

and  $\nu_0$  remain unchanged,  $\theta$  is increased from 0.0312 to 1. This means that all nonradiative processes are made radiative, so that it is reasonable to assume that  $\Delta\nu$  remains the same or increases. The resulting values of  $T_L$  and  $\eta$  are as follows:

$$\begin{aligned} T_L &\leq 1759^\circ\text{K}, \\ \eta &= 1.325/1.101 = 1.203, \\ (1 - T/T_L)^{-1} &\geq 1.206. \end{aligned} \quad (5.5)$$

Thus the thermodynamic upper limit  $\eta < (1 - T/T_L)^{-1}$  is not violated. Under these conditions the diode would have an electrical power consumption of  $\dot{E}_p \equiv jeV = 1.101 \times 10^{-3}$  W, and a refrigerating power of  $\dot{Q} = 0.203 \dot{E}_p = 2.2 \times 10^{-4}$  W. It appears, therefore, that the laws of thermodynamics do not preclude the construction of a diode having a measurable refrigerating effect and a technical efficiency significantly greater than unity.

Since quantum efficiencies at 300°K are of order 6% or less, the thermodynamic limits are at present of only academic interest at this temperature. Conditions

at low temperatures are very different. Recently, quantum efficiencies of 36% have been recorded at 77°K<sup>7</sup> and of 40% at 20°K (Carr<sup>9</sup>). It seems possible that in these cases the *internal* quantum efficiency  $\theta_i$ , i.e., the number of photons produced in the device per electron crossing it, is near unity, the losses being accounted for by internal absorption and reflection. Pilkuhn and Rupprecht<sup>10</sup> have estimated that  $\theta_i \sim 100\%$  for epitaxial GaAs diodes used as lasers at 4.2°K. One can therefore expect that improved experimental techniques may lead to external quantum efficiencies near 100%, and in such a case the thermodynamic limit would become a realistic restriction. External quantum efficiencies of 95% at 77°K have recently been reported by Lamorte *et al.*,<sup>11</sup> but this refers to a pulsed laser which is not a steady-state device and to which therefore the thermodynamic arguments of this paper do not apply.

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## Quantum Theory of an Optical Maser. II. Spectral Profile\*

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In Paper I of this series, we derived equations of motion for a quantum-laser field interacting with atomic reservoirs. In the usual region of sustained oscillation, the off-diagonal elements of the radiation density matrix  $\rho_{n,n+k}(t)$  were found to have an exponential decay associated with phase diffusion. In I we found the spectrum of the laser radiation by calculating the single time-ensemble average electric field implied by  $\rho_{n,n+1}(t)$ . This electric field was then treated as a classical variable whose Fourier analysis gave the spectrum. In the present paper, we establish the validity of this procedure by analyzing a simple model for a spectrometer. It is also shown that the same spectrum can be obtained from a two-time correlation function derived from the equations of motion.

### I. INTRODUCTION

IN the first paper of this series,<sup>1</sup> we derived the equation of motion for the density matrix of the laser field as it evolved under the influence of excited atoms (lasing medium) and a dissipation mechanism (cavity  $Q$ ). We found that the elements of the density matrix in the  $n$  representation were coupled only along lines parallel to the main diagonal. The diagonal elements of the density matrix were seen to approach a steady state while the off-diagonal terms decayed in time. It was shown, to a good approximation, that the density

matrix for a laser in sustained oscillation sufficiently above threshold obeys the equation

$$(d\rho/dt)_{n,n+k} = -i(n-n')\nu\rho_{n,n+k} - \mu_0^{(k)}\rho_{n,n+k}, \quad (1)$$

where  $\nu$  is the laser frequency. We have included the first term on the right since it is more convenient for the present purposes to work in the Schrödinger picture. The damping constant  $\mu_0^{(k)}$  had the form  $\frac{1}{2}Dk^2$ , where  $D$  is given by

$$D = \frac{1}{2}(\nu/Q)/\langle n \rangle, \quad (2)$$

and  $\langle n \rangle$  is the average number of quanta at steady state. It was seen in I that the decay of the off-diagonal elements implied by Eq. (1) is associated with phase diffusion.

