has shown no improvement. It is possible that the rate of energy release to the thermometer is limited by the thermal conductivity of the sample. Alternatively the factor α may be much less than 0.01, possibly because of clustering of free radicals.

Despite the small size of the observed response, the dependence on temperature cycling confirms the model involving resonant spin-symmetry conversion of the tunneling methyl groups which was used to account for the nuclear resonance experiments.³ At least in these preliminary experiments, though, this thermal method of detecting tunneling resonances is much less sensitive than magnetic resonance methods. It is interesting to note that the thermal method can itself be re-

garded as a kind of electron spin resonance experiment in which the resonant absorption of microwave radiation by the electrons is detected thermally, and the microwave radiation is provided by the rotating methyl groups instead of by the conventional klystron.

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Superfluorescence and Cooperative Frequency Shift

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We report on a first-principles demonstration of the superfluorescence effect in a large system. Under proper conditions, a system of two-level atoms, confined to a pencilshaped volume, and initially prepared in an uncorrelated excited state, goes through a nonexponential cooperative decay, giving rise to a superfluorescence burst. Radiation is all confined in two diffraction patterns and the central frequency presents a time-dependent shift (chirping).

Superradiance (radiation proportional to N^2 by N two-level atoms, coherently prepared in a correlated state with a macroscopic polarization) was first described by Dicke¹ assuming a system small compared to a wavelength. Rehler and Eberly² generalized the results to a large system. However the radiation process from such initial states is essentially classical, whereas cooperative spontaneous emission from an initially uncorrelated excitation (superfluorescence) is intrinsically a quantum process. In fact the system starts radiating by ordinary fluorescence and then, eventually, it evolves spontaneously toward a correlated state, in which it radiates proportionally to N^2 , always preserving large intensity fluctuations.

In previous papers³ cooperative spontaneous emission has been treated by assuming strong coupling with only one electromagnetic mode: the "Dicke end-fire mode." Emission of radiation into other modes, and escape of radiation from the active volume V_c , where atoms are enclosed, were taken into account phenomenologically, by the insertion of relaxation terms in the equations of motion for the density operator.

In this Letter we report on a derivation from first principles of a many-mode theory of cooperative decay and the superfluorescence effect. In particular, we are able to describe both cooperative and noncooperative frequency shift and damping, as well as the radiation pattern. Our physical model consists of a system of N two-level atoms, interacting only through an electromagnetic field. Atoms are fixed at the vertices of a regular cubic lattice with spacing d . The parallelepiped occupied by the atoms has dimensions L_x, L_y, L_z and volume $V_c = Md^3 = L_xL_yL_z$. We quantize the free field in a volume V, V $\gg V_c$, in the Coulomb gauge. In the dipole approximation, neglecting electrostatic dipole interaction, the Hamiltonian becomes

$$
H = \hbar \sum_{\vec{k}} \omega_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + \hbar \Omega \sum_{i} r_{3,i} + \hbar V^{-1/2} \sum_{\vec{k},i} \left[g_{\vec{k}} a_{\vec{k}} r_i^{\dagger} \exp(i\vec{k} \cdot \vec{x}_i) + \text{H.c.} \right] + \hbar V^{-1/2} \sum_{\vec{k},i} \left[g_{\vec{k}} a_{\vec{k}}^{\dagger} r_i^{\dagger} \exp(i\vec{k} \cdot \vec{x}_i) + \text{H.c.} \right], \tag{1}
$$

 \bar{k}, i
where $a_{\bar{k}}^{\dagger}$, $a_{\bar{k}}$ are field operators; $r_{3,i}$, r_i^{\dagger} , r_i^{\dagger} are spin-flip atomic operators; \bar{x}_i is the space position of atom i; $g_{\bar{k}} = \Omega(2\hbar\omega_{\bar{k}})^{-1/2}[\mu_{12}^2 - (\hat{k} \cdot \vec{\mu}_{12})^2]^{1/2}$ is the coupling constant, with μ_{12} the electric dipole matrix. We stress the fact that we keep in (I) antiresonating terms which are neglected in the rotatingwave approximation.

