

Description of n -level systems in cooperative behavior

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A number of n -level systems behaving cooperatively is described by a boson-type second-quantization formalism which has both computational and conceptual utility, and leads naturally to a classical analog. The theory is compared with that for two-level systems and illustrated by the consideration of coupling to general-type fields. Spontaneous emission at all transition frequencies is examined, with both quantum-mechanical and classical results explicitly exhibited.

The properties of macroscopic matter bear, in most instances, little resemblance to the properties of its microscopic constituents. There exist, however, cases in which a collection of atomic systems exhibits on a macroscopic scale certain significant properties exhibited by the individual atomic systems on a microscopic scale. Such a collection constitutes, for instance, the matter of lasers or masers in which the pertinent atomic systems (henceforth referred to as "molecules") are characterized by several levels; both the macroscopic system and its microscopic constituents exhibit the same (relevant) resonant frequencies, have an oscillating dipole moment associated with these frequencies, and may be regarded as nonlinear oscillators. An important characteristic of this collection is the fact that degrees of freedom can be chosen so that the excitation of only a few (out of a macroscopic number) accounts for the significant macroscopic properties. Furthermore, these few degrees of freedom may be described to a good approximation classically under many conditions of interest. Aside from the obvious advantages of such a description for a macroscopic sample, the similarity between the macroscopic and microscopic properties may be used to obtain a "classical analog" for one or more molecules. It is the purpose of the present paper to develop such a description for a collection of identical n -level molecules and briefly illustrate its usefulness both computationally and conceptually.

Consider N n -level molecules, each with energy levels $\hbar\omega_i$, $i=1, 2, \dots, n$ (spaced so that no two transition frequencies are the same) and corresponding states $|\phi_i\rangle$, that are coupled to other systems in an identical manner. (The N molecules under consideration may, of course, be part of a larger group of molecules in a given experimental situation.) We introduce the formalism of second quantization for bosons,¹ using the Fock representation and the Heisenberg picture. We associate with the collection of molecules the operator

$$|\Psi\rangle_{\text{op}} = \sum_i a_i(t) |\phi_i\rangle \quad (1)$$

and the state vectors $|\bar{n}_1 \cdots \bar{n}_1 \cdots \bar{n}_n\rangle$, where

$$a_i(0) |\bar{n}_1 \cdots \bar{n}_i \cdots \bar{n}_n\rangle = \bar{n}_i^{1/2} |\bar{n}_1 \cdots \bar{n}_i - 1 \cdots \bar{n}_n\rangle \quad (2a)$$

and

$$a_i^\dagger(0) |\bar{n}_1 \cdots \bar{n}_i \cdots \bar{n}_n\rangle = (\bar{n}_i + 1)^{1/2} |\bar{n}_1 \cdots \bar{n}_i + 1 \cdots \bar{n}_n\rangle, \quad (2b)$$

the \bar{n}_i 's being non-negative integers. The a_i 's obey the commutation relationships

$$[a_i(t), a_j^\dagger(t)] = \delta_{ij}, \quad (3)$$

all other commutators vanishing. With each pair of levels, we associate a (dimensionless) "dipole moment" operator described by the three components

$$d_{ij}^{(1)} = \frac{1}{2} (a_i a_j^\dagger + a_i^\dagger a_j), \quad (4a)$$

$$d_{ij}^{(2)} = -\left(\frac{1}{2}i\right) (a_i a_j^\dagger - a_i^\dagger a_j), \quad (4b)$$

$$d_{ij}^{(3)} = \frac{1}{2} (a_j^\dagger a_i - a_i^\dagger a_j). \quad (4c)$$

The Hamiltonian of the unperturbed (or free) collection of n -level systems is given by

$$H_0 = \sum_i \hbar\omega_i a_i^\dagger a_i. \quad (5)$$

The dynamical variables a_i and a_i^\dagger are the well-known annihilation and creation operators associated with a harmonic oscillator of (angular) frequency ω_i , and (freely) oscillate, in the Heisenberg picture, with this frequency. It follows that $d_{ij}^{(1)}$ and $d_{ij}^{(2)}$ oscillate with frequency $\omega_i - \omega_j$, while $d_{ij}^{(3)}$ is constant.

