Comparison of Quantum and Semiclassical Radiation Theories with Application to the Beam Maser*

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Summary—This paper has two purposes: 1) to clarify the relationship between the quantum theory of radiation, where the electromagnetic field-expansion coefficients satisfy commutation relations, and the semiclassical theory, where the electromagnetic field is considered as a definite function of time rather than as an operator; and 2) to apply some of the results in a study of amplitude and frequency stability in a molecular beam maser.

In 1), it is shown that the semiclassical theory, when extended to take into account both the effect of the field on the molecules and the effect of the molecules on the field, reproduces almost quantitatively the same laws of energy exchange and coherence properties as the quantized field theory, even in the limit of one or a few quanta in the field mode. In particular, the semiclassical theory is shown to lead to a prediction of spontaneous emission, with the same decay rate as given by quantum electrodynamics, described by the Einstein A coefficients.

In 2), the semiclassical theory is applied to the molecular beam maser. Equilibrium amplitude and frequency of oscillation are obtained for an arbitrary velocity distribution of focused molecules, generalizing the results obtained previously by Gordon, Zeiger, and Townes for a singel-velocity beam, and by Lamb and Helmer for a Maxwellian beam. A somewhat surprising result is obtained; which is that the measurable properties of the maser, such as starting current, effective molecular Q, etc., depend mostly on the slowest 5 to 10 per cent of the molecules.

Next we calculate the effect of amplitude and frequency of oscillation, of small systematic perturbations. We obtain a prediction

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that stability can be improved by adjusting the system so that the molecules emit all their energy $\hbar\,\Omega$ to the field, then reabsorb part of it, before leaving the cavity. In general, the most stable operation is obtained when the molecules are in the process of absorbing energy from the radiation as they leave the cavity, most unstable when they are still emitting energy at that time.

Finally, we consider the response of an oscillating maser to randomly time-varying perturbations. Graphs are given showing predicted response to a small superimposed signal of a frequency near the oscillation frequency. The existence of "noise enhancing" and "noise quieting" modes of operation found here is a general property of any oscillating system in which amplitude is limited by nonlinearity.

I. Introduction

THIS PAPER has two purposes: 1) to clarify the relationship between the quantum theory of radiation where the electromagnetic field expansion coefficients satisfy commutation relations, and the semiclassical theory where the electromagnetic field is considered as a definite function of time rather than as an operator, and 2) to apply some of the results thus obtained in a study of amplitude and frequency stability of the ammonia beam maser.

In 1), the relation between quantum electrodynamics and the semiclassical theory is shown to be quite different from that usually assumed. The semiclassical theory, when extended to take into account both the effect of the molecules on the field and the effect of the field on the molecules, reproduces almost quantitatively the same laws of energy exchange and coherence properties as the quantized field theory, even in the limit of one or a few quanta in the field cavity mode. In particular, the semiclassical theory is shown to lead to a prediction of spontaneous emission, with exactly the same decay rate as given by quantum electrodynamics, as described by the Einstein A coefficients.

There remain, however, several fundamental differences in the two theories. For example, quantum electrodynamics allows the possibility that the combined system (molecules plus field) may be in states which have properties qualitatively different than any that can be described in classical terms, even in the limit of arbitrarily high photon occupation numbers. Thus the common statement that quantum electrodynamics goes over into classical electrodynamics in the case of high quantum numbers for the field oscillators, needs to be somewhat qualified.

Having shown the essential equivalence of quantum electrodynamics and the semiclassical approach for the problems of interest, we turn to detailed calculations applying the semiclassical theory to the ammonia beam maser. Equilibrium amplitude and frequency of oscillation are obtained for an arbitrary velocity distribution of focused molecules, generalizing the results obtained previously by Gordon, Zeiger and Townes [1] for a single-velocity beam and by Lamb and Helmer [6] for a Maxwellian beam. A rather surprising result is obtained, namely that the measurable properties of the maser, such as starting current, effective molecular Q, etc., depend mostly on the slowest 5 to 10 per cent of the molecules.

Next we calculate the effect on amplitude and frequency of oscillation of small systematic perturbations. We obtain a prediction that stability can be improved by adjusting the system so that the molecules emit all their energy $\hbar\Omega$ to the field, then reabsorb part of it, before leaving the cavity. In general, the most stable operation is obtained when the molecules are in the process of absorbing energy from the radiation as they leave the cavity, the most unstable, when they are still emitting energy at that time.

Finally, we consider this response of an oscillating maser to random time-varying perturbations. Graphs are given showing predicted response to a small superimposed signal of a frequency near to the oscillation frequency. The results show a quite complicated variation as a function of the frequency difference and beam current, and resemble some results of Wiener, concerning nonlinear random phenomena.

Broadly speaking, there are two different levels of approximation used in gas maser theories published as of this writing:

1) The most common and also the crudest of these theories is the one wherein one treats the emission process of radiation from molecules as if the transition probabilities were proportional to the time. Such theories contain little that was not already contained in Einstein's 1917 paper which introduced the A and B coefficients. According to quantum mechanics, the idea of time proportional transition probabilities is an approximation, valid only when the correlation time of the radiation is short compared to the time required to accumulate an appreciable transition probability; that is, the radiation responsible for the transition must be random, with a spectrum wide compared to the line width. In an ammonia beam device the correlation time of the radiation may be of the order of 10^3 to 10^6 times the flight time of a molecule through the cavity, and thus any attempt to describe maser operation in terms of "Fermi golden rule" type of equations for the transition probabilities, *i.e.*,

$$W_{1\to 2} = \frac{2\pi}{\hbar^2} \mid H_{12} \mid^2 \rho(\omega)$$

may lead to conclusions qualitatively as well as quantitatively wrong. Most of the existing noise figure calculations are based on a treatment of this type [1], [2], and hence one cannot assess their worth until the calculation has been checked by a more rigorous theory.

2) The second method of treating the maser theoretically is that based on solving Schrödinger's time-dependent equation for a molecule as perturbed by a classically described field and finding then the expectation value of the dipole moment of the molecule and using the time derivative of this expectation value as the current source of the classical electromagnetic field.

This is essentially the calculation of Shimoda, Wang and Townes [4], and Basov and Prokhorov [5], and Lamb and Helmer [6], Feynman, Vernon and Hellwarth [7]. While this is clearly superior to the first method outline above, there are still several important approximations involved. In principle, the molecular beam should be treated as a single quantum-mechanical system, by a formalism like that of Dicke's "superradiant gas" [8]. In the theories quoted above, the molecules were ascribed independent wave functions. Also, the electromagnetic field should be quantized and the problem treated as one of quantum electrodynamics. Although the theories above lead to definite predictions for saturation and frequency pulling, it is not at all clear that they can lead to reliable predictions of fluctuation effects involved in noise figure and frequency stability. It is generally thought that the semiclassical theory should be adequate for any effects at microwave frequencies due to the smallness of the Einstein A coefficient compared to the B coefficient. However, quantization of the electromagnetic field introduces many changes in addition to the appearance of A coefficients; for instance, quantization can lead to states qualitatively different from any describable in classical terms, even in the limit of arbitrarily high photon occupation numbers per field normal mode. Such states will be shown, in the calculations to follow, to actually be the ones produced in the maser under certain idealized conditions. Thus until these calculations based on these approximations are checked in some other way, our degree of confidence in them cannot be too great.

Our approach in this paper will be, stated briefly, to first treat simple problems in which we can talk of transition probabilities with all coherence properties retained, within the formalism of quantum electrodynamics. Then we will investigate the relationship between the "modified" semiclassical ("neoclassical" theory), as employed by Shimoda, Wang, and Townes [4], and quantum electrodynamics. The relationship is not at all that which is usually assumed, *i.e.*, that quantum electrodynamics goes into semiclassical theory only in the limit of high photon occupation numbers per field-normal mode. Rather the neoclassical theory, in which expectation values of quantum mechanical operators are interpreted as *actual* values of sources in the classical Maxwell equations, and both the effect of the radiation field on the molecule and the effect of the molecule back on the field are taken into account, *does* lead to a prediction of spontaneous emission, and to only very small quantitative differences in the decay rate for the case of a few microwave photons in the cavity.

In Section IV the neoclassical theory is applied to the problem of the ammonia beam device in which the indirect "coupling" between molecules via the field is treated and a steady-state solution is obtained under the assumption of an arbitrary velocity distribution, wall losses and/or external energy coupling. The solution is obtained for the frequency stability as a function of the mean values of the square and cube of the flight time and the Q of the cavity. This solution is found to agree with that of Gordon, Zeiger and Townes [1] in the univelocity case and with the analysis of Lamb and Helmer [6] in the case of a Maxwellian distribution of velocities. Then this solution is made the basis, or unperturbed solution, in a perturbation treatment of flucutation effects to the first order in the small departure from the steady-state solution. These problems in this case become linear, so that we can analyze the effect of small periodic perturbations proportional to exp $(i\beta t)$ and superpose the solutions to give solutions for the transient response to an arbitrary small perturbation. This can represent an extra signal fed in intentionally, or it might be a randomly varying function representing thermal noise in the cavity and/or load. A "noise quieting" phenomena is seen to occur for proper values of the flight time, and graphs are drawn which exhibit the power spectrum of the thermal noise as affected by the molecular beam.

II. Quantum Electrodynamic Solutions

We approach the theory of maser operation in several stages, starting with simple, special cases for which all details of the mathematics can be worked out, then adding various features which tend in the direction of more realistic models. The mathematical form of the theory is quite similar to what one encounters in the statistical mechanics of irreversible processes. Of particular interest, however, is the extent to which the semiclassical theory is derivable from quantum electrodynamics, and we are most interested in comparing the results of this section with those obtained in Section III. Also, the effect of different statistical assumptions concerning the initial states of the molecules is interesting in this same regard.

A. Field Quantization

We first develop the formalism of field quantization in a form suitable for microwave applications. There is, of course, no need for elegant covariant formulations here; the simple approach to electrodynamics given by Fermi [9] is quite adequate for our purposes. Here the usual plane-wave expansion is not appropriate and in its place we need to use the expansion of electromagnetic fields in terms of resonant modes of the particular cavity under consideration. We use the cavity normal mode functions as defined by Slater [10]. The cavity is represented by a volume V, bounded by a closed surface S. Let $\boldsymbol{E}_a(x)$, $k_a^2=\omega_a^2/C^2$ be the eigenfunctions and eigenvalues of the boundary-value problem.

$$\begin{aligned} \nabla\times\nabla\times E - k^2 E &= 0 \qquad \text{in } V \\ n\times E &= 0 \qquad \text{on } S \end{aligned} \tag{1}$$

where $\boldsymbol{n}$ is a unit vector normal to S. The $\boldsymbol{E}_a(\mathbf{x})$ are so normalized that

$$\int_v (\boldsymbol{E}_a \cdot \boldsymbol{E}_b) dV = \delta_{ab}. \tag{2}$$

The vector functions $\boldsymbol{H}_a(\mathbf{x})$, related to $\boldsymbol{E}_a$ by

$$\nabla \times \boldsymbol{E}_a = k_a \boldsymbol{H}_a \qquad \nabla \times \boldsymbol{H}_a = k_a \boldsymbol{E}_a \tag{3}$$

are also orthonormal in V as follows:

$$\int_v (\boldsymbol{H}_a \cdot \boldsymbol{H}_b) dV = \delta_{ab}. \tag{4}$$

The electric and magnetic fields can be expanded in the following forms:

$$\boldsymbol{E}(\mathbf{x}\ t) = -\sqrt{4\pi} \sum_a p_a(t) \boldsymbol{E}_a(\mathbf{x}) \tag{5}$$

and

$$\boldsymbol{H}(\mathbf{x}\ t) = \sqrt{4\pi} \sum_a \omega_a q_a(t) \boldsymbol{H}_a(\mathbf{x}). \tag{6}$$

From these relations, we find for the total field energy

$$\mathcal{H} = \int \frac{E^2 + H^2}{8\pi} dV = \frac{1}{2} \sum_a (p_a^2 + \omega_a^2 q_a^2), \tag{7}$$

and the Maxwell equations,

$$\nabla \times \boldsymbol{E} = \frac{1}{c} \frac{\partial \boldsymbol{H}}{\partial t} \tag{8}$$

and

$$\nabla \times \boldsymbol{H} = \frac{1}{c} \frac{\partial \boldsymbol{E}}{\partial t}, \tag{9}$$

then reduce to the Hamiltonian equations of motion,

$$\dot{q}_a = \frac{\partial \mathcal{H}}{\partial p_a} = p_a, \tag{8a}$$

$$\dot{p}_a = -\frac{\partial \mathcal{H}}{\partial q_a} = -\omega_a^2 q_a, \tag{8b}$$

respectively.

