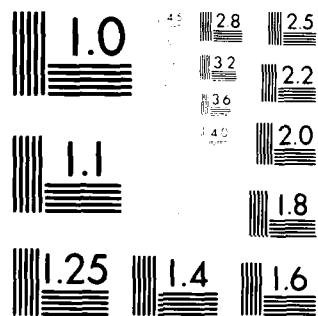AD-A085 946   ARMY MISSILE COMMAND REDSTONE ARSENAL AL RESEARCH D--ETC F/G 20/10
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RECENT ADVANCES IN COOPERATIVE
PHENOMENA AND MATTER/FIELD
INTERACTIONS

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Recent breakthroughs in the areas of swept-gain superradiance, laser induced chemistry, and optical bistability have led to a broad program which encompasses and is germanely relevant to the major areas of specialization in coherent cooperative effects being undertaken at universities under Government sponsorship. In addition to the obvious applications, a deeper and more profound understanding of advanced directed radiation and phase changes in matter/light interactions has been developed through the

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18 19 September 1979, by the board of the Soviet Journal of Quantum Elec-
tronics for publication in that journal. However, on 17 January 1980,
in keeping with President Carter's policy of technological sanctions
against Soviet Russia arising out of their invasion of Afghanistan, the
decision was made to withdraw from publication in that journal and
publish as a Technical Report.

20 appropriate utilization of basic first principles and the in-depth
analysis and study of coherent cooperative behavior.

CONTENTS

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I. INTRODUCTION

Recent efforts here in the field of Quantum Optics have led to the generation and establishment of important results and findings in the areas of swept-gain superradiance [1,2], laser induced chemistry [3] and optical bistability [4]. These areas constitute examples of collective phenomena in light-matter interactions which have incurred widespread interest and theoretical and experimental activity, not only from the standpoint of the fundamental understanding of cooperative effects, but also from the motivation stemming from obvious potential applications.

State-of-the-art results in the areas of superradiance and cooperative effects were first reported here in December 1976 as part of the US Army sponsored Symposium on New Laser Concepts. Subsequently, these presentations were published by Plenum Press as part of the text Cooperative Effects in Matter and Radiation, edited by Bowden, Howgate, and Robl [5]. In April 1977, the first swept-gain superradiance experiment [1] was performed at our laboratory using CO_2-pumped CH_3F. As part of a coordinated effort, our theoretical program has modeled superradiance in a collection of three-level molecules, where the dynamical transient effects of coherent optical pumping are explicitly treated [6]. Here, it was shown that two-photon, coherent, stimulated Raman transitions play an important role in the evolution of macroscopic collective polarization. The connection between pure superradiance and swept-gain superradiance was then explicitly established, both experimentally and theoretically.

Also, the influence of chemical reactivity and/or dissociation on laser-induced population transfer in multilevel systems has been studied [7] and simple analytical models [3,7,8,9] have been devised to determine the nature of essential fundamental processes in larger systems. Conditions under which population can be trapped in excited

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states in multilevel systems undergoing dissociation and/or reaction have been established [3]. Also, a convenient and economical computer algorithm has been developed from which a computer simulation model was established; and rules for the reduction of the effective manifold of states in a multilevel laser pumped system have been devised which can significantly increase the efficiency of and decrease the cost of machine computation [9].

Likewise, investigation of the instabilities in the ground state of the Dicke Hamiltonian [10,11] has resulted in the establishment of optical bistability as a first-order phase transition far from thermodynamic equilibrium [4] -- which is caused by virtual photon exchange in atomic pair correlations in a mean-field interaction. This observation has led to a demonstration of the precise formal connection between optical bistability, superfluorescence, super-radiance, and the BCS theory of superconductivity. To emphasize our interest and support in this dynamic new area, an International Conference on Optical Bistability is being sponsored jointly by the US Army Research Office and this installation, to be held 3-5 June 1980 in Asheville, North Carolina, USA.

It is not the purpose of this paper to review in detail our total program which is (on the whole) well documented in the open literature, but rather to focus attention on several critical areas of thrust and current momentum. With this in mind, we shall attempt in the remainder of this discussion to outline and present some of our results in several areas of current endeavor. Specifically, Section II will consider a rigorous procedure for systematically handling the general interaction of a molecular gas with the electromagnetic field, while Sections III and IV will deal more directly with specific models that enhance our understanding of and effective treatment of cooperative phenomena in light-matter interactions. Section V will

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provide a brief summary of our conclusions and future goals and direction.

