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TECHNION-ISRAEL INSTITUTE OF TECHNOLOGY
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SEPTEMBER 1976

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MACROSCOPIC DESCRIPTION OF LASER-TYPE MATTER

Annual Report

(Second year)

by

I.R. Senitzky

September 1976

EUROPEAN RESEARCH OFFICE

United States Army

London, England

GRANT NUMBER DA-ERO-75-G-012

Technion-Israel Institute of Technology

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) MACROSCOPIC DESCRIPTION OF LASER-TYPE MATTER		5. TYPE OF REPORT & PERIOD COVERED ANNUAL TECHNICAL REPORT August 1975 - August 1976
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) I. R. SENITZKY		8. CONTRACT OR GRANT NUMBER(s) DA-ERO-75-G-012
9. PERFORMING ORGANIZATION NAME AND ADDRESS TECHNION - ISRAEL INSTITUTE OF TECHNOLOGY		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 6.11.02A-1T161102B31E-00-483
11. CONTROLLING OFFICE NAME AND ADDRESS U.S. ARMY R&S GROUP (EUR) BOX 65, FPO NEW YORK 09510		12. REPORT DATE SEPTEMBER 1976
		13. NUMBER OF PAGES 37
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) UNCLASSIFIED
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

APPROVED FOR PUBLIC RELEASE: DISTRIBUTION UNLIMITED

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

LASER THEORY, QUANTUM THEORY.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

See over page.

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

Specialization of the previously developed boson-second-quantization (BSQ) formalism for the description of cooperative atomic behavior to the case of a two-level laser allows comparison with and criticism of other theories of cooperative behavior. Application of the BSQ formalism to a model of a three-level laser leads to equations of motion in terms of macroscopic variables without the use of phenomenological modifications. These equations, which can easily be generalized to more than three levels, may be interpreted both classically and quantum-mechanically, and offer the possibility of investigating a number of problems which are of both conceptual and practical interest.

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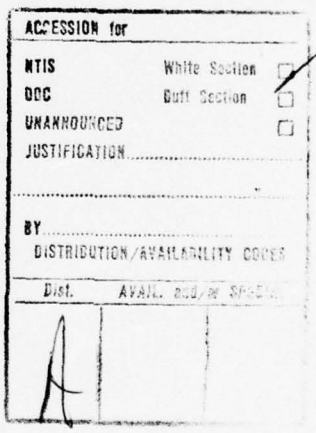

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ABSTRACT

The suitability of a boson-second-quantization (BSQ) formalism for the description of cooperative atomic behavior - such as that encountered in quantum optics or laser-type phenomena - is explained. The physical significance of BSQ energy states and atomic coherent states, as well as the corresponding first quantization descriptions are discussed. Specialization to the case in which the individual atomic systems are characterized by two levels allow comparison of the BSQ theory with other theories of cooperative behavior, not all of which can be generalized to the case of more than two levels. Passage to the classical limit, an analytical process that must be possible for macroscopic phenomena, is shown to be accomplished very simply in the BSQ formalism by conversion of the small number of collective (quantum mechanical) variables to classical random variables which are prescribed according to the quantum state of the total system. It is found that conventional semi-classical theory is a special case of the fully classical limit of the BSQ formalism, the specialization being the requirement that the system be in an atomic coherent state. The justification for the identification of coherent states with a classical description and energy states with a quantum mechanical description, found in some of the literature on two-level systems, is questioned.

The BSQ formalism is applied to a model of a three-level laser, with the dynamics of the pumping levels explicitly considered. Equations of motion in terms of macroscopic variables are obtained without the use of phenomenological modifications. These equations, which can be easily generalized to more than three levels, may be interpreted both classically and quantum-mechanically, and offer the possibility of investigating a number of problems which are of both conceptual and practical interest.

CONTENTS

I. INTRODUCTION

In a previous article[1] it was pointed out that the macroscopic description of laser-type matter can be based advantageously on boson second quantization (BSQ) theory, in which the individual atomic systems (hereafter referred to as "molecules") are considered to be bosons. The reason is associated with the fact that laser-type, or quantum-optical, phenomena are cooperative phenomena, and the boson formalism of quantum mechanics describes a system in which there exists - in a certain sense - the maximum cooperation among identical particles. Thus, for a number of particles to be describable as bosons, the total wave function must be symmetrical with respect to all particles, that is, it must exhibit the maximum amount of symmetry. This mathematical requirement can be translated (freely) into physical terms by the statement that all particles must behave similarly. The same idea underlies the well known fact that a boson field may be described, in many circumstances, classically, in contrast to a fermion field, which has no classical analog. It should be noted, however, that the use of the boson formalism in the present instance is unrelated to the intrinsic elementary character of the particles, or their statistics, since we can safely assume, as far as interactions of present interest are concerned, that the molecules have non-overlapping wave functions and are idstringuishable, obeying Maxwell-Boltzman statistics. It is only with respect to their collective behavior (say, effect on the field) that the molecules are indistinguishable. One might say that the use of BSQ focuses our attention on the essence of laser-type behavior, namely, cooperation. In the present article, the BSQ method of describing collective phenomena will be developed further.

The main emphasis will be placed on the physical significance of the description, its connection with the usual first-quantization description, and the relationship of the BSQ formalism to classical and semiclassical descriptions.

II. THE FORMALISM

For the sake of completeness, it is useful to present a summary of the formalism to be used[1]. Consider a number of identical molecules, each of which has n energy levels of interest, with all molecules coupled similarly to other systems, such as the electromagnetic field. Let the molecular energy levels be given by $h\omega_i$, i=1,2,.....,n and the corresponding one-molecule states by $|\phi_i\rangle$. The space in which the states of the system are described in the BSQ formalism is spanned by (orthonormal) basis vectors

$$|r_1 \ldots r_i \ldots r_n\rangle \qquad (2\text{-}1)$$

where the r_i's are non-negative integers.

Unless stated otherwise, the Heisenberg Picture will be used.