On quantization of the field the canonically conjugate coordinates and momenta satisfy the commutation rules,

$$[q_a, q_b] = [p_a, p_b] = 0 \tag{10}$$

and

$$[q_a, p_b] = i\hbar\delta_{ab}. \tag{11}$$

The operators C_a^*, C_a which create or annihilate a photon in the ath cavity mode are then

$$C_a^* = \frac{p_a + i\omega_a q_a}{\sqrt{2\hbar\omega_a}} \qquad C_a = \frac{p_a - i\omega_a q_a}{\sqrt{2\hbar\omega_a}} \tag{12}$$

with the commutation rule

$$[C_a\ C_b^*] = \delta_{ab}. \tag{13}$$

Denote by $\phi(n_1, n_2, \cdots)$ the state vector of the field for which there are n_1 quanta in mode 1, n_2 in mode 2, etc. The C_a operators have the properties

$$C_a \phi(\cdots, n_a, \cdots) = \sqrt{n_a}\, \phi(\cdots, n_a - 1, \cdots) \tag{14}$$

and

$$C_a^* \phi(\cdots, n_a, \cdots) = \sqrt{n_a + 1}\, \phi(\cdots, n_a + 1, \cdots) \tag{15}$$

from which we easily verify (13), and obtain the matrix elements in the n_a representation,

$$(n_a \mid C_a \mid n_a') = (n_a' \mid C_a^* \mid n_a) = \sqrt{n_a + 1}\, \delta(n_a'\ n_a + 1). \tag{16}$$

The Hamiltonian, with zero point energy removed, then reduces to

$$\mathcal{H} = \sum_a \hbar\omega_a C_a^* C_a = \sum_a \hbar\omega_a n_a. \tag{17}$$

Finally, we work out for later purposes the matrix elements of the electric field in the case of a cylindrical cavity with only the lowest TM mode excited. In this mode, the only nonvanishing component of $\boldsymbol{E}_a$ is E_{az} = (constant) $\times J_0(k_a r)$, independent of z and θ. The normalizing constant is obtained from evaluating the integral (2), with the result that on the axis of the cylinder (along which the molecules travel in an ammonia maser) the function E_{az} reduces to

$$E_{az} = \frac{1}{J_1 \sqrt{V}}. \tag{18}$$

Here $J_1 = J(u) = 0.5191$, and $u = 2.405$ is the first root of $J_0(u) = 0$. V is the volume of the cavity. The operator P_a involved in the electric field expansion is, from (12),

$$p_a = \sqrt{\frac{\hbar\omega_a}{2}} (C_a + C_a^*). \tag{19}$$

Combining (5), (16), (18), and (19), we obtain the matrix elements

$$(n \mid E \mid n') = -\left(\frac{2\pi\hbar\omega}{J_1^2 V}\right)^{1/2} [\sqrt{n}\, \delta_{n,n'+1} + \sqrt{n+1}\, \delta_{n+1,n'}] \tag{20}$$

in which we have dropped the subscript a, it being understood that (20) refers to the case where only the lowest TM mode is taken into account. For the matrix elements of electric field at points off the axis of the cylinder, this expression should be multiplied by $J_0(Kr)$.

B. Interaction with a Single Molecule

The simplest possible situation is one where we consider a lossless cavity, which has only a single resonant

mode near the natural line frequency of the molecule, and a uniform field (electric or magnetic, whichever is the one effective in field-molecule interaction) along the path of the molecules. Suppose further that only a single molecule, which has only two possible energy levels, is in the cavity. With the molecule-field interaction in the usual $(\boldsymbol{J}\cdot\boldsymbol{A})$ form, it appears that even this problem cannot be solved exactly. However, because of the simplicity of the model, we will be able to treat it more accurately than is usually done in more difficult problems, where one resorts to an expansion in powers of $(e^2/\hbar c)$. The stationary states of the system (molecule plus field) can be found to an accuracy of perhaps one part in 10^7 for radiation energy densities up to the order of those encountered in masers, by a calculation which involves nothing worse than solving quadratic equations. By use of perturbation theory still better accuracy would be feasible, but this is not done here.

Let the two possible energy levels of the molecule be denoted by E_m, and the corresponding states by $\psi_m (m=1,\ 2)$. Similarly, the number of quanta in the field oscillator will be n, and the corresponding state of the field by $\phi (n=0,\ 1,\ 2,\ \cdots)$. The state vectors $\psi_m\phi_n$ then form a basis for the system (molecule plus field). In this representation, the total Hamiltonian is

$$(mn\,|\,H\,|\,m'n') = (E_m + n\hbar\omega)\delta_{mm'}\delta_{nn'} + (mn\,|\,H_{\rm int}\,|\,m'n'). \quad (21)$$

The interaction Hamiltonian between molecule and field is taken of the form

$$H_{\rm int} = -\,\boldsymbol{\mu}\cdot\boldsymbol{E} \quad (22)$$

where $\boldsymbol{\mu}$ is the electric dipole moment of the molecule, whose component along $\boldsymbol{E}$ shall have the matrix elements

$$(mn\,|\,\mu_z\,|\,m'n') = \mu(1-\delta_{mm'})\delta_{nn'}. \quad (23)$$

Combining this with (20), we obtain the matrix elements for the interaction energy

$$(mn\,|\,H_{\rm int}\,|\,m'n') = \hbar\alpha(1-\delta_{mm'})[\sqrt{n}\,\delta_{n,n'+1} + \sqrt{n+1}\,\delta_{n+1,n'}] \quad (24)$$

where

$$\alpha = \frac{\mu}{J_1}\sqrt{\frac{2\pi\omega}{\hbar V}} \quad (25)$$

is the interaction constant. Using the value [11] $\mu = 1.47\times10^{-18}$ esu for ammonia, and a cavity 10 cm long, we find $(\alpha/\omega) = 2.08\times10^{-10}$ or, $\alpha \approx 5.0$ cps.

The interaction Hamiltonian has matrix elements of two different types: $H_{\rm int} = V + W$, where

$$V_n = (1,\ n+1\,|\,V\,|\,2,\ n) = (2,\ n\,|\,V\,|\,1,\ n+1) = \hbar\alpha\sqrt{n+1}$$

and

$$W_n = (1,\ n\,|\,W\,|\,2,\ n+1) = (2,\ n+1\,|\,W\,|\,1,\ n) = \hbar\alpha\sqrt{n+1} \quad (26)$$

all other elements being zero. The term V cannot be treated as a perturbation, for its matrix elements connect "unperturbed" states with an energy separation $(E_2 - E_1 - \hbar\omega)$ which goes through zero as the cavity is tuned exactly on the natural line frequency. On the other hand, elements of W connect states with unperturbed energy separation $(E_2 - E_1 + \hbar\omega) \approx 2\hbar\omega$. Since in typical operation conditions $(n \approx 10^6)$ we have $W_n 2\hbar\omega < 10^{-7}$, we may treat W as a small perturbation, or even neglect it entirely. We thus write the Hamiltonian as

$$H = H_0 + W$$

in which the term $H_0 = (H_{\rm mol} + H_{\rm field} + V)$ must be diagonalized exactly. This is readily done, since H_0 has a "block form" consisting of many (2×2) matrices along the main diagonal. The eigenvalues and eigenfunctions of H_0, defined by $H_0\Phi_n^{\pm} = E_n^{\pm}\Phi_n^{\pm}$, are the ground state

$$E_0 = E_1 = \hbar\omega_0 \qquad \Phi_0 = \psi_1\phi_0 \quad (27)$$

and for $n>0$,

$$E_n^{\pm} = \hbar\omega_n^{\pm} = \tfrac{1}{2}[E_1 + E_2 + (2n-1)\hbar\omega] \pm \tfrac{1}{2}[(E_2 - E_1 - \hbar\omega)^2 + 4n\hbar^2\alpha^2]^{1/2}. \quad (28)$$

We find it convenient now to define our zero molecular energy midway between the levels E_1 and E_2 such that

$$E_1 + E_2 = 0 \qquad E_2 - E_1 = \hbar\Omega$$

so that (28) now reads

$$E_n^{\pm} = \hbar\omega_n^{\pm} = (n-\tfrac{1}{2})\hbar\omega \pm \frac{\hbar}{2}[(\Omega-\omega)^2 + 4n\alpha^2]^{1/2}. \quad (28a)$$

Now

$$\Phi_n^{+} = \psi_2\phi_{n-1}\cos\theta_n + \psi_1\phi_n\sin\theta_n$$
$$\Phi_n^{-} = -\,\psi_2\phi_{n-1}\sin\theta_n + \psi_1\phi_n\cos\theta_n \quad (29)$$

where

$$\tan 2\theta_n = \frac{2\alpha\sqrt{n}}{\omega-\Omega}. \quad (30)$$

We now require the time-development matrix (in units with $\hbar=1$)

$$U(t,t') = U(t-t') = \exp[-iH(t-t')] \quad (31)$$

for which the perturbation expansion is

$$U(t) = e^{-iH_0t} - i\int_0^t e^{i(t-t')H_0}We^{it'H_0}dt' + \cdots. \quad (32)$$

The major term $U_0 = \exp(-iH_0 t)$ has the matrix elements, for $n > 0$,

$$(2, n-1 \mid U_0 \mid 2, n-1) = a_n = \cos^2\theta_n e^{-i\omega_n^+ t} + \sin^2\theta_n e^{-i\omega_n^- t}$$

$$(2, n-1 \mid U_0 \mid 1, n) = b_n = \sin\theta_n \cos\theta_n (e^{-i\omega_n^- t} - e^{-i\omega_n^+ t})$$

$$(2, n \mid U_0 \mid 2, n-1) = b_n$$

$$(1, n \mid U_0 \mid 1, n) = c_n = \cos^2\theta_n e^{-i\omega_n^- t} + \sin^2\theta_n e^{-i\omega_n^+ t} \quad (33)$$

and, for $n = 0$,

$$(1, 0 \mid U_0 \mid 1, 0) = e^{-i\omega_0 t} \quad (34)$$

where now $\omega_0 = -\Omega/2$ all other elements vanish. The transition probability for emission or absorption of one photon during time t is therefore, neglecting terms in W,

$$|b_n|^2 = \sin^2 2\theta_n \sin^2(\omega_n^+ - \omega_n^-)t/2 = \frac{n\alpha^2 \sin^2 \beta t}{\beta^2} \quad (35)$$

where

$$4\beta^2 = (\omega - \Omega)^2 + 4n\alpha^2. \quad (36)$$

The above notation has been chosen in such a way that the block form of U_0 consists of the symmetric, (2×2) unitary matrices

$$\begin{bmatrix} a_n & b_n \\ b_n & c_n \end{bmatrix} \qquad n = 1, 2, \cdots$$

along the main diagonal. The first row and column, however, contain only the single term (34).

We now consider the effect on the field of passing a single molecule through the cavity, with flight time τ. At the instant $(t = 0)$ when the molecule enters the cavity, let its state be described by the density matrix $\rho_1(0)$, and the state of the field by the density matrix $\rho_f(0)$. The initial density matrix fo the entire system is thus the direct product $\rho(0) = \rho_1(0) \times \rho_f(0)$, with matrix elements

$$(mn \mid \rho(0) \mid m'n') = (m \mid \rho_1(0) \mid m')(n \mid \rho_f(0) \mid n'). \quad (37)$$

During the interaction, ρ undergoes a unitary transformation

$$\rho(t) = U(t, 0)\rho(0)U^{-1}(t, 0) \quad (38)$$

and the density matrix $\rho_f(t)$, which describes the state of the field only, is the projection[1] of (38) onto the space of the field variables

$$(n \mid \rho_f(t) \mid n') = \sum_m (mn \mid \rho(t) \mid mn'). \quad (39)$$

[1] This formalism is developed in detail by Jaynes [12].

