SOME ASPECTS OF MASER THEORY

By
E. T. Jaynes

Topical Report

M.L. Report No. 502
May, 1958

Microwave Laboratory
W. W. HANSEN LABORATORIES OF PHYSICS
STANFORD UNIVERSITY
STANFORD, CALIFORNIA

ABSTRACT

As a preliminary to a theoretical study of the ultimate limita-
tions on noise figure and frequency stability in molecular-beam masers,
we examine the relation between quantum electrodynamics and the semi-
classical theory of radiation. Solutions according to both theories
are worked out in detail for the case of a single molecule interacting
with a single cavity oscillation mode. Comparison of the results
shows several features which make the relation of the two theories
appear rather different than is usually supposed. For example,
according to quantum electrodynamics, even in the limit of aribrarily
large photon occupation numbers, the electromagnetic field can be in
physical states qualitatively different from any describable in
classical terms, and a molecule-beam maser excited by molecules all
in the upper state should produce such a condition.

The molecular-beam maser provides an interesting example of the
Einstein-Podolsky-Rosen paradox, arising from correlations in states
of molecule and field. An attempt is made to find an experimental
situation in which effects of such correlations are observable, but
without success.

It is shown that, contrary to what is often assumed, the semi-
classical theory of radiation _can_ account for spontaneous emission of
radiation. It leads to a characteristic time just the same as pro-
vided by field quantization, when we take into account both the effect
of the field on the molecule and the effect of the molecule on the
field. The result is a system of coupled nonlinear equations whose
solutions can be found in terms of elliptic functions. Even in the
case of field intensities corresponding to one photon, this solution
reproduces almost quantitatively the same laws of energy exchange as
found in quantum electrodynamics. It is concluded that the semi-
classical theory, as extended here, is a far more reliable means of
calculating radiation processes than usually supposed.

TABLE OF CONTENTS

1. INTRODUCTION

From a theoretical standpoint, maser amplifiers and oscillators offer some of the most interesting problems in physics. There are few situations where quantum theory and classical theory are combined so closely in a single description. Furthermore, relations between the semiclassical theory of radiation, quantum electrodynamics, and the statistical mechanics of irreversible processes, are here exhibited in a very simple model accessible to both calculation and experiment.

The work here reported (which will continue after termination of the present contract) comprises the first stages of an attempt to treat maser operation entirely in terms of quantum theory. Although it is hoped that it will lead to useful results concerning noise figure and frequency stability, the primary objective is a more complete understanding of the theoretical relations mentioned above. Some of what is said below applies to solid-state masers, but we have in mind specifically the case of molecular-beam devices.

Numerous theoretical treatments of masers now exist, based on several different types of approximations and assumptions, and new ones appear almost every month. Broadly speaking, there are two different levels of approximation used in theories published to date:

(A) The most common type of theory, but also the crudest, is the one wherein the emission of radiation from molecules is considered to take place via independent and instantaneous "quantum jumps," whose probability of occurrence is proportional to the product of time of interaction and energy density of the radiation at the transition frequency. This is essentially an application of quantum theory as it existed 40 years ago. It involves little that was not already given in Einstein's famous paper of 1917, introducing the A and B coefficients. Present quantum mechanics tells us that the notion of time-proportional induced transition probabilities is only an

approximation valid when the correlation time of the radiation is short
compared to the time required to accumulate an appreciable transition
probability; in other words, that the radiation responsible for the
transitions must be random, with a spectrum wide compared to the line
width. This condition is satisfied in most optical experiments, but
in an ammonia maser 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. Under these conditions, any attempt to describe maser
operation in terms of the standard transition probability formulas,
such as

$$W_{1 \to 2} = \frac{2\pi}{\hbar^2} \left| H_{12} \right|^2 g(\omega) \tag{1-1}$$

may lead to conclusions that are not only quantitatively, but also
qualitatively wrong. Unfortunately, most of the existing noise figure
calculations are based on a treatment of this type. Of course, use
of a poor approximation does not necessarily prevent one from getting
the right answer; but one can never be sure it is right until the
calculation has been checked in other ways.

(B) A second stage of approximation, resulting in a much more
realistic treatment, is represented by the theories of Basov and
Prokhorov, Lamb, and Feynman. This is based on the semiclassical
theory of radiation, in which, instead of using Eq. (1-1), one
actually solves the time-dependent Schrödinger equation for a mole-
cule, as perturbed by a classically described radiation field. The
effect of the molecule on the field is then assumed to be the same
as the effect one would obtain classically from a dipole whose
moment is equal to the expectation value of moment of the molecule.
Such a treatment gives definite predictions for saturation and fre-
quency-pulling effects, but it is not obvious whether even this
approximation would lead to correct prediction of the fluctuation
effects involved in noise figure and frequency stability. Theories
of this type still involve many approximations, among which are:
(1) the molecules are ascribed independent wave functions, whereas
in principle one should always treat the entire system of molecules
as a single quantum-mechanical system, by a formalism like that of
Dicke's "superradiant gas." (2) In principle, one should also

quantize the radiation field and consider the problem as one of quantum electrodynamics.

It is generally thought that the semiclassical approach should be quite adequate for any effects at microwave frequencies, by virtue of the smallness of the Einstein A coefficients relative to the B coefficients. However, the relation between semiclassical theory and quantum electrodynamics is not a simple one. Quantization of the radiation field introduces many changes in addition to the appearance of A coefficients. In particular, it leads to the possibility that the electromagnetic field may be in physical states qualitatively different from any that can be described 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 be the ones actually produced in masers under certain idealized conditions.

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 will be the extent to which the semiclassical theory is derivable from quantum electrodynamics, and the effect of different statistical assumptions concerning the initial states of the molecules.

2. 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[1] is quite adequate for our purposes. Also, the usual plane wave expansion is not appropriate here, 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.[2]

---

[1]E. Fermi, Rev. Mod. Phys. 4, 131 (1932).

[2]J. C. Slater, Microwave Electronics, D. van Nostrand Co., Inc., N. Y. (1950), Chapter 4.

The cavity is represented by a volume V, bounded by a closed surface S. Let $E_a(x)$, $k_a^2 = (\omega_a^2/c^2)$ be the eigenfunctions and eigenvalues of the boundary-value problem

$$\nabla \times \nabla \times E - k^2 E = 0 \qquad \text{in } V,$$

$$n \times E \qquad \text{on } S, \tag{2-1}$$

where n is a unit vector normal to S. The $E_a(x)$ are so normalized that

$$\int_V (E_a \cdot E_b) dV = \delta_{ab}. \tag{2-2}$$

The vector functions $H_a(x)$, related to E_a by

$$\nabla \times E_a = k_a H_a, \qquad \nabla \times H_a = k_a E_a, \tag{2-3}$$

are also orthonormal in V:

$$\int_V (H_a \cdot H_b) dV = \delta_{ab}. \tag{2-4}$$

The electric and magnetic fields can be expanded in the form

$$E(x,t) = -\sqrt{4\pi} \sum_a p_a(t) E_a(x), \tag{2-5}$$

$$H(x,t) = \sqrt{4\pi} \sum_a \omega_a q_a(t) H_a(x). \tag{2-6}$$

From these relations, we find for the total field energy

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

and the Maxwell equations,

$$\nabla \times E = -\frac{1}{c} \frac{\partial H}{\partial t}, \tag{2-8}$$

$$\nabla \times H = \frac{1}{c} \frac{\partial E}{\partial t}, \tag{2-9}$$

then reduce to the Hamiltonian equations of motion,

$$\dot{q}_a = \frac{\partial \mathcal{H}}{\partial p_a} = p_a \ , \qquad\qquad (2\text{-}8a)$$

$$\dot{p}_a = -\frac{\partial \mathcal{H}}{\partial q_a} = -\omega_a^2 q_a \ , \qquad\qquad (2\text{-}9a)$$

respectively.

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

$$[q_a , q_b] = [p_a , p_b] = 0, \qquad\qquad (2\text{-}10)$$

$$[q_a , p_b] = i\hbar\delta_{ab}. \qquad\qquad (2\text{-}11)$$

The operators c_a^{*}, c_a which create or annihilate a photon in the a'th cavity mode are then

$$c_a^{*} = \frac{p_a + i\omega_a q_a}{\sqrt{2\hbar\omega_a}} \ , \qquad c_a = \frac{p_a - i\omega_a q_a}{\sqrt{2\hbar\omega_a}} \ , \qquad (2\text{-}12)$$

with the commutation rule

$$[c_a , c_b^{*}] = \delta_{ab}. \qquad\qquad (2\text{-}13)$$

Denote by $\emptyset(n_1 , n_2 , \cdots)$ 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

$$c_a \emptyset(\cdots , n_a , \cdots) = \sqrt{n_a} \ \emptyset(\cdots , n_a - 1, \cdots) \quad (2\text{-}14)$$

$$c_a^{*} \emptyset(\cdots , n_a , \cdots) = \sqrt{n_a + 1} \ \emptyset(\cdots , n_a + 1, \cdots),$$

$$(2\text{-}15)$$

from which we easily verify (2-13), and obtain the matrix elements in the n_a representation:

$$(n_a|c_a|n_a') = (n_a'|c_a^{*}|n_a) = \sqrt{n_a + 1} \ \delta(n_a', n_a + 1). \quad (2\text{-}16)$$

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

$$\mathcal{H} = \sum_a \hbar\omega_a \, c_a^* c_a = \sum_a \hbar\omega_a \, n_a . \qquad (2\text{-}17)$$

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 E_a is $E_{az} = (\text{const.}) \times J_0(k_a r)$, independent of z and θ. The normalizing constant is obtained from evaluating the integral (2-2), 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

$$E_{az} = \frac{1}{J_1 \sqrt{V}} . \qquad (2\text{-}18)$$

Here $J_1 \equiv J_1(u) = 0.5191$, 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 (2-12),

$$p_a = \sqrt{\frac{\hbar\omega_a}{2}} \, (c_a + c_a^*) . \qquad (2\text{-}19)$$

Combining (2-5), (2-16), (2-18), and (2-19), we obtain the matrix element

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

in which we have dropped the subscript a, it being understood that (2-20) 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 $J_0(kr)$.