The fundamental operators are taken to be $a_i(0)$ and $a_i^\dagger(0)$, which, when operating on the basis vectors, yield

$$a_i(0)|r_1 \ldots r_i \ldots r_n\rangle = r_1^{1/2}|r_1 \ldots r_i\text{-}1 \ldots r_n\rangle \qquad (2\text{-}2a)$$

$$a_i^\dagger(0)|r_1 \ldots r_i \ldots r_n\rangle = (r_i+1)^{1/2}|r_1 \ldots r_i+1 \ldots r_n\rangle \, . \qquad (2\text{-}2b)$$

These operators obey the commutation relations

$$[a_i(t) , a_i^{\dagger}(t)] = 1 \tag{2-3}$$

(equal-time)
with all other/commutators vanishing. The Hamiltonian describing the collection of atoms is expressed by

$$H_o = \sum_i \hbar\omega_i a_i^{\dagger} a_i \quad . \tag{2-4}$$

It is seen immediately that the a_i's and $a_i^{\dagger}$'s are the well known annihilation and creation operators associated with harmonic oscillators of frequency ω_i. The coupling of the molecules to external systems takes place through their (collective) "dipole moment", given, in dimensionless form, by

$$d_{ij}^{(1)} = \frac{1}{2} (a_j^{\dagger} a_i + a_i^{\dagger} a_j) \tag{2-5a}$$

$$d_{ij}^{(2)} = -\frac{1}{2} i(a_j^{\dagger} a_i - a_i^{\dagger} a_j) \tag{2-5b}$$

$$d_{ij}^{(3)} = \frac{1}{2}(a_j^{\dagger} a_j - a_i^{\dagger} a_i) \quad . \tag{2-5c}$$

When not coupled to ther systems, the dipole moment components $d_{ij}^{(1)}$ and $d_{ij}^{(2)}$ oscillate with frequency $|\omega_i - \omega_j|$, the resonant frequencies of the molecule, while $d_{ij}^{(3)}$ is constant. Coupling to an external system is described generally by the interaction-Hamiltonian

$$H' = h \sum_{m=1}^{3} \sum_{i<j} \gamma_{ij}^{(m)} f_{ij}^{(m)} d_{ij}^{(m)} , \qquad (2\text{-}6)$$

where the $\gamma_{ij}^{(m)}$'s are coupling constants and $f_{ij}^{(m)}$ refers to the external system (say, a field). It is convenient to introduce the reduced variables $A_i(t)$, $A_i^\dagger(t)$ defined by

$$a_j(t) = A_j(t)e^{-i\omega_j t} , \quad a_j^\dagger(t) = A_j^\dagger(t)e^{i\omega_j t} . \qquad (2\text{-}7)$$

These variables vary slowly (compared to the natural oscillation) for sufficiently weak coupling between the atoms and other systems - a condition assumed throughout - and are constant in the absence of coupling.

III. ENERGY STATES AND COHERENT STATES

The basis vectors $|r_1 \ldots r_n\rangle$ are eigenvectors of the occupation operators $n_i(0)$, where

$$n_i = a_i^\dagger a_i = A_i^\dagger A_i , \qquad (3\text{-}1a)$$

and satisfy the eigenvalue equations

$$n_i(0)|r_1 \ldots r_n\rangle = r_i|r_1 \ldots r_n\rangle . \qquad (3\text{-}1b)$$

The basis vectors are also energy states of the free (uncoupled) system. A given value of Σr_i labels a subspace of the space corresponding to all

possible values of the r_i's. The operators $a_1(0)$ and $a_i^\dagger(0)$ transfer a vector from one subspace into another, but the subspace is invariant under the operations $d_{ij}^{(m)}(0)$. Since coupling to other systems takes place only through the dipole moment, the dynamical development of the system in the Schrodinger Picture does not take the state vector out of its initial subspace. This statement is merely the expression of the fact that the total number of molecules N is conserved, and is equivalent, in the Heisenberg Picture, to the statement that $\Sigma n_i(t)$ is a constant of motion,

$$\Sigma_i n_i(t) = N \tag{3-2}$$

It is clear that any operator corresponding to a dynamical variable must contain only terms which are products of equal numbers of annihilation and creation operators. However, the basic operators $a_i(0)$ and $a_i^\dagger(0)$, individually, are useful not only for computational purposes, but for viewing an arbitrary number of n level systems - which may constitute both microscopic and macroscopic matter - in therms of n harmonic oscillators .[1]

The expectation value with respect to an energy state of a product of annihilation and creation operators is nonvanishing only if the product can be reduced, by means of the commutation relationships (2-3), to a sum of products of occupation numbers. It is then obtained immediately by use of Eq. (3-1b).

In view of the fact that the molecules are - in principle - distinguishable, it is natural to investigate the meaning of an energy state in terms of one- molecule states, or, in terms of the usual first-quantization formalism. It should be noted that the energy state $|r_1 \ldots r_n\rangle$ does <u>not</u> correspond to the first-quantization state $|\psi(1 \ldots N)\rangle$ in which the

molecules labeled 1 to r_1 are in the one-molecule energy state $|\phi_1\rangle$, those labeled r_1+1 to r_1+r_2 in the one-molecule energy state $|\phi_2\rangle$, etc., but rather, to the (normalized) sum of all states obtained from $|\psi(1\ldots N)\rangle$ by a permutation of the molecular labels. Thus, one should not think of $|r_1\ldots r_n\rangle$ as a state in which the individual molecules are in energy states, except for the case N=1 and the case $r_i=\delta_{im}N$. In terminology explained in detail in connection with two level systems,[2,3] an energy state corresponds to a first-quantization state that is correlated and incoherent. From an intuitive viewpoint, one may think of the individual molecule as being in a combination of energy states for which the phase of the dipole-moment oscillation is unknown with respect to an absolute time scale, but is correlated with respect to that of the other molecules.[2] This intuitive explanation will become clearer later, when we discuss the classical limit.

As is to be expected, phases of oscillation of dynamical variables that do not commute with the Hamiltonian are completely unknown when the system is in an energy state $|r_1\ldots r_n\rangle$. Thus, the expectation values of the oscillating components of the (total) dipole moment, the variables through which the system couples in resonant-type interactions, vanish. If we are interested in coherent states, states for which $\langle d_{ij}^{(1)}(t)\rangle$ and $\langle d_{ij}^{(2)}(t)\rangle$ exhibit oscillatory behaviour, we must seek an appropriate superposition of energy states.

