# Quantum Statistical Theory of Superradiance. I*

R. Bonifacio and P. Schwendimann
*Istituto di Fisica, Università di Milano, Milan, Italy*

and

Fritz Haake†
*Lyman Laboratory, Harvard University, Cambridge, Massachusetts* 02138

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We discuss the cooperative decay of initial atomic excitation for a pencil-shaped active volume filled with two-level atoms. As long as the length of the sample is much smaller than a certain maximal cooperation length, the atom-field interaction producing the superradiant pulse can be treated in terms of the simplest possible laser model (single mode). The basic laser master equation turns out to be exactly solvable if specified for the superradiance limit which is characterized by two conditions: (i) The photons escape from the low-$Q$ cavity so fast that they cannot feed themselves back into atomic excitation to any appreciable amount (this no-feedback condition is equivalent to the above-mentioned requirement for the length of the sample). (ii) The incoherent atomic decay due to natural relaxation is so slow that the individual atomic dipoles do not dephase before engaging themselves cooperatively in the interaction with the electromagnetic field. The present paper presents the derivation and general discussion of the equations describing the statistical properties of atoms and field in a superradiant pulse. Analytical and numerical solutions will be presented in a subsequent paper.

## I. INTRODUCTION

The cooperative spontaneous emission of radiation by a large number of atoms, first discussed and called "superradiance" by Dicke,[1] has recently attracted new interest.[2] The motivation of these efforts has been to overcome some of the limitations of Dicke's original calculation in order to render his results more liable to experimental verification. Among the limitations recently removed from the theory of superradiance, we mention especially the use of lowest-order perturbation theory and the confinement of the atoms within a volume with linear dimensions small compared to a wavelength of the emitted radiation. In the present paper we engage ourselves in the same spirit. However, our treatment of superradiance differs from that of other authors both physically and conceptionally.

We consider an ensemble of many identical two-level atoms placed in a "cavity," the length of which is assumed to be much larger than its cross-sectional dimension. As has already been pointed out by Dicke[1(b)] the "coherence brightening" occurs practically exclusively in the axial direction for such a pencil-shaped arrangement. Emission of radiation does take place in nonaxial directions, too, but to a much lesser extent and essentially unfavored by cooperative effects.[2(c),2(g)] It may therefore be looked upon as an incoherent loss mechanism for the atoms. The latter artifice greatly reduces the complexity of the problem, for it leaves us with the axial modes only as dynamical field variables. As a device which further simplifies the problem, we quantize the electromagnetic (em) field with respect to the volume of the cavity. We show that the frequency spacing between adjacent axial modes of the pencil-shaped cavity is comfortably larger than the dispersion in frequency of the emitted superradiant pulse. As a consequence we need consider only the single resonant axial mode as interacting with the atoms. We emphasize that this would of course be impossible if we quantized the em field with respect to some large laboratory volume. The escape of photons through the nonmirrored end face of the pencil into the radiation field is taken care of as a loss mechanism for the field mode in the cavity.

What we have just described as our model of a superradiant system is nothing but the well-known simplest possible laser model. Indeed, we use the "old" laser master equation[3] as a basis for our present analysis. This frequently used equation of motion for the atom plus field-density operator accounts for the coherent interaction of $N$ two-level atoms with a resonant mode of the em field in terms of the usual interaction Hamiltonian[4] and for the irreversible photon escape, as well as for the incoherent atomic decay (e.g., radiation into other field modes) in terms of suitably chosen non-Hermitian Liouville operators. However, we have to solve the laser master equation in a limit that, for good reasons, has hardly ever been considered in laser theory. We are here concerned with a low-quality cavity. At least one of the end faces of the pencil has to be nonmirrored. In contrast to the laser, the superradiant device must not be designed to lock the photons in the cavity but rather to re-

lease them as fast as they can escape according to the velocity of light. Moreover, again as opposed to what is typical for a laser, the incoherent atomic decay process must be so slow that the individual atomic dipoles do not get out of phase with each other before they can involve themselves cooperatively in the interaction with the field mode. Making these statements (which "define" the superradiant system) quantitative in Sec. II, we find the small parameter characterizing our problem. The latter measures the rate according to which photons feed themselves back into atomic excitation in terms of their escape rate. As a first step in solving the laser master equation, we eliminate the field variables from it using Zwanzig's projector technique.[5] The justification of this procedure is the fact that the integral kernel in the resulting "generalized master equation" for the reduced atomic-density operator allows for an expansion in powers of the above-mentioned small parameter. Because the parameter is small, the series can be truncated in lowest order to yield, as our main result, a "superradiance master equation." Corresponding to its being valid for the superradiance limit, it describes a cooperative decay of initial atomic excitation without allowing for any feedback of the field on the atoms. The downward transition probability according to which the decay elapses in time is seen to be just that part of the total spontaneous emission rate of a single atom which is claimed by the diffraction solid angle of the end-fire mode.[6] If we make the (unnecessary) additional assumption that the field follows the motion of the atoms adiabatically, we may replace the superradiance master equation with its Markovian version. The latter is equivalent to the former for large times but does not account for the nonadiabatic process taking place during the first few photon transient times. It has a structure well known from the theory of spin relaxation. The equations of motion for the reduced atomic-density operator derived in Sec. III completely describe the statistical behavior of the cooperatively decaying atoms but not, of course, that of the experimentally relevant em field. By further exploiting the projector technique and again using the smallness of the above-mentioned parameter, we relate the statistics of the field to that of the atoms in Sec. IV. In particular, we arrive at a "correspondence" formula which uniquely connects normally ordered field and atomic expectation values. It shows explicitly how the em field hurries to close up with the motion of the atoms during the first few photon transient times and then begins to follow the latter adiabatically. In the adiabatic regime it can be simplified to a Markovian version, as was the case for the atomic master equation. We defer the discussion of the exact analytical and numerical solution of the superradiance master equation and the evaluation of the field statistics to a separate paper. In order to elucidate the physical content of our theory in a semiquantitative manner, we show in Sec. V that it implies the well-known superradiance rate[2(a),2(c)] equations if reduced to a quasiclassical approximation. The latter assign to the emitted pulse a hyperbolic secant form.

Finally, Sec. VI is devoted to a discussion of the limit of applicability of our considerations to light pulses generated by cooperative decay of atomic excitations. It turns out that the length of the pencil of atoms has to be small compared to a certain correlation length.

## II. LASER MASTER EQUATION

We claim here and shall show in the following that the superradiant decay of an ensemble of many excited atoms can be described realistically in terms of the simplest possible laser model. The latter accounts for a single-mode em field interacting with $N$ identical resonant two-level atoms and some suitable loss and pump mechanisms. As is usual in laser theory but has been unusual in discussions of superradiance, the em field is quantized with respect to the volume of the cavity in which the atoms are placed. This artifice is of decisive importance for our considerations in that it provides the very basis for a single-mode description of the superradiant pulse. Although the latter is a transient phenomenon of very short duration, its dispersion in frequency will turn out to be comfortably smaller than the frequency spacing between adjacent axial cavity modes. Nonaxial modes may be ruled out by giving the cavity the shape of a thin pencil, i.e., making its cross-sectional dimension very small compared to its length.