The net change in the state of the field thus consists of a linear transformation,

$$(n \mid \rho_f(t) \mid n') = \sum_{k,k'} (nn' \mid G \mid kk')(k \mid \rho_f(0) \mid k') \quad (40)$$

or

$$\rho_f(\tau) = G\rho_f(0) \quad (41)$$

$$(nn' \mid G \mid kk') = \sum_{m,m',m''} (m''n \mid U \mid mk)(m'k' \mid U^{-1} \mid m''n')\sigma_{mm'} \quad (42)$$

where we have written for brevity

$$\sigma_{mm'} \equiv (m \mid \rho_1(0) \mid m'). \quad (43)$$

The sums (42) are readily evaluated with the use of (33), with the result that the only nonvanishing elements of G are

$$(nn' \mid G \mid n, n') = a_{n+1}a^*_{n'+1}\sigma_{22} + c_n c^*_{n'}\sigma_{11} \quad (44a)$$

$$(nn' \mid G \mid n+1, n') = b_{n+1}a^*_{n'+1}\sigma_{12} \quad (44b)$$

$$(n, n' \mid G \mid n, n'+1) = a_{n+1}b^*_{n'+1}\sigma_{21}, \quad (44c)$$

$$(n, n' \mid G \mid n, n'-1) = c_n b^*_{n'}\sigma_{12}, \quad (44d)$$

$$(n, n' \mid G \mid n-1, n') = b_n c^*_{n'}\sigma_{12}, \quad (44e)$$

$$(n, n' \mid G \mid n+1, n'+1) = b_{n+1}b^*_{n'+1}\sigma_{11}, \quad (44f)$$

$$(n, n' \mid G \mid n-1, n'-1) = b_n b^*_{n'}\sigma_{22}. \quad (44g)$$

These relations hold for all quantum numbers n if we understand that c_0 is not defined by (33) but by $c_0 = \exp(-i\omega_0 t)$.

To illustrate the use of this formalism, we discuss a few simple problems using (44). Consider first the case where the field is initially in its lowest state; $(0 \mid \rho_f(0) \mid 0) = 1$, all other elements of $\rho_f(0)$ vanish. Then according to (44), after a molecule with initial density matrix σ has passed through, the field density matrix has elements

$$\begin{aligned} (0 \mid \rho_f(\tau) \mid 0) &= |a_1|^2\sigma_{22} + \sigma_{11} \\ (0 \mid \rho_f(\tau) \mid 1) &= (1 \mid \rho_f(\tau) \mid 0)^* = c_0 b_1^* \sigma_{12} \\ (1 \mid \rho_f(\tau) \mid 1) &= |b_1|^2\sigma_{22}, \end{aligned} \quad (45)$$

all other elements still vanishing. If the molecule were initially in its lowest state then nothing happens, and the field remains in its ground state. If the molecule was initially in the upper state $[\sigma_{22} = 1,\ \sigma_{11} = \sigma_{12} = 0]$ we have a simple transition probability of $|b_1|^2$ for the molecule to emit one photon in passing through. If there was initially no coherence relation between upper and lower states of the molecule, then $\sigma_{12} = 0$, and ρ_f remains diagonal; no coherence between states $n = 0$ and $n = 1$ can be set up by the molecule unless there was some coherence initially between upper and lower states of the molecule.

The expectation value of electric field along the axis of the cavity, as obtained from (20), is

$$\begin{aligned}\langle E\rangle &= \text{Trace}\,(\rho_f E)\\ &= -\frac{\hbar\alpha}{\mu}\sum_n \sqrt{n+1}[(n\,|\,\rho_f\,|\,n+1)+(n+1\,|\,\rho_f\,|\,n)]\\ &= -\frac{2\alpha\hbar}{\mu}\,\text{Re}\sum_n \sqrt{n+1}(n\,|\,\rho_f\,|\,n+1). \qquad (46)\end{aligned}$$

This remains zero as long as there is no coherence among adjacent levels, even though the energy stored in the field may be large. In the case (45), we obtain for $\langle E\rangle$,

$$\begin{aligned}\langle E\rangle &= -\frac{2\alpha\hbar}{\mu}\,\text{Re}\,(c_0 b_1^*\sigma_{12})\\ &= 2\hbar\alpha^2\frac{\sin\beta t}{\mu\beta}\,\text{Re}\left[i\sigma_{12}e^{i(\Omega+\omega)t/2}\right] \qquad (47)\end{aligned}$$

where β is defined by (36) with $n=1$. Suppose now the cavity is so tuned that its resonant frequency ω is equal to Ω, then $\beta=\alpha$ and we obtain simply

$$\langle E\rangle = \frac{2\hbar\alpha}{\mu}\sin\alpha t\,\text{Re}\left[i\sigma_{12}e^{i\omega t}\right]. \qquad (47a)$$

Since $\alpha\approx 5$ cps, the term $\sin(\alpha t)$ reaches its first maximum in a quarter cycle, or about 1/20 of a second. This is the interaction time required for a molecule to emit a photon, with probability one, into a lossless cavity initially in its ground state. This shows the great enhancement of spontaneous emission probability due to the presence of the resonant cavity, for the same molecule in empty space would emit with a natural line width (full width at half-maximum intensity),

$$\Delta\omega = \frac{8\omega^3\mu^2}{3\hbar c^3} \approx 10^{-7}\ \text{sec}^{-1}, \qquad (48)$$

which leads to spontaneous emission times of the order of months at the frequencies here considered.

If the molecule and field are in arbitrary initial states, the general transformation of the field caused by passage of the molecule is, from (44),

$$\begin{aligned}&(n\,|\rho_f(t)\,|n')\\ &=\sigma_{11}[b_{n+1}b^*_{n'+1}(n+1\,|\,\rho_f(0)\,|\,n'+1)+c_nc^*_n(n\,|\,\rho_f(0)\,|\,n')]\\ &+\sigma_{12}[b_{n+1}a^*_{n+1}(n+1\,|\,\rho_f(0)\,|\,n')+c_nb^*_{n'}(n\,|\,\rho_f(0)\,|\,n'-1)]\\ &+\sigma_{12}[a_{n+1}b^*_{n'+1}(n\,|\,\rho_f(0)\,|\,n'+1)+b_nc^*_{n'}(n-1\,|\,\rho_f(0)\,|\,n')]\\ &+\sigma_{22}[a_{n+1}a^*_{n'+1}(n\,|\,\rho_f(0)\,|\,n')\\ &\qquad +b_nb^*_{n'}(n-1\,|\,\rho_f(0)\,|\,n'-1)]. \qquad (49)\end{aligned}$$

If the field density matrix is initially diagonal,

$$(n\,|\,\rho_f(0)\,|\,n') = \rho_n\delta_{nn'}. \qquad (50)$$

The only nonvanishing components of $\rho_f(t)$ are

$$\begin{aligned}(n\,|\,\rho_f(t)\,|\,n+1) &= \sigma_{11}[|\,b_{n+1}|^2\rho_{n+1}+|\,c_n|^2\rho_n]\\ &+\sigma_{22}[|\,a_{n+1}|^2\rho_n+|\,b_n|^2\rho_{n-1}] \qquad (51)\end{aligned}$$

and

$$\begin{aligned}(n\,|\,\rho_f(t)\,|\,n+1) &= (n+1\,|\,\rho_f(t)\,|\,n)^*\\ &= \sigma_{12}[b_{n+1}a^*_{n+2}\rho_{n+1}+c_nb^*_{n+1}\rho_n]. \qquad (52)\end{aligned}$$

These relations will be used in the next section.

C. Successive Single-Molecule Interactions

If several molecules pass through the cavity in succession, the Nth entering as the $(N-1)$th leaves, all with the same initial state, this generates a Markov chain,

$$\rho_f(N\tau) = G^N\rho_f(0) = G\rho_f(N\tau-\tau). \qquad (53)$$

Of particular interest is the limit $N\to\infty$.

If the density matrices of field and molecule are initially diagonal,

$$\sigma_{12}=\sigma_{21}=0 \quad (n\,|\,\rho_f(0)\,|\,n') = \rho_n\delta_{nn'}, \qquad (54)$$

then ρ_f remains diagonal for all time. In this case the entering molecules can always be described by a temperature, defined by

$$\begin{aligned}\sigma_{22} &= \sigma_{11}e^{-x} = (e^x+1)^{-1}\\ x &= \hbar\Omega/kT \qquad (55)\end{aligned}$$

and, using (51), (53) reduces to

$$\begin{aligned}\rho_n(N\tau) &= (e^x+1)^{-1}[(|\,a_{n+1}|^2+|\,c_n|^2e^x)\rho_n(N\tau-\tau)\\ &+|\,b_{n+1}|^2e^x\rho_{n+1}(N\tau-\tau)+|\,b_n|^2\rho_{n-1}(N\tau-\tau)]. \qquad (56)\end{aligned}$$

From this the limiting form of ρ_n may be found. Taking note of the fact that fact that $|a_n|^2+|b_n|^2=|b_n|^2+|c_n|^2=1$, we find that a necessary and sufficient condition for a steady state $\rho_n(N\tau)=\rho_n(N\tau-\tau)=\rho_n$, is that the quantities

$$B_n = |\,b_n|^2(\rho_{n-1}-e^x\rho_n)$$

be independent of n. Now $\Sigma_n\rho_n=1$, and so $\rho_n\to 0$ as $n\to\infty$. Consequently, $B_n\to 0$, since $|b_n|^2\le 1$. Thus B_n can be independent of n only if $B_n=0$, and the only steady-state solution is the Boltzmann distribution,

$$\rho_n = e^{-x}\rho_{n-1}, \qquad (57)$$

for all n for which $|b_n|^2\neq 0$. From (35) it is seen that b_n could vanish only for isolated special values of n.

Note that (57) is not a Boltzmann distribution with the same temperature T as that of the molecules, except in the case where the cavity is tuned exactly to the natural line frequency. The temperature of (57) is $T_f=\omega T/\Omega$. This difference would never be seen in practice because as soon as one detunes the cavity appreciably the transition probability $|b_n|^2$ becomes ex-

tremely small, and the temperature of the radiation would be determined by its interaction with the walls of the cavity, here neglected.

Nevertheless, in principle the difference is there, and we have an example of an interaction between two systems which maintains them at different temperatures. The origin of the phenomena lies in the fact that we have described the state of the molecule in terms of a temperature, which is not wholly justified, since nothing was said about their kinetic energy of translational motion. It is this translational motion which supplies or absorbs the excess energy so as to remove the above apparent violation of energy conservation. When a molecule enters or leaves the cavity it passes through a region of inhomogeneous field, and experiences a net force which very slightly changes its velocity.

In the "negative temperature" case where the entering molecules are more likely to be in the upper state, $\sigma_{22}>\sigma_{11}$, and $x<0$, the solution $B_n=$constant is still formally the only stationary one. But it now represents an infinite amount of energy in the field and could never be reached by any finite number of molecules passing through the cavity. It is, of course, only our neglect of losses which leads to such a result, and in practice the operating level quickly reaches a steady value which can be predicted by adding a phenomenological damping term to $\dot{\rho}$ in a well-known way.

As long as the density matrix σ of the entering molecules is diagonal, the density matrix of the field alone also remains diagonal; the expectation value of the electric field remains zero in spite of the fact that the number of photons present may be very large. That is, $\langle E^2\rangle$ can be very large but $\langle E\rangle$ remains zero. This is more or less to be expected since the entering molecules do not "tell" the field what phase to have. This situation raises certain questions, however, regarding the relation between quantum theory and classical theory. It is usually supposed that the condition for validity of classical electromagnetic theory is simply that the number of photons in each normal mode is large, and that then one may identify the classical electromagnetic field with the quantum-mechanical expectation value. It is seen, however, that this is a necessary but not sufficient condition, for here we have a situation where the semiclassical theory of radiation could not describe such states.

The statement, found in most books on quantum theory, that in the limit of large quantum numbers, quantum theory goes over into classical theory is somewhat misleading. Actually it is possible by coherent superposition of quantum states to construct states which are not describable in terms of classical theory at all. Thus it is that we arrive at the conclusion that classical theory is but a special case of quantum theory in the case of large quantum numbers, *i.e.*, large quantum numbers are necessary but not sufficient to insure the transition from quantum to classical theory.

The case in which two molecules pass through the cavity with flight time τ and leave just as two others enter, etc., has been worked out. The mathematics is tedious, and the result is substantially the same as the successive single-molecule interaction case.

III. Relation Between Quantum Electrodynamics and Semiclassical Radiation Theory

A. Semiclassical Electrodynamics

Now one considers that the electric field $E(t)$ is classically describable, and introduces a wave function,

$$\psi(t) = a(t)\psi_1 + b(t)\psi_2, \tag{58}$$

for the molecule alone, which develops in time according to the Schrödinger equation

$$i\hbar\dot{\psi} = (H_{\text{mol}} + H_{\text{int}})\psi \tag{59}$$

where

$$(m \mid H_{\text{mol}} \mid m') = E_m\delta_{mm'} \tag{60}$$

and

$$\begin{aligned}(m \mid H_{\text{int}} \mid m') &= (m \mid -\mu\cdot E(t) \mid m') \\ &= -\mu(1-\delta_{mm'})E(t).\end{aligned} \tag{61}$$

Schrödinger's equation (59) then reduces to

$$\begin{aligned}i\hbar\dot{a} &= E_1a - \mu E(t)b \\ i\hbar\dot{b} &= -\mu E(t)a + E_2b.\end{aligned} \tag{62}$$

These equations describe the effect of the field on the molecule.