II. INTERACTION OF A MOLECULAR GAS WITH THE ELECTROMAGNETIC FIELD

In recent years a number of papers [12] have considered theoretical models for the interaction of molecular systems with the quantum electromagnetic field. Problem areas centering around the treatment of the molecular system itself (i.e., motion of the center of mass) and restrictions pertaining to field origin and structure, etc. tend to impact the formalism such that one neither knows implicitly the approximations which have been made in the model nor how one could improve on the results of the calculations. Ideally, the basic equations of the theory should be physically "exact" in the sense that they are derived from first principles; they should be of a form which makes the physical and mathematical significance of various approximations reasonably clear; and they should permit any desired physical effect to be calculated with sufficient effort.

In order to achieve some of these goals for the case where the molecular system is a gas, a complete second-quantized Hamiltonian for a system of nonrelativistic massive charged particles interacting electromagnetically with each other and with the quantum electromagnetic field has been formulated (by C.A.C.) subject only to three _a priori_ approximations or constraints: The nuclei of the molecules can be considered as elementary particles, all massive particles can be treated nonrelativistically, and wall effects on the internal states of the molecules can be neglected. A key step in the formalism of the theory is the choice of an appropriate set of operators for the description of the molecular system. For this purpose we introduce the operators $P_k(\underline{X},t)$ which destroy a molecule in internal state k with center-of-mass position $\underline{X}$ at time t, along

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with its adjoint $P_k^\dagger(\underline{X},t)$ which creates such a molecule. A
Hamiltonian is then used to derive equations of motion for the
$P_k(\underline{X},t)$ which contain only these operators, their adjoints, and the
electromagnetic vector potential. The current term in the equation
for the electromagnetic vector potential is also expressed in terms of
these molecular operators. The resulting set of operator equations is
closed, forms a complete basis for the description of the interaction,
and has the convenient feature that in it the internal molecular
states (which are involved in electromagnetic transitions) and the
molecular center-of-mass coordinates (which play a role in transport,
density fluctuations, Doppler shifts, etc.) enter in different ways.
This fact is advantageous in allowing use of experimentally-determined
parameters in numerical calculations for specific systems, and it
permits one to see in what ways various approximations in treating the
molecular system interact with one another.

The total Hamiltonian for the system in rationalized Gaussian
units, nonrelativistic in the massive particles, is

$$
\begin{aligned}
H = \sum_m \frac{1}{2M_m} &\int d^3x \; \psi_m^\dagger(\underline{x}) \left[\frac{\hbar}{i}\underline{\nabla} - \frac{q_m}{c}\underline{A}(\underline{x})\right]^2 \psi_m(x) \\
&+ \frac{1}{2}\sum_{m,m'} q_m q_{m'} \int d^3x\, d^3x' \; \psi_m(\underline{x})\, \psi_{m'}^\dagger(\underline{x}') \frac{1}{|x-x'|} \\
&\times \psi_{m'}(\underline{x}')\psi_m(\underline{x}) + \frac{1}{2}\int d^3x \left[\frac{1}{c^2}\left(\frac{\partial A}{\partial t}\right)^2 + (\underline{\nabla}\times\underline{A})^2\right] ,
\end{aligned}
\tag{1}
$$

where $\psi_m(\underline{x},t)$ is the annihilation operator for a particle of type m
(i.e., electron, nuclei) at position $\underline{x}$ and time t, $\underline{A}(\underline{x},t)$ is the
electromagnetic vector potential in the Coulomb gauge, q_m and M_m
is the charge and mass of the mth species of particles,
respectively, etc.

We now define

$$P_k(\underline{X},t) = \int \phi_k(y) \prod_i \psi_i(\underline{x}_i,t) \prod_j d^3y_j \tag{2}$$

relevant to a molecule where $\underline{x}_i$ is the coordinate of its ith particle, $\underline{X}$ is the coordinate of its center of mass $(\underline{x}_i = \underline{X} + \sum_j b_{ij}\underline{y}_{ij})$, ϕ_k denotes the kth eigenstate, and ψ_i is the annihilation operator of its ith particle (to be distinguished from ψ_m, as m denotes specific type while i specifies a given particle). It is obvious from this definition that $P_k(\underline{X},t)$ destroys a molecule with internal coordinate k and center-of-mass coordinates $\underline{X}$ at time t, and correspondingly that $P_k^\dagger(\underline{X},t)$ creates a molecule of this kind. The field operators $P_k P_k^\top$ are convenient in studying molecular interactions with the electromagnetic field because they reflect properties of an entire molecule rather than of the individual particles that constitute the molecule.