It is well known that the laser model referred to above may be treated in terms of the following master equation for the atom plus field-density operator $W(t)$:

$$\dot{W}(t) = -iLW(t), \quad L = L_A + L_F + L_{AF} + i\Lambda_A + i\Lambda_F \,. \tag{2.1}$$

The first three terms in the Liouvillian $L$ describe the free motion and the interaction of field and atoms and read explicitly

$$\begin{aligned} L_A X &= (1/\hbar)[H_A, X], \quad H_A = \sum_{i=1}^{N} \hbar\omega R_{3i} \\ L_F X &= (1/\hbar)[H_F, X], \quad H_F = \hbar\omega a^\dagger a \\ L_{AF} X &= (1/\hbar)[H_{AF}, X], \\ H_{AF} &= \hbar g \sum_{i=1}^{N} (aR_i^+ e^{ikr_i} + a^\dagger R_i^- e^{-ikr_i}) \,. \end{aligned} \tag{2.2}$$

Here $a$ and $a^\dagger$ are photon annihilation and creation operators obeying $[a, a^\dagger] = 1$; $R_i^\pm$ and $R_{i3}$ are spin operators for the $i$th atom obeying $[R_i^+, R_i^-] = 2R_{3i}$,

$[R_{3i}, R_i^{\pm}] = \pm R_i^{\pm}$, $[R_i^{\alpha}, R_j^{\beta}] = 0$ for $i \neq j$; $\omega$ is the frequency of the atomic transition and the field mode; $g$ is the atom-field coupling constant; $k$ is the wave vector of the field mode pointing in the direction of the pencil; $r_i$ is the site of the $i$th atom. Note that the interaction Hamiltonian has been specified in the dipole and rotating-wave approximations. The remaining two terms in $L$ account for irreversible processes. The field damping Liouvillian $i\Lambda_F$ represents the irreversible escape of photons through one of the end faces of the cavity. It is defined as

$$\Lambda_F X = \kappa \{[aX, a^{\dagger}] + [a, Xa^{\dagger}]\} . \tag{2.3}$$

Assuming one end face as ideally reflecting and the other as completely nonreflecting, we have

$$\kappa = c/2l . \tag{2.4}$$

$\kappa^{-1}$ is the time a photon needs to travel twice the length $l$ of the cavity. An equivalent definition of $\Lambda_F$ is

$$\mathrm{tr}_F a^{\dagger l} a^m e^{\Lambda_F t} X = e^{-(n+m)\kappa t} \mathrm{tr}_F a^{\dagger l} a^m X . \tag{2.3'}$$

The latter displays explicitly the damping character of $\Lambda_F$. Finally, the atomic pump and damping Liouvillian refers to radiative decay of individual atomic excitations sending photons into modes other than the explicitly considered one, as well as to nonradiative decay and, eventually, an incoherent pump. It is defined in terms of downward ($\gamma_{10}$) and upward ($\gamma_{01}$) transition rates as

$$\Lambda_A X = \sum_{i=1}^{N} \{ \tfrac{1}{2}\gamma_{10}[R_i^-, XR_i^+] + [R_i^- X, R_i^+] + \tfrac{1}{2}\gamma_{01}[R_i^+, XR_i^-] + [R_i^+ X, R_i^-] \} \tag{2.5}$$

or, equivalently,

$$\mathrm{tr}_A R_i^{\pm} e^{\Lambda_A t} X = e^{-t/T_2} \mathrm{tr}_A R_i^{\pm} X , \tag{2.5'}$$

$$\mathrm{tr}_A R_{3i} e^{\Lambda_A t} X = e^{-t/T_1} \mathrm{tr}_A R_{3i} X + \sigma_0 (1 - e^{-t/T_1}) \mathrm{tr}_A X ,$$

with[7]

$$2T_1 = T_2 = (\gamma_{01} + \gamma_{10})^{-1}, \quad \delta_0 = \frac{1}{2} \frac{\gamma_{01} - \gamma_{10}}{\gamma_{01} + \gamma_{10}} .$$

The master equation (2.1) assumes a somewhat simpler appearance after subjecting it to two unitary transformations. The first of those consists of going over to the interaction representation

$$\tilde{X} = e^{i(H_A + H_F)t/\hbar} X e^{-i(H_A + H_F)t/\hbar} . \tag{2.6}$$

This changes (2.1) to read

$$\dot{\tilde{W}}(t) = -i(L_{AF} + i\Lambda_A + i\Lambda_F) \tilde{W}(t) . \tag{2.7}$$

Note that the Liouvillian remains time independent in the interaction representation. This results from our taking the atoms in resonance with the field mode. The second transformation is designed to absorb the space-dependent phase factors $e^{\pm ikr_i}$ occurring in $H_{AF}$ in a redefinition of atomic states and operators:

$$R_i^{\pm}(k) = R_i^{\pm} e^{\pm ikr_i} , \quad R_{i3}(k) = R_{i3} . \tag{2.8}$$

Obviously these "phased" operators still obey angular momentum commutation rules. The corresponding transformation on the atomic states reads

$$|-\rangle_{i,k} = |-\rangle_i e^{-ikr_i}, \quad |+\rangle_{i,k} = |+\rangle_i . \tag{2.9}$$

Here the $|\pm\rangle_i$ are the spin-$\frac{1}{2}$ eigenstates $R_{i3}|\pm\rangle_i = \pm\frac{1}{2}|\pm\rangle_i$, $R_i^+|-\rangle_i = |+\rangle_i$, $R_i^+|+\rangle_i = 0$, $R_i^-|+\rangle_i = |-\rangle_i$, $R_i^-|-\rangle_i = 0$. Of course, the phased operators act on the phased states as the unphased operators on the unphased states. The master equation (2.7) keeps its form under the transformation (2.8) and (2.9), except that the factors $e^{\pm ikr_i}$ have disappeared and that all $R_i^{\alpha}$ are replaced by $R_i^{\alpha}(k)$. We have given some emphasis to the introduction of phased states and operators because it is important to keep track of the nature of the atomic states used in a discussion of superradiance. We shall elaborate on this point in more detail after solving the master equation. Meanwhile, however, in order not to overburden the notation, we drop the index $k$ as well as the tilde and write our master equation in the form

$$\dot{W}(t) = -i(L_{AF} + i\Lambda_A + i\Lambda_F) W(t) . \tag{2.10}$$

Note that a still more compact form is gained for $L_{AF}$ by expressing it in terms of the total atomic dipole moment operators

$$R^{\pm} = \sum_{i=1}^{N} R_i^{\pm}, \quad R_3 = \sum_{i=1}^{N} R_{13} \tag{2.11}$$

according to

$$H_{AF} = g(aR^+ + a^{\dagger}R^-) . \tag{2.12}$$

The master equation (2.10) has a vast variety of solutions radically different in physical content according to different initial conditions and different relative orders of magnitude of the parameters $g$, $N$, $\kappa$, $T_1$, $T_2$, and $\sigma_0$. Therefore the proper formulation of our problem requires some further specifications.

Because we want to study a cooperative decay of initial atomic excitation and the accompanying radiation of light generated by spontaneous emission, we shall be interested in the following initial condition:

$$W(0) = W_A(0) \otimes W_F(0), \quad W_F(0) = |0\rangle\langle 0|, \quad a|0\rangle = 0 . \tag{2.13}$$

That is, we assume atoms and field to be uncorrelated and the field to be in the vacuum state at $t = 0$. For the time being we need not specify the initial value of the atomic-density operator $W_A(0)$. We shall do that in a subsequent paper in conjunction with a discussion of various experimental preparation techniques. For the sake of illustration, we

shall in the following sometimes refer to a $W_A(0)$ corresponding to the atoms being either (a) all excited or (b) in a Dicke superradiant state.

In order to establish the limit in which Eq. (2.10) can be expected to describe a cooperative decay of initial atomic excitation according to the initial condition (2.13), we now have to discuss the "orders of magnitude" of each part of the Liouvillian $L = L_{AF} + i\Lambda_F + i\Lambda_A$.

A rough estimate of the order of magnitude of the atomic pump and damping Liouvillian $\Lambda_A$ may be based on a discussion of the equation $\dot{W}_A(t) = \Lambda_A W_A(t)$. Its solution $W_A(t) = e^{\Lambda_A t} W_A(0)$ may be read off the definitions (2.5) and (2.5′) of $\Lambda_A$. We see that energy is dissipated in the loss mechanism (or incoherently pumped into the atoms) at a rate $1/T_1$. On the other hand, any initial atomic polarization $\langle R^{\pm}(0)\rangle$ is damped out at a rate $1/T_2$; in other words, the loss mechanism drives the atomic "spins" out of phase with each other in a time $T_2$. Therefore, in a rough estimate $\Lambda_A$ may be said to be of order $1/T_1 \approx 1/T_2$. Similarly, an inspection of $\dot{W}(t) = \Lambda_F W_F(t)$ or $W_F(t) = e^{\Lambda_F t} W_F(0)$ using the definitions (2.3) and (2.3′) leads us to assigning to the field damping Liouvillian $\Lambda_F$ the order of magnitude $\kappa$.