3. INTERACTION WITH A SINGLE MOLECULE

The simplest possible situation is one where we consider a loss-less cavity, which has only a single resonant mode near the natural line frequency of the molecules, 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 of the usual (J · 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 + 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 attempted here.

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

$$(mn|H|m'n') = (E_m + n\hbar\omega)\delta_{mm'}\delta_{nn'} + (mn|H_{int}|m'n') \quad (3\text{-}1)$$

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

$$H_{int} = -\vec{\mu} \cdot \vec{E} \quad (3\text{-}2)$$

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

$$(m|\mu_z|m') = \mu(1 - \delta_{mm'}). \quad (3\text{-}3)$$

Combining this with (2-20), we obtain the matrix elements for the interaction energy

$$(mn|H_{int}|m'n') = \alpha(1 - \delta_{mm'})\left[\sqrt{n}\,\delta_{n,n'+1} + \sqrt{n+1}\,\delta_{n+1,n'}\right], \quad (3\text{-}4)$$

where

$$\alpha = \frac{\mu}{J_1} \sqrt{\frac{2\pi\hbar\omega}{V}} \tag{3-5}$$

is the interaction constant. Using the value[3] $\mu = 1.47 \times 10^{-18}$ esu for ammonia, and a cavity 10 cm long, we find $(\alpha/\hbar\omega) = 2.08 \times 10^{-10}$, or, in frequency units, $\alpha \cong 5.0$ cps.

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

$$V_n \equiv (1, n + 1|V|2, n) = (2, n|V|1, n + 1) = \alpha(n + 1)^{1/2}$$

$$\tag{3-6}$$

$$W_n \equiv (1, n|W|2, n + 1) = (2, n + 1|W|1, n) = \alpha(n + 1)^{1/2}$$

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 $(E_2 - E_1 + \hbar\omega) \simeq 2\hbar\omega$. Since in typical operating 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

$$H = H_o + W,$$

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

$$E_o = E_1 = \hbar\omega_o , \qquad \Phi_o = \psi_1 \varphi_o , \tag{3-7}$$

and, for $n > 0$,

$$E_n^{\pm} = \hbar\omega_n^{\pm} = \frac{1}{2}\left[E_1 + E_2 + (2n - 1)\hbar\omega\right] \pm \frac{1}{2}\left[(E_2 - E_1 - \hbar\omega)^2 + 4n\alpha^2\right]^{1/2}$$

$$\tag{3-8}$$

---

[3]D. K. Coles, et al., Phys. Rev. 82, 877 (1951).

$$\Phi_n^+ = \psi_2 \varphi_{n-1} \cos \Theta_n + \psi_1 \varphi_n \sin \Theta_n$$

$$\Phi_n^- = -\psi_2 \varphi_{n-1} \sin \Theta_n + \psi_1 \varphi_n \cos \Theta_n$$

(3-9)

where

$$\tan 2\Theta_n = \frac{2\alpha \sqrt{n}}{\hbar\omega - (E_2 - E_1)} .$$

(3-10)

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

$$U(t,t') = U(t - t') = \exp\left[-iH(t - t')\right],$$

(3-11)

for which the perturbation expansion is

$$U(t) = e^{-iH_o t} - i \int_0^t e^{i(t-t')H_o} W e^{it'H_o} dt' + \cdots .$$

(3-12)

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

$$(2,n - 1| U_o |2,n - 1) = a_n = \cos^2 \Theta_n \, e^{-i\omega_n^+ t} + \sin^2 \Theta_n \, e^{-i\omega_n^- t}$$

$$(2,n - 1| U_o |1,n) = b_n = \sin \Theta_n \cos \Theta_n (e^{-i\omega_n^- t} - e^{-i\omega_n^+ t})$$

$$(1,n| U_o |2,n - 1) = b_n$$

(3-13)

$$(1,n| U_o |1,n) = c_n = \cos^2 \Theta_n \, e^{-i\omega_n^- t} + \sin^2 \Theta_n \, e^{-i\omega_n^+ t}$$

and, for $n = 0$,

$$(1,0| U_o |1,0) = e^{-i\omega_o t} .$$

(3-14)

All other elements vanish. The transition probability for emission or absorption of one photon during time t is therefore, neglecting terms in W,

$$|b_n|^2 = \sin^2 2\Theta_n \, \sin^2 (\omega_n^+ - \omega_n^-) \frac{t}{2} = \frac{n\alpha^2 \sin^2 \beta t}{\hbar^2 \beta^2}$$

(3-15)

where

$$4\hbar^2 \beta^2 = \left[\hbar\omega - (E_2 - E_1)\right]^2 + 4n\alpha^2. \qquad (3\text{-}16)$$

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

$$\begin{pmatrix} a_n & b_n \\ b_n & c_n \end{pmatrix}, \quad n = 1, 2, \cdot \circ \cdot$$

along the main diagonal. The first row and column, however, contain only the single term (3-14).

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 of the entire system is thus the direct product $\rho(0) = \rho_1(0) \times \rho_f(0)$, with matrix elemets

$$(mn|\rho(0)|m'n') = (m|\rho_1(0)|m')(n|\rho_f(0)|n'). \qquad (3\text{-}17)$$

During the interaction, ρ undergoes a unitary transformation

$$\rho(t) = U(t,0)\, \rho(0)\, U^{-1}(t,0), \qquad (3\text{-}18)$$

and the density matrix $\rho_f(t)$, which describes the state of the field only, is the projection[4] of (3-18) onto the space of the field variables:

$$(n|\rho_f(t)|n') = \sum_m (mn|\rho(t)|mn'). \qquad (3\text{-}19)$$

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

$$(n|\rho_f(\tau)|n') = \sum_{k,k'} (nn'|G|kk')(k|\rho_f(0)|k'), \qquad (3\text{-}20)$$

or

$$\rho_f(\tau) = G\, \rho_f(0), \qquad (3\text{-}21)$$

---

[4]This formalism is developed in more detail in the following paper: E. T. Jaynes, Phys. Rev. 108, 171 (1957).

$$(nn' | G | kk') = \sum_{m, m', m''} (m''n | U | mk)(m'k' | U^{-1} | m''n') \sigma_{mm'} , \quad (3\text{-}22)$$

where we have written for brevity

$$\sigma_{mm'} \equiv (m | \rho_1(0) | m'). \quad (3\text{-}23)$$

The sums (3-22) are readily evaluated with use of (3-13), with the result that the only nonvanishing elements of G are

$$(nn' | G | nn') = a_{n+1} a^{*}_{n'+1} \sigma_{22} + c_n c^{*}_{n'} \sigma_{11} \quad (3\text{-}24a)$$

$$(nn' | G | n+1, n') = b_{n+1} a^{*}_{n'+1} \sigma_{12} \quad (3\text{-}24b)$$

$$(nn' | G | n, n'+1) = a_{n+1} b^{*}_{n'+1} \sigma_{21} \quad (3\text{-}24c)$$

$$(nn' | G | n, n'-1) = c_n b^{*}_{n'} \sigma_{12} \quad (3\text{-}24d)$$

$$(nn' | G | n-1, n') = b_n c^{*}_{n'} \sigma_{21} \quad (3\text{-}24e)$$

$$(nn' | G | n+1, n'+1) = b_{n+1} b^{*}_{n'+1} \sigma_{11} \quad (3\text{-}24f)$$

$$(nn' | G | n-1, n'-1) = b_n b^{*}_{n'} \sigma_{22} . \quad (3\text{-}24g)$$

These relations hold for all quantum numbers n if we understand that c_o is not defined by (3-13), but by $c_o \equiv \exp(-i\omega_o t)$.

