair correlations of the atoms and photon-number fluctuations are discussed. An approximation for resonance fluorescence by a system of atoms with a broad distribution of resonance frequencies is included in an Appendix.

I. INTRODUCTION

THE experimental feasibility of stimulating atomic transitions at optical frequencies in a time which is short compared with the lifetime of the excited state for spontaneous emission has been demonstrated by the observation of photon echos[1] and self-induced transparency.[2] In this paper it will be assumed that various pulse shapes for the generation of such fast transitions are available. On the other hand, while photon echos and self-induced transparency can be explained in terms of classical properties of the electromagnetic field, this paper is concerned with phenomena which require a completely quantum-mechanical description of the radiation field.

A simple theoretical model is formulated which predicts transient effects in resonance fluorescence, and describes spontaneous emission by a system of identical atoms. If one considers incoherent scattering by individual atoms or coherent spontaneous emission by several atoms which are contained in a volume whose linear dimensions are small compared with the wavelength corresponding to the resonance frequency of the atoms, the spatial dependence of the radiation field can be ignored.

Each mode of the electromagnetic field is represented by one quantum oscillator. Different quantum oscillators with the same frequency ω_α will be distinguished by a second subscript β. The occupation number states $|n_{\alpha\beta}\rangle$ form the orthonormal set $\langle n_{\alpha'\beta'}|n_{\alpha''\beta''}\rangle=\delta_{\alpha'\alpha''}\delta_{\beta'\beta''}$. Transitions between occupation number states are described by annihilation and creation operators with the well-known properties $a_{\alpha\beta}|n_{\alpha\beta}\rangle=n_{\alpha\beta}{}^{1/2}|n_{\alpha\beta}-1\rangle$ and $a_{\alpha\beta}{}^\dagger|n_{\alpha\beta}\rangle=(n_{\alpha\beta}+1)^{1/2}|n_{\alpha\beta}+1\rangle$, or $a_{\alpha\beta}{}^\dagger a_{\alpha\beta}|n_{\alpha\beta}\rangle =n_{\alpha\beta}|n_{\alpha\beta}\rangle$. These operators obey the commutation relations $[a_{\alpha'\beta'},a_{\alpha''\beta''}{}^\dagger]=\delta_{\alpha'\alpha''}\delta_{\beta'\beta''}$.

For the present purpose it will be assumed that all atoms have the same resonance frequency ω_0 between a ground state with energy E_g and an excited state with energy E_e, such that $E_e-E_g=\hbar\omega_0$. If $|\psi_\mu{}^g\rangle$ denotes the ground state of an atom labeled by the subscript μ, and $|\psi_\mu{}^e\rangle$ is the excited state, normalization and orthogonality require $\langle\psi_\mu{}^g|\psi_\mu{}^g\rangle=\langle\psi_\mu{}^e|\psi_\mu{}^e\rangle=1$, $\langle\psi_\mu{}^g|\psi_\mu{}^e\rangle=0$, while scalar products involving different atoms such as $\langle\psi_\mu{}^g|\psi_\lambda{}^g\rangle$ are zero. In order to describe transitions between ground state and excited state we introduce pairs of Hermitian adjoint operators c_μ and $c_\mu{}^\dagger$ with the properties $c_\mu|\psi_\mu{}^e\rangle=|\psi_\mu{}^g\rangle$, $c_\mu|\psi_\mu{}^g\rangle=0$, $c_\mu{}^\dagger|\psi_\mu{}^e\rangle=0$, $c_\mu{}^\dagger|\psi_\mu{}^g\rangle=|\psi_\mu{}^e\rangle$. Consequently, $c_\mu{}^\dagger c_\mu|\psi_\mu{}^g\rangle=0$ and $c_\mu{}^\dagger c_\mu|\psi_\mu{}^e\rangle=|\psi_\mu{}^e\rangle$. Since the most general state of any atom is a linear superposition of ground state and excited state, the condition $c_\mu c_\mu{}^\dagger+c_\mu{}^\dagger c_\mu=1$ must be satisfied. It is also required that operators representing different atoms commute, for example $[c_\mu,c_\lambda] =[c_\mu,c_\lambda{}^\dagger]=0$ for $\mu\neq\lambda$. The excitation operators $c_\mu{}^\dagger$ and the deexcitation operators c_μ are related to the Pauli spin operators. If $[\sigma_i,\sigma_j]=2i\sigma_k$, $\sigma_i\sigma_j+\sigma_j\sigma_i=2\delta_{ij}$, and $\sigma_i{}^2=1$, then $c^\dagger=\sigma_+=\frac{1}{2}(\sigma_1+i\sigma_2)$ and $c=\sigma_-=\frac{1}{2}(\sigma_1-i\sigma_2)$, so that $c^\dagger c=\sigma_+\sigma_-=\frac{1}{2}(1+\sigma_3)$. All atomic operators c_μ and $c_\mu{}^\dagger$ commute with all oscillator operators $a_{\alpha\beta}$ and $a_{\alpha\beta}{}^\dagger$. Throughout this paper it will be assumed that the wave functions of different atoms do not overlap, so that the atoms are distinguishable.

The most general Hamiltonian that is of interest in

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[1] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Letters 13, 567 (1964).

[2] S. L. McCall and E. L. Hahn, Phys. Rev. Letters 18, 908 (1967).

this context has the form

$$H=\hbar\omega_0\sum_{\mu=1}^{m} c_\mu{}^\dagger c_\mu+\hbar\sum_{\alpha}\omega_\alpha\sum_{\beta=1}^{n_\alpha}(a_{\alpha\beta}{}^\dagger a_{\alpha\beta}+\tfrac{1}{2})$$
$$-\hbar g\sum_{\mu}\sum_{\alpha,\beta}(a_{\alpha\beta}{}^\dagger c_\mu+c_\mu{}^\dagger a_{\alpha\beta})$$
$$-\hbar G(t)\sum_{\mu}(c_\mu{}^\dagger e^{-i\omega_1 t}+c_\mu e^{+i\omega_1 t}).\quad(1.1)$$

At this stage the quantum oscillators are separated into groups with frequencies ω_α. The first and second term in the Hamiltonian represent the energy of the atoms and the energy of the oscillators, respectively. The third term describes the dipole interaction of the quantum oscillators with the two-level atoms.[3] The real coupling constant g represents an average over modes with different directions of propagation and polarization. For reasons of convergence of integrals it will be assumed that the interaction is confined to some frequency range $\omega_0-\gamma\leqslant\omega_\alpha\leqslant\omega_0+\gamma$, where $\gamma\ll\omega_0$. It appears reasonable to assume that atomic transitions are not strongly affected by radiation oscillators whose frequency is far from the resonance frequency of the atoms. Within the framework of a simplified model one may therefore postulate a convenient frequency dependence of the coupling constant or the density of modes without the risk of unrealistic results. In the absence of any interaction the time dependence of the operators in the Heisenberg picture is $c_\mu(t)=c_\mu(t_0)e^{-i\omega_0(t-t_0)}$, $c_\mu{}^\dagger(t)=c_\mu{}^\dagger(t_0)e^{+i\omega_0(t-t_0)}$, $a_{\alpha\beta}(t)=a_{\alpha\beta}(t_0)e^{-i\omega_\alpha(t-t_0)}$, and $a_{\alpha\beta}{}^\dagger(t)=a_{\alpha\beta}{}^\dagger(t_0)e^{+i\omega_\alpha(t-t_0)}$. High-frequency components in the solution of the equations of motion would be produced by additional terms of the form $a_{\alpha\beta}{}^\dagger c_\mu{}^\dagger+c_\mu a_{\alpha\beta}$, which are not included in the Hamiltonian. The last term in the Hamiltonian describes the interaction of the atoms with an external field whose frequency is ω_1 and whose amplitude $G(t)$ may be a slowly varying function of time. The simple form of this interaction term is justified if $\omega_1\approx\omega_0$. Under these conditions we have omitted terms of the form $c_\mu{}^\dagger e^{+i\omega_1 t}+c_\mu e^{-i\omega_1 t}$.

In the Heisenberg picture the operators are time-dependent. Let $q(t)$ stand for any one of the operators such as c_μ or $a_{\alpha\beta}$, then $i\hbar\dot{q}=[q,H]$. In this paper we will frequently use the abbreviation q for $q(t)$, but clearly identify the initial value of q at time t_0 by $q(t_0)$. Since all operators which describe the evolution of the complete system in the Heisenberg picture are generated by the same unitary transformation, it follows that all commutation relations, and identities such as $c_\mu c_\mu{}^\dagger+c_\mu{}^\dagger c_\mu=1$, do not change with time. Inspection of the expectation values in the Schrödinger picture also shows that their physical interpretation in terms of ensemble averages of certain variables remains unchanged. For example, the expectation value of $a_{\alpha\beta}{}^\dagger(t)a_{\alpha\beta}(t)$ defines the occupation number of a specific mode at time t.