Semiclassical theory as usually treated does not consider the effect of the molecule on the field. To find the effect of the molecule on the field, one calculates the expectation value of the dipole moment of the molecule from the solution of (62),

$$M(t) \equiv \langle\mu\rangle(t) = \mu(ab^* + ba^*), \tag{63}$$

and assumes that the field satisfies the classical equations of motion which would result from interaction with a dipole of moment $M(t)$. This is obtained most easily from the Hamiltonian equations of motion by addition of the interaction energy

$$-M\cdot E = +\sqrt{4\pi}\sum_a p_a(t)E_a(\mathbf{x})\cdot M(t) \tag{64}$$

to $\mathcal{H}$ in (7) of Section II, where $\mathbf{x}$ denotes the position of the molecule. The classical equations of motion are now

$$\dot{p}_a = -\frac{\partial\mathcal{H}}{\partial q_a} = -\omega_a^2 q_a$$

and

$$\dot{q}_a = \frac{\partial\mathcal{H}}{\partial p_a} = p_a + \sqrt{4\pi}\,M\cdot E_a(\mathbf{x}). \tag{65}$$

Eliminating q_a,

$$\ddot{p}_a + \omega_a^2 p_a = -\sqrt{4\pi}\,\omega_a^2 \boldsymbol{M}\cdot\boldsymbol{E}_a(\mathbf{x}). \tag{66}$$

Assuming that we have only one normal mode excited, the electric field of this mode satisfies the differential equation

$$\ddot{E} + \omega^2 E = \frac{+4\pi\omega^2}{J_1^2 V} M \tag{67}$$

where again we drop the subscript a. If the cavity has a finite Q, due to wall losses and/or energy coupled out, this is taken into account by adding a phenomenological damping term to (67), giving us

$$\ddot{E} + \frac{\omega}{Q}\dot{E} + \omega^2 E = \frac{4\pi\omega^2 M}{J_1^2 V}. \tag{68}$$

By the "semiclassical" theory we mean the system of equations (62), (63) and (68). They may be given a somewhat neater formal appearance by eliminating the amplitudes $a(t)$, $b(t)$. The result is the nonlinear system of coupled equations,

$$\ddot{M} + \Omega^2 M = -K^2 W E, \tag{69a}$$

$$\dot{W} = E\dot{M} \tag{69b}$$

and

$$\ddot{E} + \omega/Q\dot{E} + \omega^2 E = SM, \tag{69c}$$

where

$$K = 2\mu/\hbar \qquad S = 4\pi\omega^2/J_1^2 V \tag{70}$$

and

$$W = E_1|\,a\,|^2 + E_2|\,b\,|^2 - \tfrac{1}{2}(E_1 + E_2)$$

$$= \frac{\hbar\Omega}{2}\left(|\,b\,|^2 - |\,a\,|^2\right) \tag{71}$$

is the expectation value of energy of the molecule, referred to a zero lying midway between the levels E_1, E_2. In the form (69) we have an apparently classical nonlinear system, all reference to "quantum-mechanical" quantities having disappeared.

The first two equations of (69) admit a first integral,

$$\dot{M}^2 + \Omega^2 M^2 + K^2 W^2 = \text{const.} = \left(\frac{K\hbar\Omega}{2}\right)^2. \tag{72a}$$

This is readily verified by eliminating E between them. Eq. (72a) is a disguised form of the principle of conservation of probability, $|\,a\,|^2 + |\,b\,|^2 = 1$. Similarly, the last two equations of (69) can be combined, in the case $Q = \infty$, to yield the constant of the motion

$$\dot{E}^2 + \omega^2 E^2 + 2S(W - ME) = \text{constant}, \tag{72b}$$

which is easily identified as the conservation of energy statement for the system.

B. The Relation Between Semiclassical and Quantum Electrodynamic Equations of Motion

For the equation of motion of any quantum-mechanical operator we have $i\hbar\dot{F} = [F, H]$. Differentiating this, we have

$$\hbar^2\ddot{F} + [H, [H, F]] = i\hbar[\dot{H}, F] \tag{73}$$

which is exact for any operator F which has no explicit time dependence. Let us apply this identity to the electric field operator $F = E$. The total Hamiltonian $H = (H_{\text{mol}} + H_{\text{field}} + H_{\text{int}})$ has no explicit time dependence, so the right-hand side of (73) will vanish. To evaluate the double commutator, we note that H_{int} commutes with E but not with $[H_f, E]$, while H_m commutes with both. Therefore,

$$[H, [H, E]] = [H_f, [H_f, E]] + [H_{\text{int}}, [H_f, E]]. \tag{74}$$

These commutators are easily worked out, and the result is

$$[H_f, [H_f, E]] = \hbar^2\omega^2 E \tag{75}$$

$$[H_{\text{int}}, [H_f, E]] = -\hbar^2 S\mu_{\text{op}}. \tag{76}$$

Thus a special case of (73) is the operator identity

$$\ddot{E} + \omega^2 E = S\mu_{\text{op}} \tag{77}$$

which is to be compared to (12c). If we interpret (12c) as the expectation value of (20), they are seen to be identical in the limit $Q \to \infty$, provided that the expectation value of μ_{op} be defined, not in terms of $a(t)$ and $b(t)$ by means of (6), but as the expectation value taken over the complete density matrix $(mn/\rho/m'n')$, *i.e.*,

$$\langle\mu_{\text{op}}\rangle = \text{Tr}\,(\rho\mu_{\text{op}}) = \sum_{nmm'} (mn\rho m'n)(m'\,|\,\mu_{\text{op}}\,|\,m). \tag{78}$$

With this change in interpretation (69c) is seen to be an exact consequence of quantum electrodynamics.

We now write out the identity (73) for the operator $F = \mu_{\text{op}}$. This time H_{int} commutes with μ_{op} but not with $[H_m, \mu_{\text{op}}]$, while H_f commutes with both. Therefore,

$$[H, [H, \mu_{\text{op}}]] = [H_m, [H_m, \mu_{\text{op}}]] + [H_{\text{int}}, [H_m, \mu_{\text{op}}]]. \tag{79}$$

Proceeding as before, a short calculation yields the following results:

$$[H_m, [H_m, \mu_{\text{op}}]] = \hbar^2\Omega^2\mu_{\text{op}} \tag{80}$$

and

$$[H_{\text{int}}, [H_m, \mu_{\text{op}}]] = \hbar^2 K^2 H' E \tag{81}$$

where we have defined an operator

$$H' = H_{\text{mol}} - \tfrac{1}{2}(E_1 + E_2) \tag{82}$$

with matrix elements

$$(mn\,|\,H'\,|\,m'n') = \frac{\hbar\Omega}{2}(-1)^m\delta_{mm'}\delta_{nn'}, \tag{83}$$

which is just the energy of the molecule, referred to a zero lying midway between its levels E_1, E_2. Combining these relations, we find that another special case of (73) is the operator identity

$$\ddot{\mu}_{op} + \Omega^2\mu_{op} = -K^2H'E \tag{84}$$

which is to be compared to (69a). However, now when we take the expectation value of (84) we do not get (69a) in general, for in the semiclassical equation the "driving term" appears as $\langle H'\rangle\langle E\rangle$, while quantum electrodynamics yields $\langle H'E\rangle$. The difference between these terms arises from the possibility of having correlated states, a situation inherent in quantum electrodynamics but not in semiclassical theory. When the states of field and molecule are uncorrelated, the density matrix reduces to a direct product $\rho = \rho_m \rho_f$, or

$$(mn \mid \rho \mid m'n') = (m \mid \rho_m \mid m')(n \mid \rho_f \mid n') \tag{85}$$

when (85) holds, then $\langle H'E\rangle = \langle H'\rangle\langle E\rangle$. But in general, $\langle H'E\rangle \neq \langle H'\rangle\langle E\rangle$.

The possibility of obtaining "correlated states" can arise whenever two or more quantum-mechanical systems interact. Quantum electrodynamics allows the possibility of states of the combined system (molecule plus field) which are in a definite pure state, but nevertheless one cannot ascribe any definite quantum state to the molecule alone, or the field alone. This possibility forms the basis of one of Einstein's objections to quantum mechanics. The Einstein-Podolsky-Rosen [13] paradox consists of the fact that when such correlated states exist, one has the possibility of predicting with certainty either one of two noncommuting quantities of a system by making measurements which do not involve any physical interaction with it.

An interesting line of thought is based on the fact that the semiclassical theory and quantum electrodynamics predict different equations of motion for a molecule in the field, the difference arising just from those correlated states which cause the above conceptual difficulties. Thus if one could find any experimental situation in which the difference between $\langle H'E\rangle$ and $\langle H'\rangle\langle E\rangle$ leads to any observable difference in maser operations, this would constitute an indirect, but convincing, check on those aspects of quantum theory which lead to the Einstein-Podolsky-Rosen paradox. However, as will be shown, the prospects of detecting such a difference are extremely dubious, for we will see that the semiclassical theory actually reproduces many of the features which one commonly supposes can be found only with field quantization.

C. Solution of Nonlinear Semiclassical Equations

The simplest approximate solution of the coupled semiclassical equations is the one wherein we ignore the time variation of W, thereby converting the problem into a linear one, similar to the case of two coupled pendulums. The normal modes are found by assuming that E and M have a common time factor $\exp(i\nu t)$; if W = constant, then (69a) and (69c) reduce to

$$(\omega^2 - \nu^2)(\Omega^2 - \nu^2) + K^2SW = 0 \tag{86}$$

or

$$\nu^2 = \frac{\omega^2 + \Omega^2}{2} \pm \frac{1}{2}\sqrt{(\omega^2 - \Omega^2)^2 - 4K^2SW}. \tag{87}$$

We see here a new feature, not present in coupled pendulums. If $W>0$ and the cavity is tuned so closely to the natural line frequency that

$$|\omega^2 - \Omega^2| < \sqrt{4K^2SW}, \tag{88}$$

the square root in (87) becomes imaginary; one of the normal modes grows exponentially, the other decays. Now an oscillation of growing amplitude represents energy being transferred from molecule to field, and therefore we see that the semiclassical theory *does* lead to a prediction of spontaneous emission. Since W is just the energy of the molecule, we see that the condition of unstable growing oscillation is just that the molecule's wave function contains more of the upper state than the lower, $|b|^2 > |a|^2$.

Suppose that the cavity is tuned exactly to the natural line frequency, $\omega = \Omega$. Then (87) reduces to

$$\nu^2 = \omega^2 \pm i\sqrt{K^2SW} \tag{89}$$

or to an extremely good approximation,

$$\nu = \omega \pm \frac{i\sqrt{K^2SW}}{2\omega}. \tag{90}$$

If we start with the molecule nearly in the upper state then $W = \hbar\Omega/2$ and the amplitude of the field varies like

$$\exp\left(\frac{\sqrt{K^2SW}}{2\omega}t\right)e^{i\omega t} = \exp \alpha t e^{i\omega t} \tag{91}$$

where α is the interaction constant defined in (25). This is to be compared to the result (47a) describing spontaneous emission according to quantum electrodynamics. It is seen that although the two approaches lead to equations of different functional form, they predict exactly the same characteristic time $1/\alpha$ for spontaneous emission. This shows that the relation between quantum electrodynamics and the semiclassical theory of radiation is quite different from what is usually supposed. Physically, it means that whenever the molecule has a dipole moment different from zero, the fields set up by this dipole react back on the molecule and change its state in such a way that energy is delivered to the field, as long as $W>0$. These linear relations do not hold indefinitely, of course. From the conservation law (72a) it is clear that when the amplitude of the M

oscillation increases, the magnitude of W must decrease, and this will eventually put a stop to the emission process.

The change in time of the variable W, for any reasonable value of field strength, as we are concerned with here, is slow compared to the time variation of E or M, and for typical ammonia maser operating conditions, E and M go through the order of 10^7 cycles for each cycle of W. For a qualitative picture of the slow changes in the case $\omega=\Omega$, we may consider the orbits in the $(\dot{E}, \omega E)$ plane and in the $(\dot{M}, \omega M)$ plane, as in Fig. 1. Noting that the interaction energy is typically about 10^{-6} times smaller than the energy of the molecule W, the conservation of energy law (72b) reduces, in almost all cases, to

$$(\dot{E})^2 + \omega^2 E^2 + 2SW = \text{constant}, \tag{92}$$

which shows that as W increases, the orbit in the $(\dot{E}, \omega E)$ plane must shrink, and vice versa. Also, the conservation law (72a) shows that if $|W|$ increases, the M orbit must shrink, and vice versa. Therefore the direction of all secular changes is determined by the sign of W and $\dot{W}$. In the equation $\dot{W}=E\dot{M}$ we can for all practical purposes replace $E\dot{M}$ by its average over one cycle, $\overline{E\dot{M}}$, since we are interested in the trend of W over time scales of many cycles, rather than small rapid fluctuations whose effect averages to zero over a cycle. Secular changes in W depend, thus, only on the sign of $\overline{E\dot{M}}$.