Clearly, a corresponding estimate of the interaction Liouvillian $L_{AF}$ has to be based on a discussion of $\dot{W}(t) = -iL_{AF} W(t)$ or, equivalently, the Schrödinger equation $i\hbar\dot{\psi}(t) = H_{AF}\psi(t)$. In contrast to the above cases, an estimate of the order of $H_{AF}$ or $L_{AF}$ cannot simply be read from these equations. Fortunately, however, the solution of the aforementioned Schrödinger equation is known and indeed provides us with the desired estimate. $H_{AF}$ describes an oscillation of the energy back and forth between the atoms and the field mode. For the case of a large number of atoms, Bonifacio and Preparata[2(e)] have shown the frequency of this (nonlinear) oscillation to be $\sim g\sqrt{N}/\ln N$ and $\sim g\sqrt{N}$ for the initial state (2.13) with $W_A(0)$ according to (a) all atoms being excited and (b) the atoms being in a Dicke superradiant state, respectively. Clearly, we may take these values of the oscillation frequency as a rough measure for the order of magnitude of $L_{AF}$. This choice is corroborated by recent results of Scharf,[2(f)] who has calculated the eigenvalues of $H_{AF}$. For $N \gg 1$ these turn out to be nearly equidistant. This allows the general conclusion that the inverse energy difference of adjacent eigenvalues of $H_{AF}$ will be a quasiperiod of observables if only the initial state of the system is composed of sufficiently many eigenstates of $H_{AF}$. Specifically, for our initial condition (3.1) (no photons present at $t = 0$), the frequency of the oscillation thus predicted is $g\sqrt{N}/\ln N$. This is in agreement with the results of Ref. 2(e). Returning now to the master equation (2.10), we can state that each part of the Liouvillian $L$ will want to impose itself on the rate of change in time of the density operator $W(t)$ according to its order of magnitude, $\Lambda_A \sim 1/T_1 \sim 1/T_2$, $\Lambda_F \sim \kappa$, and $L_{AF} \sim g\sqrt{N}$, $g\sqrt{N}/\ln N$. If we want Eq. (2.10) to describe a cooperative decay of initial atomic excitation, it is clear that we have to require the incoherent atomic decay Liouvillian $\Lambda_A$ to be very small compared to $L_{AF}$:

$$\Lambda_A \ll L_{AF}\ , \qquad 1/T_1,\ 1/T_2 \ll g\sqrt{N},\ g\sqrt{N},\ g\sqrt{N}/\ln N. \tag{2.14a}$$

Furthermore, we stipulate that any photon, once produced, escapes from the active volume so fast that it does not have a chance to feed itself back into atomic excitation:

$$L_{AF} \ll \Lambda_F\ , \qquad g\sqrt{N},\ g\sqrt{N}/\ln N \ll \kappa\ . \tag{2.14b}$$

The physical meaning of the inequalities (2.14) will be discussed in detail in Sec. IV. As we shall see there, they constitute a necessary condition for the pencil of initially excited atoms to cooperate in the generation of a superradiant light pulse which can consistently be described in terms of our single-mode laser model. The corresponding necessary and sufficient condition which will turn out to be somewhat sharper than (2.14) will be established at the end of Sec. III and be discussed in Sec. VI also.

A quick qualitative understanding of the physical content of the master equation (2.10) and the limit (2.14) may be obtained by extracting from (2.10) classical Bloch equations and solving these in the limit (2.14). Preferring not to interrupt the quantum-mechanical argument here, we defer this classical discussion of superradiant pulses to Appendix B.

It is interesting to note that solutions of Eq. (2.10) which describe a usual stationarily operating one-mode laser are obtained in a limit just opposite to (2.14), briefly, $\Lambda_A > L_{AF} > \Lambda_F$.

## III. SUPERRADIANCE MASTER EQUATION FOR ATOMS

In a suggestive phrasing, the limit (2.14) forces the em field to follow the motion of the atoms approximately adiabatically. We therefore expect an easy way of exploiting it in terms of an approximation to consist in first eliminating the field variables from the laser master equation (2.7).

Using Zwanzig's projector formalism,[5] it is easy to derive from Eq. (2.10) a generalized master equation for the reduced atomic-density operator

$$W_A(t) = \mathrm{tr}_F\, W(t)\ . \tag{3.1}$$

As is shown in Appendix A, this equation of motion for $W_A(t)$ reads

$$\dot{W}_A(t) = \Lambda_A W_A(t) + \int_0^t ds\, K(s)\, W_A(t-s)\,, \qquad (3.2)$$

with the integral kernel

$$K(s) = -\mathrm{tr}_F L_{AF} e^{-i(1-P)(L_{AF}+i\Lambda_A+i\Lambda_F)s} L_{AF} |0\rangle\langle 0|\,. \qquad (3.3)$$

Equation (3.2) is equivalent to the laser master equation (2.10) with respect to the atomic variables. It is merely a projection of the latter into the Hilbert space of the atoms carried out with the help of the projector

$$P = |0\rangle\langle 0|\,\mathrm{tr}_F, \quad a|0\rangle = 0, \quad P(1-P) = 0 \qquad (3.4)$$

and the initial condition (2.13), $W(0) = |0\rangle\langle 0|\, W_A(0)$. Note that the definition (3.4) entails the algebraic identities

$$\Lambda_F P = P\Lambda_F = 0, \quad \Lambda_A P = P\Lambda_A\,, \qquad P(L_{AF})^{2n+1}P = 0, \quad n = 0, 1, 2, \ldots \qquad (3.5)$$

which we shall have to use below.

The main virtue of the generalized master equation (3.2) for the atomic-density operator is that it invites, by its very appearance, certain approximations which will finally render it solvable. First, let us show that the limit (2.14) allows the neglect of $\Lambda_A$ and $L_{AF}$ with respect to $\Lambda_F$ in the exponent in (3.3). This is obvious from an intuitive point of view, since (2.14) implies $\Lambda_A, L_{AF} \ll \Lambda_F$ (i.e., $1/T_1$, $1/T_2$, $g\sqrt{N} \ll \kappa$). For the sake of saving space, we demonstrate in the following that correction terms due to $L_{AF}$ are negligible, and we suppress the completely analogous discussion of corrections due to $\Lambda_A$. To that end we discard $\Lambda_A$ in $K(s)$ and represent the kernel as a power series[8] in terms of the interaction Liouvillian $L_{AF}$:

$$K(s) = \sum_{n=0}^{\infty} K_{2n}(s), \quad K_0(s) = -\mathrm{tr}_F L_{AF} U(s) L_{AF} |0\rangle\langle 0|\,,$$

$$K_{2n}(s) = (-1)^{n+1} \int_0^s ds_1 \int_0^{s_1} ds_2 \cdots \int_0^{s_{2n-1}} ds_{2n}\, \mathrm{tr}_F L_{AF} U(s-s_1) L_{AF} U(s_1-s_2) Q L_{AF} U(s_2-s_3) L_{AF} U(s_3-s_4) Q$$
$$\times \cdots Q L_{AF} U(s_{2n-2}-s_{2n-1}) L_{AF} U(s_{2n-1}-s_{2n}) Q L_{AF} U(s_{2n}) L_{AF} |0\rangle\langle 0|\,, \qquad (3.6)$$

with

$$Q = 1 - P, \quad U(s) = e^{\Lambda_F s}\,. \qquad (3.7)$$

Here we have already used the algebraic identities (3.5), the last of which entails the expansion of $K(s)$ to contain only even-order terms in $L_{AF}$. Let us discuss the order of magnitude of $K_{2n}(s)$. We assert

$$K_{2n}(s) \sim (g\sqrt{N}/\kappa)^{2n} K_0(s)\,. \qquad (3.8)$$

This can be seen as follows. According to its definition, $K_{2n}(s) \sim (L_{AF})^{2n+2}$; the order of magnitude of $L_{AF}$ has been pointed out to be $\sim g\sqrt{N}$ to within a factor of $1/\ln N$; this explains the numerator in (3.8). Next, recall that the field-damping propagator $U(s)$, according to the definitions (2.3) and (2.3′) of $\Lambda_F$, varies in time on a scale $\kappa^{-1}$. Roughly,[9] $U(s) \sim e^{-\kappa s}$. Consequently, the $2n$-fold time integral in (3.6) provides $K_{2n}(s)$ with a factor $\kappa^{-2n}$. We now see that the expansion (3.6), although obtained as a power series in the interaction Liouvillian, actually goes in terms of the parameter $(g\sqrt{N}/\kappa)^2$ [or the still smaller one $(g\sqrt{N}/\kappa \ln N)^2$]. The latter has an obvious physical meaning: It measures the rate at which photons are converted back into atomic excitation energy ($g\sqrt{N}$ or $g\sqrt{N}/\ln N$) relative to the rate at which photons escape from the cavity. As stated in (2.14) this parameter is assumed to be very small compared to unity. In contrast to the laser the superradiant laser releases the photons so fast that they cannot feed themselves back into the atoms to any appreciable amount. We may therefore safely restrict our consideration to the lowest-order term in the expansion (3.6):