We assume that the molecules couple to other systems (beginning at $t=0$) through their collective dipole moment, linearly, and that the direct coupling between them is negligible compared to the indirect coupling through the other systems. The total Hamiltonian is then given by

$$H = H_0 + \hbar \sum_m \sum_{i < j} \gamma_{ij}^{(m)} F_{ij}^{(m)} d_{ij}^{(m)} + H_1, \quad (6)$$

where $F_{ij}^{(m)}$ and H_1 refer to the other systems. Equations (3)–(6) imply that the dynamical variable K , defined by

$$K = \sum_i a_i^\dagger a_i, \quad (7)$$

is a constant of motion. From the viewpoint of second quantization (in which the molecules are the bosons)—to which we refer as Viewpoint 1— K is the number operator $_{\text{op}}\langle \psi | \psi \rangle_{\text{op}}$, and can have only one eigenvalue, N . If, however, we ignore the origin of the formalism [Eq. (1)] and consider the system to be described by Eqs. (3)–(6)—to which we refer as Viewpoint 2—then the eigenvalues of K can have any non-negative integral value. It is instructive to examine the above description for the special case of a collection of two-level systems, with levels labeled 1 and 2.

As first observed by Schwinger,²

$$d^{(m)} = l_m, \quad (8)$$

where l_m represents the three (dimensionless) components of angular momentum satisfying the commutation rules $[l_1, l_2] = i l_3$, etc. Furthermore, we obtain

$$l_1^2 + l_2^2 + l_3^2 = \frac{1}{2} K (\frac{1}{2} K + 1), \quad (9)$$

which shows that the eigenvalues of K are equal to twice the quantum numbers l of the total angular momentum. If we assume Viewpoint 2, and restrict the initial states under consideration to eigenstates of K with eigenvalues that have the same parity as N and are no larger than N , the present formalism reduces to the well-known description of a collection of N two-level systems (all exposed to the same perturbation) by means of a single angular-momentum system^{3–8} for which $l(i)$ is a measure of the collective dipole moment,^{6,7} (ii) may assume all non-negative values in integral steps with maximum value $\frac{1}{2}N$, and (iii) is equivalent to the “cooperation number” in Dicke’s theory of “superradiance”.^{4,8} Viewpoint 2, (with the above restriction of initial states) can therefore be considered a generalization of this description to one for a collection of n -level systems, with the eigenvalues of $\frac{1}{2}K$ being a generalization of l .

In the following discussion we assume Viewpoint 1, which appears as a special case in the framework of Viewpoint 2. This specialization may be regarded (in the framework of Viewpoint 2) as the one for which the similarity of behavior among the molecules (or their “cooperation”) is greatest, and the qualitative similarity between the behavior of the macroscopic sample and the individual mole-

cule is maximized. (It is also the only specialization that will allow application of the formalism to a single molecule.) All states $|\rangle$ under consideration will henceforth satisfy the relationship⁹

$$\sum_i a_i^\dagger a_i |\rangle = N |\rangle. \quad (10)$$

Equations (3)–(6) and (10) are sufficient to describe the molecular behavior (once the systems to which the collection is coupled are specified, of course). Note that the description utilizes a small number of degrees of freedom, those of n harmonic oscillators. The qualitative similarity of the macroscopic and microscopic systems is exhibited analytically by the fact that N occurs only in Eq. (10), somewhat as a normalization constant. This equation, however, has more meaning than a normalization relationship, and it is this additional meaning that must indicate any qualitative difference in behavior that may exist for different values of N . In particular, Eq. (10) implies

$$a_1^{r_1}(t) \cdots a_i^{r_i}(t) \cdots a_n^{r_n}(t) |\rangle = 0 \text{ for } \sum_j r_j > N. \quad (11)$$

The proof is given in the Appendix.

