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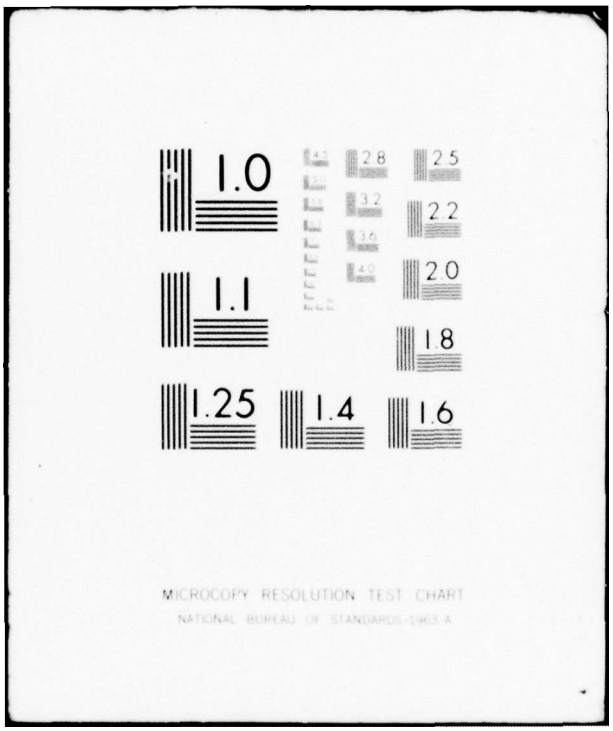
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MACROSCOPIC DESCRIPTION OF LASER-TYPE MATTER

Final Report

(Third Year)

by

I.R. Senitzky

September 1977

EUROPEAN RESEARCH OFFICE

United States Army

London, England

GRANT NUMBER DA-ERO-75-G-012

Technion-Israel Institute of Technology

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CONTENTS

I. INTRODUCTION

In several previous publications,[1,2] a formalism was developed for describing macroscopic, laser-type, phenomena produced by the cooperative behavior of a large number of n-level systems, such as molecules or atoms whose pertinent interaction with the field involves only n energy levels. This theory will be referred to in the present discussion as macroscopic quantum mechanics (MQM).

Since description of the above phenomena has also been the purpose of semiclassical radiation theory (SCT), it is of interest to examine the latter - or rather, the several forms of it - in detail, in order to bring out the essential difference between the theories. This examination is carried out in Sec. II, where the validity and applicability of three forms of SCT as well as that of MQM are examined from an orthodox quantum mechanical viewpoint.

In its development so far, MQM describes conditions under which the cooperation between the n-level systems is a maximum. Clearly, the usefulness of the theory will be enhanced if it can be generalized to describe conditions under which less than maximum cooperation exists. Such a generalization is discussed in Sec.III, where in addition, a physically meaningful method of describing quantitatively the cooperation between the n-level systems is given.

Cooperative behavior - to greater or lesser extent - may exist not only in the oscillatory process of a laser, but also in the pumping and relaxation processes. It is of interest therefore, to study the effects of cooperation in the last two processes, since they can be enhanced experimentally, if proven desirable. In Sec. IV, a four-level laser is analysed by MQM for the case of maximum

atomic cooperation.

II. SEMICLASSICAL RADIATION THEORIES AND MACROSCOPIC QUANTUM MECHANICS

We examine the validity of three forms of SCT, as well as that of MQM, from a quantum mechanical viewpoint. It will be shown that MQM may be regarded as an improved - or more general - version of some of the forms of SCT. For simplicity of discussion, the four theories will be labeled SCT I-IV, where SCT IV is MQM. They will first be defined briefly, and then examined individually.

SCTI, the oldest theory, is, in the words of Schiff,[3] a theory in which "we treat the electromagnetic field classically and the particles with which the field interacts by quantum mechanics".

SCTII, which has been used widely in the analysis of almost all coherent phenomena when the mutual interaction of atoms and field is involved,[4] couples Schrodinger's equation and Maxwell's equations; the field is described classically in both sets of equations, the atoms are described quantum mechanically by Schrodinger's equation, while the atomic - or matter - variables in Maxwell's equation (for most applications, only the polarization, or atomic dipole-moment, is of interest) are replaced by their expectation values.

SCTIII, which is the latest version of Jaynes' "neoclassical theory",[5] introduces a classical model for the atom based on its natural frequencies (or energy spectrum) and the associated oscillating dipole moments, with the latter providing the atomic polarization in the classical Maxwell's equations. An essentially similar version has been proposed by Eberly.[6]

SCTIV is the classical-limit form of a fully quantum mechanical

theory in which the atoms are described by a boson-second-quantization formalism.[7]