We now develop equations of motion in which only the operators $P_k, P_k^{\pm}$ and $\underline{A}$ appear, together with constants and functions determined by the internal molecular states. This set of three equations now forms a complete and closed set of operator equations for the description of the resulting N-molecule system interacting with the quantum electromagnetic field -- the only approximations being those three a priori constraints specified earlier. For simplicity an additional dipole approximation can now be introduced on the assumption that for states of interest the electromagnetic vector potential varies slowly enough over molecular dimensions that this variation can be neglected. In situations requiring greater accuracy, this (optional) approximation could just as well be omitted in the resulting calculation of the dynamical properties of the system.

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Our basic procedure is now to use these operator equations to generate equations for a hierarchy of Green functions. By making various factorization assumptions one may then retain as much of the exact content of these equations as is necessary and/or feasible in the case of interest. We therefore define the Green functions

$$P_{k\ell}(\underline{X},\underline{X}') = b_1 \; < P_k^\dagger(\underline{X}) \; P_\ell(\underline{X}') > \; ,$$

$$P_{k_1 k_2 k_4}(\underline{X}_1,\underline{X}_2,\underline{X}_3,\underline{X}_4) = b_2 < P_{k_1}^\dagger(\underline{X}_1) P_{k_2}^\dagger(\underline{X}_2) P_{k_3}(\underline{X}_3) P_{k_4}(\underline{X}_4) > \; , \tag{3}$$

$$\underline{a}(\underline{X}) = < \underline{A}(\underline{X}) > \; ,$$

where t is understood and b_1 and b_2 are normalization constants. Under a rational factorization procedure,

$$< P_k^\dagger P_\ell(\underline{A}) > = < P_k^\dagger P_\ell > \quad < \underline{A} > \; ,$$

$$< P_k^\dagger P_\ell \underline{A}^2 > = < P_k^\dagger P_\ell > \quad < \underline{A} >^2 . \tag{4}$$

The resulting Green function hierarchy can be truncated to include only those functions defined under expression (3). In this minimal "semi-classical" approximation (that includes also the dipole approximation), equations of motion for $P_{k\ell}(\underline{X},\underline{X}',t)$ and $\underline{a}(\underline{X},t)$ assume a representation,

$$i\hbar \frac{\partial}{\partial t} P_{k\ell}(\underline{X},\underline{X}',t)$$

$$= \left[\frac{\hbar^2}{2M} (\underline{\nabla}^2 - \underline{\nabla}'^2) - (E_k - E_\ell)\right] P_{k\ell}(\underline{X},\underline{X}') - \frac{N-1}{V} \sum_{k_1 k_2 k_3}$$

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$$\times \int d^3x'' \left[Q_{k_1 k_2 k_3 k}(\underline{X}''-\underline{X}') P_{k_1 k_2 k_3 \ell}(\underline{X},\underline{X}'',\underline{X}'',\underline{X}') \right.$$

$$\left. - Q_{\ell k_1 k_2 k_3}(\underline{X}''-\underline{X}') P_{k k_1 k_2 k_3}(\underline{X},\underline{X}'',\underline{X}'',\underline{X}') \right]$$

$$- \frac{i}{c} \sum_{k'} \left[\omega_{k'k} \, \underline{d}_{k'k} \cdot \underline{a}(\underline{X}) P_{k'\ell}(\underline{X},\underline{X}') + \omega_{\ell k'} \, \underline{d}_{\ell k'} \cdot \right.$$

$$\left. \times \underline{a}(\underline{X}') P_{kk'}(\underline{X},\underline{X}') \right] - \left(\sum_i \frac{q_i^2}{M_i c} \right) P_{k\ell}(\underline{X},\underline{X}')$$

$$\times \left[\underline{a}^2(\underline{X}) - \underline{a}^2(\underline{X}') \right] .$$

$$\left[\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \nabla^2 \right] \underline{a}(\underline{X},t) = \frac{N}{V} \frac{i}{c} \left[\sum_{k,\ell} \omega_{k\ell} \underline{d}_{k\ell} P_k (\underline{X},\underline{X}) \right]_t \qquad (5)$$

$$- \frac{N}{V} \left(\sum_i \frac{q_i^2}{M_i c} \right) \left[\underline{a}(\underline{X}) \sum_k P_{kk}(\underline{X},\underline{X}) \right]_t .$$

where $[---]_t$ is the solenoid part (zero divergence) of the vector
quantity in brackets. The role of parameters such as transition
energy $\omega_{k,k'}$, associated dipole moment $d_{k,k'}$, intermolecular
coupling $Q_{k_1 k_2 k_3 k_4}$, etc. is obvious upon inspection of the
Equations (5), which do not form a closed system because of the
presence of the functions $P_{k_1 k_2 k_3 k_4}$. If the intermolecular
interactions are to be treated, one must either include additional
Green function equations or make additional approximations to express
the $P_{k_1 k_2 k_3 k_4}$ in terms of the $P_{k_1 k_2}$.