To illustrate the use of this formalism, we discuss a few simple problems using (3-24). Consider first the case where the field is initially in its lowest state: $(0 | \rho_f(0) | 0) = 1$, all other elements of $\rho_f(0)$ vanish. Then, according to (3-24), after a molecule with initial density matrix σ has passed through, the field density matrix has elements

$$(0 | \rho_f(\tau) | 0) = | a_1 |^2 \sigma_{22} + \sigma_{11}$$

$$(0 | \rho_f(\tau) | 1) = (1 | \rho_f(\tau) | 0)^{*} = c_o b^{*}_1 \sigma_{12} \quad (3\text{-}25)$$

$$(1 | \rho_f(\tau) | 1) = | b_1 |^2 \sigma_{22} ,$$

all other elements still vanishing. If the molecule was initially in the lower state $[\sigma_{11} = 1, \sigma_{22} = \sigma_{12} = 0]$, 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 (2-20), is

$$\langle E \rangle = \mathrm{Tr}(\rho_f E) = -\frac{\alpha}{\mu} \sum_n \sqrt{n+1} \left[(n|\rho_f|n+1) + (n+1|\rho_f|n) \right]$$

$$= -\frac{2\alpha}{\mu} \mathrm{Re} \sum_n \sqrt{n+1} \ (n|\rho_f|n+1). \qquad (3\text{-}26)$$

This remains zero as long as there is no coherence between adjacent levels, even though the energy stored in the field may be large. In the case (3-25), we obtain for $\langle E \rangle$,

$$\langle E \rangle = -\frac{2\alpha}{\mu} \mathrm{Re}(c_o b_1^* \sigma_{12}) = \frac{2\alpha^2 \sin \beta t}{\mu \hbar \beta} \mathrm{Re}\left[i\sigma_{12} \ e^{i(\Omega+\omega)t/2} \right], \qquad (3\text{-}27)$$

where β is defined by (3-16) with $n = 1$, and $\Omega = (E_2 - E_1)/\hbar$ is the natural line frequency of the molecule. If the cavity is so tuned that its resonant frequency ω is equal to Ω, then $\hbar\beta = \alpha$, and we obtain simply

$$\langle E \rangle = \frac{2\alpha}{\mu} \sin\left(\frac{\alpha t}{\hbar}\right) \mathrm{Re}\left[i\sigma_{12} \ e^{i\omega t} \right]. \qquad (3\text{-}28)$$

Remembering that $\alpha \approx 5$ cps, the term $\sin(\alpha t/\hbar)$ reaches its first maximum in a quarter cycle, or about 1/20 second. This is the interaction time required for a molecule to emit a photon, with probability 1, 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) of

$$\Delta\omega = \frac{8\omega^3\mu^2}{3\hbar c^3} \sim 10^{-7} \text{ sec}^{-1}, \tag{3-29}$$

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 (3-24),

$$(n|\rho_f(t)|n') = \sigma_{11}\left[b_{n+1}b^*_{n'+1}(n+1|\rho_f(0)|n'+1) + c_n c^*_{n'}(n|\rho_f(0)|n')\right]$$

$$+ \sigma_{12}\left[b_{n+1}a^*_{n'+1}(n+1|\rho_f(0)|n') + c_n b^*_{n'}(n|\rho_f(0)|n'-1)\right]$$

$$+ \sigma_{21}\left[a_{n+1}b^*_{n'+1}(n|\rho_f(0)|n'+1) + b_n c^*_{n'}(n-1|\rho_f(0)|n')\right]$$

$$+ \sigma_{22}\left[a_{n+1}a^*_{n'+1}(n|\rho_f(0)|n') + b_n b^*_{n'}(n-1|\rho_f(0)|n'-1)\right].$$

if the field density matrix is initially diagonal,

$$(n|\rho_f(0)|n') = \rho_n\,\delta_{nn'}, \tag{3-31}$$

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

$$(n|\rho_f(t)|n) = \sigma_{11}\left[|b_{n+1}|^2\,\rho_{n+1} + |c_n|^2\,\rho_n\right] + \sigma_{22}\left[|a_{n+1}|^2\,\rho_n + |b_n|^2\,\rho_{n-1}\right], \tag{3-32}$$

$$(n|\rho_f(t)|n+1) = (n+1|\rho_f(t)|n)^* = \sigma_{12}\left[b_{n+1}a^*_{n+2}\,\rho_{n+1} + c_n b^*_{n+1}\,\rho_n\right], \tag{3-33}$$

which relations will be used in the next section.

4. SUCCESSIVE SINGLE-MOLECULE INTERACTIONS

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

$$\rho_f(N\tau) = G^N\rho_f(0) = G\,\rho_f(N\tau - \tau), \tag{4-1}$$

and particular interest attaches to the limiting form of ρ_f as $N \to \infty$.

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

$$\sigma_{12} = \sigma_{21} = 0, \qquad (n|\rho_f(0)|n') = \rho_n \delta_{nn'}, \qquad (4\text{-}2)$$

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

$$\sigma_{22} = \sigma_{11} e^{-x} = (e^x + 1)^{-1},$$

$$x = (E_2 - E_1)/kT = \hbar\Omega/kT, \qquad (4\text{-}3)$$

and, using (3-32), Eq. (4-1) reduces to

$$\rho_n(N\tau) = (e^x + 1)^{-1}\Big[(|a_{n+1}|^2 + |c_n|^2 e^x)\rho_n(N\tau - \tau)$$

$$+ |b_{n+1}|^2 e^x \, \rho_{n+1}(N\tau - \tau) + |b_n|^2 \, \rho_{n-1}(N\tau - \tau)\Big]. \qquad (4\text{-}4)$$

From this, the limiting form of ρ_n may be found. Taking note of the 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

$$B_n \equiv |b_n|^2(\rho_{n-1} - e^x \rho_n) \qquad (4\text{-}5)$$

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

$$\rho_n = e^{-x} \rho_{n-1} \qquad (4\text{-}6)$$

for all n for which $|b_n|^2 \neq 0$. From (3-15) it is seen that b_n could vanish only for certain isolated special values of n.

Note that (4-6) 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 (4-6) is $T_f = \omega T/\Omega$. This difference would never be seen in practice, for as soon as we detune the cavity appreciably the transition probability $|b_n|^2$ becomes extremely 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
phenomenon lies in the fact that for interactions of finite duration
there is no sharp distinction between "real" and "virtual" transitions,
and, perhaps more to the point, our description of the state of the
molecules in terms of a temperature was not entirely justified, since
nothing was said about their kinetic energy of translational motion.
(It is this translational motion, of course, 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 = const. 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 electric field remains exactly zero in spite
of the fact that the number of photons present may be very large.
This situation raises certain questions regarding the relation between
quantum theory and classical theory. Usually one supposes that the
condition for validity of classical electromagnetic theory is simply
that the number of photons in each field normal mode be large, and
that one may then identify the classical electromagnetic field with
the quantum-mechanical expectation value. However, here is an example
where in spite of the large photon numbers, no such interpretation is
possible, and the semiclassical theory of radiation could not be applied
to describe such states.

In almost every textbook one can find the statement that quantum theory always goes over into classical theory in the limit of large quantum numbers (or, what is the same thing, in the limit $h \longrightarrow 0$). This, however, may be misleading for the following reason. In the limit of large quantum numbers, it is possible to construct well-localized wave packets by coherent superposition of many stationary states, and by a well-known theorem, the center of gravity of such a packet then follows classical equations of motion. This means that, with sufficiently large quantum numbers, classical behavior is contained in quantum theory as a special case. But for arbitrarily large quantum numbers, it is still true that quantum theory allows the existence of a great variety of possible states (such as stationary states, or coherent superpositions representing very "broad" wave packets) whose properties cannot even be described, much less accounted for, in classical terms. So the mere excitation to large quantum numbers is no guarantee that a system will behave according to the laws of classical physics. One needs also some kind of restriction as to the type of measurements which are to be made; for example that we will make only such coarse observations that individual quantum effects could not have been seen anyway.

If the density matrix σ of the entering molecules is not diagonal, then according to (3-30) the density matrix of the field also develops off-diagonal elements. Stated intuitively, the definite phase of the dipole moment of the entering molecules "tells the field what phase to have," and results in a non-zero expectation value for E. This situation is an interesting one which could be realized experimentally (it is, for example, closely related to the "Ramsey technique" for obtaining sharp resonances). The steady-state distribution resulting from (3-30) is a difficult but soluble problem. We will not give the details here because evidence will be presented later which indicates that in this case the semiclassical theory should provide a fully reliable and more efficient way of treating the problem.