The density matrix  $\rho_{n,n'}$  represents our knowledge of the state of the system of interest and thus contains

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the information necessary to calculate expectation values of single-time Hermitian operators. That is, if we know the density matrix  $\rho$  for our system, the expectation value for an operator  $Q$  (in the Schrödinger picture) is just

$$\langle Q \rangle_t = \text{Tr}[\rho(t)Q].$$

In I, the spectrum of the laser oscillator was obtained by calculating the ensemble average electric field.

$$\langle E \rangle_t = \langle E \rangle_0 \exp(-\frac{1}{2}Dt) \sin \nu t. \quad (3)$$

This time-dependent expectation value was then Fourier analyzed, and multiplied by its complex conjugate. The resulting expression for the spectrum of the laser oscillator was found to be Lorentzian:

$$I(\omega) = |E(\omega)|^2 = E_0^2 [(\omega - \nu)^2 + (\frac{1}{2}D)^2]^{-1}. \quad (4)$$

One purpose of this paper is to establish the validity of this spectrum even when  $\langle E \rangle = 0$  because the density matrix is diagonal.

In Sec. II, we give an operational analysis of a simple spectrometer and obtain an expression for the laser spectrum in agreement with that given by Eq. (4). Before doing so, we must answer the question, "How is the spectrum of the quantized laser oscillator to be defined?" A plausible guess is that it should be deduced from the Fourier transform of the low-frequency part of the two-time correlation function

$$G(\tau) = \frac{1}{2} \langle E(t+\tau)E(t) + E(t)E(t+\tau) \rangle_{\text{ensemble}}, \quad (4')$$

where  $E(t)$  is the electric field operator at time  $t$ . There are two difficulties with this recipe for finding the linewidth: First, it is based upon the assumption that a procedure used in classical noise theory can be transferred to a quantum-mechanical problem. Second, we have not yet given a meaning to a time-dependent operator such as  $E(t)$ . The second difficulty arises because we are considering operators of the radiation field possessing a time dependence which is no longer

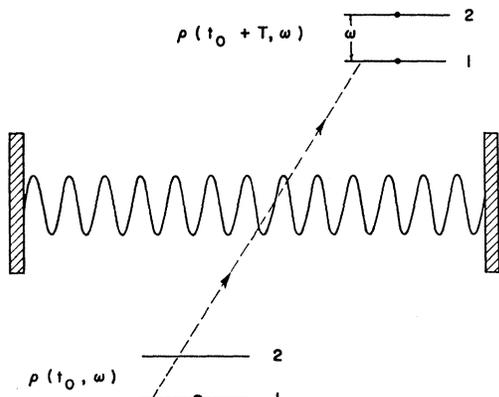


FIG. 1. Schematic illustration of spectrum analyzer. Atom enters laser cavity in ground state 1, interacts with laser radiation and emerges in linear superposition of states 1 and 2.

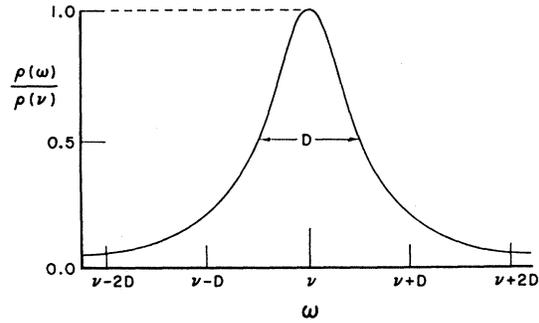


FIG. 2. Relative excitation of spectrometer atoms plotted versus atomic frequency  $\omega = (\epsilon_2 - \epsilon_1)/\hbar$ .

describable by a Hamiltonian after we have traced over the reservoir coordinates. The Appendix includes a discussion of the spectrum as inferred from a two-time correlation function using the equation of motion (1) for the density matrix.