Without the presence of the intermolecular term, Equations (5) do
become a closed set describing the evolution of the system. They
describe the processes of absorption and emission of radiation,
Doppler shifts, density fluctuations, etc., but contain no direct
field-molecule correlations and cannot produce the spontaneous

emission of radiation. To proceed to the next higher order of
accuracy one can drop the assumption that $\langle P_k^\dagger P_\ell A)$ factorizes and
define a new Green function proportional to this quantity. It is then
also necessary to introduce a Green function proportional to
$\langle P_k^\pm P, \frac{\partial A}{\partial t}\rangle$ and factorization now must be assumed for quantities of the
form $\langle P^\dagger P P^\dagger P A\rangle$, etc. This new set of equations propagates
field-molecule correlations existing in the initial state, but does
not generate new correlations; i.e., if the quantity $\langle P^\dagger P A\rangle$ factors
at the initial time, it factors at later times as well. Spontaneous
emission of radiation is not generated by the equations for this set
of Green functions, and one must go to the next higher order of
accuracy and consider a Green function proportional to $\langle P^\dagger P P^\dagger P\rangle$.
The set of equations for Green functions correct to this order is
straightforward to derive, but too cumbersome to illustrate as part of
this publication.

It should now be sufficiently clear how the technique may be
applied to calculate equations describing any desired physical effect.
Consideration of higher order effects by such methods usually must be
carried out for the simplest model systems. Nevertheless, the
hierarchy of Green function equations does allow one to determine in a
rather clear manner exactly how various physical effects are
interrelated and what the effects of various approximations may be.
Even with neglect of the intermolecular interaction term, the
semiclassical Equations (5) still contain interesting features. If
one ignores molecular motion by dropping the differential operator
$\nabla^2 - \nabla'^2$, such that it is sufficient to consider only the case of
$\underline{X}' = \underline{X}$, then one obtains a set of equations equivalent (for a finite
set of molecular levels) to the Maxwell-Schrödinger model. Thus even
in the dipole approximation one cannot consider a molecule as a point
particle in the semiclassical approximation, as evident by the
structure of the kinetic energy term in Equations (5), which cannot be

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expressed in terms of a single center-of-mass coordinate and internal molecular variables.

As one illustration of the basic theory in the semiclassical approximation (using a dipole approximation and ignoring the intermolecular interaction term), we have considered a typical three-level swept-gain superradiance model in which relaxation times and diffraction losses are introduced empirically and an initial tipping angle (Bloch model) replaces the inclusion of spontaneous emission [2,6]. In this example, we have shown that the growth of cooperative macroscopic polarization in the medium during the pumping process scales as the ratio of the pump's temporal duration $_p$ to the characteristic superradiance time $_R$, rather than to $_D$ (the delay time), as has been speculated previously in the literature [13]. The influence of the pump pulse shape, as well as area, was also determined to be critical with regard to subsequent cooperative relaxation. Also, two-photon, coherent, stimulated Raman contributions were shown to make important contributions to pulse evolution during the pump time regime. This finding, in turn, has opened up a new area of research and investigation which we are now in the process of vigorously pursuing. Other, significant and meaningful examples to illustrate the basic theory can easily be constructed, while important effects such as spontaneous emission, diffraction, etc., can be rigorously treated or accounted for solely upon the inclusion of higher order levels of approximation.

III. AN EFFECTIVE HAMILTONIAN FOR THE TREATMENT OF N
      IDENTICAL TWO-LEVEL ATOMS INTERACTING WITH THE
      ELECTROMAGNETIC FIELD

Our interests in the Dicke model via superradiance, optical bistability, etc., have led us to examine in some detail the

interaction of a set of N identical atoms with the electromagnetic
field [4,6]. In effect, the field creation and annihilation operators
$a_k, a_\ell^\dagger$ conform to standard Bose commutation relations involving two
specific modes, k and ℓ, while the atomic operators $C_{j\ell}, C_{ik}^\dagger$ are
Bose operators [14] which satisfy the commutator relations

$$\left[C_{j\ell}, C_{ik}^\dagger \right] = \delta_{ji} \, \delta_{k\ell} \quad , \quad \left[C_{j\ell}, C_{ik} \right] = 0 \quad , \tag{6}$$