Let a coherent state [4] $|\{c\}_N\rangle$ be defined by the relationship

$$|\{c\}_N\rangle = \sum_{r_1 \ldots r_n}^{(N)} \left(\frac{N!}{\pi_i r_i!} \right)^{1/2} \pi_j c_j^{r_j} |r_1 \ldots r_n\rangle \ , \qquad (3\text{-}3)$$

where the patenthetical superscript (N) in the summation indicates that the summation is to be taken only over those values of r_1's for which

$$\sum_i r_i = N \ , \qquad (3\text{-}4)$$

and $\{c\}$ stands for a set of complex numbers $c_1 \ldots c_n$ satisfying the normalization relation

$$\sum_i c_i^{\,2} = 1 \ . \qquad (3\text{-}5)$$

It can be seen easilty that $|\{c\}_N\rangle$ is normalized:

$$\begin{aligned} \langle\{c\}_N|\{c\}_N\rangle &= \sum^{(N)} \frac{N!}{\pi_i r_i!} \pi_j (|c_j|^2)^{r_j} \ , \\ &= \left(\sum_j |c_j|^2 \right)^N \ , \qquad (3\text{-}6) \\ &= 1 \end{aligned}$$

We are interested, of course, in the effect of the fundamental operators on the coherent states. For convenience, we consider the atomic system in the following discussion to be free (uncoupled). Then, from Eqs. (2-2) and (2-7) one obtains

$$\begin{aligned} A_k|\{c\}_N\rangle &= \sum^{(N)} \left(\frac{N!}{\pi_i r_i!} \right)^{1/2} \pi_j c_j^{r_j} r_k |r_1 \ldots r_k - 1 \ldots r_n\rangle \\ &= N^{1/2} c_k |\{c\}_{N-1}\rangle \ . \qquad (3\text{-}7) \end{aligned}$$

The relationships

$$\langle\{c\}_N|A_j^\dagger = \langle\{c\}_{N-1}|N^{1/2}c_j^* \quad , \tag{3-8}$$

and

$$\langle\{c\}_N|A_j^\dagger A_k|\{c\}_N\rangle = Nc_j^* c_k \tag{3-9}$$

follow, so that we have

$$\langle a_j^\dagger a_i\rangle = Nc_j^* c_i \exp[i(\omega_j - \omega_i)t] \quad , \tag{3-10}$$

$$\langle d_{ij}^{(1)}\rangle = N|c_j c_i|\cos[(\omega_j - \omega_i)t + \theta_{ji}] \quad , \tag{3-11a}$$

$$\langle d_{ij}^{(2)}\rangle = N|c_j c_i|\sin[(\omega_j - \omega_i)t + \theta_{ji}] \quad , \tag{3-11b}$$

$$\langle d_{ij}^{(3)}\rangle = \frac{1}{2}N(|c_j|^2 - |c_i|^2) \quad , \tag{3-11c}$$

where θ_{ji} is defined by

$$c_j^* c_i \equiv |c_j c_i| e^{i\theta_{ji}} \tag{3-11d}$$

Equations (3-11a) and (3-11b) display the coherence of the state $|\{c\}_N\rangle$.

The meaning of the coefficients c_i becomes more transparent when we set $N = 1$. Then, Eq. (3-3) becomes

$$|\{c\}_1\rangle = \sum_{i=1}^{n} c_i|r_1 \cdots r_j \cdots r_n\rangle \tag{3-12a}$$

where the set of numbers r_j in each term is given by

$$r_j = \delta_{ji} \quad , \tag{3-12b}$$

In first-quantization notation, this state may be written as

$$|\phi> = \sum_{i=1}^{n} c_i |\phi_i> \quad , \tag{3-13}$$

where, it is recalled, the $|\phi_i>$'s are the one-molecule energy states. Thus the state $|\{c\}_1>$ describes a coherent superposition of one-molecule energy states, with the c_i's being the superposition constants. It is not difficult to see that the state $|\{c\}_N>$ is the BSQ analog of the (first-quantization) state

$$|\psi> = \prod_{k=1}^{N} | \; |\phi^{(k)}> \tag{3-14}$$

where the superscript (k) labels the molecules, and where

$$|\phi^{(k)}> = \sum_{i=1}^{n} c_i |\phi_i^{(k)}> \quad . \tag{3-15}$$

(Such a state is said to be coherent and uncorrelated, in terminology referred to previously.[2]) In fact, the square of the absolute value of the coefficient of $|r_1 \ldots r_n>$ in the expansion of $|\{c\}_N>$ is just the probability of finding r_i molecules out of N molecules in the i'th energy state when the probability of finding one molecule in the i'th energy state is $|c_i|^2$. In the language of first quantization, $|\{c\}_N>$ described the situation in which every molecule is in the same coherent superposition of energy states. We may regard this fact as an alternate justification for the terminology "coherent state". The property of maximum symmetry or the statement that

all molecules behave similarly, is consistent with the relationship

$$\langle\{c\}_N|d_{ij}^{(m)}|\{c\}_N\rangle = N\langle\{c\}_1|d_{in}^{(m)}|\{c\}_1\rangle \ . \tag{3-16}$$

Equation (3-9) gives the expectation value of a product of one annihilation operator and one creation operator, written in normal order. One can easily obtain, by a similar method, the expectation value of the product of an arbitrary number of annihilation and creation operators written in normal order. From Eq. (3-7) we have, by repeated application of the operators A_i,

$$A_1^{w_1} \ldots A_n^{w_n}|\{c\}_N\rangle = [N(N-1)\ldots(N-R+1)]^{1/2} c_1^{w_1} \ldots c_n^{w_n}|\{c\}_{N-R}\rangle \ , \tag{3-17}$$

where the w_i's are positive integers, and $R = \Sigma_i w_i$; likewise,

$$\begin{aligned} &\langle\{c\}_N A_1^{\dagger v_1} \ldots A_n^{\dagger v_n} \\ &= \langle\{c\}_{N-R'}|[N(n-1) \ldots (N-R'+1)]^{1/2} c_1^{*v_1} \ldots c_n^{*v_n} \ , \end{aligned} \tag{3-18}$$

where the v_i's are positive integers and $R' = \Sigma v_i$. In view of the fact that subspaces corresponding to different numbers of molecules are orthogonal, we have, further,

$$\begin{aligned} &\langle\{c\}_N|\pi_i A_i^{\dagger v_i} \pi_j A_j^{w_j}|\{c\}_N\rangle \\ &= N(N-1) \ldots (N-R+1)\pi_i c_i^{*v_i} c_i^{w_i} \end{aligned} \tag{3-19}$$

for $\Sigma v_i = \Sigma w_i = R$, and

$$\langle\{c\}_N|\pi_i A_i^{\dagger v_i}\pi_j A_j^{w_j}|\{c\}_N\rangle = 0 \qquad (3\text{-}20)$$

for $\Sigma v_i \neq \Sigma w_i$. This result, together with the commutation rules, allows the calculation of the expectation value of any product of annihilation and creation operators when the system is in a coherent state.