If one solves the problem in terms of single-atom operators one has complicated equations which do not show the "phasing" of atoms due to field emission, and they have been solved only when $N=1, 2, 3$, or when the maximum linear dimension of the system, L_{max} , is such that $L_{\text{max}} \ll \lambda = 2\pi c \Omega^{-1}$ ^{4,5} For our purpose it is convenient to use collective dipole operators' defined as follows:

$$
R_{\alpha}^{\pm} = \sum_{i=1}^{N} r_i^{\pm} \exp(i\vec{\alpha} \cdot \vec{x}_i), \quad R_3 = \sum_{i=1}^{N} r_{3,i} \; ; \quad \alpha_j = 2\pi L_j^{-1} n_j, \quad -\pi d^{-1} < \alpha_j \leq \pi d^{-1}, \quad j = x, y, z. \tag{2}
$$

 R_{α}^* and R_3 have angular momentum commutation relations for each α . With the use of the collective operators, the Hamiltonian (l) becomes

$$
H = \hbar \sum_{\vec{k}} \omega_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + \hbar \Omega R_3 + \hbar V^{-1/2} \sum_{\vec{k}} \left\{ \left[g_{\vec{k}} a_{\vec{k}} R_{\vec{\alpha}}^{\dagger} f(\vec{k} - \vec{\alpha}) + \text{H.c.} \right] + \left[g_{\vec{k}} a_{\vec{k}}^{\dagger} \right] R_{\vec{\alpha}}^{\dagger} f(\vec{k} + \vec{\alpha}) + \text{H.c.} \right] \right\}, \tag{3}
$$

Here f is a "diffraction function" defined by

$$
f(\vec{k} - \vec{\alpha}) = N^{-1} \sum_{i} \exp[i(\vec{k} - \vec{\alpha}) \cdot \vec{x}_i].
$$
 (4)

The motion of the total system is governed by the Liouville equation for the density operator W: \dot{W} $=i\mathcal{L}W$. We denote by W_A the reduced density operator for atomic variables. Making use of projector techniques we can eliminate the field variables, obtaining the following equation⁷ for the density operator W_A of the atomic system alone:

$$
\dot{W}_A(t) = -i\Omega[R_3, W_A(t)] + \sum_{\vec{\alpha}, \vec{\alpha}'} \int_0^t d\tau \left\{ \alpha_{\vec{\alpha}, \vec{\alpha}'}(\tau) [R_{\vec{\alpha}}^-, W_A(t-\tau)R_{\vec{\alpha}}^+] + \text{H.c.} \right\} + \sum_{\vec{\alpha}, \vec{\alpha}'} \int_0^t d\tau \left\{ \alpha_{\vec{\alpha}, \vec{\alpha}'}(\tau) [R_{\vec{\alpha}}^+, W_A(t-\tau)R_{\vec{\alpha}}^+] + \text{H.c.} \right\}. \tag{5}
$$

Here we have performed a Born approximation and we have neglected the commutators containing R^+R^+ and $R^T R^T$ since they contain an intrinsic time dependence $\exp(\pm i2\Omega t)$. We explicitly note that neglecting these terms does not mean that we perform a rotating-wave approximation, since the antiresonant terms in (1) give rise also to the a^* terms in (5), whose effect is fully retained in the following. The limit of validity of the Born approximation' is

$$
L_{\text{max}} \ll l_c = (c V/4\pi N \lambda^2 \gamma_0)^{1/2},
$$

where l_{c} is the cooperation length of the atomic system, 3 and ${\gamma}_0^{-1}$ is the normal fluorescence decay time.