5. RELATION TO SEMICLASSICAL THEORY

One of the main objectives of this work has been to clarify the relation between the predictions of quantum electrodynamics and the semiclassical theory of maser operation mentioned in the Introduction. To define more precisely what is meant by the semiclassical theory, we write the basic equations of this approach. Here one considers the electric field $E(t)$ as classically describable (i.e., as a definite, if unknown, function of time), and introduces a wave function

$$\psi(t) = a(t)\psi_1 + b(t)\psi_2 \tag{5-1}$$

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

$$i\hbar\dot{\psi} = (H_{molecule} + H_{int})\psi, \tag{5-2}$$

where

$$(m,H_{molecule}|m') = E_m \delta_{mm'}, \tag{5-3}$$

$$(m|H_{int}|m') = (m|-\vec{\mu}\cdot\vec{E}(t)|m') = -\mu(1 - \delta_{mm'})E(t). \tag{5-4}$$

Equation (5-2) then reduces to

$$i\hbar\dot{a} = E_1 a - \mu E(t)b$$

$$i\hbar\dot{b} = -\mu E(t)a + E_2 b \tag{5-5}$$

which describes the effect of the field on the molecule. To find the effect of the molecule on the field, one calculates the expectation value of dipole moment of the molecule from the solution of (5-5),

$$M(t) \equiv \langle\mu(t)\rangle = \mu(ab^* + a^*b), \tag{5-6}$$

and assumes that the field satisfies the classical equation 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

$$-M\cdot E = +\sqrt{4\pi}\sum_a p_a E_a(x)\cdot M \tag{5-7}$$

to $\mathcal{H}$ in (2-7), where x denotes the position of the molecule. The
classical equations of motion are now

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

(5-8)

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

or, eliminating q_a ,

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

(5-9)

Finally, assuming that only the single mode (2-18) is excited, the
electric field of this mode satisfies the differential equation

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

(5-10)

where we have again dropped 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 term to (5-10), giving us

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

(5-11)

By the "semiclassical theory" we mean the system of equations (5-5),
(5-6), (5-11). They may be given a slightly neater formal appearance
by eliminating the amplitudes a(t), b(t). The result is the non-
linear system of coupled equations

$$\ddot{M} + \Omega^2 \, M = - K^2 \, W \, E(t),$$

(5-12a)

$$\dot{W} = E \, \dot{M},$$

(5-12b)

$$\ddot{E} + \frac{\omega}{Q} \, \dot{E} + \omega^2 \, E = S \, M,$$

(5-12c)

where

$$K = \frac{2\mu}{\hbar} \, , \qquad S = \frac{4\pi \, \omega^2}{J_1^2 \, V} \, ,$$

(5-13)

and

$$W \equiv E_1|a|^2 + E_2|b|^2 - \frac{1}{2}(E_1 + E_2) = \frac{1}{2}\hbar\Omega(|b|^2 - |a|^2) \qquad (5\text{-}14)$$

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 (5-12) we have an apparently classical nonlinear system, all reference to "quantum-mechanical" quantities having disappeared.

The first two of the equations (5-12) admit a first integral,

$$(\dot{M})^2 + \Omega^2 M^2 + K^2 W^2 = \text{const.} = \left(\frac{K\hbar\Omega}{2}\right)^2, \qquad (5\text{-}15a)$$

which is readily verified by eliminating E between them. Eq. (5-15a) is a highly disguised form of the "principle of conservation of probability," $|a|^2 + |b|^2 = \text{const.} = 1$. Similarly, the last two of the equations (5-12) can be combined, in the case $Q = \infty$, to yield the constant of the motion

$$(\dot{E})^2 + \omega^2 E^2 + 2S(W - ME) = \text{const.} \qquad (5\text{-}15b)$$

which expresses the conservation of energy for the system.

Now, what is the relationship between the system of equations (5-12) and our earlier ones based on quantum electrodynamics? In order to answer this, we note that Eqs. (5-12a) and (5-12c) show a strong formal resemblance to a general operator equation of motion, which is obtained by differentiating the relation $i\hbar\dot{F} = [F,H]$. The result is

$$\hbar^2\,\ddot{F} + \left[H,[H,F]\right] = i\hbar[\dot{H},F] \qquad (5\text{-}16)$$

which is exact for any operator F.

Returning to the quantum electrodynamics analysis, let us apply this identity to the electric field operator: $F = E$. The total Hamiltonian $H = (H_{mol} + H_{field} + H_{int})$ has no explicit time dependence, so the righthand side of (5-16) 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

$$\left[H,[H,E]\right] = \left[H_f,[H_f,E]\right] + \left[H_{int},[H_f,E]\right]. \qquad (5\text{-}17)$$

These commutators are readily worked out, with the results

$$\left[H_f,[H_f,E]\right] = \hbar^2\omega^2 E, \qquad (5\text{-}18)$$

$$\left[H_{int} , [H_f , E] \right] = - \hbar^2 \, S \, \mu_{op} \, . \tag{5-19}$$

Therefore, a special case of (5-16) is the operator identity

$$\ddot{E} + \omega^2 \, E = S \, \mu_{op} \tag{5-20}$$

which is to be compared to (5-12c). If we interpret (5-12c) as the expectation value of (5-20), they are seen to be identical in the limit $Q \longrightarrow \infty$, <u>provided</u> that the expectation value of μ_{op} must be defined, not in terms of $a(t)$ and $b(t)$ by means of (5-6), but as the expectation value taken over the complete density matrix $(mn|\rho|m'n')$:

$$\langle \mu_{op} \rangle = \mathrm{Tr}(\rho \mu_{op}) = \sum_{n,m,m'} (mn|\rho|m'n)(m'|\mu_{op}|m). \tag{5-21}$$

With this change in interpretation, (5-12c) is seen to be an exact consequence of quantum electrodynamics.

Next we write out the identity (5-16) for the case $F = \mu_{op}$. This time H_{int} commutes with μ_{op}, but not with $[H_m , \mu_{op}]$, while H_f commutes with both. **Therefore,**

$$\left[H , [H , \mu_{op}] \right] = \left[H_m , [H_m , \mu_{op}] \right] + \left[H_{int} , [H_m , \mu_{op}] \right] . \tag{5-22}$$

Proceeding as before, a short calculation yields the results

$$\left[H_m , [H_m \, \rho \mu_{op}] \right] = \hbar^2 \, \Omega^2 \, \mu_{op} \tag{5-23}$$

$$\left[H_{int} , [H_m , \mu_{op}] \right] \hbar^2 \, K^2 \, H'E \tag{5-24}$$

where we have defined an operator

$$H' \equiv H_{molecule} - \frac{1}{2} \, (E_1 + E_2) \tag{5-25}$$

with matrix elements

$$(mn|H'|m'n') = \frac{1}{2} \, \hbar \, \Omega(-1)^m \, \delta_{mm'} \, \delta_{nn'} \tag{5-26}$$

which is the energy of the molecule, referred to a zero lying midway between its levels E_1, E_2. Comgining these relations, we find that another special case of (5-16) is the operator identity

$$\ddot{\mu}_{op} + \Omega^2 \, \mu_{op} = - K^2 \, H'E \qquad\qquad (5\text{-}27)$$

which is to be compared to (5-12a). This time, taking the expectation
value of (5-27) does not yield (5-12a) in general, for in the semi-
classical equation the "driving force" term appears as $\langle H' \rangle \langle E \rangle$, while
according to quantum electrodynamics it should be $\langle H'E \rangle$. The differ-
ence between these quantities arises from the possibility, which exists
in quantum electrodynamics but not in the semiclassical theory, of
"correlated states." When the states of field and molecule are uncorrelated,
the density matrix reduces to a direct product $\rho = \rho_m \times \rho_f$, or

$$(mn|\,\rho\,|m'n') = (m|\rho_m|m')(n|\rho_f|n'). \qquad\qquad (5\text{-}28)$$

When (5-28) holds, it is easily shown that $\langle H'E \rangle = \langle H' \rangle \langle E \rangle$. But
if (5-28) does not hold, then in general $\langle H'E \rangle \neq \langle H' \rangle \langle E \rangle$. Before
exploring the size of this difference, we digress to consider some general
consequences of correlated states.

6. CORRELATED STATES

To describe the situation in intuitive terms, the semiclassical
theory may be regarded as based on the assumption that the electric
field has, at any time, "in reality" a definite, if unknown, value.
Similarly, one imagines the molecule as being "in reality" in a definite
if unknown quantum state ψ. However, quantum electrodynamics allows
the possibility of states of the combined system (molecule + field),
which do not admit any such description. The stationary states (3-9)
are examples wherein the system (molecule + field) is in a definite
pure state, but nevertheless one cannot ascribe any definite quantum
state to the molecule alone, or the field alone.

This situation arises in general whenever two different quantum-
mechanical systems interact, and forms the basis of one of Einstein's
objections to quantum theory. The famous Einstein-Podolsky-Rosen
paradox[5] consists in 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. The molecular-beam
maser provides a particularly neat example of this. Suppose the

---

[5]A. Einstein, B. Podolsky, and N. Rosen, Phys. Rev. **47**, 777 (1935).

field is initially in its ground state and the molecule in its upper state. When they start to interact at time $t = 0$, the state of (molecule + field) then becomes a linear combination

$$\Psi(t) = \cos \Theta_1 \; \Phi_1^+ \; e^{-i\omega_1^+ t} + \sin \Theta_1 \; \Phi_1^- \; e^{-i\omega_1^- t} \; ,$$

$$= a_1(t)\psi_2\phi_0 + b_1(t)\psi_1\phi_1 \; , \tag{6-1}$$

where the notation is the same as in Section 3. At time τ, the molecule leaves the cavity and continues on its way. But although interaction between molecule and field ceases, there remains a complete correlation between their states, for at later times than τ, molecule and field will still be jointly in the pure state

$$\Psi(t) = g_0(t)\psi_2\phi_0 + g_1(t)\psi_1\phi_1 \; , \tag{6-2}$$

where

$$g_0(t) \equiv a_1(\tau) \exp\left[\frac{-iE_2(t - \tau)}{\hbar}\right] , \tag{6-3a}$$

$$g_1(t) \equiv b_1(\tau) \exp\left[\frac{-i(E_1 + \hbar\omega)(t - \tau)}{\hbar}\right] . \tag{6-3b}$$

Now suppose we measure the energy of the molecule by passing it through an inhomogeneous field like that in the focuser. If we find the molecule in the upper state, then according to (6-2) a "reduction of the wave packet" occurs and the field is left in state ϕ_0. If we find the molecule in the lower state, on the other hand, then we know that the field must be in state ϕ_1. Thus by measuring the energy of the molecule, we can predict with certainty the result of a measurement of the energy of the field. We cannot, however, say anything at all about the phase of the electric field, since this does not commute with H_f.