It should be pointed out that the essential difference between energy states and coherent states disappears for the case $N = 1$, in which all energy states are merely special cases of coherent states; that is, they correspond to a special choice of the c_i's for which $c_i = \delta_{im}$ with $m = 1,\ldots,n$. Although such a choice of the c_i's will yield energy states for an N , it will not yield <u>all</u> energy states for $N > 1$. Additional mathematical properties of the coherent states may be found in the work of Gilmore et al.[4]

IV. SPECIALIZATION TO TWO LEVELS AND RELATION TO OTHER THEORIES OF COOPERATIVE BEHAVIOR

Much work is found in the literature on the cooperative behavior of two-level systems, of which only a part is referenced presently.[2,3,5-14] Not all of this work can be generalized easily to the case of more than two levels, as will become apparent shortly. It is of interest to examine the specialization of the present formalism to two levels and its relation to other theories. Cooperative (or macroscopic) phenomena for a collection of two level systems were first studied by Bloch[5] and by Dicke[6] using descriptions that appeared at the time to be unrelated. The relationship and physical significance of the difference between the two types of descriptions was subsequently pointed out,[2] and has been analyzed more recently in con-

siderable detail by Arrechi et al.[3] When specialized to two levels, the present coherent states are the BSQ version of fully symmetrized quantum states considered by Bloch, and the energy states are the BSQ version of fully symmetrized quantum states considered by Dicke.[2,3] The expressions for $\langle d_{12}^{(m)} \rangle$ of Eqs. (3-11) are, essentially, the dipole moments of Bloch's theory; the energy states $|r_1 r_2\rangle$ correspond to those labeled by Dicke with the "cooperation number" r and quantum number m, the correspondence being $r = \frac{1}{2}(r_1 + r_2)$ and $m = \frac{1}{2}(r_2 - r_1)$. (Only states with the maximum "cooperation number" $\frac{1}{2}N$ appear in the BSQ formalism, since these are the only fully symmetric states.)

As is well known, a two level system is formally equivalent to a spin $\frac{1}{2}$ system, and a collection of two level systems may be described in terms of angular momentum variables. In fact, the macroscopic equations developed by Bloch[5] (the "Bloch Equations") were motivated by consideration of classical angular momentum, and the collective spontaneous emmision studied by Dicke[6] ("superradiant" emission) is derived from the quantum mechanical formalism for the addition of angular momenta, with r being the total angular momentum quantum number and m being the z-component eigenvalue. As has been noted in the literature,[15] the BSQ formalism for $n = 2$ (that is, the formalism referring to two harmonic oscillators) becomes similar to that of angular momentum if we take $d_{ij}^{(m)}$, $m = 1,2,3$ to be the three components of angular momentum, respectively. Theories utilizing angular momentum for the study of cooperative behavior of two level systems cannot, however, be simply generalized to treat systems with more levels. The reason is based on a group-theoretical argument; the angular momentum operators are those of the algebra of the group SU(2), while operators of the algebra of SU(n) are needed for systems with n levels.[16]

A theory closer to the present formalism is that of Bonifacio, Kim and Scully.[9] Although they begin with a consideration of angular momentum, they relate it to the operators associated with two harmonic oscillators by means of the relationships of Eqs. (2-5) for $d_{12}^{(m)}$ and the correspondence noted above, arriving at a BSQ theory for two level systems. They define atomic coherent states in a basically different manner, however, from that in the present paper. Their method consists of putting each of the two harmonic oscillators in a harmonic-oscillator coherent state (HOC state), the state that describes an oscillating (harmonic oscillator) wave packet of well-defined phase. Since a_1 and a_2 then have independent well defined phases, it is clear that $\langle d_{12}^{(1)} \rangle$ and $\langle d_{12}^{(2)} \rangle$ will also have well defined phases. More precisely, if the two harmonic oscillators are in the HOC staes $|\alpha_1\rangle$ and $|\alpha_2\rangle$ respectively, such that

$$A_i|\alpha_i\rangle = \alpha_i|\alpha_i\rangle \quad , i = 1, 2 \quad , \tag{4-1}$$

we can write[9]

$$|\alpha_1\alpha_2\rangle = \exp[-\tfrac{1}{2}(|\alpha_1|^2+|\alpha_2|^2)] \sum_{r_1=0}^{\infty}\sum_{r_1=0}^{\infty} \frac{\alpha_1^{r_1}\alpha_2^{r_2}}{(r_1!r_2!)^{1/2}}|r_1r_2\rangle. \tag{4-2}$$

One obtains immediately,

$$\langle d_{12}^{(1)} \rangle = |\alpha_2\alpha_1|\cos[(\omega_2 - \omega_1)t + \theta_{21}] \quad , \tag{4-3a}$$

$$\langle d_{12}^{(2)} \rangle = |\alpha_2\alpha_1|\sin[(\omega_2 - \omega_1)t + \theta_{21}] \quad , \tag{4-3b}$$

$$\langle d_{12}^{(3)} \rangle = \tfrac{1}{2}(|\alpha_2|^2 - |\alpha_1|^2) \quad , \tag{4-3c}$$

where θ_{21} is defined by

$$\alpha_2^* \alpha_1 \equiv |\alpha_2 \alpha_1| e^{i\theta_{21}} , \tag{4-4}$$

Comparison with Eqs. (3-11) shows that the correspondence

$$\alpha_i \rightarrow N^{1/2} c_i \quad , \quad i = 1, 2 \tag{4-5}$$

gives the same dipole moment expectation value for the state $|\alpha_1 \alpha_2\rangle$ as for the state $|\{c\}_N\rangle$.