## II. SPECTRUM ANALYZER

We consider the following simple spectrometer. Imagine that we have a beam of two level atoms, upper state  $|2\rangle$  and lower state  $|1\rangle$  separated by an energy  $\epsilon_2 - \epsilon_1 = \hbar\omega$ . The beam is prepared with each atom initially in the lower state  $|1\rangle$ , i.e., its initial  $2 \times 2$  density matrix has only one nonvanishing element

$$\rho_{1,1}(t_0) = 1. \quad (5)$$

These atoms pass through the laser cavity and interact weakly with the laser radiation, as shown in Fig. 1. The time of flight  $T$  is much greater than  $1/D$ , so that the effective atomic linewidth is much narrower than that of the laser radiation. The fraction of excited atoms  $\rho_{2,2}$  emerging from the cavity is determined by a suitable measurement. We then prepare a new beam of slightly different atomic frequency and repeat the experiment. Finally, we plot the relative excitation of the different beams as a function of frequency, as in Fig. 2. This plot of the relative effectiveness of the laser radiation in exciting atoms with different frequencies provides us with an operational definition of the spectral profile for the laser.

We now calculate the probability that an atom of frequency  $\omega$  will make a transition to the state  $|2\rangle$ . To do this, we consider an atom injected at  $t=t_0$  in state  $|1\rangle$ , determine the density matrix for the combined spectrometer atom-field system at time  $t_0+T$ , and then trace over the radiation field. The spectrometer atom-field density matrix will evolve from one in which all the atoms in the beam are initially in their ground states,

$$\rho_{\text{atom-field}}(t_0) = \rho_{1,1}(t_0) \rho_{n,n'}(t_0) = \rho_{n,n'}(t_0), \quad (6)$$

to

$$\rho_{\text{atom-field}}(t_0+T) = \rho_{r,n;s,n'}(t_0+T), \quad r=1, 2; \quad s=1, 2. \quad (7)$$

The probability of finding an atom of frequency  $\omega$  in the upper state at  $t_0+T$  is

$$\rho_{2,2}(\omega, t_0+T) = \sum_n \rho_{2,n;2,n}(\omega, t_0+T). \quad (8)$$

We now proceed to calculate this quantity.

During the time that the spectrometer atom is in the cavity, to a very good approximation, the pumping and damping of the laser field are going on as if the spectrometer atom were absent. That is, the presence of the spectrometer atom weakly coupled to the "massive" laser field hardly affects the optical oscillator. Hence, the time rate of change of the density matrix for the spectrometer atom-laser system is given by the sum of the time derivatives in the absence of the atom plus the time derivative produced by the spectrometer atom interacting with the field,

$$d\rho/dt = (d\rho/dt)_{\text{laser}} + (d\rho/dt)_{\text{spectrometer interaction}}. \quad (9)$$

Using Eq. (1), we have

$$\begin{aligned} (d\rho/dt)_{r,n;s,n'} = & -i(n-n')\nu\rho_{r,n;s,n'} - \mu_0^{(k)}\rho_{r,n;s,n'} \\ & - i[(H_0^{\text{atom}} + V), \rho]_{r,n;s,n'}, \end{aligned} \quad (10)$$

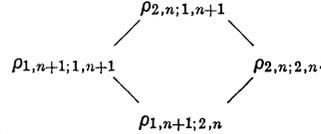
where

$$H_0^{\text{atom}} = \epsilon_2 A_2^\dagger A_2 + \epsilon_1 A_1^\dagger A_1 \quad (11)$$

is the free-atom Hamiltonian, and

$$V = -g[A_2^\dagger A_1 a + A_1^\dagger A_2 a^\dagger] \quad (12)$$

is the interaction Hamiltonian of the atom and field. Here  $a^\dagger, a, A_2^\dagger, A_2$ ; and  $A_1^\dagger, A_1$  are the creation and annihilation operators for the field and the atom in its states  $|2\rangle$  and  $|1\rangle$ , respectively. The field frequency is given by  $\nu$  and the atomic energies are  $\epsilon_1$  and  $\epsilon_2$  while the strength of the atom-field coupling is denoted by  $g$ . The equations (10) in which we are interested are those coupling  $\rho_{1,n+1;1,n+1}(t_0)$  to  $\rho_{2,n;2,n}(t_0+T)$ , i.e., we want equations of motion for the evolution