$$K(s) = K_0(s) + O(g^2 N/\kappa^2). \qquad (3.9)$$

The generalized master equation (3.2) for the atomic-density operator is then found to read

$$\dot{W}_A(t) = \Lambda_A W_A(t) + \int_0^t ds\, \kappa e^{-\kappa s} \Lambda_s W_A(t-s), \qquad (3.10)$$

with the "superradiant decay Liouvillian" $\Lambda_s$

$$\Lambda_s X = (g^2/\kappa)\{[R^-, XR^+] + [R^- X, R^+]\}\,. \qquad (3.11)$$

Equations (3.10) and (3.11) constitute the main result of this section. We emphasize that Eq. (3.10) is an "exact" consequence of the laser master equation (2.1) in the superradiance limit (2.14). It evidently is a non-Markovian equation accounting for memory effects. We shall show below that the solution $W_A(t)$ of (3.10) changes in time on a scale much larger than the photon transient time $\kappa^{-1}$. Therefore, apart from what happens for times $t \lesssim \kappa^{-1}$ (in the nonadiabatic regime), Eq. (3.10) is equivalent to its Markovian approximation[10]

$$\dot{W}_A(t) = (\Lambda_A + \Lambda_s) W_A(t), \quad t \gg \kappa^{-1}\,. \qquad (3.12)$$

This corresponds to the field following the motion of the atoms adiabatically.

In order to get some insight into the physical content of Eqs. (3.10)–(3.12) let us compare the incoherent decay Liouvillian $\Lambda_A$ and the superradiant decay Liouvillan $\Lambda_s$. As is seen from their

respective definitions, (2.5) and (3.11), they have a formally similar structure. The similarity becomes especially striking if, for the time being, we imagine them both specialized to the case of a single atom. $\Lambda_A$ accounts for upward and downward transitions at the rates $\gamma_{01}$ and $\gamma_{10}$, respectively. In $\Lambda_s$, on the other hand, there is no upward transition term whereas downward transitions do appear at the rate

$$I_1 = 2g^2/\kappa \ . \tag{3.13}$$

Clearly, upward transitions are forbidden in the superradiance limit (2.14). Note that the downward transition probabilities $\gamma_{10}$ and $I_1$ are not independent physically. As mentioned in Sec. II, $\gamma_{10}$ may have contributions from radiative as well as nonradiative decay mechanisms. The former can be calculated with Fermi's Golden Rule:

$$\begin{aligned}\gamma_{10}^{(\mathrm{rad})} &= 2g^2 \times (\text{density of modes in the cavity}) \\ &= 2g^2(4\pi\omega^2/c^3)\, s^2 l \\ &= I_1(4\pi s^2/\lambda^2) \ ,\end{aligned} \tag{3.14}$$

where $l$ and $s^2$ are the length and the end surface of the cavity. We see that $I_1$ is the photon emission probability per unit solid angle $\gamma_{10}^{(\mathrm{rad})}/4\pi$ times the diffraction solid angle $\lambda^2/s^2$ of the "end-fire" mode under consideration.[6]

However, in spite of these similarities, $\Lambda_A$ and $\Lambda_s$ describe radically different decay mechanisms for $N \gg 1$. The decay referred to by $\Lambda_s$ goes "coherently," i.e., in terms of the macroscopic dipole moment operators $R^{\pm}$, with the total dipole moment $R^2 = R_1^2 + R_2^2 + R_3^2$ ($R^{\pm} = R_1 \pm iR_2$) being a constant of the motion. On the other hand, $\Lambda_A$ describes an incoherent decay, i.e., each atom moves independently of all the others. This difference between $\Lambda_A$ and $\Lambda_s$ may also be phrased as the statement that $\Lambda_s$, in contrast to $\Lambda_A$, does not allow for an "uncorrelated" solution $W_A(t)$, i.e., a $W_A(t)$ factorizing with respect to the individual atoms. Indeed, we shall show when presenting the exact solution of the superradiant master equations that strong atom-atom correlations are characteristic for the superradiant process.

Let us emphasize that we have not yet taken full advantage of the "superradiance limit" (2.14) in simplifying Eqs. (3.2) and (3.3) to the forms (3.10) and (3.11) or to the Markovian version (3.12). We have used only $\Lambda_A$, $L_{AF} \ll \Lambda_F$, i.e., $1/T_1$, $1/T_2$, $g\sqrt{N} \ll \kappa$. Consequently, the solutions of (3.10) and (3.12) will in general not describe a cooperative decay of initial atomic excitation. This is immediately obvious from the appearance of these equations. They allow for two competing decay mechanisms, an incoherent ($\Lambda_A$) and a cooperative one ($\Lambda_s$). Clearly, we have to require the latter to dominate the former. The limit (2.14) states a necessary condition for this to be the case. We are now in a situation to state the sufficient condition: $\Lambda_A \ll \Lambda_s$. Recalling that $\Lambda_s$ is constructed (schematically) as $\int_0^\infty ds\, L_{AF} e^{\Lambda_F s} L_{AF}$ and using the orders of magnitude of $\Lambda_A$, $L_{AF}$, and $\Lambda_F$ we get $\Lambda_s \approx (g\sqrt{N})^2/\kappa$. Then $\Lambda_A \ll \Lambda_s$ may be read as

$$1/T_1, \ 1/T_2 \ll (g\sqrt{N})^2/\kappa \ . \tag{3.15}$$

This "sufficient" version of the superradiance limit will be discussed in detail in Sec. VI. It clearly allows neglecting the incoherent-decay term $\Lambda_A$ in (3.10) and (3.12). The thus simplified equations will be solved in a subsequent paper.

## IV. SUPERRADIANT FIELD STATISTICS

The master equations (3.10) and (3.12) derived above completely describe the statistical behavior of the atoms in a superradiant laser. Unfortunately, however, the latter are not directly observable laser experiments always referring to the em field output.[11] Thus we are left with the task of relating the field statistics to those of the atoms. This goal can be achieved by further exploiting the projector formalism. As is shown in Appendix A, the field-density operator $W_F(t)$ may be expressed in terms of the atomic-density operator $W_A(t)$ as

$$W_F(t) = V_F |0\rangle\langle 0| \ , \tag{4.1}$$

$$\begin{aligned}V_F(t) = 1 + \int_0^t ds\, \mathrm{tr}_A \exp[-i(1-P)(L_{AF} + i\Lambda_A + i\Lambda_F)s] \\ \times [-i(1-P)(L_{AF} + i\Lambda_A)]\, W_A(t-s) \ .\end{aligned} \tag{4.2}$$

Constructing $W_F(t)$ is equivalent to calculating all normally ordered field expectation values

$$\langle a^{\dagger l} a^m(t)\rangle = \mathrm{tr}_F\, a^{\dagger l} a^m W_F(t). \tag{4.3}$$

The evaluation of $\langle a^{\dagger l} a^m(t)\rangle$ has to be carried out, of course, for the sake of consistency, in the same limit (2.14) in which we have determined the atomic-density operator $W_A(t)$. Hence we may first neglect $\Lambda_A$ in the field "propagator" $V_F(t)$. Next, we expand $V_F(t)$ in terms of our small parameter $g\sqrt{N}/\kappa$. As in Sec. III, the series desired is formally obtained by developing the exponential $\exp[-i(1-P)(L_{AF} + i\Lambda_F)s]$ in powers of $L_{AF}$:

$$\begin{aligned}\langle a^{\dagger l} a^m(t)\rangle = \sum_{n=0}^{\infty} (-i)^{n+1} \int_0^t ds \int_0^s ds_n \int_0^{s_n} ds_{n-1} \cdots \int_0^{s_2} ds_1\, \mathrm{tr}_A \mathrm{tr}_F\, a^{\dagger l} a^m \\ \times U(s-s_n)\, QL_{AF}\, U(s_n - s_{n-1})\, QL_{AF} \cdots U(s_2 - s_1)\, QL_{AF}\, U(s_1)\, QL_{AF} |0\rangle\langle 0|\, W_A(t-s) \ .\end{aligned} \tag{4.4}$$