A. Semiclassical Theory I

It has been shown some time ago that the mutual interaction of two systems, one strictly quantum mechanical and the other strictly classical, cannot be described by a self consistent dynamical formalism.[8] In the present instance, one reason is the fact that quantum mechanical atoms generate a quantum mechanical field, so that the field, even if it is classical initially, cannot remain classical. Another reason is the fact that, to the classical system, the zero-point motion of the quantum mechanical system looks like motion that can do work, which obviosuly leads to absurd results.[8] In order to place SCTI within a quantum mechanical framework, we consider a field (in the Heisenberg Picture) which is the sum of two parts, one purely quantum mechanical and the other classical. The total field is, of course, fully quantum mechanical. The purely quantum mechanical part is due to the atoms under consideration and to the loss mechanism with which the field that is interacting with the atoms is coupled. The classical part is due to external sources which are unaffected by the atomic behavior or by the loss mechanism. (It has been shown that such an external field, no matter what its sources are, may be described classically as far as the atoms are concerned.[9]) SCTI is a valid approximation in the case where the quantum mechanical part of the field is negligible compared to the classical part, and also in the case where the questions asked refer only to atomic effects produced by the classical part. An example of the first case is the behavior of atoms in a strong laser field - assuming, of course,

that the laser operation is unaffected by the atoms under consideration. An example of the second case is (lowest order) induced emission or absorption;[10] here, only the external field determines the result - assuming that the loss mechanism does not produce a significant thermal field - since the quantum mechanical part of the field contributes, in lowest order, only to spontaneous emission. An important application - entirely valid - of SCTI is the theory of photoelectric detection.[10] In the usual approach to photoelectric phenomena, the reaction of the photoelectrons on field being detected is ignored. The induced absorption by the photosensitive atoms is investigated to lowest order in perturbation theory, in order to obtain information about the field produced by external sources. This field is the classical part of the total field, the only part needed to describe the production of photoelectrons.

B. Semiclassical Theory II

In contrast to SCTI, SCTII provides a prescription for analyzing the mutual interaction between the atoms and the field.[11] It allows the field to remain classical even though the atoms are treated quantum mechanically; however, this is accomplished by an arbitrary requirement, namely, that the matter variables in Maxwell's equations be replaced by their expectation values. The conditions for the validity of such a procedure from a quantum mechanical viewpoint will be discussed later. Presently, it is instructive to illustrate a case for which such a procedure is invalid. Let the matter under consideration consist of a single, highly excited harmonic oscillator with an electric dipole moment proportional to its displacement. One can, in principle, specify its quantum mechanical state to be an energy state

(corresponding to a high quantum number). Since, in this state, the expectation value of the dipole moment is zero, this highly excited oscillator will not radiate at all according to SCTII! The reason for such an incorrect result is not difficult to find. The description of the oscillator by means of an energy state is a statistical description. In the limit of high quantum numbers (the classical limit), the energy state becomes equivalent to a classical description in which the amplitude of oscillation is well defined but the probability distribution of the phase of oscillation is constant.[12] We see that SCTII ignores the purely statistical aspect of a quantum mechanical description, even if the description refers to a macroscopic system. (A harmonic oscillator in a high energy state is essentially such a system.) SCTII is, therefore, invalid when the statistical aspects of the description of the matter - in the sense of being nondeterministic - are significant.

C. Semiclassical Theory III

SCTIII achieves the same results as those of SCTII, for certain types of problems, without an arbitrary prescription for joining quantum mechanical and classical theories. In SCTIII the atom is described schematically by its natural frequencies and associated dipole moments, as follows:[5] Let the natural frequencies of the atom be those determined by the set of energy levels $\hbar\omega_n$, $n = 1,2,\ldots$ The (classical) Hamiltonian describing the free atom is then given by

$$H_o = \Sigma_n \hbar\omega_n a_n^* a_n \ , \qquad (1)$$

where a_n and a_n^* are independent (complex) dynamical variables.

Corresponding to each atomic frequency $|\omega_i - \omega_j|$, there exists a dipole moment, the components of which are linear superposition of the quantities

$$d_{ij}^{(1)} = \frac{1}{2}\,(a_j^* a_i + a_i^* a_j) ,$$

$$d_{ij}^{(2)} = -\frac{1}{2} i (a_j^* a_i - a_i^* a_j) , \qquad (2)$$

$$d_{ij}^{(3)} = \frac{1}{2}\,(a_j^* a_j - a_i^* a_i) .$$

The coupling between the atom and the field is assumed to be of the dipole-moment type described most generally by the interaction Hamiltonian

$$H' = \hbar \sum_{m=1}^{3} \sum_{i<j} d_{ij}^{(m)} F_{ij}^{(m)} , \qquad (3)$$

where $F_{ij}^{(m)}$ is a linear superposition of the electromagnetic field components that contain appropriate coupling constants. The total Hamiltonian for the atom and field is given by

$$H = H_o + H' + H_f , \qquad (4)$$

H_f being the classical Hamiltonian for the field only. The "canonical" equations for the atomic variables are[5]

$$i\hbar \dot{a}_n = \frac{\partial H}{\partial a_n^*} , \quad i\hbar \dot{a}_n^* = -\frac{\partial H}{\partial a_n} , \qquad (5)$$

and the equations of motion for the field variables are those of

the conventional Hamiltonian formalism for the classical electromagnetic field. It can be shown that the equations of motion for the a_n's, together with the definition of dipole moment by means of the $d_{ij}^{(m)}$'s, yield the same equations of motion for the dipole moment in SCTIII as those for the expectation value of the dipole moment in SCTII. The initial values of the a_n's in SCTIII are chosen so that the initial dipole moment obtained from Eqs. (2) is equal to the initial expectation value of the dipole moment in SCTII. Thus, SCTIII becomes a formal Hamiltonian theory that gives the same results as SCTII, as far as the field is concerned, without an ad hoc prescription to connect the quantum mechanical atomic equations with the classical field equations. SCTIII exhibits the same lack of validity as SCTII when applied to states that are not sufficiently deterministic.