We will now derive a solution for $\sum\sum a_{\alpha\beta}$ which will be used in Secs. II and III. Heisenberg's equations of motion for $a_{\alpha\beta}$ and $a_{\alpha\beta}{}^\dagger$ are

$$\dot{a}_{\alpha\beta}+i\omega_\alpha a_{\alpha\beta}=ig\sum c_\mu,\quad(1.2\text{a})$$

$$\dot{a}_{\alpha\beta}{}^\dagger-i\omega_\alpha a_{\alpha\beta}{}^\dagger=-ig\sum c_\mu{}^\dagger.\quad(1.2\text{b})$$

By means of

$$\sum_{\beta=1}^{n_\alpha}a_{\alpha\beta}=A_\alpha e^{-i\omega_0(t-t_0)},\quad(1.3\text{a})$$

$$\sum_{\mu=1}^{m}c_\mu=Ce^{-i\omega_0(t-t_0)},\quad(1.3\text{b})$$

it follows from Eq. (1.2a) that

$$\dot{A}_\alpha+i(\omega_\alpha-\omega_0)A_\alpha=ign_\alpha C,\quad(1.4)$$

where C is a slowly varying operator, since in the absence of an interaction it is constant. The formal solution of this differential equation implies

$$\sum_\alpha A_\alpha(t)=\sum_\alpha e^{-i(\omega_\alpha-\omega_0)(t-t_0)}A_\alpha(t_0)$$
$$+ig\sum_\alpha n_\alpha\int_{t_0}^{t}dt'\,C(t')e^{i(\omega_\alpha-\omega_0)(t'-t)}.\quad(1.5)$$

In the limit

$$\sum n_\alpha\rightarrow\int d\omega_\alpha\,\rho(\omega_\alpha),$$

which involves the density of radiation oscillators per unit frequency range $\rho(\omega_\alpha)$,

$$\sum_\alpha A_\alpha(t)=\sum_\alpha e^{-i(\omega_\alpha-\omega_0)(t-t_0)}A_\alpha(t_0)+igF(t),\quad(1.6)$$

where

$$F(t)=\int_{t_0}^{t}dt'\,C(t')\int_0^\infty d\omega_\alpha\,\rho(\omega_\alpha)e^{i(\omega_\alpha-\omega_0)(t'-t)}.\quad(1.7)$$

If one assumes that the main contribution to $F(t)$ originates in values of t' almost equal to t, and in values of ω_α almost equal to ω_0, then

$$F(t)=\rho(\omega_0)C(t)\int_{-\infty}^{+\infty}d\omega\,e^{-i\omega t}\int_{t_0}^{t}dt'\,e^{i\omega t'}.\quad(1.8)$$

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[3] This type of interaction is well known in quantum electronics. Simple laser models describe the interaction of many atoms with a single mode of the radiation field. Our special form of the interaction term has been used, for example, by E. Abate and H. Haken, Z. Naturforsch. **19a**, 875 (1964). The theory of dissipation or damping, and thermal noise, is based on the interaction of one particular quantum oscillator or a system of atoms with an infinite number of quantum oscillators. See, for example, I. R. Senitzky, Phys. Rev. **119**, 670 (1960); **131**, 2827 (1963); W. H. Louisell and L. R. Walker, Phys. Rev. **137**, B204 (1965); W. H. Louisell, in *Proceedings of the Third International Congress of Quantum Mechanics*, edited by P. Grivet and N. Bloembergen (Columbia University Press, New York, 1964), p. 65; H. Sauermann, Z. Physik **188**, 480 (1965); **189**, 312 (1965); and M. Lax, Phys. Rev. **145**, 110 (1966).

Since

$$\int_{-\infty}^{+\infty} d\omega\, e^{-i\omega t} \int_{t_0}^{t} dt'\, e^{i\omega t'} = \pi \tag{1.9}$$

for $t \neq t_0$, one obtains the approximate solution

$$\sum_{\alpha} A_{\alpha}(t) = \sum_{\alpha} e^{-i(\omega_\alpha - \omega_0)(t-t_0)} A_{\alpha}(t_0) + i\pi g \rho(\omega_0) C(t) \tag{1.10}$$

or, in view of Eqs. (1.3a) and (1.3b), again for $t \neq t_0$,

$$\sum_{\alpha} \sum_{\beta} a_{\alpha\beta} = \sum_{\alpha} e^{-i\omega_\alpha (t-t_0)} \sum_{\beta} a_{\alpha\beta}(t_0) + i\pi g \rho(\omega_0) \sum c_\mu . \tag{1.11}$$

The same result is obtained if we require that the interaction is confined to a frequency range $\omega_0 - \gamma \leqslant \omega_\alpha \leqslant \omega_0 + \gamma$. With the assumption $\gamma \ll \omega_0$ Eq. (1.7) can be replaced by

$$F(t) = \rho(\omega_0) \int_{t_0}^{t} dt'\, C(t') \int_{\omega_0-\gamma}^{\omega_0+\gamma} d\omega_\alpha\, e^{i(\omega_\alpha - \omega_0)(t'-t)} \tag{1.12}$$

or

$$F(t) = \rho(\omega_0) 2 \int_{t_0}^{t} dt'\, C(t') \frac{\sin\gamma(t-t')}{t-t'} . \tag{1.13}$$

We are concerned with a time interval from t_0 to t during which an observable transition and corresponding change of $C(t')$ occurs. If γ is much larger than the transition rate we can choose a time t_1 in the range $t_0 < t_1 < t$ so that $\gamma(t-t_1) \gg 1$ while $C(t')$ remains almost constant between t_1 and t. If $\gamma(t-t_1) \gg 1$ the contribution of $C(t')$ to the integral is negligible in the range $t_0 < t' < t_1$, and we obtain the approximation $F(t) = \pi\rho(\omega_0) C(t)$, which agrees with Eqs. (1.8) and (1.9). For atomic transitions at optical frequencies all the previous assumptions can be satisfied.[4]

Equation (1.11) can be applied immediately to spontaneous emission by one isolated atom. If we drop the subscript μ, it follows from the Hamiltonian in Eq. (1.1) that

$$\dot{c} + i\omega_0 c = ig[c,c^\dagger] \sum \sum a_{\alpha\beta} , \tag{1.14a}$$

$$\dot{c}^\dagger - i\omega_0 c^\dagger = -ig \sum \sum a_{\alpha\beta}^\dagger [c,c^\dagger] . \tag{1.14b}$$

By multiplication of (1.13a) with $c^\dagger$ from the left, multiplication of (1.13b) with c from the right, and addition, we obtain

$$\frac{d}{dt}(c^\dagger c) = ig(c^\dagger \sum \sum a_{\alpha\beta} - \sum \sum a_{\alpha\beta}^\dagger c) \tag{1.15}$$

because $c^\dagger[c,c^\dagger] = c^\dagger$, and $[c,c^\dagger]c = c$. Next, we substitute (1.11) and the corresponding equation for $\sum \sum a_{\alpha\beta}^\dagger$ in (1.15). This leads to

$$\frac{d}{dt}(c^\dagger c) = ig\{c^\dagger \sum e^{-i\omega_\alpha(t-t_0)} \sum a_{\alpha\beta}(t_0) - \sum e^{+i\omega_\alpha(t-t_0)} \sum a_{\alpha\beta}^\dagger(t_0) c\} - 2\pi g^2 \rho(\omega_0)(c^\dagger c) . \tag{1.16}$$

If there are no photons present in the initial state, $a_{\alpha\beta}(t_0)|0\rangle = 0$ and $\langle 0|a_{\alpha\beta}^\dagger(t_0) = 0$, so that[5]

$$\frac{d}{dt}\langle c^\dagger c\rangle = -2\pi g^2 \rho(\omega_0) \langle c^\dagger c\rangle . \tag{1.17}$$

Since $\langle c^\dagger c\rangle$ is the probability $p(t)$ of the atom being in the excited state, (1.17) describes exponential decay $p(t) = p(0) e^{-wt}$, where

$$w = 2\pi g^2 \rho(\omega_0) \tag{1.18}$$

is the transition probability per unit time for spontaneous emission by one isolated atom in the excited state. We regard this result as a strong support of the previous assumptions. It should be pointed out, however, that our model does not describe the frequency shift which is predicted by the Weisskopf–Wigner approximation.[6]

In order to establish the physical meaning of the amplitude $G(t)$ in Eq. (1.1), i.e., for a definition of this quantity in terms of fundamental constants and matrix elements, consider the simplified Hamiltonian

$$H = \hbar\omega_0 \sum_{\mu=1}^{m} c_\mu^\dagger c_\mu - \hbar G \sum (c_\mu^\dagger e^{-i\omega_1 t} + c_\mu e^{i\omega_1 t}) , \tag{1.19}$$

where G is constant. This Hamiltonian describes forced transitions which are very fast compared with spontaneous emission. We note that the equations of motion $\dot{c}_\mu + i\omega_0 c_\mu = iG[c_\mu, c_\mu^\dagger] e^{-i\omega_0 t}$ for the individual atoms are decoupled. Consequently, the external field will not introduce statistical correlations of the atoms. In particular, if the initial state is uncorrelated and defined by the state vector $|\psi_1\rangle|\psi_2\rangle \cdots |\psi_\mu\rangle \cdots |\psi_m\rangle$, then $\langle c_\mu^\dagger c_\mu\rangle = \langle \psi_\mu | c_\mu^\dagger c_\mu | \psi_\mu\rangle$. An exact solution for the more general case of different resonance frequencies is derived in the Appendix. If the atoms are in the ground state at time $t=0$, one obtains for the probability of the excited state $\langle \psi_\mu^g | c_\mu^\dagger(t) c_\mu(t) | \psi_\mu^g\rangle = p(t)$ the well-known resonance formula $p(t) = (4G^2/\Omega^2) \sin^2(\frac{1}{2}\Omega t)$, where

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[4] Following the general method described by G. Källen [in *Encyclopedia of Physics* (Springer-Verlag, Berlin, 1958), Vol. V/1, p. 274], the problem of the interaction of one two-level atom with the electromagnetic field can be solved exactly if the interaction is restricted to a finite frequency range from $\omega_0 - \gamma$ to $\omega_0 + \gamma$. It can be shown that an exponential decrease of the probability of the excited state requires that the decay constant is much smaller than γ. Otherwise, the excited state is stable [Dr. D. Dialetis (private communication)].

[5] Since the density of states $\rho(\omega_0)$ is proportional to the normalization volume V, and $g^2\rho(\omega_0)$ is independent of V, the coupling constant g approaches zero as $V \rightarrow \infty$. Consequently, a finite number of photons in the initial state has no effect in this case. No obvious conclusion can be drawn from Eq. (1.16) if the occupation numbers are different from zero in an infinite number of modes. This is the case for thermal equilibrium radiation. However, unless the temperature is very high, the average number of photons per mode is so small that the effect is negligible for our purpose.