Here we have again used the abbreviations (3.7). Note that the first nonvanishing term in the above series arises for $n+1=l+m$. This can be seen as follows. Disregard, for the time being, the factors $U(s_\mu - s_{\mu-1})Q$, which do not affect the argument; we shall concentrate on them later. Now consider the $n$th term of the series which contains $n+1$ factors $L_{AF}$. Use the cyclic invariance of the full trace $\mathrm{tr}_A \mathrm{tr}_F$ to let all $L_{AF}$ act to the left. Commuting $a^{\dagger l}a^m$ $(n+1)$ times with $H_{AF}$ gives a polynomial in $a^\dagger$, $a$, with all exponents in $a^{\dagger i}a^j$ obeying $i+j \geq (l+m)-(n+1)$ because $[a, f(a,a^\dagger)] = (\partial/\partial a^\dagger)f(a,a^\dagger)$; if $n+1 < l+m$, all terms $a^{\dagger l}a^j$ give zero contribution to the final vacuum expectation value $\mathrm{tr}_F(\cdots)|0\rangle\langle 0|$; if, however, $n+1 \geq l+m$, terms with $a^{\dagger 0}a^0$ arise whose vacuum expectation values do not vanish; therefore we have

$$\sum_{n=0}^{\infty} \to \sum_{n=l+m-1}^{\infty} \tag{4.5}$$

in (4.4). Recall now that the expansion (4.4) goes in terms of the small parameter $g\sqrt{N}/\kappa$. Therefore we may, and indeed have to, neglect all terms with $n+1 > l+m$:

$$\langle a^{\dagger l}a^m(t)\rangle = (-i)^{l+m}\int_0^t ds \int_0^s ds_{l+m-1}\int_0^{s_{l+m-1}} ds_{l+m-2}\cdots\int_0^{s_2} ds_1$$
$$\times \mathrm{tr}_A \mathrm{tr}_F\, a^{\dagger l}a^m U(s-s_{l+m-1})QL_{AF}U(s_{l+m-1}-s_{l+m-2})\cdots U(s_1)QL_{AF}|0\rangle\langle 0|W_A(t-s)\ . \tag{4.6}$$

Applying again the argument used above to prove (4.5), we see that we may replace all factors $Q=1-P$ in (4.6) with unity [this would not be possible for the higher-order terms in (4.4)]. The field-damping propagators are then taken care of step by step in the following way: The first of them on the left, $U(s-s_{l+m-1})$, is replaced with $\exp[-(l+m)\kappa(s-s_{l+m-1})]$ according to its definition (2.3′). We then let the adjacent $L_{AF}$ act to the left. The resulting commutator $[a^{\dagger l}a^m, H_{AF}]$ is a polynomial in $a^\dagger, a$ which has to be put in normal order; then again on account of (2.3′), $U(s_{l+m-1}-s_{l+m-2})$ gives place to $\exp[-(l+m-1)\kappa(s_{l+m-1}-s_{\kappa+m-2})]$ and so forth. Note that we may omit in each step all those $a^{\dagger i}a^j$ which do not have a chance of getting commuted to field $c$ numbers in the remaining steps. On combining all arising factors $\exp[-\mu\kappa(s_\mu - s_{\mu-1})]$, we get

$$\langle a^{\dagger l}a^m(t)\rangle = (-i)^{l+m}\int_0^t ds\int_0^s ds_{l+m-1}\cdots\int_0^{s_2} ds_1$$
$$\times \mathrm{tr}_A K_{l+m} W_A(t-s)\exp[-\kappa(l+m)s]$$
$$\times \exp[\kappa(s_{l+m-1}+s_{l+m-2}\cdots+s_2+s_1)], \tag{4.7}$$

with

$$K_{l+m} = \mathrm{tr}_F[[[\cdots[a^{\dagger l}a^m, H_{AF}], H_{AF}], \cdots], H_{AF}]|0\rangle\langle 0|$$
$$= \langle 0|[[[\cdots[a^{\dagger l}a^m, H_{AF}], H_{AF}], \cdots], H_{AF}]|0\rangle,$$
$$l+m \text{ commutators}\ . \tag{4.8}$$

Carrying out the $(l+m-1)$-fold integral over the $s_\mu$, we get

$$\langle a^{\dagger l}a^m(t)\rangle = \frac{(-i)^{l+m}}{(l+m-1)!\,\kappa^{l+m-1}}\int_0^t ds$$
$$\times e^{-(l+m)\kappa s}(e^{\kappa s}-1)^{l+m-1}\mathrm{tr}_A K_{l+m} W_A(t-s)\ . \tag{4.9}$$

The evaluation of the operator $K_{l+m}$ requires the calculation of a vacuum expectation value of a polynomial in $a^\dagger, a$. Some straightforward algebraic steps yield

$$\mathrm{tr}_A K_{l+m} W_A(t-s) = (-i)^l (l+m)!$$
$$\times g^{l+m}\mathrm{tr}_A R^{+l}R^{-m}W_A(t-s)\ , \tag{4.10}$$

whereupon we get our final result:

$$\langle a^{\dagger l}a^m(t)\rangle = \left(-i\frac{g}{\kappa}\right)^{l+m}(-i)^l(l+m)$$
$$\times\int_0^t ds\,\kappa e^{-(l+m)\kappa s}(e^{\kappa s}-1)^{l+m-1}\langle R^{+l}R^{-m}(t-s)\rangle\ . \tag{4.11}$$

This allows the calculation of field expectation values once the corresponding atomic expectation values are known. Note the one-to-one correspondence between normally ordered field and atomic expectation values.

Equation (4.11) is the counterpart of the non-Markovian master equation (3.10). Insofar as the latter is equivalent to its Markovian version (3.12), Eq. (4.11) may also be simplified in a completely analogous manner. If $\langle R^{+l}R^{-m}(t)\rangle$ relaxes much more slowly than its cofactor in the integrand in (4.11), its dependence on the integration variable may be neglected and the upper integration limit extended to infinity. This gives the counterpart of the Markovian equation (3.12):

$$\langle a^{\dagger l}a^m(t)\rangle = (-ig/\kappa)^{l+m}(-1)^l\langle R^{+l}R^{-m}(t)\rangle,\quad t \gg \kappa^{-1}\ . \tag{4.12}$$

This expression clearly exhibits the additional assumption that the field follows the motion of the atoms adiabatically.[12] Let us emphasize that the Markovian theory constituted by (3.12) and (4.12) does not account for what happens during the first few photon transient times $t \lesssim \kappa^{-1}$. As we shall see when presenting the solutions of the master equa-

tions (3.10) and (3.12), the nonadiabatic process taking place for $t \lesssim \kappa^{-1}$ is physically nondramatic and, moreover, in view of the smallness of $\kappa^{-1}$, extremely difficult, if not impossible, to observe experimentally. Note, however, that there is one difference between the non-Markovian expression (4.11) and the Markovian one (4.12) which is indeed striking from the conceptional point of view. The former properly reflects our initial condition: At $t=0$ the field is assumed to be in the vacuum whereupon all normally ordered field expectation values have to be, and indeed come out as, zero. On the other hand, (4.12) assigns to the $\langle a^{\dagger l} a^m \rangle$ initial values which result from (4.11) for a time of the order of $\kappa^{-1}$.