where i,j identifies the specific atomic level and k,ℓ identifies the
specific atom. In this way one attempts to treat a system which can
be identified by the existence of a many-particle Hamiltonian and
single-particle eigenstates, thereby assuring the existence of
elementary anti-commutator properties for the products $C_{j\ell}^\pm C_{i\ell}$. As
a consequence, for a two-level system (energies $\epsilon_2 > \epsilon_1$), we may
choose a vector operator $\underline{f}^i$,

$$f_1^i = (C_{2i}^\dagger C_{2i} - C_{1i}^\dagger C_{1i}) \quad , \quad f_2^i = 2^{\frac{1}{2}} C_{1i}^\dagger C_{2i} \quad ,$$

$$f_3^i = 2^{\frac{1}{2}} C_{2i}^\dagger C_{1i} \quad , \quad f_4^i = 1 \quad , \tag{7}$$

and transform to a representation obeying the commutation
(-)/anticommutation (+) relations

$$\left[f_\ell^i, f_k^i \right]_{\mp}^\pm = \sum_m S_{\ell,km}^\pm \, f_m^i \quad , \tag{8}$$

where $S_{\ell,km}^\pm$ can be established on the basis of $C_{j\ell}$, $C_{ik}^\pm$ and
their relations in a system involving only single particle eigen-
states. It is also to be noted that the three-dimensional operator
$\hat{\underline{f}}^i$ (components f_1^i, f_2^i, f_3^i) acts as an angular momentum

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operator in which f_1^i denotes the direction of diagonalization or measurement. If H^i, a Hamiltonian in $\underline{\hat{f}}^i$, has the form

$$H^i = \underline{\Gamma} \cdot \underline{\hat{f}}^i \ , \quad \Gamma_1 = E \cos\theta \ , \quad \Gamma_2 = 2^{-\frac{1}{2}} E \sin\theta \ e^{-i\alpha} \ ,$$

$$\Gamma_3 = 2^{-\frac{1}{2}} E \sin\theta \ e^{i\alpha} \ , \tag{9}$$

where $\underline{\Gamma}$ is not explicitly dependent upon time, it is a simple matter to rotate $\underline{\hat{f}}^i$ about the $\underline{\theta}$ axis to obtain a frame of reference in which H^i is diagonal. The transformation T (expressed in terms of Euler's angles), which rotates $\underline{f}^i$, then can be used to evaluate the temporal and thermal variation of $\underline{\hat{f}}^i$ in a frame of reference in which the generator H^i is not diagonal, assuming simple relations exist for this variation in that frame of reference in which H^i is diagonal. Cases in which the temporal and thermal generators are diagonal in different frames can also be adequately handled, requiring (of course) the explicit use of more than one transformation, when three different frames (i.e., $\hat{f}^i(1)$, $\hat{f}^i(2)$, $\hat{f}^i(3)$) are considered in the mathematical expression of one physical quantity. Our initial argument is then that if one can reduce a Hamiltonian for the interaction of N identical two-level atoms with an electromagnetic field to the form specified by expression (9), then one is in a position to manipulate it with relative ease and rigor.

Denoting the (internal) field by $\underline{a}\{a_k, a_k^\dagger\}$ and the atoms by $\underline{f} = \Sigma \underline{f}^i$, let the temporal variation of a general operator X be established in this environment through its Hamiltonian generator H_x and the resulting equation of motion. For complex ω, one may define also its transform

$$X(\omega) = G_x(\omega; H_x) \ , \tag{10}$$

as an analytic two-branch operator defined everywhere outside the real axis [15] and obeying an equation of motion

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$$\omega X(\omega) = (2\pi)^{-1} X + Z(\omega) \quad , \quad Z = \left[X, H_x \right] . \qquad (11)$$

As our system spans the product space of $\underline{a}$ and $\underline{f}$, H_x would assume a functional representation

$$H_x = F_x(\underline{a}, \underline{f}) \quad , \qquad (12)$$

and the temporal variation of X would then evolve from expressions (11) and (12) and the statistics for $\underline{a}$ and $\underline{f}$. An inherent problem in this configuration is the hierarchy of equations generated from relation (11) when one handles $\underline{a}$ (or the electromagnetic field) in its present representation. In this light (as a first step toward achieving expression (9)), it would be desirable to project $\underline{a}$ into the space of $\underline{f}$ such that Equation (11) is at least solvable exactly at the single-particle level with no explicit time dependence appearing in the Hamiltonian. In so doing we must abandon the statistics of $\underline{a}$ and establish a mapping of $\underline{a}$ onto $\underline{f}$ (and its statistics) that is consistent within the framework of the theory and sufficiently general to warrant its application. It will, of course, not be exact or absolute as it is a projection or approximation that in practice will depend on the constraints of modeling and interpretation. In so doing our new representation of $\underline{a}$, and thus the total system, will be in $\underline{f}$ rather than $\underline{a} \otimes \underline{f}$, and it should not be looked upon as an approximation that one introduces arbitrarily or at random.