The quantum-mechanical formalism of Eqs. (3)–(6) and (10) will be applied to a specific example later. Meanwhile we want to set up a *classical* description for the same collection of N n -level systems. The technique for transforming a classical description to a quantum-mechanical description is almost as old as quantum mechanics, and consists of changing c -number dynamical variables into q -number dynamical variables, with appropriate nonvanishing commutation relationships. It is reasonable to reverse this procedure in order to convert a quantum-mechanical description into a classical description, but in order to be able to do so, the q -number dynamical variables must have c -number analogs. Thus, $n \times n$ matrices that would ordinarily be used to describe an n -level system (of arbitrary spacing) have no direct c -number analogs. The harmonic oscillator variables, however, which we have introduced through boson second quantization, do have such analogs, namely, the corresponding classical harmonic oscillator variables.¹⁰ We therefore replace the quantum-mechanical a_i 's and a_i^\dagger 's by the corresponding variables for n classical harmonic oscillators.¹¹ Equations (3)–(6) remain formally unchanged in the classical description, except that the commutator bracket of Eq. (3) must now be interpreted as i times the Poisson bracket, and Eq. (10) becomes a straightforward normalization relationship. The classical equations of motion derived from the Hamiltonian by means of Poisson

brackets are formally identical to the Heisenberg equations of motion derived by means of commutator brackets.¹⁰

It is instructive to see how the transformation from a quantum-mechanical description to a classical description corresponds to letting \hbar approach zero, that is, taking the classical limit of the quantum-mechanical description. Where the commutator of variables that become c numbers in such a transformation is proportional to \hbar , the correspondence is obvious. In the present instance, where \hbar is introduced for dimensional reasons, and the commutator does not contain \hbar explicitly, one may look at the expression for the oscillator energies $\hbar\omega_i a_i^\dagger a_i$. For a given energy, the $a_i^\dagger a_i$'s must become infinite as \hbar vanishes. Thus, as \hbar becomes very small, the number associated with a_i become very large, so that a difference of unity between $a_i a_i^\dagger$ and $a_i^\dagger a_i$ is relatively negligible, and may be ignored. We refer to the system specified by the classical description of our collection of molecules as the *classical analog*.

The classical analog of a single molecule ($N=1$) exhibits certain important properties of the molecule. Consider a Schrödinger-picture description of the molecule, $|\psi\rangle = \sum_i c_i(t) |\phi_i\rangle$, where the c_i 's are superposition constants. If the F_{ij} 's in Eq. (6) are classical variables, then, as can be shown easily, Schrödinger's equation leads to the same equations of motion for the c_i 's as those obtained in the present formalism for the a_i 's. The *expectation value* of the dipole moment of a single molecule (in suitable units) is an expression in terms of the c_i 's identical to that given by Eqs. (4) in terms of the a_i 's. Since in the usual semiclassical analysis where account is taken of the mutual interaction between the quantum-mechanical system and the classical field,¹² the quantum-mechanical dipole moment (obtained from Schrödinger's equation) is replaced by its expectation value, this expectation value is just the dipole moment of the classical analog. Such a semiclassical analysis may, therefore, be regarded as an implicit replacement of the quantum-mechanical molecule by its classical analog. However, whereas a semiclassical analysis consists of a combination of classical theory and quantum theory with an *ad hoc* prescription for combining the two (the replacement of the quantum-mechanical dipole moment by its expectation value), use of the classical analog makes possible a complete, self-consistent, dynamical theory—starting with a Hamiltonian, for instance—in terms of c numbers.¹³

We illustrate the above formalism—both quantum-mechanical and classical—by considering a collection of n -level systems for which all possible transitions couple to fields. These fields are

quite general; they may be electromagnetic fields or acoustic fields (lattice vibrations). Their essential characteristic is the fact that each field consists of a large number of denumerable modes with a range of frequencies that are closely spaced about the pertinent transition frequency. Such fields coupled to a collection of two-level systems have been considered previously.⁷ For simplicity, we set $\gamma_{ij}^{(2)} = \gamma_{ij}^{(3)} = 0$ in Eq. (6). The field coupled to a given transition will be labeled by the corresponding indices, and fields coupled to different transitions will be considered independent. (The various fields may be different sets of modes—of the same field.) We take $\omega_j > \omega_i$ for $j > i$, and set $\omega_{ij} = |\omega_i - \omega_j|$. Using the "rotating-wave" approximation,⁷ we can write in Eq. (6),

$$\gamma_{ij}^{(1)} F_{ij}^{(1)} a_{ij}^{(1)} \approx 2^{-3/2} \sum_k \gamma_k^{(ij)} (b_k^{(ij)} a_i a_j^\dagger + b_k^{(ij)\dagger} a_i^\dagger a_j), \quad (12)$$