The coefficients $\alpha_{\alpha,\alpha'}^{\dagger}(\tau)$ are defined by

$$
\mathbf{G}_{\vec{\alpha},\vec{\alpha}}^{\dagger}(\tau) = (2\pi)^{-3} \int d^3k \ g_{\vec{\alpha}}^{\dagger}^2 f(\vec{k} - \vec{\alpha}) f(\vec{k} - \vec{\alpha}') \exp(i \ i \eta_{\vec{\alpha}}^{\dagger} \tau), \tag{6}
$$

where $\eta_k^* = \Omega \pm \omega_k$. The quantization volume V has been set equal to infinity with the replacement $\sum_{\tau=1}$ $(2\pi)^{-3}V \int d^3k$, thereby introducing irreversibility. Let us now consider the coefficients $\alpha_{\vec{\alpha},\vec{\alpha}}$, In general $\alpha \partial_{\alpha} \phi(r)$ is different from zero in a time interval $\Delta \tau$ such that

$$
\Delta \tau_{\text{max}} \sim c^{-1} L_{\text{max}}; \tag{7}
$$

i.e., the maximum time lag to be considered in (5) is of the order of the transit time of radiation in V_c . This statement is rather intuitive considering that integration (6) in a direction \hat{k} is a Fourier trans form from \vec{k} space to time space. Since the kernel $f(\vec{k} - \vec{\alpha})f(\vec{k} - \vec{\alpha}')$ has a width $\Delta k_j \sim (2\pi)^{-1}L_j$, $\alpha_{\vec{\alpha},\vec{\alpha}'}(\tau)$

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(8)

will be different from zero for $c\tau \leq (2\pi)^{-1}\Delta K_{\min} \sim L_{\max}$. Furthermore it can be shown^{9, 10} that the effect of $a \frac{1}{\alpha}, \overline{\alpha} \neq \overline{\alpha}'$, on the atomic equation of motion if negligible.

If we now assume that the atomic system evolves on a time scale τ_a such that

$$
\tau_a \gg c^{-1} L_{\text{max}}, \text{ i.e., } L_{\text{max}} \ll l_c,
$$

we can simplify Eq. (6) to a Markovian master equation:

$$
\dot{W}_A(t) = -i\Omega[R_3, W_A(t)] + \frac{1}{2}\sum_{\alpha} \left\{ \Gamma_{\alpha}^{\perp} [R_{\alpha}^{\perp}, W_A(t)R_{\alpha}^{\perp}] + \text{H.c.} \right\} + \frac{1}{2}\sum_{\alpha} \left\{ \Gamma_{\alpha}^{\perp} [R_{\alpha}^{\perp}, W_A(t)R_{\alpha}^{\perp}] + \text{H.c.} \right\},\tag{9}
$$

where

$$
\Gamma_{\alpha}^{\pm} = 2 \int_0^{\infty} \alpha \tau_{\alpha,\alpha}^{\pm}(\tau) d\tau = (2\pi)^{-3} \int g \tau_k^2 \delta(c|\vec{k}| \pm \Omega) f^2(\vec{k} - \vec{\alpha}) d^3k + i(2\pi)^{-3} P \int g \tau_k^2 (2/\pm \eta \tau_k^*) f^2(\vec{k} - \vec{\alpha}) d^3k. \tag{10}
$$

We call γ_{α}^{+} and Ω_{α}^{+} the real and imaginary part of Γ_{α}^{+} . Since γ_{α}^{+} = 0, the master equation can be written

$$
\dot{W}_A = -i(\Omega + \Omega')[R_3, W_A] + i\frac{1}{2}\sum_{\alpha} \tau_{\alpha}^2[S_{\alpha}^*, W_A] + \frac{1}{2}\sum_{\alpha} \gamma_{\alpha}^{*} \left\{ [R_{\alpha}^{*}, W_A R_{\alpha}^{*}] + \text{H.c.} \right\},\tag{11}
$$

where we have defined the cooperative polarization as

$$
S_{\overline{\alpha}} = \sum_{i \neq j} r_i^+ r_j^- \exp[i\overline{\alpha} \cdot (\overline{x}_i - \overline{x}_j)]
$$

= $R_{\overline{\alpha}}^+ R_{\overline{\alpha}}^- - (\frac{1}{2}N + R_3),$ (12)

and

$$
\Omega_{\overline{\alpha}} = \Omega_{\overline{\alpha}} + \Omega_{\overline{\alpha}} + \Omega_{\overline{\alpha}} \overline{}; \quad \Omega' = \frac{1}{2} \sum_{\overline{\alpha}} (\Omega_{\overline{\alpha}} + \Omega_{\overline{\alpha}} + \Omega_{\overline{\alpha}}).
$$