D. Semiclassical Theory IV. Macroscopic Quantum Mechanics.

In order to derive a semiclassical theory from quantum mechanics, we begin with a boson-second-quantization formalism for the description of a number of identical atoms that couple similarly through their dipole moment to the electromagnetic field. Let the relevant spectrum of each atom consist of n levels with energies $\hbar\omega_i$, i = 1, 2, ...n. The atomic state vectors that describe the entire collection lie in a space spanned by the vectors $|r_1 \ldots r_i \ldots r_n\rangle$ where the r_i's are non negative integers. The fundamental operators, from which the pertinent dynamical variables may be constructed, are - using the Heisenberg Picture - $a_1(t)$, $\ldots a_n(t)$ and $a_1^\dagger(t)$, $\ldots a_n^\dagger(t)$ such that

$$a_i(0)|r_1 \ldots r_i \ldots r_n\rangle = r_i^{\frac{1}{2}}|r_1 \ldots r_i - 1 \ldots r_n\rangle \qquad (6)$$

and

$$a_i^\dagger(0)|r_1 \ldots r_i \ldots r_n\rangle = (r_i+1)^{\frac{1}{2}}|r_1 \ldots r_i+1 \ldots r_n\rangle \quad . \qquad (7)$$

The commutation relationships are $[a_i(t), a_i^\dagger(t)] = 1$, with all other equal-time commutators vanishing. The Hamiltonian for the collection of atoms is given by the expression

$$H_o = \sum_{i=1}^{n} \hbar\omega_i a_i^\dagger a_i \quad . \qquad (8)$$

The collective atomic dipole moment is a linear superposition of the operators

$$\begin{aligned} d_{ij}^{(1)} &= \tfrac{1}{2}\,(a_j^\dagger a_i + a_i^\dagger a_j) \ , \\ d_{ij}^{(2)} &= -\tfrac{1}{2}i(a_j^\dagger a_i - a_i^\dagger a_j) \ , \qquad (9) \\ d_{ij}^{(3)} &= \tfrac{1}{2}\,(a_j^\dagger a_j - a_i^\dagger a_i) \ , \end{aligned}$$

$i \neq j$, to which we will refer as the dipole moment components. The coupling to the electromagnetic field is described by the interaction Hamiltonian

$$H' = \hbar \sum_{m=1}^{3} \sum_{i<j} f_{ij}^{(m)} d_{ij}^{(m)} \ , \qquad (10)$$

where the $f_{ij}^{(m)}$'s are linear superpositions of the components of the electromagnetic field (considered to be Heisenberg-Picture operators) that contain appropriate coupling constants. The total Hamiltonian for atom and field is given by

$$H = H_o + H' + H_f \ , \tag{11}$$

H_f being the field Hamiltonian, which we do not need in explicit form.

Using the notation $n_i \equiv a_i^\dagger a_i$, we note that the basis vectors are eigenvectors of $n_i(0)$ with eigenvalue r_i, and that $\Sigma_i n_i(t)$ is a constant of motion, such that[7]

$$[\Sigma_i n_i(t)] \, | \, r_1 \ldots r_n \rangle = N | r_1 \ldots r_n \rangle \ , \tag{12}$$

where $N = \Sigma_i r_i$. We take the state of the atomic collection (the initial state, in the Schrodinger Picture) to be that for which N is the number of atoms under consideration. The bosons in this description are, therefore, the atoms themselves, and any state in this description corresponds to a fully symmetrized many-atom state in a first quantization formalism.

The basis vectors are, clearly, energy states, that is, eigenvectors of $H_o(0)$ with eigenvalues $\Sigma_i r_i \hbar\omega_i$. It is useful to define another set of states, coherent states,[7,13-15] described by the complex set of numbers $c_1 \ldots c_n$ (designated as $\{c\}$, for brevity), with $\Sigma_i |c_i|^2 = 1$, as follows:

$$|\{c\}_N\rangle = \sum_{r_1 \ldots r_n}^{(N)} \left(\frac{N}{r_1! \ldots r_n!} \right)^{\frac{1}{2}} c_1^{r_1} \ldots c_n^{r_n} | r_1 \ldots r_n \rangle \ , \tag{13}$$

the superscript (N) indicating that the summation is taken over all values of $r_1, \ldots r_n$ for which $\Sigma_i r_i = N$. These states obey the simple relationship

$$a_k(0)|\{c\}_N> = N^{\frac{1}{2}}c_k|\{c\}_{N-1}> \ . \tag{14}$$

In the case of two levels, and translated into a first quantization formalism, the energy states have been referred to in the literature as correlated incoherent states,[16] or as fully symmetrized Dicke states,[14] and the coherent states have been referred to as uncorrelated coherent states,[16] or as fully symmetrical Bloch states.[14] It can be shown that, for the free atoms (uncoupled from the field),

$$\begin{aligned} &<r_1\ldots r_n|d_{ij}^{(m)}|r_1\ldots r_n> = 0 \ , \quad m = 1,2, \\ &<r_1\ldots r_n|d_{ij}^{(m)}|r_1\ldots r_n> = \tfrac{1}{2}(r_j - r_i) \ , \end{aligned} \tag{15}$$