where $b_k^{(ij)}$ and $b_k^{(ij)\dagger}$ are the annihilation and creation operators, respectively, for the k th mode of the ij th field (and transform into classical variables similarly to a_i and a_i^\dagger , respectively). It is convenient to use the "reduced" variables $A_i, B_k^{(ij)}$, defined by

$$a_i = A_i e^{-i\omega_i t}, \quad b_k^{(ij)} = B_k^{(ij)} e^{-i\omega_k^{(ij)} t}, \quad (13)$$

with $\omega_k^{(ij)}$ being the frequency of the (free) k th mode of the ij th field. (In the absence of coupling, the reduced variables are constant.) The equations of motion for the molecules become

$$\dot{A}_i = 2^{-1/2} \left(\sum_{k>i} \mathcal{G}^{(ik)\dagger} A_k - \sum_{k<i} A_k \mathcal{G}^{(ki)} \right), \quad (14a)$$

where

$$\mathcal{G}^{(ij)}(t) = \frac{1}{2} i \sum_k \gamma_k^{(ij)} B_k^{(ij)}(t) e^{-i(\omega_k^{(ij)} - \omega_{ij})t}. \quad (14b)$$

The equation of motion for each field depends only on the variables of the coupled pair of harmonic oscillators, and is similar to that treated in Ref. (7). Neglect of radiative frequency shifts and approximation based on the dense spacing of modes yields⁷

$$\mathcal{G}^{(ij)} \approx \mathcal{G}_0^{(ij)} + 2^{-1/2} \alpha_{ij} A_i^\dagger A_j, \quad (15a)$$

where

$$\alpha_{ij} = \frac{1}{4} \pi [\gamma^{(ij)}(\omega_{ij})]^2 \rho^{(ij)}(\omega_{ij}), \quad (15b)$$

$\rho^{(ij)}(\omega_{ij})$ being the density of modes at ω_{ij} of the ij field, and $[\gamma^{(ij)}(\omega_{ij})]^2$ being the average of $[\gamma_k^{(ij)}]^2$ over all modes of the ij field with frequency

$\omega_k^{(ij)}$ near ω_{ij} . $\mathcal{Q}_0^{(ij)}$ is obtained from $\mathcal{Q}^{(ij)}$ by replacing $B_k^{(ij)}(t)$ with $B_k^{(ij)}(0)$, and represents the free-field value of $\mathcal{Q}^{(ij)}$. Substitution from Eq. (15a) into Eq. (14a) gives a set of equations for the (collective) molecular variables only, since $\mathcal{Q}_0^{(ij)}$ is *prescribed*. Equations (14) and (15) are valid both classically and quantum mechanically. [Note that, quantum mechanically, $\mathcal{Q}^{(ij)}$ commutes with the (equal time) molecular variables, and $\mathcal{Q}_0^{(ij)}$ does not.]

We consider the case in which the fields are initially unexcited, and enquire about the time development of the occupation number expectation values $\langle A_i^\dagger A_i \rangle$. The initial condition with respect to the fields implies $\mathcal{Q}_0^{(ij)} = 0$, classically, and $\langle \mathcal{Q}_0^{(ij)} \rangle = \langle |\mathcal{Q}_0^{(ij)\dagger} \rangle = 0$, quantum mechanically. Equations (14) and (15) then yield

$$\langle \dot{n}_i \rangle = \sum_{k>i} \alpha_{ik} \langle n_k (n_i + \lambda) \rangle - \sum_{k<i} \alpha_{ki} \langle n_i (n_k + \lambda) \rangle, \quad (16)$$

where the summation is over k only, $n_i \equiv A_i^\dagger A_i$, λ is equal to 1 quantum mechanically and to 0 classically, and the expectation-value brackets are dropped classically. For $N=1$, Eqs. (16) assume a much simpler form quantum mechanically, since, from Eq. (11), $\langle n_i n_k \rangle$ vanishes, and only the linear terms remain. If the occupation numbers are initially well defined, Eqs. (16) yield, quantum mechanically, for $N > 1$, the (second-order) perturbation-theory relationship

$$\langle \dot{n}_i \rangle_{t=0} = \sum_{k>i} \alpha_{ik} \langle n_k(0) \rangle (\langle n_i(0) \rangle + 1) - \sum_{k<i} \alpha_{ki} \langle n_i(0) \rangle (\langle n_k(0) \rangle + 1). \quad (17)$$