Equation (11) generalizes all previous results on cooperative spontaneous emission, as will be de-
scribed in detail in a forthcoming paper.¹⁰ Here $\,$ scribed in detail in a forthcoming paper. 10 Here we state only the main results. The time evolution of W_A contains three different contributions:

(1) The first term gives a constant frequency shift Ω' to the atomic transition, and can be eliminated by a renormalization of the naked frequency. Since $\sum_{\alpha} f^2(\vec{k} - \vec{\alpha}) = 1$, one can easily see from (10) that Ω' is nothing else than the Bethe part of the Lamb shift of a two-level atom.

(2) The second unitary term describes a cooperative effect, since it vanishes for a single
atom.¹¹ It gives rise to a time-dependent c atom.¹¹ It gives rise to a time-dependent cooperative frequency shift ("chirping") which is proportional to the total number of atoms. More precisely, calculating from the master equation (9) the equation of motion for $\langle R_{\alpha}^{+}(t) \rangle$, one obtains the so-called "super-Bloch-vector" equations derived semiclassically for a single atom by Jaynes¹² or for N atoms in a small volume by Stroud et or for N atoms in a small volume by Stroud et
 $al.^{13}$ These equations lead immediately to a timedependent frequency for $\langle R_{\alpha}^{+} \rangle$ given by

$$
\omega_{\alpha}^{\pm t}(t) = \pm \left[\Omega + \Omega' + \Omega_{\alpha}^{\pm} \langle R_3(t) \rangle \right]. \tag{13}
$$

We stress the fact that, in a large volume, Ω_{σ} is strongly dependent on the geometry of V_c and on the wave vector $\vec{\alpha}$. We have thus demonstrated that a cooperative chirping takes place also for volumes larger than a wavelength, provided

that the superfluorescence conditions stated later are met. This frequency shift is the macroscopic consequence of an effective dipole-dipole interaction potential induced by the radiation field spontaneously radiated by the atomic system during the cooperative decay.

(3) The third term gives a nonunitary time evolution. It contains both the Wigner-Weisskopf exponential decay as well as the cooperative superradiant decay. In fact from the master equation (11) we obtain exactly

$$
\langle \dot{R}_3 \rangle = -\sum \frac{1}{\alpha} \gamma_{\alpha} \langle S_{\alpha}^{\perp} \rangle - \gamma_0 \langle \frac{1}{2} N + \langle R_3 \rangle \rangle, \tag{14}
$$

where $\gamma_0 = \sum \frac{1}{\alpha} \gamma_{\alpha}^*$ turns out to be the Wigner-Weisskopf decay constant.

The radiation rate per unit solid angle¹⁴ in the direction \hat{k} can be shown to be

$$
I(\hat{k}) = \sum_{\hat{\alpha}} \gamma(\hat{k}) f^2 \left(\hat{k} \frac{\Omega}{c} - \vec{\alpha}\right) \left\langle S_{\hat{\alpha}} \right\rangle + \gamma(\hat{k}) \left(\frac{N}{2} + \left\langle R_3 \right\rangle\right), \quad (15)
$$

where $\gamma(\hat{k})$ is the normal fluorescence radiation rate:

$$
\gamma(\hat{k}) = \pi^{-1} \int g \tau_k^{2} \delta(c |\vec{k}| - \Omega) d^3 k.
$$

It can be verified that our equations and definitions are consistent with the energy conservation law: $I_T = \int I(\hat{k}) d\Omega_k = -\langle R_s \rangle$. The statements we have made so far are generally valid for any initial state.