and

$$\begin{aligned} &<\{c\}_N|d_{ij}^{(1)}|\{c\}_N> = N|c_jc_i|\cos[(\omega_j-\omega_i)t+\theta_{ji}] \\ &<\{c\}_N|d_{ij}^{(2)}|\{c\}_N> = N|c_jc_i|\sin[(\omega_j-\omega_i)t+\theta_{ji}] \\ &<\{c\}_N|d_{ij}^{(3)}|\{c\}_N> = \tfrac{1}{2}N(|c_j|^2 - |c_i|^2) \ , \end{aligned} \tag{16}$$

where θ_{ji} is defined by $c_j^*c_i = |c_jc_i|\exp(i\theta_{ji})$. Thus, $<d_{ij}^{(1)}>$, for instance, vanishes for an energy state but oscillates with frequency $|\omega_j-\omega_i|$ and well defined phase for a coherent state (provided c_i, $c_j \neq 0$). On the other hand, if we look at $<d_{ij}^{(1)2}>$, we obtain[7]

$$<r_1\ldots r_n|d_{ij}^{(1)2}|r_1\ldots r_n> = \tfrac{1}{4}[2r_ir_j+r_i+r_j] \ , \tag{17}$$

and

$$\overline{\langle\{c\}_N|d_{ij}^{(1)2}|\{c_N\}\rangle} = \frac{1}{4} N[2(N-1)|c_i|^2|c_j|^2+|c_i|^2+|c_j|^2] , \quad (18)$$

where the bar indicates time average. We see that for $r_k = N|c_k|^2$, the terms of order N^2 are equal.

It may be remarked that the physical significance of treating atoms as bosons in the present manner lies in their cooperative behavior. Complete symmetrization in a first quantization formalism - as is required by such a treatment - may be regarded, intuitively, as maximum cooperation among the atoms under consideration. The present method may be generalized to the case of less than maximum atomic cooperation by dividing a number of atoms into two or more collections, each collection being bosons of a different kind.

The collection of atoms is described completely, in the present formalism, by the coordinates of n harmonic oscillators. In order to investigate the classical-limit conditions, we merely need to consider the classical-limit conditions pertaining to the harmonic oscillators. These are well known to be the limit of high (energy) quantum numbers. Since a description need not be associated with a definite energy state, a more general formulation can be expressed by the inequalities,

$$\langle n_j\rangle >> 1 , \quad j = 1, 2, \ldots n . \quad (19)$$

When these inequalities are fulfilled, the oscillators may be treated classically, the commutators $[a_j, a_j^\dagger]$ are relatively negligible (the last statement can be considered an alternate formulation of the classical-limit condition), and the operators a_j and $a_j^\dagger$ may be

treated as c -number variables. For clarity, where necessary, we will use a tilde henceforth to indicate such variables.

One also needs to look at the field under these limiting conditions. If the atoms under consideration are classical, the field they generate is classical. External fields are also classical, since they must be prescribed.[9] The only part of the field that remains quantum mechanical is the zero-point field. However, classical systems must not "see" the zero point field, since, formally, it appears to them as a field which can do work.[8] Thus, as far as the field that interacts with the collection of atoms under consideration is concerned, it may be described classically if the atoms are described classically.

Thus, the Hamiltonian of Eq. (11) becomes a classical Hamiltonian. Equations of motion for the dynamical variables are obtained according to classical dynamics by means of Poisson brackets. Noting that $a_j = 2^{-\frac{1}{2}}(q_j + ip_j)$, where q_j and p_j are the dimensionless coordinate and momentum of the j'th oscillator, such that $H_o = \frac{1}{2}\Sigma_j \hbar\omega_j (q_j^2 + p_j^2)$, the canonical equations of motion for $\tilde{a}_j$ and $\tilde{a}_j^*$ become

$$\frac{d\tilde{a}_j}{dt} = \frac{1}{ih}\frac{\partial \tilde{H}}{\partial \tilde{a}_j^*} , \qquad \frac{d\tilde{a}_j^*}{dt} = -\frac{1}{ih}\frac{\partial \tilde{H}}{\partial \tilde{a}_j} , \tag{20}$$

while the canonical equations of motion for the field variables become essentially equivalent to the classical Maxwell's equation. In order to complete this theory, which was obtained as the classical limit of a fully quantum mechanical theory, we need, in addition to the equations of motion, a method of prescribing initial conditions that is consistent with the classical-limit procedure.