The definition of Bonifacio et al. in terms of HOC states can be generalized immediately to the case of an arbitrary number of levels, of course, the state now being labeled $|\{\alpha\}\rangle$, where $\{\alpha\}$ stands for the collection of complex numbers $\alpha_1 \ldots \alpha_n$. A generalization of the expansion in terms of energy states analogous to Eq. (4-2), is obvious. However, there exists a basic difference between the two types of coherent atomic states, that is, between the states $|\{c\}_N\rangle$ and $\{\alpha\}\rangle$. The expansion of $|\{\alpha\}\rangle$ in terms of energy states contains states of arbitrarily high energy. One cannot, therefore, define $|\{\alpha\}\rangle$ in terms of basis vectors that refer to a system with a fixed number (or even a finite number) or molecules. [This fact is recognized in Ref. (9).] Furthermore, the state $|\{\alpha\}\rangle$ contains more information than is necessary to obtain expectation values of dynamical variables, which (as mentioned previously] must consist of terms that can be reduced to the form $\pi_i a_i^{\dagger v_i} \pi_j a_j^{w_j}$ with $\Sigma v_i = \Sigma w_j$. It is easy to see that an infinite number of <u>linearly independent</u> states $|\{\alpha'\}\rangle$, where $\alpha_i' = \alpha_i e^{i\theta}$, with θ arbitrary (but independent of i), yield the same expectation value of dynamical variables as the state $|\{\alpha\}\rangle$.

[Note that in the case of the state vector $|\{c\}_N\rangle$, multiplication of the c_i's by a common phase factor does not yield a different state vector.] It should also be noted that expectation values of products other than $a_i^\dagger a_j$ are not the same for the states $|\{\alpha\}\rangle$ and $|\{c\}_N\rangle$ under the correspondence (4-5), those for $|\{\alpha\}\rangle$ being given by

$$\langle\{\alpha\}|\pi_i A_i^{\dagger\nu_i} \pi_j A_j^{w_j}|\{\alpha\}\rangle = \pi_i \alpha_i^{*\nu_i} \alpha_i^{w_i} \quad , \tag{4-6}$$

while those for $|\{c\}_N\rangle$ being given by Eqs. (3-19) and (3-20). Only for $\Sigma w_i = \Sigma v_i$ and $N \to \infty$ do these expectation values become equal. The algebraic aspects of the relationship between the states $|\{\alpha\}\rangle$ and $|\{c_N\}\rangle$ are discussed in detail by Gilmore, Bowden and Narducci,[4] who show that the states $|\{\alpha\}\rangle$ can be used as generating functions for the states $|\{c_N\}\rangle$.

V. CLASSICAL LIMIT

The BSQ formalism is particularly suitable for passage to the classical limit. As in the case of boson fields, the classical limit is approached by letting the number of bosons become large. The mathematical - or formal - reason for this procedure is the fact that the difference between the numbers associated with $a_i^\dagger a_i$ and $a_i a_i^\dagger$ becomes relatively negligible. Thus, if the commutator of a_i and $a_i^\dagger$ can be ignored, a_i and $a_i^\dagger$ may be regarded as c-numbers, and the quantum mechanical formalism reduces to a classical formalism. In other words, as the occupation number[17] associated with a given harmonic oscillator becomes large, the oscillator becomes classical.

(Note that the last statement is essentially an expression of the Correspondence Principle). Since, according to Eq. (3-2), $\Sigma n_i = N$, a necessary condition for appraoch to the classical limit is the condition $N \gg 1$. If the occupation numbers of interest in the interaction under consideration are all significant fractions of N, then $N \gg 1$ is also a sufficient condition.

The numbers associated with the occupation-number operator n_i are determined by the state, of cours. For an energy state $|r_1 \ldots r_n\rangle$, which is also an eigenstate of n_i, the number associated with n_i is clearly its eigenvalue r_i, and if all r_i's are large, the behavior of the system is approximately classical. In general, however, the state under consideration need not be an energy state, and then the numbers associated with the occupation-number operators are not uniquely indicated. For instance, the coherent state of Eq. (3-3) consists of a superposition of energy states, which, for a given level (or oscillator) have occupation-number eignvalues varying from zero to N, respectively. The relative importance of the various energy states in the superposition is determined by the absolute values of the superposition constants, which, in turn, are determined by the set of numbers $c_1 \ldots c_n$. A reasonable measure of the significant occupation numbers - a weighted average - is provided by the expectation values of the occupation-number operator. In the case in which the state is not an eigenstate of n_i, we will, therfore, use $\langle n_i \rangle$ as a measure of the numbers associated with n_i. The argument concerning the classical limit can now be stated as follows: If the difference between $\langle a_i^\dagger a_i \rangle$ and $\langle a_i a_i^\dagger \rangle$ is relatively negligible, then (as a reasonable approximation) a_i may be treated as a c-number, and the i-th harmonic oscillator may be regarded as classical; furthermore, the entire molecular system may be regarded as classical, if all the pertinent harmonic oscillators are classical.

It is seen that many types of quantum mechanical states may be regarded as approximately classical, since the only requirement is that which calls for all the $\langle n_i \rangle$'s to be sufficiently large. The question arises: What method of classical description should be used for a state which meets this requirement? One might, perhaps, wonder why this question should arise at all, since, classically, one generally specifies all the pertinent coordinates. However, a precise specification of all the coordinates is not the only type of classical description possible. In quantum mechanics, a precise description is impossible and a description in terms of a quantum mechanical state must be interpreted statistically. In classical theory, a precise description is possible, but so is a statistical description, the latter being as fully classical as the former, with the precise description, in fact, being a special case of the statistical description. The problem consists, therefore, of finding the classical statistical description which is the classical limit of the quantum state under consideration.