Equations (17) describe spontaneous emission (or relaxation, for an acoustic field). They also exhibit the physical meaning of α_{ik} as the spontaneous emission rate—or spontaneous transition-probability rate—of a molecule from an upper occupied level (the k th) into a lower unoccupied level (the i th). Equations (16) do not constitute a set of differential equations for the quantum-mechanical $\langle n_i \rangle$'s, since, in general, $\langle n_k n_i \rangle \neq \langle n_k \rangle \langle n_i \rangle$. Only for the case $N=1$ (in which the nonlinear terms vanish) do we obtain such a set, the solution of which, for two levels, yields the same result of exponential decay as the Weisskopf-Wigner theory¹⁴ (in which only two levels are considered). It is interesting to note that only the linear terms of Eqs. (16) and (17) are contained in quantum-mechanical rate equations; the bilinear terms describe cooperative—or coherent—effects that are ignored in such equations.

The details of the solution of Eqs. (16) are beyond the scope of the present discussion. However, a few additional remarks about these equations are in order. A method for the *exact* solution of the quantum-mechanical version can be set up along the lines used in solving an angular-momentum coupled-moments equation in Ref. 7. This method requires a separate calculation for each N , and becomes increasingly complicated as N increases. In their classical version (which describes the classical analog coupled to a classical field) Eqs. (16) provide a set of nonlinear differential equations for the occupation numbers n_i . Likewise, if we *approximate* in the quantum-mechanical version by setting $\langle n_i n_k \rangle \approx \langle n_i \rangle \langle n_k \rangle$, we obtain a somewhat different set of nonlinear equations, to which we may refer as the semi-quantum-mechanical approximation⁷ (since the nonvanishing value of λ comes from commutators). These two sets differ significantly only when n_i , for $i < k$, is not larger than unity. It is reasonable to conclude that as far as spontaneous emission is concerned the classical-analog behavior approximates that of the quantum-mechanical system when n_i is larger than unity, a conclusion consistent with a more detailed examination of spontaneous emission from two-level systems in Ref. 7. One may also argue that a large value of $\langle n_i \rangle$ is associated with high quantum numbers of the i th harmonic oscillator, and justifies classical treatment according to the correspondence principle. The joining of the quantum-mechanical perturbation-theory solution (in the region where the classical solution is invalid) to the classical solution (in the region where it becomes valid) constitutes another method of approximation. Statistical properties of the quantum-mechanical solution can be retained in this method.⁷

The special case of a collection of three-level systems emitting spontaneously has been discussed in the literature.^{15,16} In Ref. 15, a group-theoretical treatment [utilizing SU(3) group properties] yields a result equivalent to the specialization of Eqs. (17) to three levels. The authors, however, use this result to produce one that is, essentially, a differential equation for $\langle n_i \rangle$ obtained by specializing Eqs. (16) to three levels and replacing $\langle n_k n_i \rangle$ by $\langle n_k \rangle \langle n_i \rangle$ (thus making the semi-quantum-mechanical approximation), without noting the approximation involved. In Ref. 16, the interaction between a three-level system and a radiation field is analyzed by semiclassical theory. The resulting equations—as is to be expected—are equivalent to the classical version of Eqs. (16) specialized to the three-level case.

Equations (14) and (15) are applicable to interactions with excited fields also, of course, and

may be used to obtain information about dipole moments as well as occupation numbers (or energies). Further study along these lines will not only yield additional information about a collection of n -level systems interacting cooperatively with fields, but will exhibit explicitly the difference between the quantum-mechanical and classical results, and indicate the extent of the usefulness of the classical analog.