In the quantum mechanical theory, initial conditions are

described by means of a quantum state. What is the classical limit of such a description? As is well known, a quantum-state description yields information in a statistical form, in general. In the classical limit, therefore, a quantum-state description must become a statistical description, in which $\tilde{a}_j(0)$ and $\tilde{a}_j^*(0)$ are considered to be random variables. One method of providing such a description is that of specifying moments. Consider the moments $\langle O\{\Pi_i a_i^{\dagger v_i}(0) a_i^{w_i}(0)\}\rangle$, where O is an ordering operator that provides for some specific ordering arrangement of the operators inside the curly bracket. Moments corresponding to different arrangements may have different values because of non-vanishing commutators. In the classical limit, these commutators are relatively negligible, provided

$$v_j + w_j << \langle n_j \rangle \ , \tag{21}$$

for all j. We restrict the number and kind of specified moments by these inequalities (thus making the statistical description somewhat non-unique - or somewhat approximate) and specify the various moments of the random variables $\tilde{a}_j$, $\tilde{a}_j^*$ by

$$\langle \Pi_i \tilde{a}_i^{*v_i}(0) \tilde{a}_i^{w_i}(0) \rangle = \langle \psi | O\{\Pi_i a_i^{\dagger v_i}(0) a_i^{w_i}(0)\} | \psi \rangle \tag{22}$$

where $|\psi\rangle$ is the (initial) state of the collection of atoms, and O indicates an arbitrary ordering arrangement that may be chosen for convenience of evaluation. [Note that all ordering arrangements of moments subject to inequality (21) yield results that differ by negligible amounts, in the classical limit.] Probability distributions consistent with this definition of moments provide an alternate statistical description.

Comparison of the equations of motion of SCTIV with those of SCTIII shows that they are identical; however, the method of determining initial conditions in the two theories is different. While in SCTIII the initial conditions for the dipole moments are obtained by equating their values to the corresponding quantum-mechanical expectation values, in SCTIV the initial conditions consist of a statistical description obtained from the quantum mechanical state. SCTIII and SCTII are, thus, a special case of SCTIV; they may be applied only in those situations where not only the classical-limit conditions for the application of SCTIV are met, but also where the (initial) state of the collection of atoms describes the atomic dipole moment in a sufficiently deterministic manner.

A simple argument shows that the above conditions for the application of SCTII - IV are both necessary and sufficient. If we begin with SCTIII as a classical Hamiltonian theory, and convert it, by usual methods, into a quantum theory, we obtain precisely the boson-second-quantization formalism, from which we derived SCTIV. Thus, only this formalism can give, in the classical limit, a theory in which SCTIII may be included, and if SCTIII (and SCTII, for which SCTIII is the Hamiltonian formalism) is to be derived from quantum mechanics, it must meet the aforementioned conditions. Needless to say, these conditions do not allow the application of SCTII - IV to a single atom.

It is instructive to illustrate the application of SCTIV to an energy-state description and to a coherent-state description. For simplicity, we ignore the reaction of the field as well as the effect of external fields. For an energy state that meets the classical-limit conditions ($r_i >> 1$ for all r_i). Eq. (22) yields, using an

ordering arrangement in terms of n_i's for easy evaluation,

$$\langle \Pi_i \tilde{a}_i^{*\nu_i}(0) \tilde{a}_i^{w_i}(0) \rangle = \Pi_i r_i^{\nu_i} \delta_{w_i \nu_i} \tag{23}$$

A simple probability distribution consistent with these moments is given by

$$\tilde{a}_j(0) = r_j^{\frac{1}{2}} e^{-i\theta_j} , \tag{24}$$

where θ_j has a uniform probability distribution. Note that the θ_j's and, therefore, the $a_j(0)$'s, are <u>independent</u> random variables. For a coherent state which meets the classical limit conditions, Eq. (22) yields, using a normal ordering arrangement for easy evaluation,

$$\begin{aligned} \langle \Pi_i \tilde{a}_i^{*\nu_i}(0) \tilde{a}_i^{w_i}(0) \rangle &= [N(N-1)\ldots(N-V+1)] \Pi_i c_i^{*\nu_i} c_i^{w_i} \delta_{VW} \\ &\simeq N^V \Pi_i c_i^{*\nu_i} c_i^{w_i} \delta_{VW} , \end{aligned} \tag{25}$$

where $V = \Sigma_i \nu_i$, $W = \Sigma_i w_i$, and the inequality (21) was utilized in the approximation. A simple probability distribution consistent with these moments is

$$\tilde{a}_j(0) = N^{\frac{1}{2}} c_j e^{i\theta} , \tag{26}$$

where θ is a random variable with a uniform probability distribution, but it has the <u>same</u> value for all $a_j(0)$'s. The $a_j(0)$'s are, therefore, <u>dependent</u>, random variables. Consider, now, the dipole moment of the collection of atoms. If we take $r_j = N|c_j|^2$, the amplitude of

oscillation of $d_{ij}^{(1)}$ and $d_{ij}^{(2)}$ will have the same value for both the energy state and the coherent state. However, the phase of oscillation for the energy state will be a random variable with all values equally probable, while that for the coherent state will be well defined. It is clear that, in the case of these two states, SCTII - III can be applied only to the coherent state (in the classical limit) while SCTIV can be applied to both the coherent state and the energy state. The former is an example of a deterministic description (of the pertinent matter variables in Maxwell's equation) while the latter is an example of a statistical - in the sense of non-deterministic - description.

The conclusion of the present section may be expressed, loosely speaking, by the statement that SCTII - IV are applicable only to those situations where the atoms belong to one or more groups in each of which they behave cooperatively and their number is large; SCTII - III are further restricted to cases where the description of the dipole moment is deterministic. Since SCTIV is the most general form of a classical-limit theory for quantum mechanical systems in cooperative behavior, it is reasonable to refer to it as macroscopic quantum mechanics.