The conversion of a quantum mechanical description to a classical description, thus, consists of converting the operators a_i and $a_i^\dagger$ to classical random variables, which we designate by $\tilde{a}_i$ and $\tilde{a}_i^*$, respectively. The dynamical meaning (or definition) of these variables is the same in both descriptions.[18] Let the corresponding (classical) reduced variables be $\tilde{A}_i$ and $\tilde{A}_i^*$. These variables will be specified as follows: Consider an arbitrary product of (reduced) annihilation and creation operators $O\{\pi_i A_i^{\dagger\nu_i} A_i^{w_i}\}$, where O indicates a certain ordering of these operators. For a given state, the expectation values of this product will depend, of course, on O, but, as mentioned previously, the difference for different ordering arrangements of the operators becomes negligible in the classical limit. Implied in this statement is the assumption

$$\sum_i v_i + \sum_i w_i \ll N. \tag{5-1}$$

(otherwise, terms arising from commutators may not be relatively negligible.) Now, classical random variables $\tilde{A}_i$, $\tilde{A}_j^*$ can be described by their moments $\langle \pi_i \tilde{A}^{*v_i} \tilde{A}_i^{v_i} \rangle_{av}$, which are averages over an ensemble inherent in the statistical description. We prescribe these moments (in the classical limit) by the relationship

$$\langle \pi_i \tilde{A}_i^{*v_i} \tilde{A}_i^{w_i} \rangle_{av} = \langle 0 \{ \pi_i \tilde{A}_i^{\dagger v_i} \tilde{A}_i^{w_i} \} \rangle , \tag{5-2}$$

where $\tilde{0}$ is a particular ordering arrangement of the operators that may be chosen arbitratily, or, better yet, chosen for simplicity of calculation of the expectation value (since, within the range of approximation, all ordering arrangements yield the same result). For an energy state, the operators may be ordered so that the product becomes a product of occupation-number operators. (If the product cannot be so ordered, the expectation value vanishes.) Thus, the energy state $|r_1 \ldots r_n\rangle$ in the classical limit yields the statistical description in terms of moments (in hybrid but obvious notation)

$$\langle r_1 \ldots r_n | \pi_i \tilde{A}_i^{*v_i} \tilde{A}_i^{w_i} | r_1 \ldots r_n \rangle_{cl} = \pi_i r_i^{w_i} \delta_{w_i v_i} \tag{5-3}$$

For a coherent state, normal ordering allows a simple computation of the expectation values, by means of Eqs. (3-19) and (3-20), and we obtain as the classical limit, the moments

$$\langle\{c\}_N|\pi_i \tilde{A}_i^{*\nu_i} \tilde{A}_i^{w_i}|\{c\}_N\rangle_{c\ell} = N^R \pi_i c_i^{*\nu_i} c_i^{w_i} \delta_{\Sigma\nu_i,\ \Sigma w_i} \tag{5-4}$$

where the inequality (5-1) has been utilized in approximating the right side.

Instead of specifying random variables by a set of mements, one can specify them by a probability distribution. [Note that the present procedure does not allow specification of an <u>infinite</u> set of moments for finite N, due to the inequality (5-1); this is consistent with the fact that the calssical-limit approximation improves as N increases.] It is evident that the set of moments $\langle r_1 \ldots r_n|\pi_i A_i^{*\nu_i} A_i^{w_i}|r_1 \ldots r_n\rangle_{c\ell}$ are consistent with an <u>independent</u> statistical description of each oscillator by the expression

$$\tilde{A}_j = r_j^{1/2} e^{-i\theta_j} \tag{5-5}$$

where θ_j has a uniform probability distribution, that is, the probability of finding θ_j in a unit interval between 0 and 2π is given by

$$P(\theta_j) = (2\pi)^{-1} \tag{5-6}$$

and the corresponding joint probability is given by

$$P(\theta_1 \ldots \theta_n) = P(\theta_1) \ldots P(\theta_n) = (2\pi)^{-n}, \tag{5-7}$$

Likewise, the set of moments $\langle\{c\}_N|\pi_i A_i^{*\nu_i} A_i^{w_i}|\{c\}_N\rangle_{c\ell}$ are consistent with a dependent statistical description of each oscillator, in which

$$\tilde{A}_i = N^{1/2} c_i e^{i\theta} \tag{5-8}$$

where θ has a uniform probability distribution

$$P(\theta) = (2\pi)^{-1} \tag{5-9}$$

It should be noted that for the coherent state, in contrast with the energy state, we have the same random phase for all the oscillators. In the dipole moment expectation value, where only phase differences are significant, the random phase of each oscillator plays no role for the coherent state, but produces a value of zero for the energy state. Finally, we look, in the classical limit, at the state $|\{\alpha\}\rangle$. (Here the question of finite N, or limited number of moments, does not enter.) From Eq. (4-6), we obtain

$$\langle\{\alpha\}|\pi_i \tilde{A}_i^{*v_i} \tilde{A}_i^{w_i}|\{\alpha\}\rangle_{c\ell} = \pi_i \alpha_i^{*v_i} \alpha_i^{w_i} , \tag{5-10}$$

It is clear that this set of moments is consistent with a completely deterministic description of each harmonic oscillator, given by

$$\tilde{A}_i = \alpha_i \tag{5-11}$$

It should be emphasized that energy states and coherent states are equally "calssical", in the sense of having a classical limit. This point of view is different from that sometimes expressed in the literature[3,9] to the effect that coherent states are "calssical" and energy states are

"qauntum mechanical". If the (physically meaningless) arbitrary phase factor in Eq. (5-8) is ignored, one can say only that, in the classical limit, coherent states offer a deterministic (classical) description and energy states offer a statistical (classical) description. Neither state, however, can be said to be closer to its classical limit. As pointed out previously, the mere presence of statistics is no indication of quantum mechanics.

The above-mentioned association of coherent states with classical theory and energy states with quantum theory is based by some authors[3] on the fact that coherent states can be produced dynamically by a classical (c-number), deterministically described, perturbation acting on the quantum mechanical ground state of the N-molecule system. It appears most reasonable, nevertheless, to judge the quantum mechanical aspects of a state - as contrasted with the classical aspects - by the significance of the expectation value of the previously-discussed commutators (which are related to the uncertainty principel, or to the disturbance of the system produced by the measurement process). It is obvious that this significance is greater for coherent states of small $\langle n_i \rangle$ than for energy states of large $\langle n_i \rangle$, in contradiction to the above-mentioned association.

VI. "SEMICLASSICAL" THEORY

The classical limit of the BSQ formalism is closely related to semiclassical radiation theory (SCT). In view of the wide use of SCT and the important role it has played in many calculations, it is instructive to examine this relationship. The usual form of SCT[19] consists of replacing

the dipole moment in volume V, in classical equations of motion for the radiation field, by ΔN times the quantum mechanical expectation value of a molecular dipole moment, ΔN being the number of molecules in ΔV. For a single molecule described in the Schrodinger picture (and first-quantization formalism) by the state $|\phi\rangle = \Sigma c_i(t)|\phi_i\rangle$, the expectation value of the dipole moment is given by

$$\langle d_{ij}^{(1)} \rangle = \frac{1}{2} [c_j^*(t)c_i(t) + c_i^*(t)c_j(t)], \tag{6-1a}$$

$$\langle d_{ij}^{(2)} \rangle = -\frac{i}{2}[c_j^*(t)c_i(t) - c_i^*(t)c_j(t)], \tag{6-1b}$$

$$\langle d_{ij}^{(3)} \rangle = \frac{1}{2} [c_j^*(t)c_j(t) - c_i^*(t)c_i(t)]. \tag{6-1c}$$

The replacement of the classical dipole moment by a quantum mechanical expectation value is an ad hoc prescription which connects a quantum mechanical description of atomic systems with a classical description of the electromagnetic field.