In order to effectively stabilize our system and make it accessible to statistical interpretation, we shall now introduce a phenomenological damping coefficient γ_k (rate of photon escape from the quantization volume V) for the kth photon mode. This device will effectively account for the reflectivity from cavity mirrors which limit the quantization volume, etc. and will emphasize the

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Markovian nature of the system for times long compared with some characteristic relaxation time $\tau_c = \gamma_k^{-1}$. The electromagnetic field we (in turn) interpret to exhibit both internal and external origins, with the latter potentially identifiable as an applied field in a coherent state, with field amplitude α and carrier frequency ω_o. The generator H_{a_k}, we shall assume to induce an equation of motion for a_k of the form

$$i\, \partial a_k / \partial t = \Omega_k a_k + \underline{\lambda}^k \cdot \underline{f} \quad , \quad \gamma_k = -\,\mathrm{Im}(\Omega_k) \quad , \tag{13}$$

with a similar equation for the conjugate $a_k^\dagger$. Taking the transform of Equation (13), using expression (11), then establishes the requirement that if

$$(\omega - \Omega_k)a_k(\omega) \rightarrow 0 \quad , \text{ as } \quad (\omega - \Omega_k) \rightarrow 0 \quad , \tag{14}$$

then [16]

$$a_k = -\, 2\pi\, \underline{\lambda}^k \cdot \underline{f}(\Omega_k)_a \quad , \tag{15}$$

with a similar statement for $a_k^\dagger$. Thus $\underline{f}$ acts as an operator whose temporal variation provides a means for transmitting the statistics of $\underline{a}$ (contained in $H_{\underline{a}}$) into $\underline{f}(\Omega_k)$ and its conjugate, and, as such, Equation (15) must still be identified as existing in $\underline{a} \otimes \underline{f}$. Requirement (14), on the other hand, implies that $\underline{f}$ is capable of supporting $\underline{a}$ to the extent that the interaction $\underline{af}$ will prevent $a_k(\Omega_k)$ from tending to become singular or otherwise unconstrained in its integral over time. In any event, the field $\underline{a}$ has now been correlated to the atoms $\underline{f}$ through the interaction (coefficients λ_i^k); and the external field (or source) may, in turn, either drive the atoms or be coupled

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directly to the internal field $\underline{a}$ through the coefficient λ_4^k as it appears in Equation (15).

Substituting Equation (15) into expression (12) and defining [17]

$$H_{\underline{f}} = F_{\underline{f}}(\underline{f}(\Omega_k)_a,\underline{f}) \quad , \quad F_{\underline{f}} = \tfrac{1}{2}(F_a + F_{a^+}) \quad , \tag{16}$$

one is now led to insert this expression into the transform (10) thereby obtaining

$$f(\Omega_k) = G_{\underline{f}}(\Omega_k; F_{\underline{f}}(\underline{f}(\Omega_k)_a,\underline{f})) \quad . \tag{17}$$

In general, expression (17), as an operator equation, cannot be directly evaluated, and one must go to some statistical average or measurement in order to extract information useful for physical interpretation. The statistical average of the product of $\underline{f}(\Omega_k)$ and an arbitrary parameter Q, or $\langle \underline{f}(\Omega_k) Q \rangle$, is a likely basis for evaluation. One is now in a position to reduce expression (17) to the space of $\underline{f}$ by defining the respective mappings

$$\langle \underline{f}(\Omega_k)Q \rangle = A(\Omega_k) \langle \underline{f}Q \rangle \quad , \quad \underline{f}(\Omega_k)_a = B(\Omega_k)\underline{f} \quad , \tag{18}$$

where Ω_k is now arbitrary in nature and $\underline{f}$ could just as well be represented by its single-particle version $\underline{f}^1$. For the general case of many particles, relations (18) can only be identified in the realm of approximations to the system and the mappings (matrices) A and B are impacted by modeling considerations. For restrictive situations A can be taken identical to B and the statistical average of expression (17) becomes an equation for self consistency in B. In the case of a

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many-particle system, the substitution of $\underline{f}(\Omega_k)$, as approximated by relation (18) into H_f, as defined by expression (11), does not immediately produce $\overline{H^i}$, as defined in expression (9). One must first resort to a "mean-field" form of approximation in order to decouple operator products involving different atoms (i.e., $f_\ell^i f_k^j$, $i \neq j$). Thus, in the fabrication of a single atom Hamiltonian H^i, the operator f_k^j must be replaced by its statistical average $\langle f_k^j \rangle$. The mean-field approximation is certainly justified in models which assume that all of the atoms are contained in a volume much smaller than an atom resonance wavelength. In summary, the three steps necessary for the fabrication of an H^i are: (a) the correlation of a_k to $f(\Omega_k)$; (b) the mapping of $\underline{f}(\Omega_k)$ into $\underline{f}$; and (c) the factoring of $f_\ell^i f_k^i$ in a mean-field approximation. Of primary importance is, of course, the choice of A and B which impacts one's ability to successfully model any systems considered.