III. ATOMIC COOPERATION LESS THAN MAXIMUM

The MQM formalism of the preceding Section describes a collection of n-level systems among which there exists maximum cooperation. We will consider now the case in which the cooperation is less than maximum, but one in which macroscopic dipole-moment effects may nevertheless exist. It is instructive to consider a specific example which illustrates this case. Consider a large number of three level

systems, with energies, E_1, E_2, E_3 (where $E_1<E_2<E_3$) in equilibrium with a thermal reservoir. Let the two lower levels be sufficiently close together so that their population is of the same order of magnitude, and let the third level be sufficiently high so that its population is essentially negligible. How much cooperative behavior exists in this atomic collection? Assuming, for simplicity, that the population difference between the two lower levels may be neglected, then, as far as these two levels are concerned, there exists no cooperative behavior whatsoever. This is obvious from the application of any of the several criteria for cooperation among two-level systems: 1) Description in terms of one-atom states contains no well-defined symmetry properties; 2) the cooperation number[14] is zero; 3) There exists no macroscopic dipole moment, and none can be generated by the application of a resonant pulse that acts equally on all the molecules; 4) The Bloch vector[14] is zero. If, however, we consider the pair of levels E_1, E_3 or E_2, E_3, then we have maximum cooperation among an effective number of atoms $\frac{1}{2}N$, where N is the total number of atoms, according to any of the criteria above. Thus, there does exist cooperation among the three-level systems, but not the maximum cooperation that is described by the BSQ formalism for one kind of bosons.

Since, by definition, bosons of the same kind cooperate completely, the method of describing less than complete cooperation clearly lies in the use of more than one kind of bosons for the description of the atomic collection. We label the kind of boson by an index within a square bracket, either as a superscript or subscript, depending on notational convenience. The Hamiltonian is then given by

$$H_o = \sum_{ki} \hbar\omega_i a^\dagger_{i[k]} a_{i[k]} \quad . \tag{27}$$

The operators $a_{i[k]}$ are to be considered to operate only on the states of the bosons of the k'th kind, and are c-numbers with respect to all other kinds of bosons. The total dipole moment $D^{(m)}_{ij}$ is the sum of the dipole moments of the individual kinds,

$$D^{(m)}_{ij} = \sum_k d^{(m)[k]}_{ij} \quad . \tag{28}$$

We return to the collection of three-level systems described above. Consider two kinds of bosons, and let their combined state be given by

$$|\psi\rangle = |\tfrac{1}{2}N,\ 0,\ 0\rangle_{[1]} |0,\ \tfrac{1}{2}N,\ 0\rangle_{[2]} \quad . \tag{29}$$

In this state, both kinds of bosons are equal in number, with all of the first kind being in the state that corresponds to E_1, and all of the second kind being in the state that corresponds to E_2. It is clear that the oscillatory moment, interpreted according to MQM, that is, as a (classical) random variable in accordance with Eq. (24), is zero, since $d^{(m)[k]}_{ij}$ is zero for the values m = 1, 2 . Consider now a prescribed resonant field acting on the collection of three -level systems. In accordance with Eq. (10), we take the interaction Hamiltonian to be

$$H'_{ij} = \hbar \sum_{k=1}^{2} f_o (a^\dagger_{j[k]} a_{i[k]} + a^\dagger_{i[k]} a_{j[k]}) \cos\omega_{ij} t \quad , \tag{30}$$

where, for simplicity, we have assumed that $f_{ij}^{(2)} = f_{ij}^{(3)} = 0$, have chosen a specific phase for the driving field, and have set $h\omega_{ij} = |E_i - E_j|$. If we now let

$$\omega_{ij} = \omega_{12} ,$$

then, according to MQM, it is easy to show (using the rotating-wave approximation) that

$$d_{12}^{(1)[1]} = -\frac{1}{2} N \sin \Omega t \cos \Omega t \sin \omega t ,$$
$$d_{12}^{(1)[2]} = \frac{1}{2} N \sin \Omega t \cos \Omega t \sin \omega t , \qquad (31)$$

where $\Omega^2 = \frac{1}{4} f_o^2$; we have, thus, $D_{12}^{(1)} = 0$. Similarly, it can be shown that $D_{12}^{(2)} = D_{12}^{(3)} = 0$. One obtains, therefore

$$D_{12}^{(m)} = 0 .$$

It is seen that there exists no oscillating dipole moment, nor can one be generated by a resonant field, at frequency ω_{12}. This is consistent with the microscopic picture of saturation of the pair of levels E_1 and E_2.

As far as the level-pairs E_1, E_3 and E_2, E_3 are concerned, the situation is entirely different. Using the same method as above, we have, for $\omega_{ij} = \omega_{i3}$, $i = 1, 2$,

$$D_{i3}^{(1)} = d_{i3}^{(1)[1]} = -\tfrac{1}{2} N \sin \Omega t \cos \Omega t \sin \omega_{i3} t \ ,$$

$$D_{i3}^{(2)} = d_{i3}^{(2)[i]} = \tfrac{1}{2} N \sin \Omega t \cos \Omega t \cos \omega_{i3} t \ , \qquad (32)$$

$$D_{i3}^{(3)} = d_{i3}^{(3)[i]} = \tfrac{1}{4} N (\sin^2 \Omega t - \cos^2 \Omega t) \ .$$