Another form of SCT, developed by Jaynes under the name of "neo-classical" theory, achieves the same result without the ad hoc procedure by starting from a classical Hamiltonian that utilizes a classical model of an n-level molecule.[20] This model is described by precisely the classical version of the present BSQ formalism, the molecular behavior being specified by a_i and $a_i^\dagger$, treated classically, and normalized to $\Sigma a_i^\dagger a_i = 1$. The (classical) Hamiltonian for a single molecule is that of Eqs. (2-4) - (2-6), from which equations of motion may be derived by means of Poisson brackets in the usual manner. It is seen that the dipole moment, given

by Eqs. (2-5), has the same form in terms of the a_i's as the quantum mechanical expectation value of the dipole moment in terms of the c_i's. Furthermore, one can show that the (classical) equations of motion of the a_i's are formally identical with the (Schrodinger picture) equations of motion for the c_i's. If the initial values of the a_i's equal, respectively, to those of the c_i's (except for a multiplicative phase constant that is independent of i), then the classical version of the present BSQ formalism for a single molecule constitutes SCT.

Reference to a single molecule is significant in the examination of SCT. One might pose the following question: Consider a BSQ description of N molecules; do we obtain SCT by converting the a_i's to classical variables (with the appropriate normalization, of course)? The answer depends - perhaps surprisingly, at first glance - on the type of state used to describe the system. For a coherent-state description, the answer is positive, and for an energy-state description, the answer is negative. The reason is due to the fact that, in the classical limit, coherent states allow a deterministic description of the dipole moment of the system, while the energy states do not. The latter, in the classical limit, yield information only in a statistical form about an ensemble of N-molecule systems; SCT, however, requires that the expectation value with respect to the state under consideration specify the dipole moment of a single N-molecule system. On the other hand, as far as coherent states are concerned, the same dipole-moment expectation value is obtained for an N-molecule system in the state $|\{c\}_N\rangle$ as for as N molecules (with each) in the one-molecule state $|\{c\}_1\rangle$; this relationship, for arbitrary $\{c\}$

is the one required by SCT. Since SCT consists of the replacement in the BSQ formalism for a single molecule of the q-number variables by c-number variables, it must be regarded, from a quantum mechanical viewpoint, as the fully classical limit of a quantum mechanical formalism, valid only when such a limiting procedure is justified. Furthermore, it is a restricted form of describing the classical limit, applicable to coherent states but not to energy states, and, thus, a special case of the more general form developed in Sec. V.

VII. APPLICATION OF THE FORMALISM TO A THREE-LEVEL LASER

We utilize the special features of the present formalism by applying it to a model of a three-level laser. Let the laser matter consist of N identical three-level molecules, with energy levels $h\omega_i$, $i = 1,2,3$, ordered so that $\omega_i < \omega_j$ for $i < j$, with $\omega_{ij} \equiv |\omega_i - \omega_j|$. The matter is therefore described by three oscillators of complex amplitude a_1, a_2, a_3. We consider single mode operation of the cavity, the mode frequency being ω_{12}. The loss constant associated with this mode is ξ, which is defined in terms of the quality factor Q by $2\xi = \omega_{23}/Q$. The field of a damped cavity mode has been described in detail previously[21], and only the pertinent results will be used here.

The complex amplitudes (or annihilation and creation operators) of the radiation oscillator associated with the cavity mode are designated by b_{23} and b_{23}^+, respectively, and are expressed in terms of reduced amplitudes B_{23} and B_{23}^+ by

$$b_{23}(t) = B_{23}(t)\, e^{-i\omega_{23}t}, \quad b^\dagger(t) = B_{23}^\dagger(t)\, e^{i\omega_{23}t}, \tag{7-1}$$

With each atomic transition we associate a relaxation "field" (it may be electromagnetic or acoustic, or it may represent some other loss mechanism) described by the (non-hermitian) dynamical variables $\mathcal{Q}_{ij}(t)$, where the pair of indices corresponds to the transition to which the relaxation field is coupled, the order of the indices having no significance. These relaxation fields have also been described in detail previously[12b], and as in the case of the cavity field, only the required results will be

used. The relaxation constant associated with the (i,j) transition is specified by α_{ij}. The pumping will be described, formally, as that of a prescribed field of arbitrary coherence properties.

The above brief description of the system is made precise by the following specification of the terms whose sum constitutes the interaction Hamiltonian,

$$H'_{12}\ (\text{relaxation}) = -\frac{i}{\sqrt{2}}\hbar\,(A_1A_2^\dagger \mathcal{Q}_{12} - \mathcal{Q}_{12}^\dagger A_2A_1^\dagger) \tag{7-2}$$

$$H'_{13}\ (\text{relaxation}) = -\frac{i}{\sqrt{2}}\hbar\,(A_1A_3^\dagger \mathcal{Q}_{13} - \mathcal{Q}_{13}^\dagger A_1^\dagger A_3) \tag{7-3}$$

$$H'_{23}\ (\text{relaxation}) = -\frac{i}{\sqrt{2}}\hbar\,(A_2A_3^\dagger \mathcal{Q}_{23} - \mathcal{Q}_{23}^\dagger A_2^\dagger A_3) \tag{7-4}$$

$$H'_{23}\ (\text{cavity}) = \hbar\,\gamma_{23}(A_2A_3^\dagger B_{23} + B_{23}^\dagger A_2^\dagger A_3) \tag{7-5}$$

$$H'_{13}\ (\text{pump}) = i\hbar\,(\mathcal{P}A_1A_3^\dagger - \mathcal{P}^\dagger A_3A_1^\dagger) \tag{7-6}$$