We now specialize to the situation in which the system starts from an initially uncorrelated state with a population difference:

$$
\langle R_3(0)\rangle=\tfrac{1}{2}\overline{N}\leq\tfrac{1}{2}N;\quad \langle S_\alpha^\bullet(0)\rangle=0.
$$

Let us define the threshold length $l_T=(4\pi)^{-1}\lambda^{-2}$ $\times (V_c/\overline{N})(\gamma_T/\gamma_0)$, where γ_T is the total decay rate (including inhomogeneous broadening). It can be shown that if $L_{\rm max} < l_{T}$, then all S_{α} are zero at all times, and the system radiates by normal fluorescence; if $L_{\text{max}} \gg l_T$, and the Fresnel number rescence; if $L_{\text{max}} \gg \ell_T$, and the Fresher number
 $F = (\lambda L_{\text{max}})^{-1} L_{\text{min}}^2$ is such that $F \sim 1$ (pencil-shaped $F = (\lambda L_{\text{max}})^{-1} L_{\text{min}}^2$ is such that $F \sim 1$ (pencil-shape volume), only two $S_{\tilde{\alpha}}$ survive, specifically, $S_{\pm} \frac{1}{\tilde{\alpha}}$ volume), only two 3α survive, specifically, 3α α
 $\approx \frac{1}{2}[(\frac{1}{2N})^2 - \langle R_3 \rangle^2]$, with $|\vec{\alpha}_0| = c^{-1}\Omega$ and $\hat{\alpha}_0$ directed along L_{max} . In the latter case the system starts radiating by normal fluorescence, but then the radiation rate increases becoming proportional to \overline{N}^2 , and the radiation is all condensed into two opposite diffraction patterns centered around the % opposite diffraction patterns centered around the $L_{\rm max}$ direction, the so-called "end-fire modes." 15 This can be seen by substituting the above expression for $S_{\pm} \frac{1}{\alpha_0}$ in (14) and (15) and solving for $\langle R_{\rm s}(t) \rangle$. In this case the results for the one-mode model, concerning the conditions for superfluorescence and the time behavior of the radiated intensity and of R_3 , turn out to be correct.

From conditions (8) and $L_{\text{max}} \gg l_T$, we confirm that the superfluorescence effect takes place in a pencil-shaped volume only if

$$
l_T \ll L_{\text{max}} \ll l_C, \tag{16}
$$

where the left-hand side ensures that the dephasing atomic process occurs on a time scale much larger than the characteristic times of cooperative emission, while the right-hand side guarantees that the decay takes place always in a vacuum of photons, so that stimulated processes do not play any role.

Our results do not agree with the statement that that $L_{\text{max}} \gg l_{\text{r}}$ is the only requirement for superthat $L_{\text{max}} \gg l_{\textit{T}}$ is the only requirement for supe
fluorescence, made by Skribanowitz *et al.*¹⁶ in the interpretation of their experimental work, where more pulses were obtained. Since in their experimental conditions $F \sim 1$, but $l_T \ll L_{\text{max}} \sim l_c$, the right-hand side of (16) is violated. Hence we cannot interpret their results with this theory, because the Born and Markov approximations are no longer valid. On the other hand, we make the following suggestion: If the experimental situation is changed to satisfy condition (16), singlepulse superfluorescence will occur, as described in this paper and in Ref. 3.

In a forthcoming paper⁹ the condition $L_{\text{max}} \ll l_c$ will be relaxed, obtaining ringing (i.e., oscillation in radiated intensity) as is observed in the experiment of Ref. 16. Ringing in non-Markovian superfluorescence is due to the fundamental role of stimulated emission and absorption processes when L_{max} ^{\sim} l_c . Furthermore in Ref. 9 it will be shown that for $L_{\text{max}} \gg l_c$ the superfluorescence effect disappears.

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e-Expansion Solution of Wilson's Incomplete-Integration Renormalization-Group Equations

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Wilson's incomplete-integration renormalization-group equations have been solved in $4 - \epsilon$ dimensions for an arbitrary cutoff function. The two relevent exponents are computed to order ϵ and the exponent η is computed to order ϵ^2 . To order ϵ the exponents agree with the sharp-cutoff renormalization group. In order ϵ^2 , however, η appears to depend on the choice of the cutoff function.