## V. SUPERRADIANCE RATE EQUATIONS

Because a thorough discussion of the solutions of the superradiance master equations (3.10) and (3.12) and their physical implications would require quite some labor as well as space, we defer it to a separate paper. A semiquantitative insight into the underlying physics may, however, quickly be obtained by treating Eq. (3.12) in a quasiclassical approximation. The latter amounts to neglecting the quantum-mechanical dispersion of the total inversion operator $R_3$:

$$\langle (R_3)^2 \rangle - (\langle R_3 \rangle)^2 = 0 \ . \tag{5.1}$$

Let us write down the equations of motion for the radiated intensity

$$I(t) = 2\kappa \langle a^\dagger a(t) \rangle = (2g^2/\kappa) \langle R^+ R^-(t) \rangle = I_1 \langle R^+ R^-(t) \rangle \tag{5.2}$$

and for $\langle R_3(t) \rangle$ which follow from Eqs. (3.12) and (5.1):

$$\langle \dot{R}_3(t) \rangle = I(t) - (1/T_1)[\langle R_3(t) \rangle + \tfrac{1}{2}N] \ , \tag{5.3}$$

$$\dot{I}(t) = 2I_1 I(t)[\langle R_3(t) \rangle - R_{3T}] \ , \tag{5.4}$$

with $R_{3T} = \frac{1}{2} + \kappa/2g^2 T_2$ .

For simplicity we have assumed that the atomic pump and loss mechanism described by $\Lambda_A$ is at zero temperature ($\gamma_{01}=0$). Commenting on the above superradiance rate equations[2(a), 2(c)] let us make the following remarks.

(i) Their main difference from the well-known laser rate equations consists in the absence of a back-reaction term $-2I_1 I(t) \langle R_3(t) \rangle$ in (5.3). This is a consequence of our condition $\kappa \gg g\sqrt{N}$ (or $g\sqrt{N}/\ln N$), which we have already phrased as a no-feedback condition.

(ii) Equation (5.3) describes the decay of atomic excitation according to radiation into the end-fire mode $[-I(t)]$ and natural relaxation (i.e., especially radiation into other modes of the em field). Note that it fails to be a balance equation for the total internal energy $\langle R_3 \rangle + \langle a^\dagger a \rangle$ by the absence of a term $d/dt \langle a^\dagger a(t) \rangle$ on the left-hand side; such an exact energy balance is indeed entailed by the original laser master equations, (2.1) and (2.10), which do not yet have the superradiance limit (2.14) built in; however, we have

$$\left| \frac{d}{dt} \langle a^\dagger a \rangle \right| \ll 2\kappa \langle a^\dagger a \rangle = I(t) \ ,$$

if the characteristic time $\tau_p$ during which the superradiant process takes place, fulfills $\tau_p \gg \kappa^{-1}$. The latter condition will turn out to be equivalent to the superradiant-limit condition. Therefore Eq. (5.3) may indeed be looked upon as the exact energy balance for the superradiant limit.

Let us now use the limit (3.15), i.e., require the superradiant pulse elapse in a time very short compared to the "natural" atomic relaxation times. We then have to look for a solution of the rate equations (5.3) and (5.4) for $1/T_1,\ 1/T_2 = 0$. In this case it is easily shown that the pulse has the well-known hyperbolic secant form

$$\langle R_3(t) \rangle = \frac{1}{2} - \frac{N+1}{2} \tanh \frac{t - t_m}{\tau_p} \ ,$$
$$I(t) = I_1 \left( \frac{N+1}{2} \right)^2 \operatorname{sech} \frac{t - t_m}{\tau_p} \ . \tag{5.5}$$

The width $\tau_p$ of the pulse is given by

$$\tau_p = \kappa / g^2 (N+1) \ . \tag{5.6}$$

The time $t_m$ at which the pulse reaches its (superradiant) maximum intensity $I_1[\frac{1}{2}(N+1)]^2$ depends on the initial condition. For the fully excited initial state

$$\langle R_3(0) \rangle = \tfrac{1}{2}N, \quad I(0) = I_1 \langle R^+ R^-(0) \rangle = I_1 N,$$

we find

$$t_m = \tfrac{1}{2} \tau_p \ln N \ , \tag{5.7a}$$

and for the Dicke superradiant initial state

$$\langle R_3(0) \rangle = 0 \ ,$$
$$I(0) = I_1 \langle R^+ R^-(0) \rangle = I_1 \tfrac{1}{2}N (\tfrac{1}{2}N + 1) \ , \tag{5.7b}$$
$$t_m = -\tau_p \ln[(N+2)/N] \approx -\tau_p / N \ .$$

As will be shown in the subsequent paper, the shape of the radiated intensity $I(t)$ obtained in this section is qualitatively correct. In particular, the characteristic times $\tau_p$ and $\tau_m$ will turn out to be in agreement with the corresponding values obtained by solving the superradiance master equation exactly. It is only the statistical features of the emission process which have to be expected and indeed turn out to be falsified by the present quasiclassical treatment which suppresses the quantum fluctuations of the atomic energy [see (5.1)].

It is interesting to see that the characteristic times $\tau_p$ and $t_m$ of the emission processes could

have been determined correctly by an order-of-magnitude argument based on the discussion at the end of Sec. II. Recall that the superradiant decay Liouvillian $\Lambda_s$ was constructed as $\int_0^\infty ds\, L_{AF} \times e^{\Lambda_F s} L_{AF}$ [see (3.6), (3.9), and (3.10)]. According to the order of magnitude of $\Lambda_F$ and $L_{AF}$ it should therefore be of order $(g\sqrt{N})^2/\kappa$ (to within a factor of $\ln N$). This is indeed the inverse width $\tau_p$ of the pulse given by (5.6).

## VI. LIMIT OF VALIDITY

Let us finally return to the discussion of the superradiance limits (2.14) and (3.15). In anticipation of what will be shown presently, we have already stated this limit to be the necessary [(2.14)] and sufficient [(2.14) and (3.15)] condition (a) for the cooperation of the atoms in generating the superradiant pulse to be unimpeded by natural relaxation and (b) for the emitted pulse to be consistently describable in terms of our single-mode model. To prove that now let us first combine (2.14) and (3.15) to obtain

$$\Lambda_A \ll \Lambda_s \ll \Lambda_F \,, \qquad 1/T_1,\ 1/T_2 \ll g^2 N/\kappa \ll \kappa \,, \tag{6.1}$$

where the order of magnitude of $\Lambda_s$ is specified up to factors $\ln N$. Upon dimensional arguments, the magnitude $g^2N/\kappa$ of $\Lambda_s$ is, at least roughly, the inverse characteristic time $\tau_p^{-1}$ of the cooperative decay described by $\Lambda_s$. This is corroborated by the result (5.6) of the semiclassical analysis in Sec. V. Thus the left-hand inequality in (6.1) may be read as $\tau_p \ll T_1,\ T_2$, which is indeed statement (a) above: Natural atomic relaxation is not allowed to drive the individual atoms out of phase with each other before the cooperative decay is over. Next, we observe that the right-hand inequality in (6.1) implies that the width of the frequency spectrum of the emitted pulse ($\sim 2\pi/\tau_p$) is much smaller than the frequency spacing of adjacent axial modes of the cavity ($2\pi c/l$). This is readily seen using $\kappa = c/2l$. Equivalently, $\kappa = c/2l \gg \tau_p^{-1}$ may be read as $l \ll c\tau_p$, i.e., the length $l$ of the sample is required to be small compared to the spatial extension of the radiated pulse. Thus the envelope of the pulse is essentially constant over the length of the sample. This shows again that the right-hand inequality in (6.1) may be understood as ensuring that we need not worry about propagation of the pulse along the active volume (i.e., many-mode effects).

Let us rewrite the superradiance limit (6.1) in terms of measurable quantities. To that end we express the coupling constant $g$ in terms of the natural linewidth of an isolated atom given by (3.14):

$$\gamma_{10}^{(\mathrm{rad})} \equiv \gamma = 2g^2 4\pi V/c\lambda^2 \,. \tag{6.2}$$

Assuming a constant density $\rho = N/V$ of atoms in the active volume and using $\kappa = c/2l$, we may read (6.1) as a condition for the length $l$ of the sample:

$$l_{\mathrm{thr}} \ll l \ll l_c \,. \tag{6.3}$$

Here the upper (cooperation) limit $l_c$ is given by

$$l_c = (2\pi c/\gamma\lambda^2\rho)^{1/2} \tag{6.4}$$

and the lower (threshold) limit[13] $l_{\mathrm{thr}}$ by

$$l_{\mathrm{thr}} = \frac{4\pi(\gamma_\perp/\gamma)}{\lambda^2\rho} \,, \quad \text{with } \gamma_\perp \equiv \frac{1}{T_2} \approx \frac{1}{T_1} \,. \tag{6.5}$$

The labels "threshold" and "cooperative" indicate the physical meaning of the corresponding quantities. $l \ll l_c = c\tau_p$ ensures that the envelope of the emitted pulse varies slowly over the length of the sample and, consequently, that the atoms at one end of the cavity see the same field as those at the other end ("no propagation").[14] $l \gg l_{\mathrm{thr}}$ stems from the left-hand inequality in (6.1). Since it is well known that $\kappa\gamma/g^2$ is the threshold inversion of a laser, this can be read as the requirement that our system is very far above threshold at $t = 0$.