V. F. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930).

$\Omega=[(\omega_1-\omega_0)^2+4G^2]^{1/2}$. Obviously, if in the case of exact resonance $\omega_1=\omega_0$ the interaction is turned off at time $t_0=\pi/G$, each atom is completely excited. In the Schrödinger picture this would correspond to a state vector $|\psi(t_0)\rangle=|\psi_1^e(t_0)\rangle|\psi_2^e(t_0)\rangle\cdots|\psi_\mu^e(t_0)\rangle\cdots|\psi_m^e(t_0)\rangle$ for the complete system. In Sec. III this is the initial state of a system of identical atoms prior to the onset of spontaneous emission.

At the conclusion of this introduction we return to the general Hamiltonian in Eq. (1.1) and the corresponding equations of motion (1.2a) and (1.2b). By multiplication of (1.2a) with $a_{\alpha\beta}^\dagger$ from the left, and multiplication of (1.2b) with $a_{\alpha\beta}$ from the right, we obtain after addition of the equations, and summation,

$$\frac{d}{dt}\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta} =ig\Big(\sum\sum a_{\alpha\beta}^\dagger\sum c_\mu-\sum c_\mu^\dagger\sum\sum a_{\alpha\beta}\Big). \quad (1.20)$$

Substitution of the approximate solution (1.11) and the corresponding expression for $\sum\sum a_{\alpha\beta}^\dagger$ leads to

$$\frac{d}{dt}\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta}=ig\Big\{\sum_\alpha e^{i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}^\dagger(t_0)\sum c_\mu -\sum c_\mu^\dagger\sum_\alpha e^{-i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}(t_0)\Big\} +2\pi g^2\rho(\omega_0)\sum_{\lambda=1}^{m} c_\lambda^\dagger\sum_{\mu=1}^{m} c_\mu. \quad (1.21)$$

The expectation value of the expression in curly brackets vanishes if no photons are present in the initial state. For these initial conditions

$$\frac{d}{dt}\langle\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta}\rangle=w(\langle\sum c_\mu^\dagger c_\mu\rangle+\langle\sum_{\lambda\neq\mu}\sum c_\lambda^\dagger c_\mu\rangle), \quad (1.22)$$

where according to Eq. (1.18) $w=2\pi g^2\rho(\omega_0)$. This result is completely independent of the presence of an external field. For a single atom, Eq. (1.22) reduces to

$$W(t)=wp(t), \quad (1.23)$$

where $p(t)$ is the probability of the atom to be in the excited state, and $W(t)=(d/dt)\langle\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta}\rangle$ is the total rate of emission of photons. Since the maximum value of $p(t)$ is 1, it follows that the emission rate of one isolated atom cannot exceed w, even if the amplitude of the external field is very large. This is an important consequence of our model.

In the absence of an external field, the last term in the Hamiltonian (1.1) can be removed. From the equations of motion for c_μ and $c_\mu^\dagger$ it follows in analogy with Eq. (1.15) that

$$\frac{d}{dt}\sum c_\mu^\dagger c_\mu =ig\Big(\sum c_\mu^\dagger\sum\sum a_{\alpha\beta}-\sum\sum a_{\alpha\beta}^\dagger\sum c_\mu\Big). \quad (1.24)$$

By comparison of (1.24) with (1.20) we see that in the absence of an external field the operators $\sum c_\mu^\dagger c_\mu=M(t)$ and $\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta}=N(t)$ satisfy the conservation law

$$\dot{M}(t)+\dot{N}(t)=0, \quad (1.25)$$

which implies that the sum of the probabilities of excited states, or the total number of excited atoms plus the total number of photons, is constant.

II. RESONANCE FLUORESCENCE

In this section we will apply the basic approximation in Eq. (1.11) to a description of transient effects in resonance scattering. We will not be concerned with the line shape of the scattered radiation which has been the subject of other recent publications.[7] Our approach is consistent with the fact that the spectral resolution is limited by $\Delta\omega\simeq 1/\tau$ if the observation of the transient effects requires a time resolution τ. We consider a system which consists of a single atom with resonance frequency ω_0, a continuum of radiation oscillators, and an external field with frequency ω_1 and amplitude $G(t)$. For this problem, the subscript μ in the Hamiltonian (1.1) can be omitted, and Eq. (1.11) assumes the simpler form

$$\sum\sum a_{\alpha\beta}=\sum_\alpha e^{-i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}(t_0)+i\pi g\rho(\omega_0)c, \quad (2.1a)$$

$$\sum\sum a_{\alpha\beta}^\dagger=\sum_\alpha e^{+i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}^\dagger(t_0)-i\pi g\rho(\omega_0)c^\dagger. \quad (2.1b)$$

Heisenberg's equations of motion for the operators c and $c^\dagger$ are

$$\dot{c}+i\omega_0 c=i[c,c^\dagger]\{g\sum\sum a_{\alpha\beta}+G(t)e^{-i\omega_1 t}\}, \quad (2.2a)$$

$$\dot{c}^\dagger-i\omega_0 c^\dagger=-i\{g\sum\sum a_{\alpha\beta}^\dagger+G(t)e^{i\omega_1 t}\}[c,c^\dagger]. \quad (2.2b)$$

By multiplication of (2.2a) with $c^\dagger$ from the left, and multiplication of (2.2b) with c from the right we obtain

$$\frac{d}{dt}(c^\dagger c)+w(c^\dagger c)=iG(t)(c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}) +ig\Big\{c^\dagger\sum_\alpha e^{-i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}(t_0) -\sum_\alpha e^{i\omega_\alpha(t-t_0)}\sum_\beta a_{\alpha\beta}^\dagger(t_0)c\Big\}, \quad (2.3)$$

where we have introduced the transition rate for spontaneous emission by an excited atom, $w=2\pi g^2\rho(\omega_0)$. It will be assumed again that the initial state is the vacuum state for which $a_{\alpha\beta}(t_0)|0\rangle=0$ and $\langle 0|a_{\alpha\beta}^\dagger(t_0)=0$. For any initial state of the atom the expectation value $\langle c^\dagger c\rangle=p(t)$ is the probability of finding the atom in the excited state at time t. Consequently,

$$\dot{p}(t)+wp(t)=iG(t)\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle. \quad (2.4)$$

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[7] Maurice C. Newstein, Phys. Rev. **167**, 89 (1968).

Differentiating again and using (2.1a), (2.1b), (2.2a), and (2.2b), we find

$$\ddot{p}(t)+w\dot{p}(t)=2G^2\langle[c,c^\dagger]\rangle +i(\dot{G}-\tfrac{1}{2}wG)\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle +G(\omega_1-\omega_0)\langle c^\dagger e^{-i\omega_1 t}+ce^{i\omega_1 t}\rangle . \quad (2.5)$$

By means of Eq. (2.4) and the identity $\langle[c,c^\dagger]\rangle=1-2p$, this can be expressed in the form

$$\ddot{p}(t)+(\tfrac{3}{2}w-\dot{G}/G)\dot{p}(t)+(\tfrac{1}{2}w^2+4G^2-w\dot{G}/G)p(t) =2G^2+G(\omega_1-\omega_0)\langle c^\dagger e^{-i\omega_1 t}+ce^{i\omega_1 t}\rangle . \quad (2.6)$$

At exact resonance $\omega_1=\omega_0$,

$$\ddot{p}(t)+(\tfrac{3}{2}w-\dot{G}/G)\dot{p}(t) +(4G^2+\tfrac{1}{2}w^2-w\dot{G}/G)p(t)=2G^2 . \quad (2.7)$$

We will consider two specializations of this equation which appear to be of practical interest. First, suppose $G(t)$ is zero for $t<0$, and equal to the constant value G for $t>0$. In this case

$$p''(\varphi)+\tfrac{3}{2}p'(\varphi)+(4\gamma+\tfrac{1}{2})p(\varphi)=2\gamma , \quad (2.8)$$

where the primes denote derivatives with respect to the variable $\varphi=wt$, and $\gamma=(G/w)^2$. The solution of this equation for $\gamma>1/64$ is

$$p(\varphi)=\frac{2\gamma}{4\gamma+\frac{1}{2}}\left\{1-e^{-(3/4)\varphi}\left[\cos\tfrac{1}{4}(64\gamma-1)^{1/2}\varphi +\frac{3}{(64\gamma-1)^{1/2}}\sin\tfrac{1}{4}(64\gamma-1)^{1/2}\varphi\right]\right\} . \quad (2.9)$$

For $\gamma\gg1$ this can be approximated by

$$p(\varphi)\cong\tfrac{1}{2}\{1-e^{-(3/4)\varphi}\cos(2(G/w)\varphi)\} . \quad (2.10)$$

We note that $p(\varphi)$ approaches $\frac{1}{2}$ as $t\to\infty$. It may be surprising that the probability of the excited state can approach a constant value in spite of the fact that an external field with a large amplitude continues to act on the atom. This is due to spontaneous emission which changes the relative phase of the dipole moment with respect to the external field in a random way, so that the statistical average of the probability of the atom to be in the excited state eventually becomes independent of time. For $\gamma=1/64$ the solution is

$$p(\varphi)=(1/18)\{1-e^{-(3/4)\varphi}(1-\tfrac{3}{4}\varphi)\} . \quad (2.11)$$

Finally, for $\gamma<1/64$,

$$p(\varphi)=\frac{2\gamma}{4\gamma+\frac{1}{2}}\left\{1+\frac{2}{(1-64\gamma)^{1/2}}e^{-(3/4)\varphi} \times\left[\tfrac{1}{4}(3-(1-64\gamma)^{1/2})\exp(-\tfrac{1}{4}(1-64\gamma)^{1/2}\varphi) -\tfrac{1}{4}(3+(1-64\gamma)^{1/2})\exp(\tfrac{1}{4}(1-64\gamma)^{1/2}\varphi)\right]\right\} . \quad (2.12)$$

Numerical solutions for $\gamma=1$, $\frac{1}{4}$, and $\frac{1}{16}$ are shown in Fig. 1. According to Eq. (1.28) the rate of emission is determined by $W(t)=wp(t)$.