One sees that

$$\sum_m D_{i3}^{(m)2} = (\tfrac{1}{4} N)^2 \ , \quad i = 1, 2, \qquad (33)$$

which means that an oscillating dipole moment corresponding to a cooperation number - or a Bloch-vector magnitude - of $\frac{1}{4}$ N can be generated at <u>either</u> frequency ω_{13} or ω_{23} . (It should be noted that in MQM the cooperation number and the magnitude of the Bloch vector are the same quantity.) This is half the cooperation number of that in the case of maximum cooperation. Had there existed no (empty) third level, the cooperation number, as far as the two saturated levels are concerned, would have been zero. By a suitable combination of resonant pulses, one can generate an oscillating dipole moment also at frequency ω_{12} in the present case. Using first a pulse of frequency ω_{13} and of duration r such that $\Omega\tau = \frac{1}{2}\pi$, we can transfer the ground-level population to the third level; then, an oscillating moment can be generated at frequency ω_{12} by a field of frequency ω_{12} . The cooperation number associated with the levels E_1, E_2 under these conditions is also $\frac{1}{4}$ N.

It is seen that the number $\frac{1}{4}$ N is associated with the state of the collection of three-level systmes, rather than with any particular pair of levels, and may be regarded as a cooperation number for the state under consideration. If our purpose is the generation

of an oscillating dipole moment by the application of a resonant field, or by the application of succession of resonant pulses at the various resonant frequencies, then the number $\frac{1}{4}$ N may be regarded as a figure of merit of this state. We can use this operational procedure to define the cooperation number - or figure of merit - of a given state of a collection of n-level systems (where the cooperation is less than maximum). This cooperation number is given by the maximum (dimensionless) dipole moment, or the maximum of the quantity $\{\Sigma_m D_{ij}^{(m)2}\}^{\frac{1}{2}}$, that can be obtained by using a series of resonant pulses. No restriction is to be made on the pulses that may be used.

The above discussion has illustrated the fact that less than maximum cooperation among n-level systems may be described by the consideration of more than one kind of bosons. It has also led to a quantitative measure of the cooperation for a given state. One may naturally enquire about the number of kinds of bosons that are needed to describe a given state. Although no general theory has yet been developed in this connection, it is reasonable to conjecture that the number of kinds of bosons needed for an arbitrary state is not larger than n.

IV. COOPERATIVE ATOMIC BEHAVIOR IN A FOUR-LEVEL LASER

We consider the operation of a four-level laser under the idealized condition of maximum atomic cooperation. This cooperation is assumed to exist not only in the oscillation - or "lasing" - process, but also in the pumping and relaxation processes. Cooperative atomic behavior in the last two processes - to greater or lesser extent - is not impossible to achieve, in principle, for

certain types of lasers. In the case of optical pumping, for instance, uniformity of the pumping field will produce such an effect, while in the case of relaxation by spontaneous emission, the introduction of resonators at the relaxation frequency will do likewise. First, the equations of motion for the dynamical variables in the BSQ formalism will be given. These equations are fully quantum mechanical. Then, taking the classical limit, we obtain the MQM description. Certain properties of the steady-state solution which indicate the special features introduced by the cooperative behavior will be discussed.

Consider N "four-level" atoms (or molecules) with energy levels $i = 1, 2, 3, 4$, ordered so that $\omega_i < \omega_j$ for $i<j$, with $\omega_{ij} = |\omega_i - \omega_j|$. According to the BSQ formalism,[1] the laser matter is described by four oscillators of complex amplitude a_i. Let the 23 transition be coupled to the (lossy) cavity mode of frequency ω_{23}, with which we associate the photon annihilation and creation operators b and $b^\dagger$ respectively.[17] The 14 transition couples to the pump (as well as to a relaxation mechanism), while the 12 and 34 transitions are coupled to relaxation mechanisms, as indicated chematically in Fig. 1.

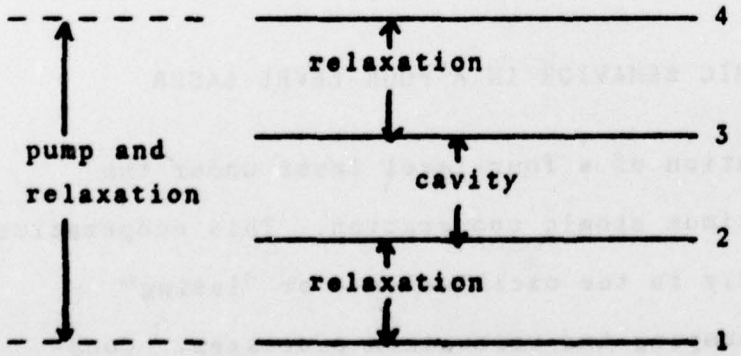

Fig. 1. Schematic depeription of laser under consideration.