APPENDIX

Consider the vector

$$|1\rangle \equiv a_1^{r_1}(t) \cdots a_i^{r_i}(t) \cdots a_n^{r_n}(t) |0\rangle,$$

where

$$\sum_i r_i = N,$$

and $|0\rangle$ is a vector satisfying the relationship

$$K(t)|0\rangle \equiv \sum_i a_i^\dagger(t) a_i(t) |0\rangle = N |0\rangle.$$

The commutation relationship of Eq. (3) yields

$$[K, a_1^{r_1} \cdots a_n^{r_n}] = - \left(\sum_i r_i \right) a_1^{r_1} \cdots a_n^{r_n},$$

so that

$$K|1\rangle = a_1^{r_1} \cdots a_n^{r_n} \left(K - \sum_i r_i \right) |0\rangle = 0,$$

and

$$\langle 1|K|1\rangle = 0.$$

Now, $\langle 1|K|1\rangle$ consists of a sum of terms, each one of which is the norm of a vector; each term must therefore vanish. For a suitable choice of r_i 's, one of these terms will be the norm of the vector

$$|2\rangle \equiv a_1^{p_1} \cdots a_i^{p_i} \cdots a_n^{p_n} |0\rangle,$$

where the p_i 's are an arbitrary set of numbers satisfying

$$\sum_i p_i = N + 1.$$

Since any vector on the left side of Eq. (11) can be written as a product of a_i 's operating on $|2\rangle$, Eq. (11) is proved.

¹See, for instance, H. A. Kramers, *Quantum Mechanics* (North-Holland, Amsterdam, 1957), p. 331, for a discussion of boson second quantization.

²J. Schwinger, in *Quantum Theory of Angular Momentum*, edited by L. C. Biedenhorn and H. Van Dam (Academic, New York, 1965).

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⁸F. T. Arecchi *et al.*, *Phys. Rev. A* **6**, 2211 (1972).

⁹Since we are using the Heisenberg picture, this is a restriction on initial states. In view of the fact that K is a constant of motion (in the Heisenberg picture), however, the same relationship is valid in the Schrödinger picture for states at any time if it is valid initially.

¹⁰The a_j 's and a_j^\dagger 's are defined in terms of the (dimensionless) coordinates q_j and momenta p_j of the respective harmonic oscillators by $a_j = 2^{-1/2}(q_j + ip_j)$, $a_j^\dagger = 2^{-1/2}(q_j - ip_j)$, where, quantum mechanically, we have $[q_i, p_i] = i$. The Hamiltonian of each oscillator is given, both classically and quantum mechanically, by $\frac{1}{2}\hbar\omega_i \times (q_i^2 + p_i^2) = \hbar\omega_i (a_i^\dagger a_i + \frac{1}{2})$. Note that \hbar occurs here for

dimensional reasons only, so that classical equations of motion obtained by means of Poisson brackets read $\dot{f}(q_j, p_j) = \hbar^{-1}\{f, H\}$, where the Poisson bracket $\{, \}$ is specified in terms of the q_j 's and p_j 's.

¹¹A classical description of the two-level case has been considered previously. In Refs. (6) and (7), it is obtained by regarding the angular-momentum variables as c numbers. In Ref. 5, the authors utilize the two-harmonic-oscillator expression for l_m of Eq. (8), assume Viewpoint 2, and approach a classical description by introducing coherent states for each harmonic oscillator. Presently, we are not concerned with the quantum states of the n -harmonic oscillators, since any state has a statistical classical counterpart and there is no need to minimize the statistics in the classical description; in this connection see A. Messiah, *Quantum Mechanics* (Wiley, New York, 1961), Ch. XII.

¹²W. E. Lamb Jr., *Phys. Rev.* **134**, A1429 (1964); see also H. Haken, *Handbuch der Physik* (Springer, Berlin, 1970), Vol. XXV/2c, p. 173.

¹³The most recent version of Jaynes's "neoclassical" theory, presented as a fundamental substitute for quantum electrodynamics by E. T. Jayne's in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), is identical to a theory in which the electromagnetic field is treated classically

and the (single) atom is replaced by its classical analog. Thus, from an orthodox quantum-mechanical viewpoint, the above version of the neoclassical theory may be regarded as the classical limit of quantum electrodynamics in which the atom is formally described by boson second quantization.

¹⁴V. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930).

¹⁵Y. C. Cho, N. A. Kurnit, and R. Gilmore, in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973).

¹⁶M. S. Crisp and E. T. Jaynes, Phys. Rev. 179, 1253 (1969).