For a second specialization of Eq. (2.7) let us assume that the amplitude $G(t)$ has a time dependence of the form

$$G(t)=K/\cosh(\Omega t) . \quad (2.13)$$

In principle, this pulse shape could be obtained experimentally in an active medium under conditions where spontaneous emission is negligible, i.e., for $w\ll\Omega$, and the frequency could be shifted in a tunable device to the resonance frequency ω_0 of atoms which have a spontaneous emission rate $w\simeq\Omega$. Experimental variations of the ratio K/Ω can be achieved with lenses.

The initial state is now at time $t_0=-\infty$. This requires no changes of the derivations. It is convenient, however, to introduce the new variable $\xi=\tanh(\Omega t)$. In terms of this new variable Eq. (2.7) can be written in the form

$$(1-\xi^2)p''(\xi)+(\tfrac{3}{2}\lambda-\xi)p'(\xi) +[4\sigma^2+(\tfrac{1}{2}\lambda^2+\lambda\xi)/(1-\xi^2)]p(\xi)=2\sigma^2 , \quad (2.14)$$

where $\lambda=w/\Omega$, and $\sigma=K/\Omega$. The initial condition for $p(\xi)$ is $p(-1)=0$. Since $\dot{p}(t)=(1-\xi^2)p'(\xi)$, it follows from Eq. (2.4) that

$$p'(\xi)=-\lambda\frac{p(\xi)}{(1-\xi^2)}+i\sigma\frac{\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle}{(1-\xi^2)^{1/2}} . \quad (2.15)$$

Applying l'Hospital's rule to the first term on the right-hand side, we find

$$(1+\tfrac{1}{2}\lambda)p'(-1)=i\sigma\lim_{\xi=-1}\frac{\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle}{(1-\xi^2)^{1/2}} . \quad (2.16)$$

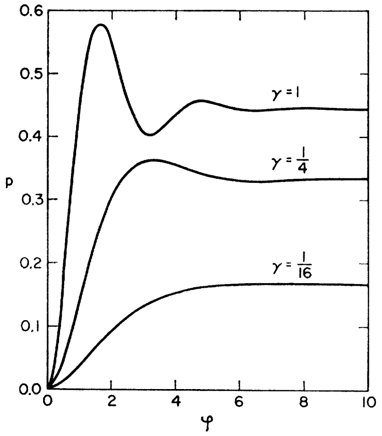

FIG. 1. Probability of the excited state of an atom in the presence of an external field with resonance frequency and constant amplitude as a function of $\varphi=wt$, where w is the transition rate for spontaneous emission in the excited state. The parameter γ is proportional to the square of the amplitude of the external field.

By means of

$$\frac{d}{dt}\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle =-\tfrac{1}{2}w\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle-2iG(t)[1-2p(t)], \quad (2.17)$$

which follows from Eqs. (2.1), (2.2), and $[c,c^\dagger]=1-2c^\dagger c$, and by repeated applications of l'Hospital's rule we obtain

$$(1+\tfrac{1}{2}\lambda)p'(-1)=2\sigma^2-\tfrac{1}{2}\lambda i\sigma \lim_{\xi=-1}\frac{\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle}{(1-\xi^2)^{1/2}}. \quad (2.18)$$

The limit on the right-hand side can be eliminated by means of Eq. (2.16). The result is

$$p'(-1)=2\sigma^2/(1+\tfrac{1}{2}\lambda)^2. \quad (2.19)$$

It should be pointed out that repeated applications of l'Hospital's rule give

$$(1+\tfrac{1}{2}\lambda)p'(-1) =2\sigma^2[1-(\tfrac{1}{2}\lambda)+(\tfrac{1}{2}\lambda)^2-(\tfrac{1}{2}\lambda)^3+\cdots]. \quad (2.20)$$

In view of the restricted convergence of this series, Eq. (2.19) holds only for $\lambda<2$. Numerical solutions of (2.14) also require the second derivative

$$p''(-1)=p'(-1)\frac{2-(\frac{1}{2}\lambda)-(\frac{1}{2}\lambda)^2-8\sigma^2}{6+5(\frac{1}{2}\lambda)+(\frac{1}{2}\lambda)^2}. \quad (2.21)$$

Spontaneous emission is negligible if $\lambda=w/\Omega\ll1$. For $\lambda=0$, Eq. (2.14) is replaced by

$$(1-\xi^2)p''(\xi)-\xi p'(\xi)+\nu^2p(\xi)=\tfrac{1}{2}\nu^2, \quad (2.22)$$

where $\nu=2\sigma$. If ν is an integer, the solution of (2.22) can be expressed in terms of the Chebyshev polynomials $T_\nu(\xi)$:

$$p(\xi)=\tfrac{1}{2}[1+(-1)^{\nu+1}T_\nu(\xi)]. \quad (2.23)$$

This solution satisfies the initial conditions $p(-1)=0$ and $p'(-1)=2\sigma^2=\frac{1}{2}\nu^2$. If ν is not an integer, (2.22) may be solved by the change of variable $\xi=\sin\varphi$. Then

$$d^2p/d\varphi^2+\nu^2p(\varphi)=\tfrac{1}{2}\nu^2. \quad (2.24)$$

The initial conditions are $p(-\frac{1}{2}\pi)=0$ and $p'(-\frac{1}{2}\pi)=0$. The corresponding solution is

$$p(\varphi)=\tfrac{1}{2}[1-\cos\nu(\varphi+\tfrac{1}{2}\pi)]. \quad (2.25)$$

Some plots for various values of ν are shown in Fig. 2. These results are related to those of McCall and Hahn.[2] Their θ_0 is defined by $\theta_0=\pi\nu=2\pi\sigma$.

In general, when λ is different from zero, i.e., if spontaneous emission must be taken into account, Eq. (2.14) must be solved numerically. Some computer results are shown in Fig. 3 for $\nu=2$ and several values of $\lambda=w/\Omega$. The rate of emission is determined by Eq. (1.23).

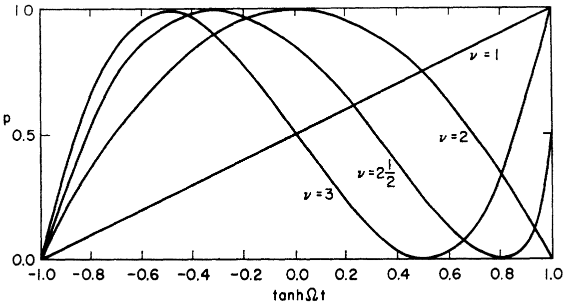

FIG. 2. Probability of the excited state of an atom in the presence of an external field which has the exact resonance frequency and whose amplitude varies as $K/\cosh\Omega t$. The parameter ν is proportional to K/Ω. It is assumed that Ω is much larger than the transition rate for spontaneous emission.

In order to obtain an equation for $p(t)$ when $\omega_1\neq\omega_0$, we differentiate both sides of Eq. (2.6). Since

$$\frac{d}{dt}\langle c^\dagger e^{-i\omega_1 t}+ce^{i\omega_1 t}\rangle=-\tfrac{1}{2}w\langle c^\dagger e^{-i\omega_1 t}+ce^{i\omega_1 t}\rangle -i(\omega_1-\omega_0)\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle, \quad (2.26)$$

it follows that

$$p^{(3)}+(\tfrac{3}{2}w-\dot{G}/G)p^{(2)} +\left[\tfrac{1}{2}w^2+4G^2-w\dot{G}/G-\frac{d}{dt}(\dot{G}/G)\right]p^{(1)} +\left[8\dot{G}G-w\frac{d}{dt}(\dot{G}/G)\right]p =4\dot{G}G-(\tfrac{1}{2}w-\dot{G}/G)G(\omega_1-\omega_0)\langle c^\dagger e^{-i\omega_1 t}+ce^{i\omega_1 t}\rangle -(\omega_1-\omega_0)^2iG\langle c^\dagger e^{-i\omega_1 t}-ce^{i\omega_1 t}\rangle. \quad (2.27)$$

The remaining expectation values are eliminated by means of Eqs. (2.4) and (2.6). The resulting third-order linear and inhomogeneous differential equation is greatly simplified if the amplitude G is constant. In terms of the variable $wt=\varphi$, and the abbreviations

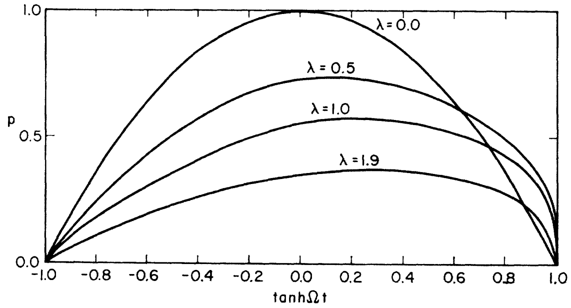

FIG. 3. Probability of the excited state as in Fig. 2, in the special case $\nu=2$, for several values of the parameter $\lambda=w/\Omega$, where w is the transition rate for spontaneous emission in the excited state.

$G^2/w^2=\gamma$, $(\omega_1-\omega_0)^2/w^2=\Delta$, we obtain

$$p'''(\varphi)+2p''(\varphi)+(5/4+4\gamma+\Delta)p'(\varphi)+(\tfrac{1}{4}+2\gamma+\Delta)p(\varphi)=\gamma. \qquad (2.28)$$

In the limit $\varphi\rightarrow\infty$, the solution approaches the equilibrium value

$$p(\infty)=G^2[(\omega_1-\omega_0)^2+2G^2+\tfrac{1}{4}w^2]^{-1}. \qquad (2.29)$$

The corresponding rate of emission or scattering is $wp(\infty)$. This can be compared with the result of time-dependent perturbation theory. Agreement is achieved if in the denominator of Eq. (2.29) the term $2G^2$ is negligible compared with $\frac{1}{4}w^2$. If G^2 is very large compared with $(\omega_1-\omega_0)^2$ and w^2, the probability of the excited state approaches the value $\frac{1}{2}$, so that the final emission rate is $\frac{1}{2}w$.