One of the most beautiful but puzzling features of the renormalization-group approach is the fact that apparently *ad libitum* changes in the parameters of the physical system and the renormalization group itself do not affect the results of the theory. This of course is the theoretical basis of the physical fact of the universality of critical phenomena in a wide variety of systems and substances. Invariance of exponents and other quantities with respect to changes in parameters of the system have found their explanation in the concepts of relevant and irrelevant observables introduced by Kadanoff. ' The effect of changes in the renormalization group itself are less well understood. Wegner² and Jona-Lasinio³ have given a very general definition of a renormalization group and have given conditions under which two renormalization groups give the same exponents. DiCastro⁴ has shown in the context of the ϵ expansions that the renormalization group (RNG) of Gell-Mann and Low and the renormalization group of Wilson (sharp cutoff) yield the same exponents. It is the purpose of this Letter to carry out the ϵ expansion for yet another renormalization group, Wilson's incomplete-integration RNG. '

We start with Wilson's incomplete-integration RNG equation,

$$
\frac{\partial H}{\partial t} = \int_{k} \left(\frac{d}{2} \sigma_{k} + k \cdot \nabla_{k} \sigma_{k} \right) \frac{\partial H}{\partial \sigma_{k}} + \int_{k} \left[b + \beta(k) \right] \left(\frac{\delta H}{\delta \sigma_{k}} \frac{\delta H}{\delta \sigma_{k}} + \frac{\delta^{2} H}{\delta \sigma_{k} \delta \sigma_{k}} + \sigma_{k} \frac{\delta H}{\delta \sigma_{k}} \right),
$$

where b is a constant and $\beta(k)$ is any positive increasing analytic function of k^2 with $\beta(0) = 0$. The effective cutoff on the wave vector is e^{-t} and $\beta(k)$ determines the shape of this cutoff. σ_n is the Fourier component of the spin field corresponding to wave vector k . We attempt to determine the fixed-point Hamiltonian H^* such that

$$
\partial H / \partial t \big|_{H^*H^*} = 0,
$$

and we make the expansion

$$
H^* = -\frac{1}{2} \int_k U_2^*(k) \sigma_k \sigma_{-k} - \frac{1}{4!} \int_k \int_{k_1} \int_{k_2} U_4^*(k_1 k_1, k_2, k_3) \sigma_k \sigma_{k_1} \sigma_{k_2} \sigma_{k_3} - \dots
$$

We fix $\beta(k)$ and vary b so as to determine U_{2n}^* which are analytic. We make the following Ansatz for b and U_{2n}^* :

$$
b = 1 + b_1 \epsilon + b_2 \epsilon^2 + \dots; \quad U_2^* = U_{20}^* + U_{21}^* \epsilon + U_{22}^* \epsilon^2 + \dots; \quad U_4^* = U_{41}^* \epsilon + U_{42}^* \epsilon^2 + \dots; \quad U_6^* = U_{62}^* \epsilon^2 + \dots
$$

We skip the explicit writing of the arguments of the U_{2n}^* when it is not difficult to see what they should be. We have chosen $b_0 = 1$ in order that $U_{20}^*(k)$ be analytic with an expansion that begins with k^2 .

The above expansion enables us to truncate the RNG equations. To order ϵ^2 , we have

$$
k \cdot \nabla_k U_{20}^* = 2[1 + \beta(k)](1 - U_{20}^*)U_{20}^*; \quad k \cdot \nabla_k U_{21}^* = 2[1 + P(k)]U_{21}^* + 2b_1(1 - U_{20}^*)U_{20}^* + \int_{k_1} [1 + \beta(k_1)]U_{41}^*;
$$

\n
$$
k \cdot \nabla_k U_{22}^* = 2[1 + P(k)]U_{22}^* - 2[1 + \beta(k)](U_{21}^*)^2 + 2b_1(1 - 2U_{20}^*)U_{21}^* + 2b_2(1 - U_{20}^*)U_{20}^*
$$

\n
$$
+ \int_{k_1} [1 + \beta(k_1)]U_{42}^* + b_1 \int_{k_1} [1 + \beta(k_1)]U_{41}^* + \int_{-1}^{\epsilon} [1 + \beta(k_1)]U_{41}^*;
$$