Multiplying (6.3) with the density $\rho$ and the cross section $s^2$ of the sample, we get an equivalent condition for the number of atoms $N$:

$$N_{\mathrm{thr}} \ll N \ll N_c \,, \tag{6.6}$$

with[14]

$$N_{\mathrm{thr}} = 4\pi \frac{\gamma_\perp}{\gamma} \frac{s^2}{\lambda^2}, \quad N_c = \frac{s^2}{\lambda}\left(\frac{2\pi c\rho}{\gamma}\right)^{1/2} \,. \tag{6.7}$$

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## APPENDIX A

For the sake of completeness we give a brief outline of the projector technique. For a thorough account the reader is referred to Zwanzig's work.[5]

Let the density operator $W(t)$ of some system obey the equation of motion

$$\dot{W}(t) = -iLW(t) \,. \tag{A1}$$

$L$ is an operator acting in the Hilbert space to which $W(t)$ belongs. In this space the scalar product is defined as

$$(W_1, W_2) = \mathrm{tr} W_1^* W_2 \,. \tag{A2}$$

If the system under consideration is closed, its dynamics can be described in terms of a Hamiltonian $H$ and $L$ has the Liouville–von Neumann form

$$LX = (1/\hbar)[H, X] \,. \tag{A3}$$

Then $L$ is Hermitian with respect to the scalar

product (A2), corresponding to (A1) having time-reversal symmetry. However, $L$ can also be, and indeed will be, non-Hermitian if the system described by $W(t)$ is open and therefore undergoes an irreversible motion.

Suppose now that $W(t)$ can be split into a "relevant" and an "irrelevant" part with the help of a projection operator

$$W_r = PW, \quad W_i = (1-P)W, \quad W = W_r + W_i ,$$
$$P^2 = P \quad \text{or} \quad P(1-P) = 0 . \tag{A4}$$

We may then obtain from (A1) two coupled equations of motion for $W_r(t)$ and $W_i(t)$:

$$\dot{W}_r(t) = -iPLW(t) = -iPLW_r(t) - iPLW_i(t) , \tag{A5a}$$

$$\dot{W}_i(t) = -i(1-P)LW(t)$$
$$= -i(1-P)LW_r(t) - i(1-P)LW_i(t) . \tag{A5b}$$

Formally integrating (A5b) we express the irrelevant part in terms of the relevant part:

$$W_i(t) = e^{-i(1-P)Lt}W_i(0) - i\int_0^t ds\, e^{-i(1-P)Ls}(1-P)LW_r(t-s) . \tag{A6}$$

Inserting this into (A5a) we get Zwanzig's "generalized master equation" for the relevant part:

$$\dot{W}_r(t) = -iPLW_r(t) - \int_0^t ds\, PLe^{-i(1-P)Ls}(1-P)LW_r(t-s) - iPLe^{-i(1-P)Lt}W_i(0) . \tag{A7}$$

The irrelevant part is eliminated except for the occurrence of its initial value, which formally gives rise to an inhomogeneity.

Adapting the above to our needs, we identify Eq. (A1) with our atom-field master equation (2.10). We want the generalized master equation (A7) to become an equation of motion for the reduced atomic-density operator $W_A(t) = \mathrm{tr}_F W(t)$. This is achieved by defining the projector $P$ as

$$P = F\,\mathrm{tr}_F, \quad \mathrm{tr}_F F = 1 . \tag{A8}$$

Indeed, after applying the partial trace $\mathrm{tr}_F$ to (A7) we get

$$\dot{W}_A(t) = -i\,\mathrm{tr}_F LFW_A(t) - \int_0^t ds\,\mathrm{tr}_F Le^{-i(1-P)Ls}(1-P)LFW_A(t-s) - i\,\mathrm{tr}_F Le^{-i(1-P)Lt}(1-P)W(0) . \tag{A9}$$

We fix the "parameter" $F$ in $P$ so that the inhomogeneity in (A9) vanishes identically. In view of our initial condition $W(0) = |0\rangle\langle 0|\, W_A(0)$, we simply have to put

$$F = |0\rangle\langle 0|, \quad P = |0\rangle\langle 0|\,\mathrm{tr}_F . \tag{A10}$$

Identifying now $L$ with $L_{AF} + i\Lambda_A + i\Lambda_F$ as in (2.10), and using the algebraic identities following from the definitions (A8) and (A10) of $P$,

$$P\Lambda_A = \Lambda_A P, \quad P\Lambda_F = \Lambda_F P = 0,$$
$$P(L_{AF})^{2n+1}P, \quad n = 0, 1, 2, \ldots \tag{A11}$$

we bring (A9) to the form (3.2).

Suppose we have solved for $W_A(t)$ and want to construct the field-density operator $W_F(t) = \mathrm{tr}_A W(t)$. This can be done using the formal integral (A6) for the irrelevant part:

$$W_F(t) = \mathrm{tr}_A[PW(t) + (1-P)W(t)]$$
$$= |0\rangle\langle 0| + \mathrm{tr}_A(1-P)W(t) . \tag{A12}$$

Inserting (A6) and using again $(1-P)W(0) = 0$ and the identities (A11), we at once arrive at (4.1).

## APPENDIX B

For the sake of comparison and in order to facilitate a qualitative visualization of the preceding arguments, let us briefly discuss a classical treatment of the superradiant pulse. This may be based on the original laser master equation (2.10) by extracting from it equations of motion for $\alpha(t) \equiv i\langle a(t)\rangle$, $r_1(t) \equiv \langle R^+(t)\rangle$, $r_3(t) \equiv \langle R_3(t)\rangle$ and factoring all expectation values to products of $\alpha$, $r_1$, and $r_3$. One finds

$$\dot{r}_1 = 2g\alpha r_3 - (1/T_2)r_1 ,$$
$$\dot{r}_3 = -2g\alpha r_1 - (r_3 - N\sigma_0)/T_1 , \tag{B1}$$
$$\dot{\alpha} = gr_1 - \kappa\alpha .$$

Now $r_1$ and $r_3$ may be interpreted as transverse and longitudinal components of the classical Bloch vector. Let us find the conditions under which the Bloch equations (B1) describe a cooperative decay of initial atomic excitation. First, observe that the field variable $\alpha(t)$ may be eliminated by integrating the last equation in (B1):

$$\alpha(t) = g\int_0^t ds\, e^{-\kappa s}r_1(t-s) , \tag{B2}$$

where $\alpha(0) = 0$ has been used as initial condition. Assuming, subject to later proof of consistency, that $r_1(t)$ changes very slowly on a scale $\kappa^{-1}$, we may approximate (B2) by

$$\alpha(t) = (g/\kappa)r_1(t) \quad \text{for } t \gg \kappa^{-1} . \tag{B3}$$

Clearly, this amounts to assuming that $\kappa$ is so big that the field can follow the motion of the atoms adiabatically. Equation (B3) is the classical analog of the quantum-mechanical result (4.12).