On the other hand, for $\dot{G}=0$ and $w=0$, Eq. (2.27) implies

$$\ddot{p}(t)+[(\omega_1-\omega_0)^2+4G^2]p(t)=C, \qquad (2.30)$$

where C is a constant of integration. The initial values are $p(0)=0$ and $\dot{p}(0)=0$, according to (2.3). Furthermore, in view of (2.6) the initial value of the second derivative is determined by $\ddot{p}(0)=2G^2$, so that $C=2G^2$. In terms of the abbreviation $[(\omega_1-\omega_0)^2+4G^2]^{1/2}=\Omega$, the solution is $p(t)=(4G^2/\Omega^2)\sin^2(\frac{1}{2}\Omega t)$. This special result has already been mentioned in Sec. I.

III. SPONTANEOUS EMISSION BY A SYSTEM OF IDENTICAL ATOMS

The mutual interaction of a system of atoms through the radiation field has been studied by several investigators. The original concept of coherent spontaneous emission is due to Dicke.[8] In order to explain the onset of laser oscillations, completely quantum-mechanical models were proposed which describe the interaction of many atoms with a single mode or discrete set of modes of the radiation field. Among the first dynamical solutions which are not based on perturbation theory are those obtained by Buley and Cummings.[9] Other investigations by Schwabl and Thirring,[10] and by Ernst and Stehle[11] are concerned with spontaneous emission into a continuum of modes for various geometries of a macroscopic body which contains the radiating atoms.

In this section we consider the interaction of a continuum of radiation oscillators with several identical atoms which are contained in a volume whose linear dimensions are much smaller than the wavelength of the emitted radiation. The objective is a description of the evolution of the system from an initial state in which all atoms are excited and no photons are present. We formulate the problem for an arbitrary number of atoms m, and present explicit solutions for small values of m.

The model is based on the assumption that all atoms are contained in a volume which is so small that the phase of a radiation oscillator with the resonance frequency ω_0 is the same at the position of each atom. We assume that the atoms are distinguishable, i.e., that their wave functions do not overlap. These requirements could be satisfied by several identical atoms in a single molecule, or by clusters of identical atoms in a crystal lattice. The existence of a statistical dependence of different atoms will be demonstrated For example, the probability of a simultaneous observation of two atoms in the excited state is different from the squared probability of one particular atom in the excited state, at the same time.

The Hamiltonian for this problem is obtained from Eq. (1.1) by omission of the last term:

$$H=\hbar\omega_0\sum_{\mu=1}^{m}c_\mu{}^\dagger c_\mu+\hbar\sum_{\alpha}\omega_\alpha\sum_{\beta=1}^{n_\alpha}(a_{\alpha\beta}{}^\dagger a_{\alpha\beta}+\tfrac{1}{2})-\hbar g\sum_{\mu}\sum_{\alpha,\beta}(a_{\alpha\beta}{}^\dagger c_\mu+c_\mu{}^\dagger a_{\alpha\beta}). \qquad (3.1)$$

For the radiation oscillators we adopt the approximate solution (1.11). We consider the following set of operators: $P_1=c_1{}^\dagger c_1$, $P_2=c_1{}^\dagger c_1c_2{}^\dagger c_2$, and, in general, for $n\leqslant m$,

$$P_n=(c_1{}^\dagger c_1)(c_2{}^\dagger c_2)\cdots(c_n{}^\dagger c_n). \qquad (3.2)$$

Also, we define $Q_1=c_1{}^\dagger c_2$, $Q_2=c_1{}^\dagger c_2{}^\dagger c_3c_4$, and, in general, for $2n\leqslant m$,

$$Q_n=(c_1{}^\dagger c_2{}^\dagger\cdots c_n{}^\dagger)(c_{n+1}c_{n+2}\cdots c_{2n}). \qquad (3.3)$$

Finally, we introduce for $j+2k\leqslant m$

$$R_{j,k}=P_j(c_{j+1}{}^\dagger c_{j+2}{}^\dagger\cdots c_{j+k}{}^\dagger)(c_{j+k+1}c_{j+k+2}\cdots c_{j+2k}). \qquad (3.4)$$

This definition of $R_{j,k}$ implies $R_{0,k}=Q_k$ and $R_{j,0}=P_j$. By means of the equations of motion for these operators one can derive a system of coupled first-order linear differential equations for the expectation values $\langle\psi(0)|P_n(t)|\psi(0)\rangle=p_n(t)$, $\langle\psi(0)|Q_n(t)|\psi(0)\rangle=q_n(t)$, and $\langle\psi(0)|R_{j,k}(t)|\psi(0)\rangle=r_{j,k}(t)$, where the initial state

$$|\psi(0)\rangle=\prod_{\alpha,\beta,\mu}|0\rangle_{\alpha\beta}|\psi_\mu{}^e\rangle \qquad (3.5)$$

is a product of the ground-state vectors $|0\rangle_{\alpha\beta}$ of the radiation oscillators and the excited-state vectors $|\psi_\mu{}^e\rangle$ of the atoms. The procedure is based on the fact that the atoms which are represented by the operators can be relabeled without changing the expectation values. Only the expectation values $p_n(t)$ have a simple physical meaning: They determine the probability of a simultaneous observation of n selected atoms in the excited state, irrespective of the state of the other atoms and the radiation oscillators. In particular, $p_1(t)$ is the probability of observing one atom in the excited state,

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[8] R. H. Dicke, Phys. Rev. 93, 99 (1954).

[9] E. R. Buley and F. W. Cummings, Phys. Rev. 134, A1454 (1964).

[10] F. Schwabl and W. Thirring, Ergeb. Exakt. Naturw. 36, 219 (1964).

[11] V. Ernst and P. Stehle, Phys. Rev. 176, 1456 (1968).

provided no observation is made of the state of other atoms and the radiation field. The remaining functions $q_n(t)$ and $r_{j,k}(t)$ are required to determine $p_1(t)$.

The total emission rate follows from the conservation law (1.25):

$$W_m(t)=-\frac{d}{dt}\sum_{\mu=1}^{m}\langle\psi(0)|c_\mu{}^\dagger c_\mu|\psi(0)\rangle, \tag{3.6}$$

or, in view of the fact that all atoms are equivalent,

$$W_m(t)=-m\dot{p}_1(t). \tag{3.7}$$

On the other hand, according to Eq. (1.22),

$$W_m(t)=w[mp_1(t)+m(m-1)q_1(t)]. \tag{3.8}$$

It will be shown that (3.7) and (3.8) are equivalent.

As a consequence of the equations of motion $i\hbar\dot{P}_n=[P_n,H]$,

$$\dot{P}_n(t)=ig\{\sum_{\nu=1}^{n}[P_n,c_\nu{}^\dagger]\sum_{\alpha,\beta}a_{\alpha\beta}+\sum_{\alpha,\beta}a_{\alpha\beta}{}^\dagger\sum_{\nu=1}^{n}[P_n,c_\nu]\}, \tag{3.9}$$

where we have used the fact that P_n commutes with c_μ and $c_\mu{}^\dagger$ for $\mu>n$. From the expectation values of $\sum\sum a_{\alpha\beta}$ and $\sum\sum a_{\alpha\beta}{}^\dagger$ from Eq. (1.11) for an initial state with no photons present, it follows that

$$\dot{p}_n(t)=-\pi g^2\rho(\omega_0)$$

$$\times\langle\sum_{\nu=1}^{n}[P_n,c_\nu{}^\dagger]\sum_{\mu=1}^{m}c_\mu-\sum_{\mu=1}^{m}c_\mu{}^\dagger\sum_{\nu=1}^{n}[P_n,c_\nu]\rangle. \tag{3.10}$$

First, we recall that $2\pi g^2\rho(\omega_0)=w$ is the transition probability per unit time for spontaneous emission by one isolated atom in the excited state. We again use the variable $wt=\varphi$ and denote the derivative with respect to φ by a prime. Next we use the identity

$$[P_n,c_\nu{}^\dagger]c_\mu-c_\mu{}^\dagger[P_n,c_\nu]=(c_1{}^\dagger c_1)\cdots c_\nu{}^\dagger\cdots(c_n{}^\dagger c_n)c_\mu$$
$$+c_\mu{}^\dagger(c_1{}^\dagger c_1)\cdots c_\nu\cdots(c_n{}^\dagger c_n) \tag{3.11}$$

for $\mu=\nu$ or $\mu>n$, and zero otherwise. Since the double sums in (3.10) have n terms with $\mu=\nu$ and $n(m-n)$ terms with $\mu>n$, we obtain

$$p_n'(\varphi)+np_n(\varphi)=-n(m-n)r_{n-1,1}(\varphi). \tag{3.12}$$

Here we have used the fact that the expectation value of a product of n different factors $c_\lambda{}^\dagger c_\lambda$ is p_n, and that the expectation value of a product of $n-1$ different factors $c_\lambda{}^\dagger c_\lambda$ with an additional factor $c_\mu{}^\dagger c_\nu$ is $r_{n-1,1}$, provided $\mu\neq\nu$, and no λ is equal to μ or ν. We note that for $n=m$ Eq. (3.12) implies $p_m'(\varphi)=-mp_m(\varphi)$. For the initial value $p(0)=1$, the solution of this equation is $p_m(\varphi)=e^{-m\varphi}$, so that the probability of an observation of all atoms in the excited state decreases exponentially. In general, the solution of (3.12) requires additional sets of equations for the functions $r_{j,k}$ and q_n.