But instead of measuring the energy of the molecule, we could decide to measure instead its dipole moment $\vec{\mu}$. According to quantum mechanics, we must obtain one of the eigenvalues $\pm\mu$ of the operator μ_{op}. Now the eigenstates of μ_{op}, defined by $\mu_{op} \mathcal{X}_\pm = \pm \mu \mathcal{X}_\pm$, are

$$\chi_{\pm} = \frac{1}{\sqrt{2}} \, (\psi_1 \pm \psi_2), \qquad (6\text{-}4)$$

so that the wave function (6-2) can be written equally well as

$$\Psi(t) = \chi_{+} \left[\frac{g_o(t)\phi_o + g_1(t)\phi_1}{\sqrt{2}} \right] + \chi_{-} \left[\frac{g_o(t)\phi_o - g_1(t)\phi_1}{\sqrt{2}} \right].$$
$$(6\text{-}5)$$

Suppose our measurement at time t_1 gives the result $(+\mu)$. Then we know that at this same instant the field must have been in the state

$$\phi_{+}(t) = g_o(t_1)\phi_o + g_1(t_1)\phi_1 \, , \qquad (6\text{-}6)$$

and therefore, at any time $t > \tau$, the field is described by the pure state

$$\phi_{+}(t) = g_o(t_1)\phi_o + g_1(t_1)\phi_1 \, e^{-i\omega(t - t_1)}. \qquad (6\text{-}7)$$

From this, using the relations of Section 3, we find the expectation value of electric field to be

$$\langle E(t) \rangle_{+} = - \frac{2\alpha}{\mu} \, \text{Re}\left[g_o(t_1)g_1^{*}(t_1) e^{i\omega(t-t_1)} \right]$$

$$= \frac{\alpha \sin 2\Theta_1}{\mu} \, \text{Re}\left\{ \left[\cos 2\Theta_1 (1 - \cos 2\beta\tau) + i \sin 2\beta\tau \right] \right.$$

$$\left. e^{i(\omega-\Omega)(t_1-\tau)} \, e^{i\omega(t-t_1)} \right\}. \qquad (6\text{-}8)$$

For simplicity, let the cavity be tuned exactly on the natural line frequency $(\omega = \Omega)$, and choose the time of interaction τ so that $4\beta\tau = \pi$. Then $\Theta_1 = \pi/4$, and (6-8) reduces to

$$\langle E(t) \rangle_{+} = - \frac{\alpha}{\mu} \sin \omega(t - t_1). \qquad (6\text{-}9)$$

If, on the other hand, measurement of μ_{op} at time t_1 yields the result $(-\mu)$, then we know that the field must, at time t_1, have been in the state

$$\phi_{-}(t_1) = g_o(t_1)\phi_o - g_1(t_1)\phi_1 \, , \qquad (6\text{-}10)$$

and, repeating the above argument, we obtain instead

$$\langle E(t) \rangle_- = + \frac{\alpha}{\mu} \sin \omega(t - t_1), \quad t > \tau. \qquad (6\text{-}11)$$

The remarkable thing about (6-9) and (6-11) is that they still contain t_1. Merely by choosing the time at which we measure μ, we can "force" the electric field to have any phase we please, except for an uncertainty of $180°$! The measurement of μ can, in principle, be carried out at a time when the molecule is arbitrarily far from the cavity, so there can be no question of any physical interaction which could influence the field in the cavity.

This example allows us to see the Einstein-Podolsky-Rosen paradox in a particularly excruciating form. Clearly, once we have seen the full implications of this situation, then we must give up the notion that physical systems have "real," objective states which exist independently of human knowledge, or else we must decide that quantum theory is not valid when applied to such correlated states.

Modern physics is firmly committed to the view that the test of a theory is not whether it contradicts preconceived philosophical notions, but whether it contradicts experimental facts. However difficult this situation may be conceptually, it will have no effect on quantum theory unless it should prove possible to reduce it to a question of experimental fact, involving experiments which can actually be carried out in the laboratory.

Is there any possibility that the molecular-beam maser might provide such a crucial test of quantum electrodynamics? This would seem to be a question well worth studying. However, we have to note the following. Direct approaches involving single-molecule measurements, as visualized above, even if feasible in the laboratory, probably would not have any bearing on these questions. The reason for this has been pointed out many times by Bohr. The essential thing will always be, not the mode of description furnished by quantum theory, but its predictions as to observable effects. We have, it is true, the choice of making definite predictions as to the energy of the field or of being able to say something about its phase. But these are different experiments, requiring mutually exclusive experimental arrangements.

We could imagine an arrangement whereby after measuring energy or moment of the first molecule, we send a second molecule through the cavity as a "probe" to examine the state of the field left by the first molecule. When the second molecule emerges, we can measure its energy or moment, and quantum theory will predict certain correlations between the results of the two measurements, of the form, "in a certain calculable fraction of those cases where the first molecule yields a moment $(+\mu)$, we should find the second molecule to have a moment $(-\mu)$, $\cdots$, etc." If experiment should show a significantly different degree of correlation, then of course we would have something. But similar correlations would be expected on the basis of almost any reasonable causal theory one might propose as an alternative to quantum electrodynamics, and it seems very unlikely that any such experiment would result in a clear-cut decision.

A more promising kind of approach would appear to be based on the result noted in Section 5: the semiclassical theory and quantum electrodynamics predict different equations of motion for a molecule in the field. This difference, furthermore, arises just from the possibility of those correlated states which cause the above conceptual difficulties. Consequently, 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 operation, this would constitute an indirect, but very convincing, check on those aspects of quantum theory which lead to the Einstein-Podolsky-Rosen paradox. In the next section we look more closely at this difference.

7. EQUATIONS OF MOTION

We have seen that the semiclassical theory leads to the following equation for describing the effect of the field on the state of a molecule:

$$\ddot{M} + \Omega^2 M = - K^2 W E, \qquad (5\text{-}12a)$$

where M, W are the dipole moment and energy of the molecule, E the electric field, and $K \equiv 2\mu/\hbar$. If the quantities M, W, E are interpreted as expectation values, this is equivalent to the equation

$$\frac{\partial^2}{\partial t^2} \langle \mu \rangle + \Omega^2 \langle \mu \rangle = - K^2 \langle H' \rangle \langle E \rangle . \qquad (7\text{-}1)$$

In quantum electrodynamics, the operator equation of motion (5-27) leads instead to

$$\frac{\partial^2}{\partial t^2} \langle \mu \rangle + \Omega^2 \langle \mu \rangle = - K^2 \langle H'E \rangle . \qquad (7\text{-}2)$$

We now write out the right-hand sides of (7-1) and (7-2) in terms of the density matrix $(mn| \rho |m'n')$. From the relations of Sections 3 and 5, we have

$$\langle H' \rangle = \sum_{\substack{m,n \\ m',n'}} \frac{\hbar \Omega}{2} (-)^m \, \delta_{mm'} \, \delta_{nn'} (m'n'| \rho |mn) ,$$

$$= \frac{\hbar \Omega}{2} \sum_{n} [(2n| \rho |2n) - (1n| \rho |1n)] ; \qquad (7\text{-}3)$$

$$\langle E \rangle = \sum_{\substack{m,n \\ m',n'}} \left\{ - \frac{\alpha}{\mu} \left[\sqrt{n} \, \delta_{n,n'+1} + \sqrt{n+1} \, \delta_{n+1,n'} \right] \delta_{mm'} \right\} (m'n'| \rho |mn) ,$$

$$= - \frac{2\alpha}{\mu} \, \mathrm{Re} \sum_{m,n} \sqrt{n+1} \; (mn| \rho |m,n+1) ; \qquad (7\text{-}4)$$

$$\langle H'E \rangle = \sum_{\substack{m,n \\ m',n'}} \left\{ - \frac{\hbar \Omega \alpha}{2\mu} (-)^m \delta_{mm'} \left[\sqrt{n} \, \delta_{n,n'+1} + \sqrt{n+1} \, \delta_{n+1,n'} \right] \right\} (m'n'| \rho |mn) ,$$

$$= - \frac{\hbar \Omega \alpha}{\mu} \, \mathrm{Re} \sum_{n} \sqrt{n+1} \left[(2,n| \rho |2,n+1) - (1,n| \rho |1,n+1) \right] .$$

$$(7\text{-}5)$$

It should be noted that a direct comparison of (7-1) and (7-2) is not really justified; for the transition from (5-12a) to (7-1) is not.