Although we have mentioned the existence of an applied external field, we have as yet not taken into account the impact of its potential explicit dependence on time. Any explicit time dependence must be removed initially prior to other considerations, otherwise fabrication of H^i (as previously envisioned) is jeopardized, and statistical and equilibrium properties of a given system cannot be effectively treated. A convenient solution is to canonically transform to a doubly rotating frame of reference rotating at the carrier frequency ω_0 and discuss statistics and thermodynamics in the rotating frame. In this frame the applied electromagnetic field appears as a dc field. After canonical frequency renormalization, the transformed Hamiltonian H_T will assume the form UHU^{-1} for an appropriate transformation operator U. The existence of a given U may impose certain constraints or restrictions on the generality of H, particularly in regard to the preservation of counterrotating terms which may defy manipulation. In order to discuss thermodynamic equilibrium properties in the presence of a time-dependent external

field, one must introduce an effective or "spin" temperature.
Agreement with nonequilibrium models would then exist in the limit of
zero effective temperature. The combination of effective or "spin"
temperature and transformation to a frame rotating at the carrier
frequency ω_0 has been employed effectively by two of us (CMB and
CCS) in the treatment of first- and second-order phase transitions in
the extended Dicke model [4].

IV. LIGHT-ATOM INTERACTION IN COOPERATIVE PHENOMENA IN
QUANTUM OPTICS

As introduced in the previous section, one of the principal
theoretical efforts in quantum optics in our group is concerned with
the atom-atom interaction via the electromagnetic field - an area that
has drawn considerable attention in the literature for some time. A
complete review on this subject is given by Milonni and Knight [18].
A second area of emphasis and study in our program is an investigation
of the thermodynamic properties of the Dicke model [17-19], defined by
the Hamiltonian

$$H = \sum_i g(\sigma_i^+ a + \sigma_i^- a^+) + \sum_i \varepsilon \sigma_i^z ,$$

(19)

where σ_i^+ σ_i^- and σ_i^z are the spin operators for atom i and $a^\dagger(a)$
are the field creation (annihilation) operators for the resonant
single mode.

It was first shown by Hepp and Lieb [19] that a second-order phase
transition appears at a critical temperature T_c if $\hbar\omega/8\pi d^2\rho < 1$,
where d is the atomic dipole moment matrix element, ω the frequency,
and $\rho = N/V$ the density of atoms in a cavity. Despite considerable

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progress made by Wang and Hioe [20] and Gilmore and Bowden [21] in the simplification of the calculation and the study of the effect of the nonlinear term in H, it was still not clear as to the physical mechanism which causes the phase transition. Subsequently, we have used various different formalisms to study the problem and it is now clear from our work [10,11] that the phase transition of the atomic system is simply caused by the atom-atom pair interaction induced by the radiation field,

$$H_{int} = \sum_{i,j} \bar{g} \, \sigma_i^+ \, \sigma_j^- \quad , \tag{20}$$

where $\bar{g} \sim g^2/\epsilon$.

Equation (20) shows that the atomic interaction in the Dicke model is an infinitely long range force, since $\bar{g}$ has no spatial dependence. This conclusion is valid for a single mode of the radiation field. Otherwise, we must assume that the volume V_g containing the atoms is much smaller than the resonant wavelength, which therefore is much smaller than the field quantization or cavity volume V. When the inter-atomic distance becomes larger than the resonant wavelength, then $\bar{g}$ becomes equivalent to the Van der Waal force between two atoms [18]. These observations seem to have been neglected in the literature. We also have extended our calculations to three-level atoms in the Dicke model and have shown the existence of two second-order phase transitions.

Optical Bistability (OB) [22] has drawn a great deal of attention in the last few years after the pioneering experimental work by Gibbs, McCall, and Venkatesan, and McCall's own theoretical effort [23].