Whenever the E motion is advanced in phase over the M motion, we have $\overline{E\dot{M}}\geq 0$. In this case, W will slowly increase and the E orbit will shrink. The M orbit will then grow if $W<0$, shrink if $W>0$. If the M motion is advanced in phase over the E motion, all these changes are reversed. The situation is summarized by the orbit diagrams of Fig. 2. Or again, let us assume that W is given as some periodic function of time, so that we can summarize these same conclusions graphically as in Fig. 3.

Whenever the E orbit is expanding, energy is being delivered from the molecule to the field, and the necessary and sufficient condition for this is that the M motion be advanced in phase over the E motion. Thus in order to understand the long time course of events, one must study the secular changes in relative phases of the E and M motion.

To this end introduce the slowly varying complex amplitudes X and Y, defined by

$$\dot{E} + i\omega E = X(t)e^{i\omega t} \tag{93}$$

and

$$M + i\omega M = Y(t)e^{i\omega t}. \tag{94}$$

The quantities depicted in Fig. 2 are just the complex numbers (93) and (94). Noting the properties,

$$(\dot{E})^2 + \omega^2 E^2 = |X|^2 \tag{95}$$

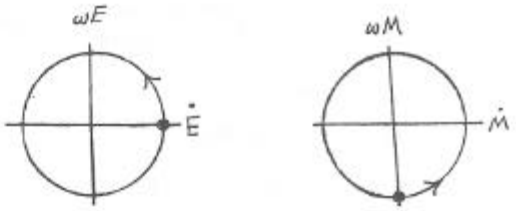


Fig. 1—Closed orbits in the phase space of the E and M oscillators. The dots indicate that the E motion is 90° ahead of the M motion in the phase.

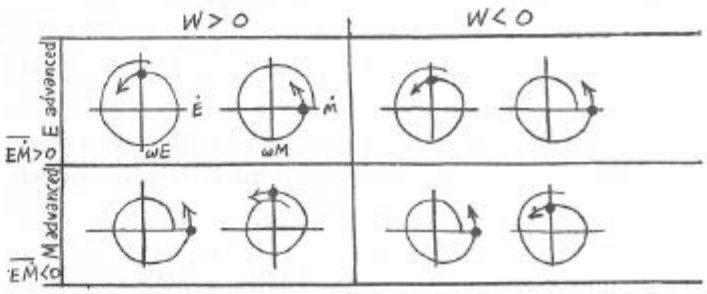


Fig. 2—Secular changes in orbits for the four combinations of signs of $\overline{E\dot{M}}$ and W.

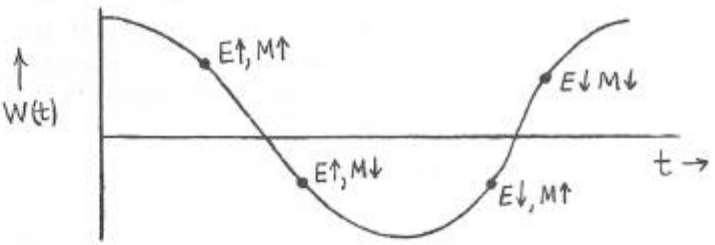


Fig. 3—Representation of the orbit diagrams in another way.

and

$$\ddot{E} + \omega^2 E = \dot{X}e^{i\omega t} \tag{96}$$

and similarly for M, we can write the equations of motion (69) in the form, for the case $\omega=\Omega$,

$$2i\omega\dot{X} = S(Y - Y^*e^{-2i\omega t}), \tag{97a}$$

$$2i\omega\dot{Y} = -K^2W(X - X^*e^{-2i\omega t}), \tag{97b}$$

$$4i\omega\dot{W} = XYe^{2i\omega t} + XY^* - X^*Y - X^*Y^*e^{-2i\omega t}. \tag{97c}$$

The conservation laws become

$$|Y|^2 + K^2W^2 = \text{constant} = \left(\frac{K\hbar\Omega}{2}\right)^2 \tag{98a}$$

and

$$|X|^2 + 2SW = \text{constant}. \tag{98b}$$

Now the functions X and Y are slowly varying functions of time, and again it is their average change over many cycles, rather than the very small rapid fluctuations at frequency 2ω, which are of interest. Thus the oscillating terms in (97) can be dropped, since their average over a cycle is negligible compared to their dc components.

The system of equations determining secular changes of both amplitude and phase is, therefore,

$$2i\omega\dot{X} = SY, \tag{99a}$$

$$2i\omega\dot{Y} = -K^2WX, \tag{99b}$$

$$4i\omega\dot{W} = XY^* - X^*Y. \tag{99c}$$

It is easily verified that the conservation laws (98) are exact consequences of (99). Differentiating (99c) once more and making use of the conservation laws, we can eliminate X and Y, obtaining the equation

$$4\omega^2\ddot{W} - 3SK^2W^2 + K^2CW + SK^2\left(\frac{\hbar\Omega}{2}\right)^2 = 0 \tag{100}$$

where C is the constant of the motion (98b). A first integral of (100) can be obtained immediately by multiplication with $\dot{W}$ and integrating

$$2\omega^2(\dot{W})^2 - SK^2W^3 + \frac{K^2CW^2}{2} + SK^2\left(\frac{\hbar\Omega}{2}\right)^2 W = \text{constant}. \tag{101}$$

This equation has the form of the Hamilton-Jacobi equation for motion of a particle in a particular potential well. For any motion in which either of the points $W=\pm(\hbar\Omega/2)$ is accessible, we have the constant on the right-hand side of (101) equal to

$$\frac{K^2C}{2}\left(\frac{\hbar\Omega}{2}\right)^2.$$

This is easily seen from (98a), for if $W=\pm(\hbar\Omega/2)$, then $Y=0$ and $\dot{W}=0$. For any such motion the cubic polynomial in (101) factors. To see this most easily, introduce the change of variable $(\hbar\Omega/2)z=W$. Then (101) takes the form,

$$b\dot{z}^2 - (z^3 - z - az^2 + a) = b\dot{z}^2 - [(z-1)(z+1)(z-a)] = 0, \tag{102}$$

where

$$b = \frac{4\omega^2}{SK^2\hbar\omega} = \frac{1}{2\alpha^2} \qquad a = \frac{C}{S\hbar\omega}. \tag{103}$$

The solution is

$$\sqrt{2}\,\alpha t = \int_{z(0)}^{z(t)} \frac{dz}{\sqrt{(1-z)(1+z)(a-z)}}. \tag{104}$$

The z motion is therefore periodic between turning points represented by singularities of the integrand. If $a>1$, these turning points are at $z=\pm1$, while if $a<1$ they are at $z=-1$ and $z=a$. Now we consider evaluation of the constant $a=C/\hbar\omega S$. From (5), (9a) and (17) we have

$$\dot{E}^2 + \omega^2E^2 = \frac{4\pi\omega^2}{J_1^2V}(2n\hbar\omega)$$

where n is the number of photons stored in the cavity. Now, examination of (72b) with the small interaction term neglected gives us

$$\frac{4\pi\omega^2}{J_1^2V}(2n\hbar\omega) + S\hbar\omega = C = S\hbar\omega(2n+1)$$

if we assume there are n molecules in the field when the molecule is in its upper state, $W=+(\hbar\omega/2)$. Thus $a=(2n+1)$. There is in this theory of course no restriction on n to be an integer. The smallest value which a can attain is represented by zero energy in the field and the molecule in its ground state, $a=-1$, or $n=-1$. When n is negative this of course means that the total energy is insufficient for the molecule to get into its upper state, and this is the physical reason why the turning point of the z motion then occurs at $z=a$. The integral (104) is one of the standard forms defining elliptic functions. Using the standard notation sn (u, k), the solution for the case $n\geq0$ is

$$z(t) = -1 + 2sn^2\left(\sqrt{n+1}\,\alpha t + Q, \frac{1}{\sqrt{n+1}}\right) \tag{105}$$

where

$$Q \equiv sn^{-1}\left(\sqrt{\frac{z(0)+1}{2}}, \frac{1}{\sqrt{n+1}}\right) \tag{106}$$

is the initial phase of the motion. In the limit of large n, the elliptic functions approach trigonometric functions, as is seen most easily from (104). If $a\gg1$, then (104) reduces to

$$\sqrt{2}\,\alpha t \approx \frac{1}{\sqrt{a}}\int \frac{dz}{\sqrt{1-z^2}} = \frac{1}{\sqrt{a}}\sin^{-1}z(t) + \text{constant},$$

or

$$z(t) \approx \sin(2\sqrt{n}\,\alpha t + \theta). \tag{107}$$

The case $a=1$, $n=0$ is a special one, for the integrand of (104) then develops a first-order pole at $z=1$. The solution (105) is still valid but is no longer periodic; in fact sn $(u, 1)$ is equal to tanh u which approaches ±1 asymptotically as $u\to\pm\infty$. This represents a case where the energy in the field exactly disappears just as the molecule gets into its upper state, and the final stages of the solution then represent the "shrinking normal mode" of (90), where E is 90° ahead of M. (This phase relation is in fact maintained throughout the part of the motion (105) in which z increases. Throughout the decreasing part, E is 90° behind M.)

The point $W=\hbar\Omega/2$, $z=1$ is a metastable point of the orbit in this case, for if we start out with exactly the initial comditions $z=1$, $E=M=0$ then nothing happens. All time derivatives remain zero and the molecule does not emit. However if there is the slightest change in this initial condition, the growing normal mode of (91) will be started up (unless the phase relations between M and E happen to be just the value for the pure shrinking mode), and eventually the energy of the

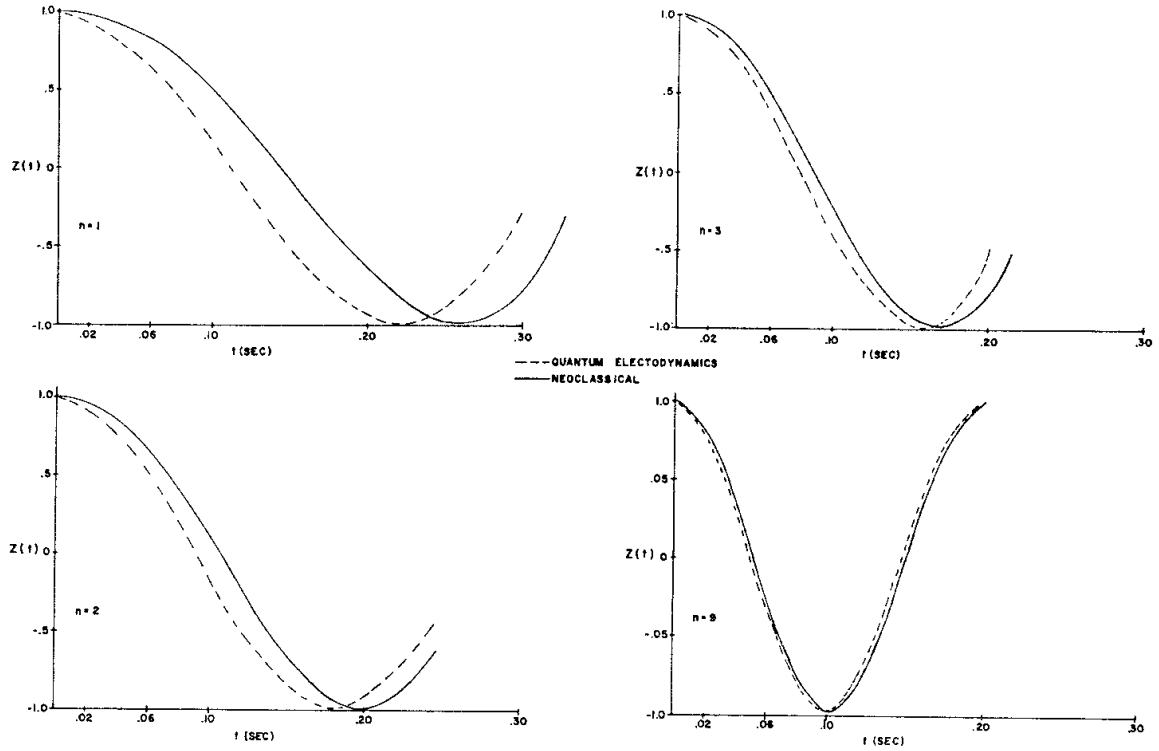


Fig. 4—Energy as a function of time for several n's.

molecule spills out entirely into the field when we reach the lower turning point $z=-1$. The molecule then reabsorbs the energy $\hbar\omega$ from the field, passing back to the metastable point $z=1$ but requiring an infinite time to do so. Fig. 4 shows $z(t)$ as a function of time for several values of the parameter n, the number of photons in the cavity, and these are compared with the corresponding quantum electrodyanmics curves. It is seen that for a few photons, the correspondence is almost exact. Even in the case of one or two quanta, the semiclassical theory gives solutions reproducing almost quantitatively everything that is found in the quantum electrodynamics analysis. Even the "quantum jumps" are still with us, but here they show up as perfectly continuous processes, where an instability develops in the solution of the nonlinear equations and an amount of energy $\hbar\omega$ is more or less rapidly transferred from molecule to field.