In a fully consistent semiclassical theory the variation of M, W, E, would be determined from solving Eqs. (5-12) instead of "borrowing" the solutions of the quantum electrodynamics problem as is implied by (7-3) and (7-4). Therefore the following arguments cannot claim full validity. It is apparent from the above equations, however, that in general the difference between (7-1) and (7-2) could be very great. For example, only diagonal elements of ρ contribute to $\langle H' \rangle$, while no diagonal elements contribute to $\langle H'E \rangle$.

Because of this, it is easy to invent special initial density matrices (for example, the one representing the field in the ground state and an entering molecule with $\sigma_{12} \neq 0$) for which the effect of correlations is decisive. We can have $\langle H' \rangle \langle E \rangle = 0$, but $\langle H'E \rangle \neq 0$ throughout the motion. However, these seem to represent idealized cases which could hardly be set up experimentally. Realistic situations are those where the magnitude of the field is well determined on a percentage basis, but with uncertainties still large compared to effects of a single photon; for example, where all elements of $(mn| \rho |m'n')$ corresponding to $n \simeq 10^6 \pm 10^3$ are significantly large, other elements being small. Under these conditions, however, the major contributions to $\langle H' \rangle$, $\langle E \rangle$, and $\langle H'E \rangle$ all come from the same range of quantum numbers, and unless there is some very fine-grained variation of elements of ρ within this range (which would represent a far more detailed state of information than we ever have in practice), it turns out that the difference between $\langle H' \rangle \langle E \rangle$ and $\langle H'E \rangle$ will be negligible.

Thus, although in principle the difference is there and this point perhaps deserves further study, the writer has been unable to find any realistic situation in which it could become a question of experimental fact. The results of the following section make the prospects appear still more 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.

8. SOLUTIONS OF THE NONLINEAR SEMICLASSICAL EQUATIONS

In this section we look more closely at the system of equations (5-12) representing the semiclassical theory. It is convenient to eliminate the constants K, S by the change of variables

$$x(t) = K \, E(t),$$

$$y(t) = KS \, M(t), \qquad (8\text{-}1)$$

$$z(t) = K^2 S \, W(t),$$

whereupon the equations of motion reduce to

$$\ddot{x} + \omega^2 x = y, \qquad (8\text{-}2a)$$

$$\ddot{y} + \Omega^2 y = -zx, \qquad (8\text{-}2b)$$

$$\dot{z} = x\dot{y}, \qquad (8\text{-}2c)$$

and the conservation laws (5-15) become

$$(\dot{y})^2 + \Omega^2 y^2 + z^2 = \text{const.}, \qquad (8\text{-}3)$$

$$(\dot{x})^2 + \omega^2 x^2 + 2(z - xy) = \text{const.} \qquad (8\text{-}4)$$

These quantities are not dimensionless, but involve only time. Their numerical values will therefore depend on our unit of time. The constant in (8-3) is

$$\left(\frac{8\pi \, \mu^2 \, \omega^2 \, \Omega}{\hbar \, J_1^2 \, V} \right)^2 = 3.4 \times 10^{51} \ \sec^{-8}. \qquad (8\text{-}5)$$

Noting that $(2.76 \times 10^6)^8 = 3.4 \times 10^{51}$, we see that if we choose our unit of time as 0.36 microsecond (call it a subsecond), this constant becomes numerically equal to unity, and (8-3) becomes

$$(\dot{y})^2 + \Omega^2 y^2 + z^2 = 1 \ \text{subsec}^{-8}, \qquad (8\text{-}6)$$

i.e., the projection of the orbit onto the space of $\dot{y}$, Ωy, z always lies on the unit sphere. There is no limitation imposed on numerical values of the field x, but the amplitudes in typical maser operation $(n \lesssim 10^6$ quanta) correspond to

$$|x| \lesssim \frac{2\mu}{\hbar} \sqrt{\frac{2\pi n \hbar \omega}{J_1^2 \, V}} = 5.3 \times 10^4 \ \sec^{-1} = 2 \times 10^{-2} \ \text{subsec}^{-1}$$

$$(8\text{-}7)$$

Therefore, since $|\overset{\circ}{y}| \leq 1$, we see from (8-2c) that z will always be slowly varying; under the conditions of interest here, any appreciable change in z can take place only in times of the·order of tens to thousands of subseconds.

We thus have a very convenient time scale for our problem, for the frequencies $(\omega/2\pi)$, $(\Omega/2\pi)$ are about 10^4 cycles per subsecond, and the flight time of a molecule through the cavity is of the order of 1000 subseconds. On this time scale, the oscillations of field (x) and dipole moment (y) are still very rapid, while secular changes due to their interaction are very slow. Because of this clean separation into fast and slow changes, one might hope to get a fairly complete understanding of the solutions of (8-2) in spite of their nonlinear character.

The simplest approximate solution is the one wherein we ignore the time variation of z, thereby converting the problem into a linear one, similar to the case of two coupled pendulums. The two normal modes are found by assuming that x and y have a common time factor $\exp(i\nu t)$; if $z = $ const., the Eqs. (8-2a), (8-2b) then reduce to

$$(\omega^2 - \nu^2)(\Omega^2 - \nu^2) + z = 0, \qquad (8\text{-}8)$$

or

$$\nu^2 = \frac{\omega^2 + \Omega^2}{2} \pm \frac{1}{2}\sqrt{(\omega^2 - \Omega^2)^2 - 4z} \, . \qquad (8\text{-}9)$$

We see here a new feature, not present in the case of coupled pendulums: if $z > 0$ and the cavity is tuned so closely to the natural line frequency that

$$|\omega^2 - \Omega^2| < 2\sqrt{z} \, , \qquad (8\text{-}10)$$

the square root in (8-9) 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, contrary to what is usually supposed, <u>the semiclassical theory does lead to a prediction of spontaneous emission.</u> Since z is just the energy of the molecule, in unconventional units, we see that the condition for existence of unstable growing oscillations is that the

molecule's wave function $\psi = a\psi_1 + b\psi_2$ must contain more of the upper state than the lower: $|b|^2 > |a|^2$.

We work out some further details for the case that the cavity is tuned exactly on the natural line frequency: $\omega = \Omega$. Then (8-9) reduces to

$$\nu^2 = \omega^2 \pm i \sqrt{z} , \qquad (8\text{-}11)$$

or, to an extremely good approximation,

$$\nu = \omega \pm \frac{i\sqrt{z}}{2\omega} . \qquad (8\text{-}12)$$

If we start with the molecule nearly in the upper state, then $z \approx 1$ subsec^{-4}, and the amplitude of the field varies like

$$\exp\left[\frac{\sqrt{z}\ t}{2\omega}\right] e^{i\omega t} = \exp\left(\frac{\alpha t}{\hbar}\right) e^{i\omega t} , \qquad (8\text{-}13)$$

where α is the interaction constant defined in Eq. (3-5). This is to be compared to the result (3-28) 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 $(\hbar/\alpha)$ for spontaneous emission.

This appears to be one of the most important results of the work here reported, and it shows that the relation between quantum electro-dynamics and the semiclassical theory of radiation is of a quite different type than is usually assumed. Physically, it means that when the molecule has any 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 $z > 0$. These linear relations, of course, cannot hold indefinitely. From the conservation law (8-6) it is clear that when the amplitude of the y oscillation increases, the magnitude of z must decrease, and this will eventually put a stop to the emission process.

For a qualitative picture of the secular changes in the case $\omega = \Omega$, we may consider the orbits in the $(\overset{\circ}{x}, \omega x)$ plane and in the $(\overset{\circ}{y}, \omega y)$ plane, as in Fig. 1.
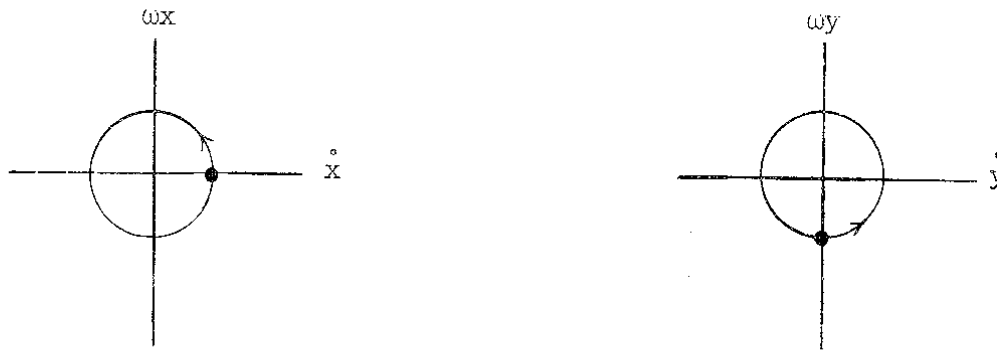
FIG. 1.—Closed orbits in the phase space of the x and y oscillators. The dots indicate that the x motion is $90°$ ahead of the y motion in phase.