and by the expressions for $\mathcal{Q}_{ij}$ and B_{23},

$$\mathcal{Q}_{ij} \simeq \mathcal{Q}_0^{(ij)} + \frac{1}{\sqrt{2}}\,\alpha_{ij}\,A_i^\dagger A_j\,, \quad i < j \tag{7-7}$$

$$B_{23} \simeq B_0^{(23)} + i\,\frac{\gamma_{23}}{\xi}\int_0^t dt' A_2^\dagger(t')A_3(t')\,e^{-\xi(t-t')}\,, \tag{7-8}$$

where $\mathcal{Q}_0^{(ij)}$ and $B_0^{(23)}$ describe $\mathcal{Q}_{ij}$ and B_{23} in the absence of the atoms. (In other words, they account for vacuum and thermal effects.) The coupling between atoms and cavity is assumed to begin at $t = 0$. If we ignore thermal effects by assuming the cavity and relaxation mechanisms to be at zero temperature, then quantum mechanically,

$$B_0^{(23)}|> \;=\; <|B_0^{(23)} = 0 \tag{7-9}$$

$$\mathcal{Q}_0^{(jk)}|> \;=\; <|\mathcal{Q}_0^{(jk)\dagger} = 0 \tag{7-10}$$

$$<B_0^{(23)}(t_1)B_0^{(23)\dagger}(t_2) \;=\; e^{-\xi|t_1-t_2|} \tag{7-11}$$

$$<\mathcal{Q}_0^{(jk)}(t_1)\mathcal{Q}_0^{(jk)\dagger}(t_2) \;=\; 2\,\alpha_{jk}\,\delta(t_1-t_2)\;, \tag{7-12}$$

while classically, $B_0^{(23)}$ and $\mathcal{Q}_0^{(ij)}$ vanish. $\mathcal{B}(t)$ is the prescribed pumping "field" and, may be, most generally, a stochastic function of time, that is, the description of a random process. For nonzero temperatures, where thermal effects need to be considered, the specification of $B_0^{(23)}$ and $\mathcal{Q}_0^{(ij)}$ can be suitably modified both quantum mechanically and classically[12b,21].

Equations of motion are obtained from the interaction Hamiltonian of Eqs. (7-2) - (7-6), as follows:

$$\dot{A}_1 \;=\; \frac{1}{\sqrt{2}}\mathcal{Q}^{\dagger}_{12}A_2 + \frac{1}{\sqrt{2}}\mathcal{Q}^{\dagger}_{13}A_3 - \mathcal{B}^{\dagger}A_3, \tag{7-13}$$

$$\dot{A}_2 \;=\; -\frac{1}{\sqrt{2}}A_1\mathcal{Q}_{12} + \frac{1}{\sqrt{2}}\mathcal{Q}^{\dagger}_{23}A_3 - i\,\gamma_{23}B^{\dagger}_{23}A_3 \tag{7-14}$$

$$\dot{A}_3 \;=\; -\frac{1}{\sqrt{2}}A_1\;_{13} - \frac{1}{\sqrt{2}}A_2\;_{23} - i\,\gamma_{23}A_2B_{23} + \mathcal{B}A_1\;. \tag{7-15}$$

These equations, upon substitution from Eqs. (7-7) and (7-8) become

$$\dot{A}_1 = \frac{1}{\sqrt{2}}\mathcal{Q}_0^{(12)\dagger}A_2 + \frac{1}{2}\alpha_{12}A_1 n_2 + \frac{1}{\sqrt{2}}\mathcal{Q}_0^{(13)}A_3 + \frac{1}{2}A_1 n_3 + \mathcal{B}^* A_3 \tag{7-16}$$

$$\dot{A}_2 = -\frac{1}{\sqrt{2}}A_1\mathcal{Q}_0^{(12)} - \frac{1}{2}\alpha_{12}(n_1+\lambda)A_2 + \frac{1}{\sqrt{2}}\mathcal{Q}_0^{(23)}A_3 + \frac{1}{2}A_2 n_3$$

$$- i\gamma_{23}B_0^{(23)\dagger}A_3 - \frac{\gamma_{23}^2}{\xi^2}\int_0^t dt' A_2(t')A_3^\dagger(t')A_3(t)e^{-\xi(t-t')} \tag{7-17}$$

$$\dot{A}_3 = -\frac{1}{2}A_1\mathcal{Q}_0^{(13)} - \frac{1}{2}\alpha_{13}(n_1+\lambda)A_3 - \frac{1}{2}A_2\mathcal{Q}_0^{(23)} - \frac{1}{2}(n_2+\lambda)n_3$$

$$- i\gamma_{23}A_2 B_0^{(23)} + \frac{\gamma_{23}^2}{\xi^2}\int_0^t dt' A_2(t)A_2^\dagger(t')A_3(t')e^{-\xi(t-t')}$$

$$+\mathcal{B}A_1 \tag{7-18}$$

where $n_i = A_i^\dagger A_i$, and λ stands for unity or zero for quantum mechanical or classical interpretation of the equations, respectively. To these equations of motion we must add the normalization condition

$$n_1 + n_2 = n_3 = N \tag{7-19}$$

Equations (7-16) - (7-19) are the complete laser equations for the atomic variables. Once the latter are known, the cavity field is obtained immediately from Eq. (7-8). As indicated above, these equations can be interpreted both classically and quantum mechanically. Although they appear rather complicated, they can be considerably simplified for special conditions. Classically, they are simplified not only by the fact that λ vanishes and all variables commute, but also by the fact that all terms

with subscript zero vanish. Quantum mechanically, also, the latter terms vanish if we take the expectation value with respect to the vacuum field. (Whether this expectation value is taken at this point or at a later point in the calculation depends on the question to be answered). In the steady-state, the integrals occurring in the equation can be carried out explicitly, the derivatives vanish, and the integro-differential equations become algebraic equations. It should be noted that the interaction Hamiltonian and the equations of motion can be easily generalized to apply to the case of more than three levels.

The above equations of motion offer a rich supply of information relative to a number of questions of physical interest. One question, for instance, concerns the role of quantum mechanics, (as contrasted with classical mechanics) in laser operation. Another question, which is of considerable practical significance, concerns the influence of the pumping process on the stability (or noise properties) of the laser output. The answer to the latter question may be sought in the present theory because the dynamics of the pumping levels are explicitly considered.

The study of solutions of these equations will be considered in a subsequent publication.

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