Consider the equations of motion of the operators Q_n which are defined in Eq. (3.2). The commutator of Q_n with the first term in the Hamiltonian is zero, as can be seen by inspection of the equation

$$\sum_{\mu=1}^{m}[Q_n,c_\mu{}^\dagger c_\mu]=-\sum_{\nu=1}^{n}(c_1{}^\dagger\cdots c_\nu{}^\dagger\cdots c_n)(c_{n+1}\cdots c_{2n})$$
$$+(c_1{}^\dagger\cdots c_n{}^\dagger)\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda\cdots c_{2n}). \tag{3.13}$$

Consequently,

$$\dot{Q}_n(t)=ig\{\sum_{\lambda=1}^{2n}[Q_n,c_\lambda{}^\dagger]\sum_\alpha\sum_\beta a_{\alpha\beta}$$
$$+\sum_\alpha\sum_\beta a_{\alpha\beta}{}^\dagger\sum_{\lambda=1}^{2n}[Q_n,c_\lambda]\} \tag{3.14}$$

or

$$\dot{Q}_n(t)=ig\{(c_1{}^\dagger\cdots c_n{}^\dagger)$$
$$\times\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots[c_\lambda,c_\lambda{}^\dagger]\cdots c_{2n})\sum\sum a_{\alpha\beta}$$
$$-\sum\sum a_{\alpha\beta}{}^\dagger\sum_{\nu=1}^{n}(c_1{}^\dagger\cdots[c_\nu,c_\nu{}^\dagger]\cdots c_n{}^\dagger)$$
$$\times(c_{n+1}\cdots c_{2n})\}. \tag{3.15}$$

By means of Eq. (1.11) for $\sum\sum a_{\alpha\beta}$ and the corresponding equation for $\sum\sum a_{\alpha\beta}{}^\dagger$ we obtain for an initial state with no photons

$$q_n'(\varphi)=-\tfrac{1}{2}\langle(c_1{}^\dagger\cdots c_n{}^\dagger)$$
$$\times\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots[c_\lambda,c_\lambda{}^\dagger]\cdots c_{2n})\sum_{\mu=1}^{m}c_\mu$$
$$+\sum_{\mu=1}^{m}c_\mu{}^\dagger\sum_{\nu=1}^{n}(c_1{}^\dagger\cdots[c_\nu,c_\nu{}^\dagger]\cdots c_n{}^\dagger)$$
$$\times(c_{n+1}\cdots c_{2n})\rangle. \tag{3.16}$$

In view of $c_\mu{}^\dagger c_\mu{}^\dagger=c_\mu c_\mu=0$ and $[c_\mu,c_\mu{}^\dagger]=1-2c_\mu{}^\dagger c_\mu$, Eq. (3.16) can be written in the form

$$q_n'(\varphi)+nq_n(\varphi)=-\tfrac{1}{2}\langle\sum_{\nu=1}^{n}(c_1{}^\dagger\cdots c_\nu{}^\dagger c_\nu\cdots c_n{}^\dagger)\sum_{\lambda=n+1}^{2n}\{c_{n+1}\cdots(1-2c_\lambda{}^\dagger c_\lambda)\cdots c_{2n}\}$$
$$+(c_1{}^\dagger\cdots c_n{}^\dagger)\sum_{\lambda=n+1}^{2n}\{c_{n+1}\cdots(1-2c_\lambda{}^\dagger c_\lambda)\cdots c_{2n}\}\sum_{\mu=2n+1}^{m}c_\mu$$
$$+\sum_{\nu=1}^{n}\{c_1{}^\dagger\cdots(1-2c_\nu{}^\dagger c_\nu)\cdots c_n{}^\dagger\}\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda{}^\dagger c_\lambda\cdots c_{2n})+\sum_{\nu=1}^{n}\{c_1{}^\dagger\cdots(1-2c_\nu{}^\dagger c_\nu)\cdots c_n{}^\dagger\}(c_{n+1}\cdots c_{2n})\rangle. \tag{3.17}$$

Since expectation values with the same number of factors c_μ and $c_\nu^\dagger$ representing different atoms are equal, it follows that

$$\begin{aligned} q_n'(\varphi)+n(m+1-2n)q_n(\varphi)=-\tfrac{1}{2}\langle \sum_{\nu=1}^{n}(c_1^\dagger\cdots c_\nu^\dagger c_\nu\cdots c_n^\dagger)\sum_{\lambda=n+1}^{2n}\{c_{n+1}\cdots(1-2c_\lambda^\dagger c_\lambda)\cdots c_n\} \\ -2(c_1^\dagger\cdots c_n^\dagger)\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda^\dagger c_\lambda\cdots c_{2n})\sum_{\mu=2n+1}^{m}c_\mu-2\sum_{\mu=2n+1}^{m}c_\mu^\dagger\sum_{\nu=1}^{n}(c_1^\dagger\cdots c_\nu^\dagger c_\nu\cdots c_n^\dagger)(c_{n+1}\cdots c_{2n}) \\ +\sum_{\nu=1}^{n}\{c_1^\dagger\cdots(1-2c_\nu^\dagger c_\nu)\cdots c_n^\dagger\}\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda^\dagger c_\lambda\cdots c_{2n})\rangle. \quad (3.18) \end{aligned}$$

By evaluating the expectation values of those terms which contain the factor 1, and using the definition (3.4), we obtain

$$\begin{aligned} q_n'(\varphi)+n(m+1-2n)q_n(\varphi)=-n^2r_{1,n-1}(\varphi)+\langle(c_1^\dagger\cdots c_n^\dagger)\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda^\dagger c_\lambda\cdots c_{2n})\sum_{\mu=2n+1}^{m}c_\mu \\ +2\sum_{\nu=1}^{n}(c_1^\dagger\cdots c_\nu^\dagger c_\nu\cdots c_n^\dagger)\sum_{\lambda=n+1}^{2n}(c_{n+1}\cdots c_\lambda^\dagger c_\lambda\cdots c_{2n})+\sum_{\mu=2n+1}^{m}c_\mu^\dagger\sum_{\nu=1}^{n}(c_1^\dagger\cdots c_\nu^\dagger c_\nu\cdots c_n^\dagger)(c_{n+1}\cdots c_{2n})\rangle. \quad (3.19) \end{aligned}$$

The remaining expectation values can be expressed in terms of $r_{1,k}$ and $r_{2,n-1}$. The final result is

$$\begin{aligned} q_n'(\varphi)+n(m+1-2n)q_n(\varphi)=2n^2r_{2,n-1}(\varphi) \\ +2n(m-2n)r_{1,n}(\varphi)-n^2r_{1,n-1}(\varphi). \quad (3.20) \end{aligned}$$

An application of the same method to the equations of motion of the operators $R_{j,k}$ leads after lengthy manipulations to the differential equation

$$\begin{aligned} r_{j,k}'(\varphi)+[j+k(m+1-2k)]r_{j,k}(\varphi) \\ =-k^2[r_{j+1,k-1}(\varphi)-2r_{j+2,k-1}(\varphi)] \\ +(m-j-2k)[2kr_{j+1,k}(\varphi)-jr_{j-1,k+1}(\varphi)]. \quad (3.21) \end{aligned}$$

The set of Eqs. (3.12), (3.20), and (3.21) is sufficient for the determination of $p_1(\varphi)$. It should be pointed out that (3.12) is defined for $1\leqslant n\leqslant m$. Equation (3.20) holds for $2\leqslant 2n\leqslant m$, and in Eq. (3.21) $1\leqslant j$, $1\leqslant k$ subject to the condition $j+2k\leqslant m$. Furthermore, we recall that $r_{0,k}=q_k$ and $r_{j,0}=p_j$. The total number of equations is $m+\frac{1}{4}(m^2-1)$ if m is an odd number, and $m+\frac{1}{4}m^2$ if m is even.

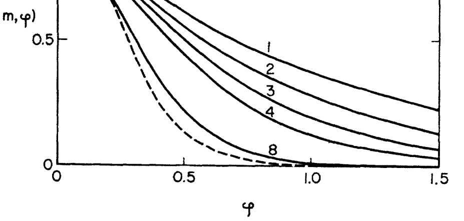

FIG. 4. Probability $p(m,\varphi)$ of the observation of one particular atom in the excited state as a function of the variable $\varphi=wt$, where w is the transition rate for spontaneous emission by one isolated atom in the excited state, for several values of the number of atoms m. The total emission rate is $W_m(t)=-mw(d/d\varphi)p(m,\varphi)$. The dashed curve corresponds to the approximation in Eq. (3.28) for $m=8$.

The initial values for completely excited atoms are $p_n(0)=1$, $q_n(0)=0$, and $r_{j,k}(0)=0$. Explicit solutions have been obtained for $m=1, 2, 3, 4$, and 8. In order to distinguish between different values of the number of atoms m, let $p_1(\varphi)=p(m,\varphi)$ denote the probability of the observation of one particular atom in the excited state. The corresponding solutions for $m=1$ to 4 and $m=8$, are

$$p(1,\varphi)=e^{-\varphi}, \quad (3.22)$$

$$p(2,\varphi)=e^{-2\varphi}(\varphi+1), \quad (3.23)$$

$$p(3,\varphi)=e^{-3\varphi}(4\varphi-1+2e^{-\varphi}), \quad (3.24)$$

$$p(4,\varphi)=e^{-4\varphi}[9\varphi-\tfrac{7}{2}+(3\varphi+\tfrac{9}{2})e^{-2\varphi}], \quad (3.25)$$

$$\begin{aligned} p(8,\varphi)=e^{-8\varphi}[(49\varphi-323/20) \\ +(175\varphi-147/4)e^{-6\varphi} \\ +(189\varphi-147/20)e^{-10\varphi} \\ +(35\varphi+245/4)e^{-12\varphi}]. \quad (3.26) \end{aligned}$$

These results are shown in Fig. 4. The initial slope $p'(m,0)=-1$ is independent of m, and $p''(m,0)=2-m$.