The semiclassical analysis gives a very interesting description of the process of spontaneous emission. Consider a large number of molecules, as nearly as possible in the upper state. In practice, of course, we cannot prepare them *exactly* in the upper state, but there will be a certain probability distribution of initial values of amplitude for the growing normal mode. A molecule with an initial value $M_+(0)$ will at time t have an M amplitude of $M_+(0)e^{\alpha t}=M_+(t)$.

If we agree to say that when this reaches the value K, the molecule is actively emitting energy, then, no matter what the probability distribution of initial values, provided only that this distribution is a continuous function in the neighborhood of the metastable point $W=\hbar\Omega/2$, we find that the number of molecules emitting at time t is proportional to $\exp(-2\alpha t)$. We can see this by a simple argument which runs as follows: We shall say that the molecule has reached stage K of the emission process when the amplitude of the oscillation reaches a value in the range, $K<M_+(0)e^{\alpha t}<(K+\delta K)$. We now ask, "How many atoms will be in stage K at time t?" Clearly, all those for which $M_+(0)$ lies in the range, $Ke^{-\alpha t}<M_+(0)<(K+\delta K)e^{-\alpha t}$. If the initial probability distribution in the phase space of the molecule is constant, this would be proportional to the area of the annular ring, $2\pi r dr=2\pi(Ke^{-\alpha t})(\delta Ke^{-\alpha t})\sim e^{-2\alpha t}$. Thus the "law of radioactive decay" or "time proportional transition probabilities" appears in this analysis as a consequence of the existence of metastable states. The time constant of the decay law is independent of the method of preparation of the molecules, and only depends on the interaction constant of the molecules with the electromagnetic field. The situation is exactly like that of a large number of pencils nearly perfectly balanced on their points. The time required for any one pencil to fall over depends on how close it was to vertical at $t=0$. If the probability distribution of initial states is continuous in the neighborhood of this metastable point, then we have a decay law with a time constant which depends only on the laws of mechanics, not on the method of preparation of initial states.

Mathematically, this semiclassical theory as expounded in this section is exactly the same as that already used by Shimoda, Wang and Townes [4]. The new feature is the realization that this formalism ac-

counts for effects which all standard textbooks describe as requiring field quantization for their explanation. Because of this success, and the fact that the correspondence with quantum electrodynamics continues to strengthen as this formalism is applied to a larger group of problems, it is felt that this formalism deserves independent status as a physical theory in its own right, and we suggest it be called the neoclassical theory of electrodynamics.

Conceptually, the neoclassical theory amounts to reinterpreting the quantities usually denoted as expectation values of energy and dipole moment as *actual* values, the latter serving as the source of classical electromagnetic fields. These fields are then inserted in the Hamiltonian of the molecule and the reaction of the molecule to the field calculated according to the Schrödinger equation. Thus a general problem would be governed by the set of coupled equations, $i\hbar\dot{\psi}=H(A_u)\psi$ and $\Box A_u+4\pi\langle j_u\rangle=0$, where $\langle j_u\rangle$ is the current density operator, the expectation value here being interpreted as a classical current density.

Having now convinced ourselves of the efficacy of this method, we now turn to the application of these equations to the problem of the ammonia beam maser.

IV. Application of Neoclassical Radiation Theory to the Ammonia Maser

A. Ideal Steady-State Solution

Our starting point for obtaining the ideal steady-state solution for the ammonia maser will be (69), *i.e.*,

$$\ddot{M}_i + \Omega^2 M_i = -K^2 W_i E(t) \tag{108a}$$

$$\ddot{E} + \omega^2 E + \frac{\omega}{Q}\dot{E} = SM_i \tag{108b}$$

$$\dot{W}_i = E\dot{M}_i \tag{108c}$$

where now the subscript i refers to the ith molecule. If now all of the molecules are subjected to the same field $E(t)$ as in the Stanford [6] ammonia maser, we can simply define the total moment and energy

$$M(t) \equiv \sum_i M_i(t), \tag{109a}$$

$$W(t) \equiv \sum_i W_i(t). \tag{109b}$$

We see that (108a)–(108c) are still satisfied by these quantitites, in particular,

$$\ddot{E} + \omega^2 E + \frac{\omega}{Q}\dot{E} = SM = S\sum_i M_i. \tag{110}$$

The conservation law (72a) is still valid in the sense that the left-hand side is still constant; but, of course, the value of the constant now depends on the initial conditions. Since the problem of N molecules in the cavity is hardly any more difficult in this formalism than that of one molecule, we can see the advantage of this formalism over the quantum electrodynamics approach, where two molecules in the cavity at a time would involve solving cubic equations, that of three molecules, quartic equations, and so forth.

As our first application, we consider the case where molecules enter the cavity all in the upper state, $W_i=\hbar\Omega/2$, with the same velocity and with a uniform rate of A molecules per unit of time. We wish to find how, under these circumstances, the steady-state frequency and amplitude of oscillation depend on the experimentally controllable parameters A, ω, Q.

Denote by t_i the time at which the ith molecule enters the cavity. It is readily verified by substitution that the solution of (108a) with the initial conditions $M_i(t_i)=\dot{M}_i(t_i)=0$ is

$$M_i(t) = -\frac{K^2}{\Omega}\int_{t_i}^{t} W_i(t')E(t')\sin\Omega(t-t')dt'. \tag{111}$$

Using this, (108c) can be written as an integral equation. A time integration yields

$$W_i(t) - W_i(t_i) = \int_{t_i}^{t} dt''E(t')\dot{M}(t')$$

$$= -K^2\int_{ti}^{t} dt''E(t')\int_{ti}^{t''} dt'W_i(t')E(t')\cos\Omega(t''-t'). \tag{112}$$

Interchanging the order of integration in (112), we find that $W_i(t)$ satisfies an integral equation of Volterra form,

$$W_i(t) - W_i(t_i) = \int_{t_i}^{t} G(t,t')W_i(t')dt', \tag{113}$$

with the kernel,

$$G(t,t') = -K^2\int_{t'}^{t} dt''E(t'')\cos\Omega(t''-t')E(t'). \tag{114}$$

We now assume the electric field is given by

$$E(t) = 2a\sin\nu t \tag{115}$$

where a and ν are parameters to be determined by the condition that (110), (111) and (113) be self-consistent. It is clear from (108c) that the exact solution of $W_i(t)$ contains terms oscillating at frequencies of the order of $(\Omega+\nu)$. While these terms may contribute appreciably to $\dot{W}_i$, their effect on W_i averages to zero in times of the order of one cycle of the RF. Since we are interested in the long time drift in W_i, rather than these small rapid fluctuations, we neglect terms in (114) of frequency $(\Omega+\nu)$. Their contribution to W_i is of the relative order of magnitude

$$\frac{(\Omega-\nu)}{(\Omega+\nu)} \lesssim 10^{-7}$$

in all cases of practical interest. With this approximation (114) reduces to

$$G(t,t') \approx -K^2a^2 \frac{\sin(\Omega-\nu)(t-t')}{(\Omega-\nu)} \tag{116}$$

and the slowly varying part of $W_i(t)$ satisfies the integral equation

$$\dot{W}_i(t) = W_i(t_i) - K^2a^2 \int_{t_i}^{t} W_i(t') \frac{\sin(\Omega-\nu)(t-t')}{(\Omega-\nu)} dt'. \tag{117}$$

The exact solution of (117) with the initial condition $W_i(t_i) = \hbar\Omega/2$ is

$$W_i(t) = \frac{\hbar\Omega}{2\lambda^2}\left[(\Omega-\nu)^2 + a^2K^2\cos\lambda(t-t_i)\right] \tag{118}$$

where

$$\lambda^2 \equiv (\Omega-\nu)^2 + (Ka)^2. \tag{119}$$

As a check, and an illustration of some of our previous remarks, we note that (118) and (119) agree with the results found from quantum electrodynamics, and (35) and (36), and also with the result of others who have treated the problem by direct integration of Schrödinger's equation.

The total dipole moment of all the moledules in the cavity is

$$\begin{aligned} M(t) &= A\int_{t-\tau}^{t} M_i(t)dt_i \\ &= -\frac{AK^2}{\Omega}\int_{t-\tau}^{t} dt'E(t')\sin\Omega(t-t') \\ &\quad\cdot\int_{t-\tau}^{t'} dt_i W_i(t') \end{aligned} \tag{120}$$

where we have used (111) and inverted the order of integration. With the solution (118) for $W_i(t)$, this becomes

$$\begin{aligned} M(t) = -\frac{Aa\hbar\Omega K^2}{\Omega\lambda^2}\int_0^{\tau}\left[(\Omega-\nu)^2 q + \frac{a^2K^2}{\lambda}\sin\lambda q\right] \\ \cdot\sin\nu(q+t-\tau)\sin\Omega(\tau-q)xdq \end{aligned} \tag{121}$$

where $q=(\tau-t+t')$. As a function of q, the last factor of the integrand contains oscillating terms of frequencies $(\Omega\pm\nu)$, and again the relative contribution of the high-frequency term will be of the order of 10^{-7} or smaller under all conditions of interest. Neglecting this small term, (121) reduces to

$$\begin{aligned} M(t) = \frac{Aa\hbar\Omega K^2}{\Omega\lambda^2}\Big[(1-\cos\lambda\tau)\cos\nu t \\ -\frac{(\Omega-\nu)}{\lambda}(\lambda\tau-\sin\lambda\tau)\sin\nu t\Big]. \end{aligned} \tag{122}$$

Details from this point on will be considered later, as a special case of a more general solution.

B. Velocity Distribution

In the preceding section, all the molecules were assumed to have the same flight time τ. To find the effect of a velocity distribution, we have only to note that the analysis leading to (122) is still valid, and it gives the contribution to total moment of those molecules with flight time in the range $d\tau$, provided that we replace A by $nf(\tau)d\tau$, where n is the total number of molecules entering the cavity per unit time, and $f(\tau)d\tau$ is the fraction of entering molecules with flight times in the range $d\tau$, normalized so that

$$\int_0^{\infty} f(\tau)d\tau = 1. \tag{123}$$

The total dipole moment of all molecules in the cavity is then obtained by one more integration of (122), as follows:

$$\begin{aligned} M(t) = n\gamma\Big\{[1-c(\lambda)]\cos\nu t \\ -\frac{(\Omega-\nu)}{\lambda}[\lambda\bar{\tau}-S(\lambda)]\sin\nu t\Big\} \end{aligned} \tag{124}$$

where we have defined

$$\gamma = \frac{a\hbar\Omega K^2}{\Omega\lambda^2} \tag{125}$$

for convenience, and where

$$\bar{\tau} = \int_0^{\infty}\tau f(\tau)d\tau \tag{126}$$

is the mean flight time, and

$$c(\lambda) \equiv \int_0^{\infty}\cos\lambda\tau f(\tau)d\tau \tag{127}$$

and

$$s(\lambda) \equiv \int_0^{\infty}\sin\lambda\tau f(\tau)d\tau \tag{128}$$

are the Fourier transforms of the flight time distribution.