Introducing the reduced variables A and B such that

$$a_j(t) = A_j(t)\exp(-i\omega_j t), \quad b(t) = B(t)\exp(-i\omega_{23} t) ,$$

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

$$H'_{ij}(\text{relaxation}) = -\frac{i}{\sqrt{2}}\hbar(A_i A_j^\dagger \mathcal{Q}_{ij} - \mathcal{Q}_{ij}^\dagger A_j A_i^\dagger) , \quad i < j ,$$

$$H'_{23}(\text{cavity}) = \hbar\gamma(A_2 A_3^\dagger B + B^\dagger A_3 A_2^\dagger) , \tag{34}$$

$$H'_{14}(\text{pump}) = -i\hbar(\mathcal{B} A_1 A_4^\dagger - \mathcal{B}^\dagger A_4 A_1^\dagger) ,$$

and by the expressions for $\mathcal{Q}_{ij}$ and B

$$\mathcal{Q}_{ij} = \mathcal{Q}_o^{(ij)} + \frac{1}{\sqrt{2}}\alpha_{ij} A_i^\dagger A_j , \quad i < j , \tag{35a}$$

$$B = B_o - i\gamma\int_o^t dt' A_2^\dagger(t') A_3(t') e^{-\xi(t-t')} . \tag{35b}$$

The symbols occurring in these equations have the following meaning: $\mathcal{Q}_{ij}$ is the relaxation "field" associated with the ij relaxation mechanism,[13] α_{ij} is the corresponding relaxation constant, and $\mathcal{Q}_o^{(ij)}$ is the unperturbed relaxation field; B_o is the cavity field in absence of the molecules, and ξ is the cavity relaxation constant; $\mathcal{B}$ is the prescribed (reduced) pumping field with arbitrary coherence properties. If we ignore, for simplicity, thermal effects, then, classically, the quantities with subscript zero

vanish, and quantum mechanically, they represent the "vacuum" field described by[12,13]

$$B_o|> = \mathcal{Q}_o|> = 0 \ ,$$

$$<|B_o(t_1)B_o^\dagger(t_2)|> = e^{-\xi|t_1-t_2} \ , \tag{36}$$

$$<|\mathcal{Q}_o^{(jk)}(t_1)\mathcal{Q}_o^{(jk)\dagger}(t_2)|> = 2\alpha_{jk}\,(t_1-t_2) \ ,$$

where $|>$ is the ground state of the pertinent system. The resulting equations of motion for the laser matter (the collection of atoms or molecules) are

$$\dot{A}_1 = \frac{1}{\sqrt{2}}\mathcal{Q}_{12}^\dagger A_2 + \frac{1}{\sqrt{2}}\mathcal{Q}_{14}^\dagger A_4 + \mathcal{B}^\dagger A_4 \tag{37a}$$

$$\dot{A}_2 = -\frac{1}{\sqrt{2}} A_1 \mathcal{Q}_{12} - i\gamma B^\dagger A_3 \tag{37b}$$

$$\dot{A}_3 = \frac{1}{\sqrt{2}}\mathcal{Q}_{34}^\dagger A_4 - i\gamma A_2 B \tag{37c}$$

$$\dot{A}_4 = -\frac{1}{\sqrt{2}} A_3 \mathcal{Q}_{34} - \frac{1}{\sqrt{2}} A_1 \mathcal{Q}_{14} - A_1 \mathcal{B} \tag{37d}$$

To these, we must add the normalization condition

$$\Sigma A_i^\dagger A_i = N$$

We will discuss only a very simple special case of these equations, that of a <u>classical</u> system in which A_2 and A_3, as well as the pumping strength $|\mathcal{B}|$, are constant (and non-vanishing).

In other words, conditions are assumed to be such that only the classical macroscopic properties play a significant role in the behavior of this composite system (where, as stated previously, maximum cooperation exists), and the cavity field is in a steady state.

Using Eqs. (35), letting the quantities with subscript zero vanish, ignoring the commutators of all variables, carrying out the integration in Eq. (35b), taking $t >> \xi^{-1}$, and using the notation $n_i = A_i^\dagger A_i$, one can show that Eqs. (37) become

$$\begin{aligned} &\alpha_{12} n_1 n_2 + \alpha_{14} n_1 n_4 = D \ , \\ &\alpha_{12} n_1 = (2\gamma^2/\xi) n_3 \ , \\ &\alpha_{34} n_4 = (2\gamma^2/\xi) n_2 \ , \\ &\alpha_{34} n_3 n_4 + \alpha_{14} n_1 n_4 = D \ , \end{aligned} \tag{38}$$

where

$$D \equiv -(\beta A_1 A_4^\dagger + \beta^* A_1^\dagger A_4) \ .$$

Only three of these equations are obviously independent.

We will not investigate the general solution, but rather show that these equations may be interpreted as describing a classical parametric oscillator. The classical steady-state field in the cavity is, from Eq. (35b), given by

$$B(\text{classical}) = -\, i(\gamma_{23}/\xi)A_2^\dagger A_3 \ .$$

This should be regarded as the signal, at frequency ω_{23}. As for the idler, there exist <u>two</u> idler oscillations, at frequencies ω_{12} and ω_{34}, with atomic amplitudes proportional to $A_1^\dagger A_2$ and $A_3^\dagger A_4$, respectively. It is clear from Eqs. (38) that D must be a real positive quantity, which means that the phases of A_1 and A_4 must depend on the phase of $\mathcal{B}$. There is nothing in the equations, however, which determines the phase of the signal oscillation; it is arbitrary, and independent of the pump phase, as it should be. We see, therefore, that a "classical" laser, in which there exists a maximum of atomic cooperation in the non-lasing processes, is a parametric oscillator.

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