It follows from Eqs. (3.12) and (3.20) that $q_1'(\varphi)+p_2'(\varphi)=p_1'(\varphi)$, or $q_1(\varphi)=p_1(\varphi)-p_2(\varphi)$. If this expression for $q_1(\varphi)$ is substituted in Eq. (3.12) with $n=1$, i.e., $p_1'(\varphi)+p_1(\varphi)=-(m-1)r_{0,1}(\varphi)=-(m-1)q_1(\varphi)$, one obtains

$$p_1'(\varphi)+mp_1(\varphi)=(m-1)p_2(\varphi). \quad (3.27)$$

This result is exact. It should be noted, however, that $p_2(\varphi)\neq p_1^2(\varphi)$. For example, in the case of $m=2$, one finds $p_2(\varphi)=e^{-2\varphi}=1-2\varphi+\cdots$, while $p_1^2(\varphi)=e^{-4\varphi}(1+\varphi)^2=(1+\varphi)^2=1-2\varphi+\cdots$. If the probability correlations are ignored and $p_2(\varphi)$ is replaced by $p_1^2(\varphi)$, the corresponding solution of Eq. (3.27) is

$$p(m,\varphi)=[1+m^{-1}(e^{m\varphi}-1)]^{-1}. \qquad (3.28)$$

In Fig. 4 this approximation is shown for $m=8$ by a dashed curve. For any value of $m>1$ the exact solution decreases more slowly than the approximate solution (3.28). An inspection of Eq. (3.28) shows, however, that the derivatives $p'(m,0)$ and $p''(m,0)$ are correct.

It has been pointed out that Eqs. (3.7) and (3.8) for the total emission rate are equivalent. This statement can now be confirmed. By means of the identity $q_1(\varphi)=p_1(\varphi)-p_2(\varphi)$, Eq. (3.8) can be written in the form $W_m(t)=wm[mp_1-(m-1)p_2]$, or in view of (3.27) $W_m(t)=-m\dot{p}_1(t)$, in agreement with Eq. (3.7).

If the atoms are initially in a superposition of ground state and excited state, the differential equations (3.12), (3.20), and (3.21) are still valid, provided the state vector of the complete system is a direct product of the state vectors of the individual atoms, and the amplitudes of the ground state and the excited state are the same for all atoms. This condition implies that the atoms are initially uncorrelated and that the dipole moments of all atoms are in phase and have the same magnitude. The initial values of p_n, q_n, and $r_{j,k}$ can be expressed in terms of $p_1(0)$. In general, $p_n(0)=p_1^n(0)$, $q_n(0)=p_1^n(0)[1-p_1(0)]^n$, and $r_{j,k}(0)=p_1^{j+k}(0)\times[1-p_1(0)]^k$. From Eq. (3.27) it follows that $p_1'(0)=-p_1(0)-(m-1)p_1(0)[1-p_1(0)]$. The preparation of initially uncorrelated states by means of a strong external field has already been discussed in Sec. I.

The original requirement that all atoms have exactly the same resonance frequency can be relaxed. If Γ is the actual spread of resonance frequencies, and τ is the time scale for return of the atoms to the ground state, the condition $\Gamma\tau\ll 1$ will ensure that the dipole moments remain in phase. For a rough estimate, we may use the approximation (3.28) which predicts for $p(\tau)=\frac{1}{2}$ the value $\tau=\ln(m+1)/wm$.

The following remarks are concerned with photon-number fluctuations. The operator which represents the total number of photons at time t is $N(t)=\sum\sum a_{\alpha\beta}^\dagger a_{\alpha\beta}$. According to Eq. (1.25), $N(t)=M(0)+N(0)-M(t)$, where $M(t)=\sum c_\mu^\dagger c_\mu$ represents the total number of excited atoms. Let us assume that in the initial state all atoms are excited and no photons are present, so that $\langle M(0)\rangle=m$ and $\langle N(0)\rangle=0$. Then, $\langle N^2(t)\rangle-\langle N(t)\rangle^2=\langle M^2(t)\rangle-\langle M(t)\rangle^2$. Since $\langle M(t)\rangle=mp_1(t)$ and $\langle M^2(t)\rangle=mp_1(t)+m(m-1)p_2(t)$, we conclude that

$$\langle N^2(t)\rangle-\langle N(t)\rangle^2 \\ =m^2[p_2(t)-p_1^2(t)]+m[p_1(t)-p_2(t)]. \qquad (3.29)$$

Obviously, the mean-square fluctuation of the photon number is zero at time $t=0$, and approaches zero as $t\to\infty$. In the special case of a single atom $\langle N^2(t)\rangle-\langle N(t)\rangle^2=e^{-wt}(1-e^{-wt})$. In the general case $m\neq 1$, one can eliminate $m(m-1)p_2(t)$ from Eq. (3.29) by means of Eq. (3.27).

Our theoretical model is based on the assumption that m identical atoms are contained in a volume whose extension is much smaller than the wavelength of the emitted radiation. In a real experiment it will be difficult to observe the emission from a single system or "cluster" of m atoms in a sufficiently small volume. It may be possible, however, to study the emission from a macroscopic sample which contains a large number of similar systems, for example, pairs of atoms, under conditions where the distance between the clusters is very large compared with the wavelength. It is only required that the clusters do not form a regular lattice with spacings exactly equal to a small integer number of wavelengths. If these conditions can be realized, it should be expected that Z identical clusters will contribute to the total emission rate $W(t)=ZW_m(t)$. Assuming statistical independence of the emission from different clusters, the mean-square fluctuation of the total number of photons emitted in the time interval from zero to t, i.e., the total number of photons present at time t is Z times the expression in Eq. (3.29).

APPENDIX

It is instructive to derive an approximate solution for scattering by a very large number of atoms with a broad distribution of resonance frequencies. In contrast with the methods described in Secs. II and III, the reaction of the radiation oscillators on the atoms will now be neglected. For this purpose we consider the Hamiltonian

$$H=\hbar\sum_{\lambda}\omega_\lambda\sum_{\mu=1}^{m_\lambda}c_{\lambda\mu}^\dagger c_{\lambda\mu}+\hbar\sum_{\alpha}\omega_\alpha\sum_{\beta=1}^{n_\alpha}(a_{\alpha\beta}^\dagger a_{\alpha\beta}+\tfrac{1}{2}) \\ -\hbar g\sum_{\alpha,\beta}\sum_{\lambda,\mu}(a_{\alpha\beta}^\dagger c_{\lambda\mu}+c_{\lambda\mu}^\dagger a_{\alpha\beta}) \\ -\hbar G\sum_{\lambda,\mu}(c_{\lambda\mu}^\dagger e^{-i\omega_1 t}+c_{\lambda\mu}e^{+i\omega_1 t}), \qquad (A1)$$

which represents two-level atoms with various resonance frequencies ω_λ in the presence of an external field with frequency ω_1 and constant amplitude G. The simple form of this Hamiltonian is justified if for all resonance frequencies $(\omega_\lambda-\omega_1)/\omega_1\ll 1$ and if the corresponding wavelengths are very large compared with the dimensions of the volume which contains all atoms.

The equations of motion of the operators $a_{\alpha\beta}$ and $c_{\lambda\mu}$ are

$$\dot{a}_{\alpha\beta}+i\omega_\alpha a_{\alpha\beta}=ig\sum_{\lambda,\mu}c_{\lambda\mu}, \qquad (A2)$$

$$\dot{c}_{\lambda\mu}+i\omega_\lambda c_{\lambda\mu}=i[c_{\lambda\mu},c_{\lambda\mu}^\dagger]\{g\sum_{\alpha,\beta}a_{\alpha\beta}+Ge^{-i\omega_1 t}\}. \qquad (A3)$$

From (A2) and the corresponding equation for $a_{\alpha\beta}^\dagger$ it follows that

$$\frac{d}{dt}\sum_{\alpha,\beta}(a_{\alpha\beta}{}^{\dagger}a_{\alpha\beta})$$

$$=ig\{\sum_{\alpha,\beta}a_{\alpha\beta}{}^{\dagger}\sum_{\lambda,\mu}c_{\lambda\mu}-\sum_{\lambda,\mu}c_{\lambda\mu}{}^{\dagger}\sum_{\alpha,\beta}a_{\alpha\beta}\}. \quad \text{(A4)}$$

In analogy with Eq. (1.11),

$$\sum_{\alpha,\beta}a_{\alpha\beta}=\sum_{\alpha}e^{-i\omega_{\alpha}t}\sum_{\beta}a_{\alpha\beta}(0)+i\pi g\rho(\omega_0)\sum_{\lambda,\mu}c_{\lambda\mu}, \quad \text{(A5)}$$

provided the density of states $\rho(\omega)$ is essentially constant over the whole range of resonance frequencies, and all resonance frequencies are approximately equal to ω_0. If no photons are present in the initial state

$$\frac{d}{dt}\langle\sum_{\alpha,\beta}a_{\alpha\beta}{}^{\dagger}a_{\alpha\beta}\rangle=2\pi g^2\rho(\omega_0)\langle\sum_{\lambda,\mu}c_{\lambda\mu}{}^{\dagger}\sum_{\sigma,\nu}c_{\sigma\nu}\rangle. \quad \text{(A6)}$$