These equations are

$$(d\rho/dt)_{2,n;2,n} = -i[V_{2,n;1,n+1}\rho_{1,n+1;2,n}(t) - \text{c.c.}], \quad (13)$$

$$\begin{aligned} (d\rho/dt)_{1,n+1;2,n} = & -[i(\nu-\omega) - \mu_0^{(1)}]\rho_{1,n+1;2,n} \\ & - i[V_{1,n+1;2,n}\rho_{2,n;2,n} + \rho_{1,n+1;1,n+1}V_{1,n+1;2,n}], \end{aligned} \quad (14)$$

$$\rho_{2,n;1,n+1} = \rho_{1,n+1;2,n}^*. \quad (15)$$

We now proceed with the perturbation calculation. Noting that at  $t=t_0$  the atom-field density matrix factors and that the atom is in the lower state at that time, we have the initial conditions given by Eq. (5). The first nonvanishing contribution to  $\rho_{2,n;2,n}$  is given by

$$\rho_{2,n;2,n}(\omega) = i \int_{t_0}^{t_0+T} dt' [V_{2,n;1,n+1}\rho_{1,n+1;2,n}^{(1)}(t') - \text{c.c.}]. \quad (16)$$

The argument  $\omega$  in Eq. (16) indicates that we are considering an atom having that atomic frequency. The off-diagonal elements  $\rho_{1,n+1;2,n}^{(1)}(t')$  are calculated from Eq. (14) and are given by

$$\rho_{1,n+1;2,n}^{(1)}(t') = -i \int_{t_0}^{t'} dt'' \exp\{-i[(\nu-\omega) - i\mu](t'-t'')\} V_{1,n+1;2,n}\rho_{n,n}(t_0), \quad (17)$$

where

$$\mu = \mu_0^{(1)}. \quad (17')$$

Inserting (17) into (16) we find

$$\rho_{2,n;2,n}(\omega) = - \int_{t_0}^{t_0+T} dt' \int_{t_0}^{t'} dt'' \{|V_{2,n;1,n+1}|^2 \exp[i(\omega-\nu) - \mu](t'-t'') + \text{c.c.}\} \rho_{n,n}(t_0). \quad (18)$$

As indicated in Eq. (2), we must sum this expression over  $n$  to find the probability that the atom of frequency  $\omega$  has absorbed a laser photon in a time  $T$ . We find

$$\rho_{2,2}(\omega) = -[g^2 \sum_{n=0}^{\infty} n\rho_{n,n}(t_0)] \left\{ \int_{t_0}^{t_0+T} dt' \int_{t_0}^{t'} dt'' \exp[i(\omega-\nu) - \mu](t'-t'') + \text{c.c.} \right\} \approx (2g^2\mu T) \langle n \rangle [(\omega-\nu)^2 + \mu^2]^{-1} \quad (19)$$

which is in exact agreement with the spectrum given by Eq. (4), since  $\mu = \frac{1}{2}D$ .

#### ACKNOWLEDGMENT

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#### APPENDIX

In this paper we designed an experimental "apparatus" to measure the laser spectrum. This device was then analyzed using the equation of motion for the density matrix  $\rho_{n,n'}(t)$  of the laser radiation. Such an operational procedure for obtaining the spectral profile has the merit of being understandable physically.

The spectral profile for a system may be defined<sup>2,3</sup> formally as the Fourier transform of the correlation function given by

$$G(t) = \frac{1}{2} \text{Tr} \{ [a^\dagger(t)a(0) + a(t)a^\dagger(0)]\rho(0)_{\text{total}} + [a^\dagger(0)a(t) + a(0)a^\dagger(t)]\rho(0)_{\text{total}} \}, \quad (\text{A1})$$

where  $\rho_{\text{total}}$  is the density matrix for the total system of field plus reservoirs. This Appendix is intended to show that the linewidth implied by Eq. (A1) agrees with that of Eq. (19).