Next, by introducing polar coordinates $r_1(t) = r(t)\sin\varphi(t)$ and $r_3(t) = r(t)\cos\varphi(t)$ we see that the total magnitude $r(t)$ of the Bloch vector is not conserved owing to the incoherent atomic relaxation terms $(1/T_1,\ 1/T_2)$. This well-known feature of the Bloch equations parallels the fact that the quan-

tum-mechanical master equations (2.10), (3.10), and (3.12) do not allow for the operator $\vec{R}^2$ to be conserved as long as the incoherent decay Liouvillian $\Lambda_A$ is present. Clearly, optimal cooperation of the atoms in building up the field $\alpha(t)$ can be expected only if the decay of $r(t)$ is negligible. In other words, we are interested in a solution of the Bloch equations describing a pulse whose characteristic times are much smaller than the incoherent atomic decay times $T_1$ and $T_2$. If such a solution exists, it can be obtained from (B1) with $1/T_1$ and $1/T_2 = 0$. Then, with the help of

$$r_1(t) = r\sin\varphi(t)\ ,\quad r_3(t) = r\cos\varphi(t)\ , \tag{B4}$$

we find the equation of motion for the Bloch angle $\varphi(t)$ to read

$$\ddot{\varphi}(t) + \kappa\dot{\varphi}(t) = 2rg^2\sin\varphi(t)\ . \tag{B5}$$

This is the equation for a damped nonlinear pendulum. Having already assumed implicitly that $\varphi(t)$ changes slowly on a scale $\kappa^{-1}$ [in replacing (B2) by (B3)], we may as well neglect $\ddot{\varphi}$ as compared to $\kappa\,\dot{\varphi}(t)$:

$$\dot{\varphi}(t) = r(2g^2/\kappa)\sin\varphi(t)\ . \tag{B6}$$

Formally, (B5) and (B6) [as well as (B2) and (B3)] are equivalent if the pendulum is heavily overdamped, i.e., if $\kappa \gg g(2r)^{1/2}$. This condition is precisely the right-hand side of our superradiance limit (2.14). We still have to obtain the condition allowing the neglect of $1/T_1$ and $1/T_2$ in the Bloch equations (B1). This is done most easily by comparing (B6) with the semiclassical rate equations (5.3) and (5.4). The latter may be stated as

$$\dot{\varphi}(t) = (2g^2/\kappa)(r+\tfrac{1}{2})\sin\varphi(t) \tag{B7}$$

if $1/T_1 = 1/T_2 = 0$ and the transformation

$$\langle R_3(t)\rangle - \tfrac{1}{2} = (r+\tfrac{1}{2})\cos\varphi(t) \tag{B8}$$

are used. Since the semiclassical equation (B7) is identical to the classical equation (B6) for $r \gg 1$, the requirement that the pulse be short compared to $T_1$, $T_2$ immediately leads to the full superradiance-limit condition discussed in Sec. VI.

There is a slight difference between the semiclassical theory [(B7) and (B8) and Sec. V] and the classical one [(B4) and (B6)] worth pointing out. For the fully excited initial state $\langle R_3(0)\rangle = r_3(0) = r$, the latter assigns to the Bloch angle the unstable equilibrium point of the pendulum $\varphi(0) = 0$. The physical reason for this shortcoming is, of course, the complete suppression of quantum fluctuations by the classical Bloch equations. On the other hand, the semiclassical rate equations account for the triggering of the pulse by spontaneous emission: For $\langle R_3(0)\rangle = r$ according to (B8), $\varphi(0) = \arccos(r-\frac{1}{2})/(r+\frac{1}{2})$, which is small but finite. Let us point out that this advantage of the semiclassical over the classical approach is less drastic than it might appear. Both approaches will turn out to be inappropriate for the fully excited initial state, as will be shown in the subsequent paper.

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†On leave of absence from Institut für Theoretische Physik, Universität Stuttgart, Stuttgart, Germany.

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[7]It is easy to generalize $\Lambda_A$ to account also for purely phase destroying mechanisms so as to have $T_2 < \frac{1}{2}T_1$; for this and other refinements as well as for a derivation of the master equation, the reader is referred to Weidlich and Haake, Ref. 3.

[8]The expansion (3.6) may be obtained by first Laplace transforming $K(s)$ and then using the operator identity $1/(A-B) = 1/A + (1/A)\,B\,(1/A) + (1/A)\,B\,(1/A)\,B\,(1/A) + \cdots$.

[9]This argument can be made quantitative; see the considerations in Sec. II following Eq. (4.6).

[10]A similar equation has been considered in Ref. 2(d) for the case of the atoms confined to a volume with linear dimensions small compared to the wavelength of the emitted radiation.

[11]From an "economical" point of view it would there-

fore seem more rational to eliminate the experimentally irrelevant atoms from Eq. (2.10) and discuss a generalized master equation for the reduced field-density operator $W_F(t) = \mathrm{tr}_A W(t)$. This has actually been done for the laser by Haake, Ref. 5, which in many respects parallels the present paper. However, elimination procedures are ultimately motivated not by economical considerations but rather the physically sound approximations they eventually give rise to. As may be seen from Sec. 4 of Haake's paper, the integral kernel in the generalized master equation for $W_F(t)$ allows for a convergent expansion if $1/T_1$, $1/T_2 \gg \kappa$ (which is usually fulfilled for a laser) but does not for the superradiance limit (2.14).

[12] This formula has also been obtained for the far field in Ref. 2(c).

[13] Recall that $\gamma_\perp = \gamma + \gamma^{\mathrm{coll}}$; if no collision broadening, i.e., nonradiative decay is present, $\gamma_\perp$ equals the natural linewidth $\gamma$ of the atomic transition.

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# Theory of Defects in Quantum Crystals

C. M. Varma
*Bell Telephone Laboratories, Murray Hill, New Jersey* 07974
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The classical theory for mass defects in crystals is inapplicable to quantum crystals, where zero-point motion leads to a force-constant renormalization on the introduction of mass defects. We develop a variational theory for handling such problems which simplifies the renormalization so that the problem reduces to that of a single frequency-dependent mass defect, to be calculated in a self-consistent fashion. Various anomalies observed in the properties of quantum crystals are discussed in light of the present theory. The theory may also be used to calculate in a simple fashion the properties of extended defects in classical crystals.

## I. INTRODUCTION

The traditional theory of lattice dynamics[1] assumes that the deviations of particles about their mean positions in the crystal are small relative to the equilibrium interparticle separation, and that the ratio of these quantities is a legitimate small expansion parameter in a perturbation treatment. In isotopes of helium and hydrogen, such an approach breaks down.[2] Because of the small mass of the atoms and their weak attractive interaction, the zero-point kinetic energy is of the same order of magnitude as the potential energy, and the rms deviation is as large as 30% of the equilibrium interparticle separation. Since the peculiarity of these crystals is rooted in the large zero-point motion, they have been called *quantum crystals*. The work on these crystals in the past few years has resulted in a clear understanding of the theoretical framework in which these crystals ought to be regarded. From the point of view of this paper, the salient points in this development have been the following: Nosanow[3] clearly formulated the problem of short-range correlations in such solids and showed that the single-particle variational wave functions are approximately Gaussian. Brenig[4] and Fredkin and Werthamer[5] showed through the random-phase approximation that the elementary excitations of even such highly anharmonic crystals are phonons. Assuming phonon wave functions (generalized Gaussians in coordinate space), Koehler[6] obtained a concise formula for the phonon frequencies by a variational calculation. Horner[7] obtained the result of Koehler through an elegant perturbation resummation and also showed how one may go beyond the noninteracting quasiparticle picture in such crystals. The result of these theoretical investigations may be summarized by saying that at least for long wavelengths, the excitations in quantum crystals are phonons, whose frequencies however should be determined self-consistently from force constants that are the thermal average of the second derivatives of some effective interparticle potential which includes the effect of short-range correlations. More precisely, the frequency $\omega_{\vec{k}\lambda}$ of a mode with wave vector $\vec{k}$ and polarization $\lambda$ is given by

$$\omega^2_{\vec{k}\lambda} = \frac{1}{m} \sum_{i<j,\alpha\beta} \epsilon^*_{\lambda\alpha}(\vec{k})\, \epsilon_{\beta\lambda}(\vec{k})\,(1 - e^{i\vec{k}\cdot\vec{R}_{ij}}) \times \langle \nabla_{i\alpha} \nabla_{j\beta} v(r_{ij}) \rangle \,, \qquad (1.1)$$

where $m$ is the mass of an atom; $i$, $j$ label the lattice sites; $\alpha$, $\beta$ label the Cartesian coordinates; $\vec{R}_{ij}$ is the vector joining $i$ to $j$; $\epsilon_{\lambda\alpha}(\vec{k})$ is a polarization vector; $v(r_{ij})$ is the effective potential between