For a large amplitude G one can solve (A3) with $g=0$ and substitute the approximate solutions for $c_{\lambda\mu}{}^{\dagger}$ and $c_{\sigma\nu}$ in (A6). Radiation damping is thereby neglected. For $g=0$ the equations of motion are

$$\dot{c}_{\lambda\mu}+i\omega_{\lambda}c_{\lambda\mu}=iG[c_{\lambda\mu},c_{\lambda\mu}{}^{\dagger}]e^{-i\omega_1 t}; \quad \text{(A7)}$$

by means of $c_{\lambda\mu}(t)=q_{\lambda\mu}(t)e^{-i\omega_1 t}$ one obtains

$$\dot{q}_{\lambda\mu}+i(\omega_{\lambda}-\omega_1)q_{\lambda\mu}=iG[q_{\lambda\mu},q_{\lambda\mu}{}^{\dagger}], \quad \text{(A8a)}$$

$$\dot{q}_{\lambda\mu}{}^{\dagger}-i(\omega_{\lambda}-\omega_1)q_{\lambda\mu}{}^{\dagger}=-iG[q_{\lambda\mu},q_{\lambda\mu}{}^{\dagger}], \quad \text{(A8b)}$$

and it follows immediately that

$$\ddot{q}_{\lambda\mu}+i(\omega_{\lambda}-\omega_1)\dot{q}_{\lambda\mu}=2G^2(q_{\lambda\mu}{}^{\dagger}-q_{\lambda\mu}), \quad \text{(A9a)}$$

$$\ddot{q}_{\lambda\mu}{}^{\dagger}-i(\omega_{\lambda}-\omega_1)\dot{q}_{\lambda\mu}{}^{\dagger}=2G^2(q_{\lambda\mu}-q_{\lambda\mu}{}^{\dagger}). \quad \text{(A9b)}$$

From the last two equations one obtains two coupled differential equations for $S_{\lambda\mu}=q_{\lambda\mu}+q_{\lambda\mu}{}^{\dagger}$ and $D_{\lambda\mu}=q_{\lambda\mu}-q_{\lambda\mu}{}^{\dagger}$. The final solution can be written in the form

$$c_{\lambda\mu}(t)=e^{-i\omega_1 t}\{f_{\lambda}(t)c_{\lambda\mu}(0)$$
$$+g_{\lambda}(t)c_{\lambda\mu}{}^{\dagger}(0)+h_{\lambda}(t)[c_{\lambda\mu}(0),c_{\lambda\mu}{}^{\dagger}(0)]\}, \quad \text{(A10)}$$

where

$$f_{\lambda}(t)=\cos\Omega_{\lambda}t+2\frac{G^2}{\Omega_{\lambda}{}^2}(1-\cos\Omega_{\lambda}t)$$
$$-\frac{i}{\Omega_{\lambda}}(\omega_{\lambda}-\omega_1)\sin\Omega_{\lambda}t, \quad \text{(A11)}$$

$$g_{\lambda}(t)=(G^2/\Omega_{\lambda}{}^2)2(1-\cos\Omega_{\lambda}t), \quad \text{(A12)}$$

$$h_{\lambda}(t)=\frac{G}{\Omega_{\lambda}}\left\{\frac{\omega_{\lambda}-\omega_1}{\Omega_{\lambda}}(1-\cos\Omega_{\lambda}t)+i\sin\Omega_{\lambda}t\right\}, \quad \text{(A13)}$$

with $\Omega_{\lambda}=[(\omega_{\lambda}-\omega_1)^2+4G^2]^{1/2}$. This solution satisfies the requirement $c_{\lambda\mu}{}^2(t)=0$, and $c_{\lambda\mu}(t)c_{\lambda\mu}{}^{\dagger}(t)+c_{\lambda\mu}{}^{\dagger}(t)c_{\lambda\mu}(t)=1$. If all atoms are initially in the ground state so that the initial state of the whole system is $|\psi(0)\rangle=\prod_{\lambda,\mu}|\psi_{\lambda\mu}{}^{g}\rangle$, then

$$\langle\psi(0)|\sum_{\lambda,\mu}c_{\lambda\mu}{}^{\dagger}\sum_{\sigma,\nu}c_{\sigma\nu}|\psi(0)\rangle$$
$$=\sum_{\lambda}m_{\lambda}\{|g_{\lambda}(t)|^2+|h_{\lambda}(t)|^2\}$$
$$+\sum_{\lambda}m_{\lambda}h_{\lambda}{}^{*}(t)\sum_{\sigma}m_{\sigma}h_{\sigma}(t), \quad \text{(A14)}$$

where m_{λ} denotes the number of atoms with resonance frequency ω_{λ}. In terms of a normalized frequency distribution $\chi(\omega_{\lambda})$ for the total number of atoms m, one obtains in the limit

$$\sum m_{\lambda}\rightarrow m\int d\omega_{\lambda}\,\chi(\omega_{\lambda})$$

for the total emission rate in (A6):

$$W_m(t)=wm4G^2\int_0^{\infty}d\omega_{\lambda}\,\chi(\omega_{\lambda})\frac{\sin^2\frac{1}{2}\Omega_{\lambda}t}{\Omega_{\lambda}{}^2}$$
$$+wm^2G^2\left[\int_0^{\infty}d\omega_{\lambda}\,\chi(\omega_{\lambda})\frac{\omega_{\lambda}-\omega_1}{\Omega_{\lambda}{}^2}(1-\cos\Omega_{\lambda}t)\right]^2$$
$$+wm^2G^2\left[\int_0^{\infty}d\omega_{\lambda}\,\chi(\omega_{\lambda})\frac{\sin\Omega_{\lambda}t}{\Omega_{\lambda}}\right]^2, \quad \text{(A15)}$$

where $w=2\pi g^2\rho(\omega_0)$ is the transition probability per unit time for spontaneous emission by one completely excited atom. The first integral in (A15) times $4G^2$ represents an average over all resonance frequencies of the probability of the excited state. The additional factor wm determines the incoherent contribution to the emission by all atoms. If the width of the frequency distribution $\chi(\omega_{\lambda})$ is large compared with G, one can replace $\chi(\omega_{\lambda})$ by the constant value $\chi(\omega_1)$ and integrate over the variable $\omega_{\lambda}-\omega_1=\omega$ from $-\infty$ to $+\infty$. The second integral in (A15) vanishes in this approximation, so that

$$W_m(t)=w\left\{K2G\frac{2}{\pi}\int_{-\infty}^{+\infty}d\omega\frac{\sin^2\frac{1}{2}t(\omega^2+4G^2)^{1/2}}{\omega^2+4G^2}\right.$$
$$\left.+K^2\left[\frac{1}{\pi}\int_{-\infty}^{+\infty}d\omega\frac{\sin t(\omega^2+4G^2)^{1/2}}{(\omega^2+4G^2)^{1/2}}\right]^2\right\}, \quad \text{(A16)}$$

where $K=m\pi\chi(\omega_1)G$ is a dimensionless constant. Since

$$\frac{2}{\pi}\frac{d}{dt}\int_{-\infty}^{+\infty}\frac{\sin^2\frac{1}{2}t(\omega^2+4G^2)^{1/2}}{\omega^2+4G^2}d\omega$$
$$=\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{\sin t(\omega^2+4G^2)^{1/2}}{(\omega^2+4G^2)^{1/2}}d\omega=J_0(2Gt), \quad \text{(A17)}$$

one obtains in terms of zero-order Bessel functions

$$W_m(t)=w\left\{K\int_0^{2Gt} J_0(x)dx+K^2J_0^2(2Gt)\right\}. \quad (A18)$$

This result includes the incoherent and coherent contributions of emission. In the limit $t\to\infty$, the total emission rate approaches the value $W_m(\infty)=wK$. In the special case of a Lorentzian frequency distribution $\chi(\omega_\lambda)=(\Gamma/\pi)[(\omega_\lambda-\omega_0)^2+\Gamma^2]^{-1}$, K has the value

$$K=mG\Gamma[(\omega_1-\omega_0)^2+\Gamma^2]^{-1},$$

and in particular, $K=m(G/\Gamma)$ for $\omega_1=\omega_0$. The conditions for the validity of the approximation are $w\ll G\ll\Gamma$, and $mw\ll\Gamma$.

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Effect of Pressure on the Néel Temperature of Cobaltous Oxide*

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The effect of pressure on the Néel temperature of CoO powder samples is measured up to 150 kbar by the thermal scanning of Mössbauer absorption. The hyperfine field H, which is known at room temperature as a function of pressure and at 1 atm as a function of temperature, is represented as a function of $\epsilon=1-T/T_N$. The constant-temperature data follow the theoretical law $H\sim\epsilon^{1/3}$ over a larger range of ϵ than the constant-pressure data. This result is discussed within the present model for the magnetic properties of CoO.

I. INTRODUCTION

MÖSSBAUER measurements of the hyperfine field on Fe^{57} in CoO at room temperature and pressures up to 250 kbar [1] indicated a marked increase of the Néel temperature in agreement with earlier pressure studies on CoO in the range 0–6 kbar.[2] This agreement seemed surprising, since the hyperfine field studies used powder samples and it is known[3] that the Néel temperature in CoO at atmospheric pressure depends sensitively on grain size, stress, and imperfections. In addition to this question, direct measurements of the Néel temperature in CoO over a larger pressure range were expected to show how far the hyperfine H at constant temperature and high pressures follows the theoretical $\frac{1}{3}$-power law[4,5] $H\propto[1-T/T_N(P)]^{1/3}$ and how the high-pressure results compare with studies at 1 atm and lower temperatures.[6]

II. EXPERIMENT

The change of the Néel temperature in CoO under high pressures was measured by thermal scanning of Mössbauer absorption.[7,8] Using the values of the isomer shift and linewidth measured previously by the high-pressure Mössbauer technique,[1] the thermal scanning curves indicate that the transition is broadened because of a spread of local Néel temperatures in the powder samples, which were prepared for consistency according to the earlier procedure,[1] and exhibit a single-line spectrum at 1 atm, identical with the atmospheric spectrum exhibited in Fig. 2 of Ref. 1. The mean value of the Néel temperature in these samples at atmospheric pressure was (264±6)°K. Despite this rather large uncertainty, the changes of the Néel temperature under pressure are determined by the thermal scanning method with more accuracy,[8] and the uncertainty is still small compared with the effect of pressures in the range 0–150 kbar.

Furthermore, the hyperfine field data at 20 kbar,[1] which correspond to $T_N(20\text{ kbar})=(245\pm3)°K$, relate the measured changes to an absolute scale and allow a direct comparison with the earlier results on bulk material at lower pressures[2] (see Fig. 1).

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