Noting that xy is typically about 10^{-6} times smaller than z, the conservation law (8-4) reduces, in almost all cases, to

$$(\overset{\circ}{x})^2 + \omega^2 x^2 + 2z = \text{const.}, \qquad (8\text{-}14)$$

which shows that when z increases, the orbit in the $(\overset{\circ}{x}, \omega x)$ plane must shrink, and vice versa. Similarly the conservation law (8-6) shows that if $|z|$ increases, the y-orbit must shrink, and vice versa. Therefore the direction of all secular changes is determined by the sign of z and $\overset{\circ}{z}$. In the equation $\overset{\circ}{z} = x\overset{\circ}{y}$ we can for all practical purposes replace $x\overset{\circ}{y}$ by its average $\overline{x\overset{\circ}{y}}$ over one cycle, since we are interested in the trend of z over time scales of many cycles, rather than small rapid fluctuations whose effect averages to zero over a cycle. Secular changes in z thus depend only on the sign of $\overline{x\overset{\circ}{y}}$.

Now whenever the x motion is advanced in phase over the y motion, we have $\overline{x\overset{\circ}{y}} > 0$. In this case, z will slowly increase, and the x-orbit will shrink. The y-orbit will then grow if $z < 0$, shrink if $z > 0$. If the y motion is advanced in phase over the x motion, all these secular changes are reversed. Thus the situation may be summarized by the orbit diagrams of Fig. 2. The situations depicted in the column labelled $z > 0$ are just the growing and shrinking normal modes of Eq. (8-12).

Whenever the x-orbit is expanding, energy is being delivered from the molecule to the field, and the necessary and sufficient condition for this is that the y motion be advanced in phase over the x motion.
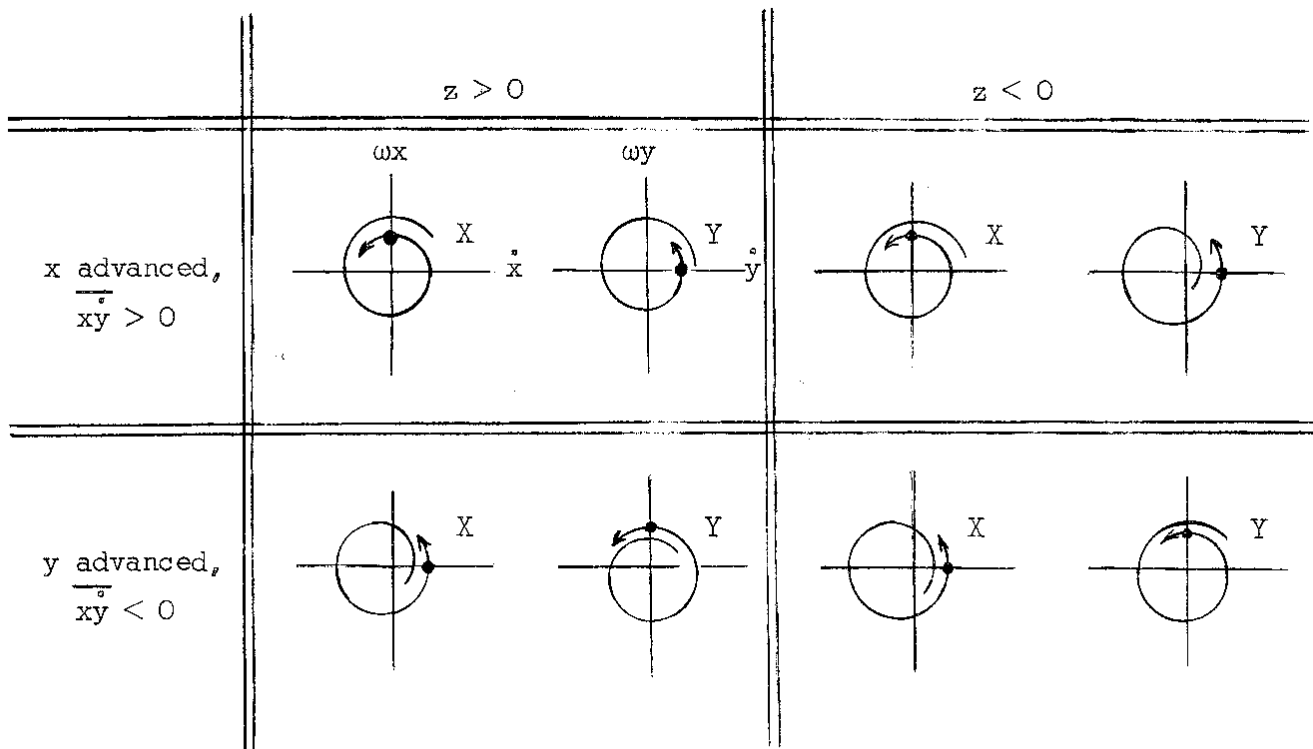
	z > 0	z < 0

FIG. 2.—Secular changes in orbits for the
four combinations of signs of $\overline{x\dot{y}}$ and z.

Thus in order to understand the long-time course of events,
we must study the secular changes in relative phase of x and y.
To this end, introduce the slowly varying complex amplitude X, Y,
defined by

$$\dot{x} + i\omega x = X(t)\ e^{i\omega t}, \qquad (8\text{-}15)$$

$$\dot{y} + i\omega y = Y(t)\ e^{i\omega t}. \qquad (8\text{-}16)$$

If we regard the above orbit diagrams as complex planes, the quantities
depicted are just the complex numbers (8-15) and (8-16). Noting the
properties

$$(\dot{x})^2 + \omega^2 x^2 = |X|^2, \qquad (8\text{-}17)$$

$$\ddot{x} + \omega^2 x = \dot{X}\ e^{i\omega t}, \qquad (8\text{-}18)$$

and similarly for Y, we can write the equations of motion (8-2) in the form:

$$2i\omega\dot{X} = Y - Y^* e^{-i2\omega t}, \tag{8-19a}$$

$$2i\omega\dot{Y} = -z\left[X - X^* e^{-i2\omega t}\right], \tag{8-19b}$$

$$4i\omega\dot{z} = XY^* - X^*Y + XY e^{i2\omega t} - X^*Y^* e^{-i2\omega t}, \tag{8-19c}$$

which are exact for the case $\omega = \Omega$. The conservation laws become simply

$$|Y|^2 + z^2 = 1, \tag{8-20}$$

$$|X|^2 + 2z = \text{const.} \tag{8-21}$$

Now the quantities X, Y are slowly varying functions of time, and again it is their average change over many cycles, rather than the very small rapid fluctuation at frequency 2ω, which interest us. Therefore the oscillating terms in (8-19) may be dropped, since their average over a cycle is negligible compared to that of the "DC" terms. The system of equations determining secular changes of both amplitude and phase is therefore

$$2i\omega\dot{X} = Y, \tag{8-22a}$$

$$2i\omega\dot{Y} = -zX, \tag{8-22b}$$

$$4i\omega\dot{z} = XY^* - X^*Y. \tag{8-22c}$$

It is readily verified that the conservation laws (8-20), (8-21) are exact consequences of (8-22). Differentiating (8-22c) once more are making use of the conservation laws, we can eliminate X and Y, obtaining the equation

$$4\omega^2\ddot{z} - 3z^2 + 2az + 1 = 0, \tag{8-23}$$

where $2a = |X|^2 + 2z$ is the constant of the motion (8-21). A first integral of (8-23) is obtained immediately:

$$2\omega^2(\dot{z})^2 - z^3 + az^2 + z = \text{const.} = c, \tag{8-24}$$

which has the form of the Hamilton-Jacobi equation for motion of a particle in a particular potential well. For any motion in which

- 33 -

either of the points $z = \pm 1$ is accessible, we have $c = a$. To see this, note that if $|z| = 1$, we have $Y = 0$ from (8-20), and hence $\dot{z} = 0$. But then (8-24) reduces to $a = c$. For any such motion, the cubic polynomial in (8-24) factors, and the solution is

$$\frac{t}{\omega \sqrt{2}} = \int_{z(0)}^{z(t)} \frac{dz}{\sqrt{(1 + z)(1 - z)(a - z)}} . \qquad (8\text{-}25)$$