To obtain the conditions for a self-consistent solution, we substitute (124) into (110) and equate the coefficients of $\cos\nu t$, $\sin\nu t$. We obtain the relations

$$\frac{\omega\nu}{Q} = \frac{Sn\gamma}{2a}[1-c(\lambda)] \tag{129}$$

and

$$\nu^2-\omega^2 = \frac{Sn\gamma}{2a}\frac{\Omega-\nu}{\lambda}[\lambda\bar{\tau}-s(\lambda)]. \tag{130}$$

The starting current n_0 is determined by (129) for small λ. From (127) we have

$$\lim_{\lambda\to 0}\frac{1-c(\lambda)}{\lambda^2}=\int_0^\infty \frac{1}{2}\tau^2 f(\tau)d\tau=\frac{\overline{\tau^2}}{2} \tag{131}$$

so that it is the mean-square flight time which determines the starting current, as follows:

$$n_0=\frac{4a\omega\nu}{Q\lambda^2 j\overline{\tau^2}S}\approx\frac{\hbar J_1^2 V}{Q\overline{\tau^2}4\pi\mu^2}. \tag{132}$$

Similarly, we have from (128),

$$\lim_{\lambda\to 0}\frac{\lambda\bar{\tau}-S(\lambda)}{\lambda^3}=\frac{1}{6}\overline{\tau^3}, \tag{133}$$

so that if we define new functions,

$$F(\lambda)\equiv\frac{6[\lambda\bar{\tau}-s(\lambda)]}{\lambda^3\overline{\tau^3}} \tag{134}$$

and

$$G(\lambda)\equiv\frac{2[1-c(\lambda)]}{\lambda^2\overline{\tau^2}}, \tag{135}$$

we have $F(0)=G(0)=1$. Previous writers [1] have expressed their results in terms of an "effective Q" of the molecular beam. The appropriate definition here would be

$$Q_m\equiv\frac{\Omega\overline{\tau^3}}{6\overline{\tau^2}}\approx 10^6. \tag{136}$$

Our conditions (129) and (130) then assume the forms

$$n_0/n=G(\lambda) \tag{137}$$

and

$$(\nu-\Omega)\cong(\omega-\Omega)\frac{Q}{Q_m}\frac{G(\lambda)}{F(\lambda)} \tag{138}$$

if we neglect terms of order $(Q/Q_m)^2$. These relations are to be used graphically as follows: For a given velocity distribution, the functions $F(\lambda)$ and $G(\lambda)$ can be calculated and plotted. Then from (137) one determines λ as a function of the beam current. The frequency pulling factor (G/F) of (138) is then determined. Finally, the amplitude of oscillation a is determined for given beam current n and cavity tuning ω, by use of (138) and (119). For constant beam current, *i.e.*, constant λ, the graph of amplitude vs frequency of oscillation is a half ellipse, the amplitude reaching a maximum for perfect tuning of the cavity to the molecule line, in which case $Ka=\lambda$. Oscillations can persist over a frequency band $(\Omega-\lambda)<\nu<(\Omega+\lambda)$.

In the case of a single molecular velocity, $f(\tau)=\delta(\tau-\tau_0)$, these relations reduce to those of Shimoda, Wang, and Townes [4], and in the case of a Maxwellian velocity distribution, where the fraction of molecules per unit time with velocities in the range dv is proportional to $v^3\exp(-v^2/v_0^2)\,dv$, or

$$f(\tau)\sim\exp(-L^2/v_0^2\tau^2)/\tau^5 \tag{139}$$

(L = length of cavity, $v_0^2=2KT/m$), they reduce to the theory of Lamb and Helmer [6]. In the Maxwellian case our dimensionless parameters F, G, Q_m become identical with those defined by Lamb and Helmer.

The assumption of a Maxwellian distribution is certainly a reasonable first approximation, but it is probably not very accurate because the quality of focusing is velocity dependent, the focuser having the property of focusing the small velocity molecules much more effectively than the high-speed ones; and, except for extremely strong focusing voltage, the distribution of flight times may be biased considerably more in favor of large τ than is indicated by (139). This could have an important effect on stability, as may be seen in the following.

In the case of a Maxwellian distribution, half of the mean-square flight time $\overline{\tau^2}$, which determines the starting current, is contributed by the slowest 12 per cent of the molecules. Half of the third moment $\overline{\tau^3}$, which determines the effective molecular Q, and hence the long time frequency stability, is due to the slowest 1.9 per cent. Any further biasing in favor of higher τ would have a considerable effect on $\overline{\tau^2}$, and a very large effect on $\overline{\tau^3}$. For this reason, effects of fluctuations in beam current may not be of the relative order of magnitude $1/\sqrt{N}$, where N is the total number of molecules in the cavity. Fluctuations in the experimentally significant quantities may be determined almost entirely by the slowest 5 per cent of the molecules, with corresponding greater relative variation.

The effect on the frequency-pulling function G/F of a "truncated" Maxwellian distribution has been worked out for the case where the velocity distribution is taken to be Maxwellian up to some v_{max} and zero thereafter, where v_{max} was taken arbitrarily to be $\frac{1}{2}v_0$. The results were compared to that of a Maxwellian distribution. A region of stability still appears, as in the analysis of Lamb and Helmer, but it occurs for smaller values of λ (about a factor of two) which corresponds to smaller values of beam flux.

V. Fluctuation Effects

The steady-state relations found in Section IV form the starting point for investigations of fluctuation effects, by perturbation methods in which we expand in powers of the small departure from the previous solutions. If we calculate only to the lowest nonvanishing order, these problems become linear. But in this case we can analyze the effect of small periodic perturbations, proportional to $\exp(i\beta t)$ and superpose the solu-

tions to find the transient response to an arbitrary small perturbation. Thus, consider the effect of an additional signal $F(t)$ impressed on the cavity. This might be an extra signal intentionally fed in, or it might be a randomly varying function representing thermal noise generated in the cavity and/or load. The equation of motion for the electric field then becomes

$$\ddot{E} + \omega^2 E + \frac{\omega}{Q}\dot{E} = SM(t) + F(t) \tag{140}$$

and this $F(t)$ causes a change of E_1 in the electric field. Suppose now that $F(t)$ contains the time factor $\exp(i\beta t)$; then if β is not too close to the oscillation frequency ν, the change in electric moment of the ith molecule will satisfy

$$\ddot{M}_{1_i} + \Omega^2 M_{1_i} = -K^2 W_i E_1 \tag{141}$$

where we have set

$$E = E_0 + E_1 = 2a \sin \nu t + E_1 \tag{142}$$

and

$$M_i = M_{0_i} + M_{1_i} \tag{143}$$

$$W_i = W_{0_i} + W_{1_i} \tag{144}$$

where the subscript "0" denotes the unperturbed or steady-state solutions of Section IV. Here we have dropped a term EW_{1_i} on the grounds that it will not have an appreciable component of frequency β. Under these conditions, the change in total moment of all molecules in the cavity will be simply

$$M_1 = \sum_i M_{1_i} = \frac{-K^2 \overline{W} E_1}{\Omega^2 - \beta^2} \tag{145}$$

where $\overline{W}$ is the average energy of all molecules in the cavity. Combining these relations, we find the electric field fluctuation to be given by

$$E_1 = \frac{F(t)(\Omega^2 - \beta^2)}{\left[\left(\omega^2 - \beta^2 + \frac{i\omega\beta}{Q}\right)(\Omega^2 - \beta^2) + K^2 S\overline{W}\right]}. \tag{146}$$

If $\overline{W} = 0$, this reduces, as it must, to the response of the cavity alone. The effect of the molecules is, in this approximation, to suppress the magnitude of the electric field fluctuations E_1, for those frequency components which lie close to the natural line frequency.

If the period of the beat frequency $(\beta - \omega)$ is comparable to the flight time, however, then one should take into account the term W_{1_i}; generally, perturbations of any type can lead to the greatest effects when their frequency is related in this way to the flight time. Since the theory remains linear, flucutations due to any cause are readily calculated. We now proceed to the calculation of electric field fluctuation in which we retain the term W_{1_i}.

Keeping terms to only the first order in the perturbation, we have to solve the system of linear equations with time varying coefficients,

$$\ddot{E}_1 + \frac{\omega}{Q}\dot{E}_1 + \omega^2 E_1 = SM_1(t) + F(t) = S\sum_i M_{1_i}(t) + F(t), \tag{147}$$

$$\ddot{M}_{1_i} + \Omega^2 M_{1_i} = -K^2 W_{1_i} E_0 - K^2 W_{0_i} E_1, \tag{148}$$

and

$$\dot{W}_{1_i} = E_1 \dot{M}_{0_i} + E_0 \dot{M}_{1_i}. \tag{149}$$

From (149) we have

$$W_{1_i}(t) = \int_{t_i}^{t} E_0(t')\left[-K^2 \int_{t_i}^{t'} (W_{1_i}(t'')E_0(t'') + W_{0_i}(t'')E_1(t'')) \cos \Omega(t' - t'')dt''\right]dt' + \int_{t_i}^{t} E_1(t')\left[-K^2 \int_{t_i}^{t'} W_{0_i}(t'')E_0(t'') \cos \Omega(t' - t'')dt''\right]dt' \tag{150}$$

where we have used

$$M_{1_i}(t) = -\frac{K^2}{\Omega}\int_{t_i}^{t} \sin \Omega(t - t') \cdot [W_{1_i}(t')E_0(t') + W_{0_i}(t')E_1(t')]dt'. \tag{151}$$

We specialize to the case of the tuned cavity, $\omega = \Omega$, since this will not significantly affect the results and renders the mathematics very much less tedious. Now, assuming a solution of the form

$$E_1(t) = a_1 e^{i\beta t} + a_2 e^{i(\beta - 2\Omega)t} \tag{152}$$

we are able to get self-consistent solutions for (147)–(149). Using (152) and the unperturbed solutions of the preceding chapter for the case of a single molecular flight time, we have for (150),

$$W_{1_i}(t) = \frac{Kh\Omega}{4\Omega'}(a_1 - a_2)(e^{-i\Omega' t} - e^{-i\Omega' t_i}) \sin \lambda(t - t_i). \tag{153}$$

We have dropped terms of frequency $\Omega + \beta$, as they do not contribute appreciably to W_1. Here $\Omega' \equiv \Omega - \beta$. Now we put this result into (151) and after integrating over all the molecules, *i.e.*,

$$M_1(t) = A\int_{t-\tau}^{t} M_{1_i}(t)dt, \tag{154}$$

and, inverting the order of integration, we have

$$M_1(t) = -\frac{K^2A}{\Omega}\int_{t-\tau}^{t} dt' E_0(t') \sin \Omega(t-t') \int_{t-\tau}^{t} W_{1_i}(t')dt_i - \frac{K^2A}{\Omega}\int_{t-\tau}^{t} dt' E_1(t') \sin \Omega(t-t') \cdot \int_{t-\tau}^{t'} W_{0_i}(t')dt_i. \quad (155)$$

Note that $\Omega_1 \approx -\Omega_2$ if $\Omega - \beta \leq \pm 10\lambda$. The expressions (157) and (158) can be simplified somewhat by remembering that we can replace A by the starting current A_0 divided by $G(\lambda)$ for the univelocity case, *i.e.*,

$$A = A_0/G(\lambda) = \frac{4\Omega^2}{Q\tau^2(\hbar\Omega K^2 S)} \frac{\lambda^2\tau^2}{2(1-\cos\lambda\tau)} \quad (163)$$

from (132) and (135). Thus,

$$C = \frac{i\lambda^2(\Omega^2/Q)[\Omega' \cos \Omega'\tau/2 \sin \lambda\tau/2 - \lambda \cos \lambda\tau/2 \sin \Omega'\tau/2]}{2\Omega'(\Omega'^2 - \lambda^2) \sin \lambda\tau/2} \quad (164)$$

and

$$B = C + \frac{i\lambda^2\Omega^2/Q[(\cos\lambda\tau - \cos\Omega'\tau) + i/\lambda(\Omega' \sin\lambda\tau - \lambda \sin\Omega'\tau)]}{2\Omega'(\Omega'^2 - \lambda^2)(1-\cos\lambda\tau)}. \quad (165)$$

The result of the integrations is

$$SM_1(t) = a_1Be^{i\beta t} + a_1Ce^{i(\beta-2\Omega)t} - a_2Be^{i(\beta-2\Omega)t} - a_2Ce^{i\beta t} \quad (156)$$

where we have defined

$$C = \frac{AK^2Sh i}{4(\Omega'^2 - \lambda^2)} e^{i\Omega'\tau/2} \cdot \left[\cos \Omega'\tau/2(1-\cos\lambda\tau) - \frac{\lambda}{\Omega'} \sin \Omega'\tau/2 \sin\lambda\tau\right] \quad (157)$$

and

$$B = C + \frac{AK^2Sh i}{4(\Omega'^2 - \lambda^2)} \cdot \left[\cos\lambda\tau + i\frac{\Omega'}{\lambda}\sin\lambda\tau - \cos\Omega'\tau - i\sin\Omega'\tau\right]. \quad (158)$$

Inserting this expression into (147) we find

$$a_1 = \frac{F(\Omega_2 + B)}{[(\Omega_1 - B)(\Omega_2 + B) + C^2]} \quad (159)$$

and

$$a_2 = \frac{FC}{[(\Omega_1 - B)(\Omega_2 + B) + C^2]} \quad (160)$$

where we define

$$\Omega_1 = \Omega^2 - \beta^2 + i\Omega\beta/Q \quad (161)$$

and

$$\Omega_2 = \Omega^2 - (\beta - 2\Omega)^2 + i\frac{\Omega}{Q}(\beta - 2\Omega). \quad (162)$$

We see here the result of having time-varying coefficients in our linear equations (147)–(149). If we feed in a signal of frequency β, the field given back contains the frequencies β and $(2\Omega - \beta)$, *i.e.*, the frequency β and the reflection of β about Ω.