Theoretical models [24,25] for the analytical interpretation of the
hysteresis of the input field E_I versus the transmitted field E_T
are based on the coupled Maxwell-Bloch equations. The theoretical
results obtained show that the relation E_T versus E_I is quali-
tatively consistent with experimental observations for the absorptive
case. OB based on this model [26] is essentially a saturation
phenomenon similar to magnetic resonance, and (as a matter of fact)
the condition for OB implies specifically that the atomic absorption
must be saturable.

In the spirit of the study of atom-field interactions, we propose
a new model for OB which is assumed to be represented by a thermo-
dynamic ensemble in the rotating frame of the external field. After
transformation to the rotating frame of reference, we obtain an
atom-atom interaction similar to Equation (20), except that

$$\bar{g} = g_0^2 \frac{(\omega_c - \omega_0 - \sigma)}{(\omega_c - \omega_0 - \sigma)^2 + \gamma^2}, \tag{21}$$

where ω_c is the cavity frequency of the Fabry-Perot interferometer, γ
is the photon escape rate, and

$$\sigma = \left[(\epsilon - \omega_0)^2 + \Delta^2\right]^{\frac{1}{2}} \tag{22}$$

is the renormalized atomic level separation with Δ to be deter-
mined self-consistently from the equation of motion. Distinctive
characteristics of our model are that we take Δ into account in the
operator equation for $a^\dagger(a)$ and study OB in the limit of large T_1

and T_2 -- the small absorption limit. The resultant equation for E_T versus E_I is then given by

$$E_T = E_I + 2 E_T \bar{g} \frac{\tanh (\sigma/2KT_s)}{\sigma} , \qquad (23)$$

where T_s is the effective temperature directly analogous to the "spin temperature" in the rotating frame, and Δ (as specified in Equation (22)) is simply E_T.

In the limit $\omega_c = \omega_o = \varepsilon$, the condition for perfect tuning, Equations (21) and (22) reduce to the well-known formula (for establishing OB) given in the previous literature [24,25] with different coupling constant C. Although our work may be used to explain the dispersive effect better than the previous works [24,25], we should emphasize our fundamental difference in approach and physical mechanism to explain OB. It seems our model (by using the Green's function approach) requires T_1 and T_2 to be sufficiently large so that each atomic level is not critically broadened; whereas the Maxwell-Bloch equations assume small T_1 and T_2 for observation of OB.

Experimentally, the Stark shift Δ (in our model) can easily be observed in the light spectrum; likewise the change in the linewidth discussed in the Maxwell-Bloch model can also be observed easily. We hope some experimental work in this area will appear soon to clarify the situation. Regardless of the outcome as to which model is more adequate to describe OB, we believe it interesting that a first-order phase transition can indeed be deduced from the Dicke model when an external field is introduced to modulate the atomic interaction. At present, we are working on the effect of introducing finite T_1 and

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T_2 into our model and including the spatial dependence of the interaction discussed earlier. Likewise, an attempt will be made to extend our modeling activities to larger and more complex systems.

V. CONCLUSION

In the preceding sections we have outlined the scope of our program in cooperative phenomena and matter/field interactions and detailed some specific areas of current thrust and momentum. We have tried to emphasize the development of appropriate building blocks from scientific first principles and the establishment of effective techniques for the structuring and analysis of theory and data. The theory developed in Section II and depicted in Equation (5) is exceedingly general and definitive both in its scope and content. Its application to the case of swept-gain superradiance is but one illustration of its general appeal. Even here a rigorous treatment of effects, such as relaxation, diffraction, and spontaneous emission, is available upon consideration of higher levels of detail and approximation.

Concurrently, our investigations into OB have characterized that phenomenon from a purely thermodynamic point of view. As a dipole-dipole pair correlation, the effective interaction is identical to the retarded dipole-dipole interaction that gives rise to cooperative radiation reaction and frequency shifts in superfluorescence [27] and is the same effective interaction responsible for the existence of the "superradiant" phase transition in thermodynamic equilibrium [11,19]. Likewise, the interaction is also identical in form to the interaction coupling Cooper pairs in the BCS theory of superconductivity [11,28]. Our results provide not only a description of OB as a first-order phase transition, but also, give conditions for OB in terms of characteristic material and cavity

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parameters, and incorporate the effects of dispersion as well as absorption. In Sections III and IV we have attempted to present certain basic aspects of this research that impact the development of the formalism and provide insight into its structure and implications.

In future research, we will increase the rigor and complexity of our present modeling applications and generalize our theoretical activity to the consideration and understanding of new systems and new and related phenomena and effects. Applications involving strong oscillating fields, multiple-level systems, a combined temporal and spacial variation, the rigorous treatment of relaxation, diffraction, spontaneous emission, etc. will be considered both from the standpoint of evaluation and implementation.

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