In order to use (A1) we must ask, what is the time dependence of  $a(t)$ ? Clearly it is not that of a free field,<sup>4,5</sup> for then there would be no linewidth ( $\delta$  function spectrum). To obtain the temporal evolution of the correlation function  $G(t)$ , we must recall how "noise" entered the problem in the first place. We considered the radiation field to be acted upon by the pumping and damping atoms (reservoirs) and then traced the density matrix over the reservoir states. After contraction, the radiation field cannot be described by a state vector but is in a mixture requiring a density matrix for its specification. Thus we have extended the definition (A1) to include the reservoir states over which we will later trace. The time dependence of the operator  $a(t)$  is now given by

$$a(t) = U^\dagger(t)a(0)U(t), \quad (\text{A2})$$

where  $U(t)$  is the time development operator for the combined laser-reservoir system. The correlation function with the atomic reservoirs included, but traced over, may now be written in terms of these time-dependent operators. In the following discussion, we restrict our attention to the quantity

$$g(t) = \text{Tr} [a^\dagger(t)a(0)\rho_{\text{total}}(0)], \quad (\text{A2}')$$

since the other terms of (A1) are similarly obtained. Writing (A2') in terms of (A2), we have

$$g(t) = \text{Tr}_\rho \text{Tr}_R \{ [U^\dagger(t)a^\dagger(0)U(t)a(0)]R(0)\rho(0) \} \\ = \text{Tr}_\rho \{ \text{Tr}_R [U^\dagger(t)a(0)U(t)R(0)]a^\dagger(0)\rho(0) \}, \quad (\text{A3})$$

where  $R(0)$  is the reservoir density matrix which is uncoupled from the radiation field  $\rho(0)$  at  $t=0$ . Equa-

tion (A3) is seen to have a simple form if we define a new operator

$$\mathcal{Q}(t) = \text{Tr}_R [U^\dagger(t)a(0)U(t)R(0)], \quad (\text{A4})$$

for now Eq. (A3) is

$$g(t) = \text{Tr}_\rho [\mathcal{Q}^\dagger(t)\mathcal{Q}(0)\rho(0)]. \quad (\text{A5})$$

We obtain an explicit form for the time dependence of the operators  $\mathcal{Q}(t)$  and  $\mathcal{Q}^\dagger(t)$  by recalling that the density matrix is given by

$$\rho_{n,n'}(t) = \{ \text{Tr}_R [U(t)R(0)\rho(0)U^\dagger(t)] \}_{n,n'}. \quad (\text{A6})$$

However, we may obtain the time dependence of this quantity by integrating Eq. (1)

$$\rho_{n,n'}(t) =$$

$$\rho_{n,n'}(0) \exp \{ -[\frac{1}{2}D(n-n')^2 + i\nu(n-n')]t \}. \quad (\text{A7})$$

Comparing (A6) and (A7) we see that  $U(t)_{n,n'}$  is in fact diagonal. Hence, we may write (A6) as

$$\rho_{n,n'}(t) = \rho_{n,n'}(0) \text{Tr}_R [U(t)_{n,n}R(0)U^\dagger(t)_{n',n'}]. \quad (\text{A8})$$

A comparison of (A7) and (A8) then shows that

$$\text{Tr}_R [U(t)_{n,n}R(0)U^\dagger(t)_{n',n'}] \\ = \exp [ -\frac{1}{2}D(n-n')^2t + i\nu(n-n')t ], \quad (\text{A9})$$

and we may write the time dependence of the matrix elements of the  $\mathcal{Q}$  operator (A4) as

$$[\mathcal{Q}(t)]_{n,n+1} = a(0)_{n,n+1} \text{Tr}_R [U(t)_{n,n}R(0)U^\dagger(t)_{n+1,n+1}] \\ = a(0)_{n,n+1} \exp [ -(\frac{1}{2}D + i\nu)t ]. \quad (\text{A10})$$

We may now write Eq. (A3) as

$$G(t) = \text{Tr}_\rho [a^\dagger(0)a(0)\rho(0)] \cos \nu t \exp (-\frac{1}{2}Dt). \quad (\text{A11})$$

It is clear that the Fourier transform of (A11) and the other terms in Eq. (A1) will give the spectral profile of Eq. (19). Accordingly, the decay of the two-time correlation function for the laser radiation (A11) is identical to the decay of the single time averaged electric field as given by Eq. (3).

This Appendix can be regarded as a demonstration of the Onsager regression hypothesis.<sup>6</sup> Lax<sup>2</sup> has shown that Onsager's original statement for an equilibrium system is true even for nonequilibrium situations, provided the system is Markoffian. Louisell and Marburger<sup>7</sup> have extended the proof to show "that this result is always true, but that only under the conditions stated by Lax are the equations of motion the macroscopic ones."

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