If we plot the amplitudes $|a_1|^2$ and $|a_2|^2$ of the frequencies β and $(2\Omega - \beta)$ as a function of the parameter k, such that $\Omega' = k\lambda$, for various values of the flight time τ, we observe a "quieting" effect for certain values of the flight time. For small values of the flight time, the effect of the molecular beam is to amplify any input signal, *e.g.*, thermal noise. The region of greatest stability, that at which "noise quieting" would be the greatest, appears at $\lambda\tau = 3\pi/2$. See Fig. 5, pp. 107–108.

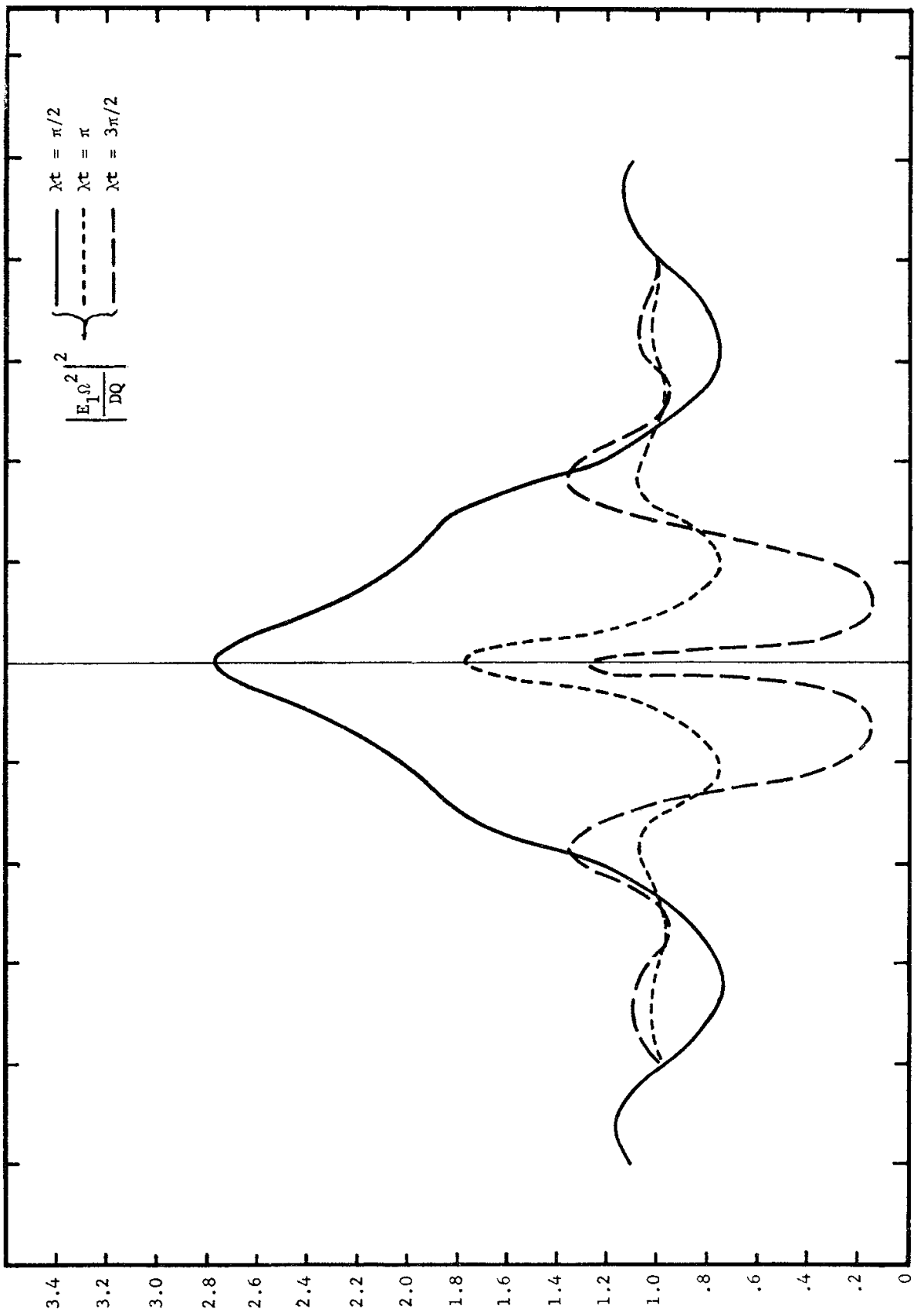
This analysis was carried out under the assumption of a uniform velocity distribution and a tuned cavity. It is clear enough that these restrictions do not greatly impair the generality of the results obtained. The analysis for an untuned cavity would merely have the effect of shifting the axis in the plots of Fig. 5 by the amount $\Omega - \nu$, so that one would have plots symmetrical about ν instead of Ω. Also, very plausible speculation leads one to conclude that the only effect of a velocity distribution, which mathematically would appear as integrals over τ in B and C in the denominators of (159) and (160), would be to smooth out the "wiggles" which appear near the "bare" cavity response value $FQ/\Omega^2 = 1$.

One can recast the electric field,

$$E(t) = 2a \sin \Omega t + \int_{-\infty}^{+\infty} a_1 e^{i\beta t} d\beta + \int_{-\infty}^{+\infty} a_2 e^{i(2\Omega-\beta)t} d\beta + cc \quad (166)$$

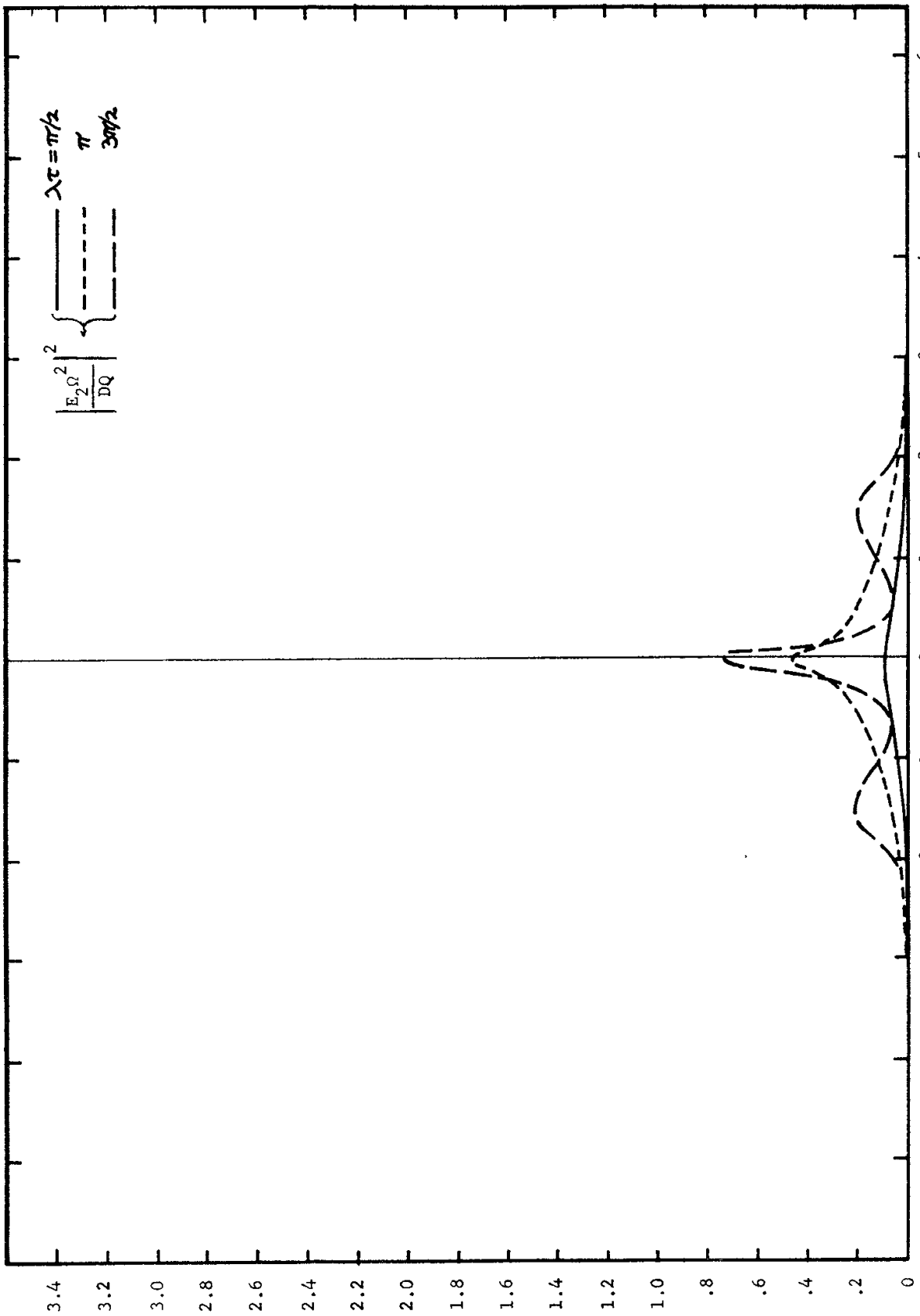
in the form

$$E(t) = A(t) \sin[\Omega t + \phi(t)] \quad (167)$$


$\left|\frac{E_1\Omega^2}{DQ}\right|^2$
$\lambda t = \pi/2$
$\lambda t = \pi$
$\lambda t = 3\pi/2$
k→
$\Omega-\beta = k\lambda$
0
1
2
3
4
5
6
-1
-2
-3
-4
-5
.2
.4
.6
.8
1.0
1.2
1.4
1.6
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4

(a)

Fig. 5.


$\left|\frac{E_2\Omega^2}{DQ}\right|^2$
$\lambda\tau = \pi/2$
π
$3\pi/2$
k→
$\Omega-\beta = k\lambda$

(b)

Fig. 5.

where the power spectra of $A(t)$ and $\phi(t)$ are known from the knowledge of a_1 and a_2. One might then argue that we can obtain $\langle\phi^2\rangle$ and thus have an answer for the frequency stability; but we must remember that we have used a first-order perturbation method, which is valid only for small momentary departures from the steady-state solution—the frequency instability due to thermal noise in the walls probably causes the phase to wander in a manner closely akin to Brownian motion phenomena; and we cannot expect to predict cumulative phenomena by use of perturbations. However, since there is "restoring force" on the amplitude, it can never wander very far from the steady-state value, and we would expect that this analysis could lead to a good prediction of the amplitude stability due to a random perturbation such as thermal noise generated in the cavity or walls.

The criterion for the accuracy of any frequency standard, or "clock," is not so much how far the oscillation frequency drifts from its nominal value in a given time, but rather how well we are able to *predict* how it will wander. Here one would be concerned with the wander of the phase of the output under the perturbation of 1) the thermal radiation from the cavity walls and 2) the unavoidable random fluctuations in beam composition. The analysis given above indicates that under conditions likely to be realized in the foreseeable future, the former effect will be by far the most important.

By analogy with the classical Einstein treatment of Brownian motion, one expects that the phase ϕ will be uncertain in the following sense: While $\langle\phi(t)\rangle=0$, $\langle\phi^2(t)\rangle=Kt$. The constant K would then be a reasonable measure of the oscillation stability.

The analysis given above is not, however, adapted to answering questions of this type. In assuming that the actual output could be represented in the form

$$E(t) = 2a \sin \nu t + E_1(t)$$

with $|E_1| \ll a$, we have in effect restricted the theory to cases (or time intervals) such that the phase wandering ϕ is at most of order E_1/a. A more general theory of random processes is therefore needed before questions about very long time behavior can be answered.

Wiener [15] has given an interesting discussion of the response of nonlinear systems to a random perturbation. His curves are very similar to those of Fig. 5, which arise, for instance, in the analysis of the alpha rhythm of brain waves. It appears that the phenomena predicted by Fig. 5 is a general property of any nonlinear system which is attempting to stabilize itself.

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