The z-motion is therefore periodic, between turning-points represented by singularities of the integrand. If $a > 1$, these turning-points are at $z = \pm 1$, while if $a < 1$, they are at $z = -1$ and $z = a$. Noting that the condition for the total energy stored in the field to be just $\hbar \omega$ is that $|X|^2 = 4$, we see from (8-21) that

$$\frac{a + 1}{2} = n + 1 \qquad (8\text{-}26)$$

where n is the number of quanta in the field when the molecule is in its upper state; therefore $(n + 1)$ is the number when the molecule is in its ground state. There is in this theory no restriction of n to integer values. The smallest value which a can attain is represented by zero energy in the field and the molecule in its ground state, or $(n + 1) = 0$. (When $n < 0$, 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 in (8-25) is one of the standard forms defining elliptic functions. Using the standard notation $sn(u, k)$, the solution for the case $n \geq 0$ is

$$z(t) = -1 + 2 \, sn^2 \left[\sqrt{n + 1} \, \frac{\alpha t}{\hbar} + Q, \quad \frac{1}{\sqrt{n + 1}} \right], \qquad (8\text{-}27)$$

where

$$Q \equiv sn^{-1} \left[\sqrt{\frac{z(0) + 1}{2}} , \quad \frac{1}{\sqrt{n + 1}} \right] \qquad (8\text{-}28)$$

is the initial phase of the motion. In the limit of large n, the elliptic functions approach trigonometric functions, as is seen most

easily directly from (8-25). If $a \gg 1$, then (8-25) reduces to

$$\frac{t}{\omega \sqrt{2}} \simeq \frac{1}{\sqrt{a}} \int^{} \frac{dz}{\sqrt{1-z^2}} = \frac{1}{\sqrt{a}} \sin^{-1} z(t) + \text{const.,}$$

or

$$z(t) \simeq \sin\left(2 \sqrt{n} \ \frac{at}{\hbar} + \Theta\right). \tag{8-29}$$

The case $a = 1$, $n = 0$ is a special one, for the integrand of (8-25) then develops a first-order pole at $z = 1$. The solution (8-27) is still valid but it is no longer a periodic solution, for $\text{sn}(u,1)$ is qualitatively like $\tanh u$; it approaches ± 1 asymptotically as $u \longrightarrow \pm \infty$. 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 (8-27) then represent the "shrinking normal mode" of (8-12), where X is 90° ahead of Y. (This phase relation is in fact maintained throughout the part of the motion (8-27) in which z increases; throughout the decreasing part, X is 90° behind Y).

The point $z = 1$ is a metastable point of the orbit in this case, for if we start out with exactly the initial conditions $z = 1$, $X = Y = 0$, 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 (8-13) will be started up (unless the phase relation between X and Y happens to be exactly the value for the pure shrinking mode), and eventually the energy of the 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 $z = 1$ according to the solution (8-27), but requiring an infinite time to do so.

The constant of integration c in (8-24) is related to the relative phase of the X and Y motions, and other values than $c = a$, as in (8-25), lead to more general solutions. To show this, note that at $z = 0$, (8-22c) and (8-24) combined give

$$4\omega^2 (\dot{z})^2 = 2c = |\text{Im}(XY^*)|^2 = |X|^2 |Y|^2 \sin^2 \Theta(0), \tag{8-30}$$

where $\Theta(z)$ is the relative phase of the X and Y motions, at a time when z has the specified value. But in the case $z = 0$, the conservation laws give $|Y|^2 = 1$, $|X|^2 = 2a$, and therefore

$$c = a \sin^2 \Theta(0). \qquad (8\text{-}31)$$

For arbitrary relative phase angle $\Theta(0)$, the turning points of the z motion are the two lowest roots of the equation

$$z^3 - az^2 - z + c = (z - z_1)(z - z_2)(z - z_3) = 0, \qquad (8\text{-}32)$$

or

$$(1 - z^2)(a - z) = a \cos^2 \Theta(0). \qquad (8\text{-}33)$$

Let us order the roots so that $z_1 \le z_2 \le z_3$. Then, if $a > 1$ and $c < a$, we have the relations

$$-1 < z_1 \le z \le z_2 < 1 < a < z_3, \qquad (8\text{-}34)$$

and z oscillates periodically between z_1 and z_2. The molecule never gets entirely in the ground state or entirely in the upper state. The solution $z(t)$ is a generalization of (8-27):

$$\sqrt{\frac{z - z_1}{z_2 - z_1}} = \text{sn}\left[\sqrt{\frac{z_3 - z_1}{2}} \frac{at}{\hbar} + Q, \sqrt{\frac{z_2 - z_1}{z_3 - z_1}} \right] \qquad (8\text{-}35)$$

which again approaches a sinusoidal oscillation in the limit of many quanta, $a \gg 1$.

For arbitrary values of z, the relative phase of the X and Y motions is given by combining (8-30) with (8-24) and the conservation laws, with the result

$$\sin^2 \Theta(z) = \frac{(z - z_1)(z - z_2)(z - z_3)}{(z + 1)(z - 1)(z - a)}. \qquad (8\text{-}36)$$

This reduces to the value $\Theta(z) = \pm 90^\circ$ as previously noted, in the case $c = a$.

To summarize the above results, a close analysis of the semiclassical theory for a simple special case reveals many unexpected and remarkable features. It is usually supposed that the semiclassical theory of

radiation cannot account for spontaneous emission; indeed this belief was historically one of the main reasons for introducing field quantization. However, we see that when we take into account simultaneously both the effect of the molecule on the field _and_ the effect of its radiated field reacting back on the molecule, we are led to a nonlinear system of coupled equations whose solutions are readily found, and which exhibit almost all the properties usually associated with field quantization.

In the above analysis the semiclassical theory has been put to a far more severe test than would be the case in practical maser calculations, and it has met the test very well. Even in the case of field intensities corresponding to one or two quanta, the semiclassical theory gives solutions reproducing almost quantitatively everything that is found in the quantum electrodynamics analysis. The characteristic times of interaction processes turn out just the same. The "quantum jumps" are still with us, but they now appear 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 between molecule and 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, 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 $Y_+(0)$ will, at time t, have a Y amplitude of

$$Y_+(0) \, \exp\left(\frac{\alpha t}{\hbar}\right) = Y_+(t) \; .$$

Let us agree to say that when this reaches the value K, the molecule is actively emitting energy. The, no matter what the probability distribution of initial values, provided only that this distribution is a continuous function in the neighborhood of the metastable state $z = 1$, we find that the number of molecules emitting at time t is porportional to $\exp(-2\alpha t/\hbar)$. Thus, the "law of radioactive decay," or "time-proportional transition probabilities" appears in this analysis

as a simple 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 depends only on the interaction constant 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 time $t = 0$. 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.

9. CONCLUSION

The above analysis is evidently no more than a very preliminary survey of the relations between different theories which might be used in studying the ultimate limitations imposed by quantum theory on the noise figure and frequency stability of molecular beam maser amplifiers and oscillators. The limitation to a single molecule in the cavity is easily removed in the semiclassical theory, but with more difficulty in the quantum electrodynamics analysis. For example, the energy levels in the case of two molecules present involve solution of a general cubic equation, for three molecules we have a quartic equation, etc. Analyses of the type given in Section 3, therefore, are quite impractical in the case of a large number of molecules. However, the following reasoning shows that there must be some as yet undiscovered way of circumventing this complication.

Even though a complete theoretical discussion would require in principle keeping track of perhaps $10^{10^{10}}$ different parameters, the purpose of the theory is only to predict the values of three or four quantities which are to be measured. Therefore, it must be that the overwhelming majority of the microscopic parameters are irrelevant to the particular predictions we are after. If this were not so, the experimental measurements could not be reproducible, for when we repeat an experiment, we surely do not repeat all the details of $10^{10^{10}}$ different parameters. In other words, a form of statistical mechanics must be applicable, in which we calculate averages over all possible microscopic conditions which conform to the knowledge we have of the

state of the system. Any quantity which emerges from this treatment with a very sharply peaked probability distribution must have practically the same value for _each_ of the possible microscopic states, and this value can be predicted with confidence without having to go into microscopic details. Viewed in this way, the maser problem becomes one of the theory of irreversible statistical mechanics, and this approach is now being pursued.

The unexpected success of the semiclassical theory, on the other hand, suggests that it may provide a much more efficient formalism for handling such problems. The introduction of many molecules, cavity losses, etc., is easily accomplished in this treatment, and the same statistical considerations just mentioned apply equally well. There seem to be excellent grounds for expecting that this extended semi-classical theory would reproduce every feature of experimental significance which could be found in the quantum electrodynamics analysis.

The relationships demonstrated here between quantum electrodynamics and the semiclassical theory of radiation evidently carry implications far beyond the field of maser theory. Almost any example of the interaction of matter and radiation would seem to be tractable by these methods.

Laser Amplifier References

Pao Yok Han J. Opt. Soc. Am. <u>52</u>, 871 (1962) NMR Tech.

A. Yariv & J. P. Gordon IEEE <u>51</u>, 4 (1963)
(3 level) claim $\frac{N_2}{N_2 - N_1}$ extra factor noise

O. S. Heavens Appl. Optics Supp. 1, 1 (1962)
Spont. Em. into cavity. also:

Purcell PR 69, 681 (1946)

Senitzky PR 111, 3 (1958)

V. N. Smiley IEEE 51, 120 (1963) Fabry-Perot
with negative abs. coeff.
<u>actually built</u> one

Kislink & Boyle IRE <u>49</u>, 1563 (1961) Ruby

Shawlow & Townes PR <u>112</u>, 1940 (1958)