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Propagational effects in an *ab initio* theory of super-radiance from extended systems

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Abstract. We report a comprehensive *ab initio* all-mode second-quantized theory of super-radiance from extended systems. Spatially-dependent chirps during the cooperative motion are not consistent with the assumption of a single or symmetrical two-mode solution and the pure superfluorescent regime of Bonifacio and Lugiato is not a possibility. Numerical integration of an associated set of c-number equations shows that super-radiance is characterized in general by a train of pulses in agreement with the two experiments. Propagational effects have an important effect on the cooperative radiative emission.

Dicke (1954) first discussed super-radiance (SR), the cooperative spontaneous emission of radiation from excited many-atom systems. The first observations of SR by Skribanowitz *et al* (1973) only partially confirm Dicke's prediction that a rod of inverted dielectric emits a single directed pulse of sech^2 intensity. They observed radiation at $\lambda = 50$–200 μm from inverted pairs of adjacent molecular rotational levels in a rod of HF gas at densities $n \sim 10^{11}$–10^{12} molecules cm^{-3}. After a delay ($\sim 1{\cdot}2$ μs) radiation was observed from one end of the rod in a series of short (~ 100 ns) pulses of decreasing intensity. Gross *et al* (1976) now report similar emission at $\lambda \sim 3$ μm from the optically-pumped $^5S_{1/2}$ state of Na: in these experiments $n \sim 10^9$–10^{10} molecules cm^{-3}, delays are of the order of nanoseconds, and fewer pulses appear in each pulse train.

Theories of SR now divide between those which predict a 'pure superfluorescent regime' (PSR) with SR much as in Dicke's original discussion‡, and those which do not. The PSR is defined by (BL) $k^{-1} \leqslant \tau_R \ll T_2^*$ in which $k^{-1} \equiv 2Lc^{-1}$ is the round trip time, and $\tau_R \equiv (2\pi p^2 n \hbar^{-1} k_s L)^{-1}$ (ck_s is the atomic resonance frequency, p the dipole matrix element, L a maximal length in the dielectric).

In this paper we report a comprehensive *ab initio* all-mode second-quantized theory of SR from extended systems of non-degenerate two-level atoms which shows: (i) that spatially-dependent chirps during the cooperative motion *invalidate* the single-mode approximation which could otherwise define a PSR; (ii) by approximating the operator equations by c-number equations and explicitly treating the passage of the radiation across the boundary of a finite medium, that k^{-1} does not define the lower limit to a PSR; (iii) that observable SR will be restricted to high-gain systems ($T_2^* \gg \tau_R$) and will be

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‡ For example, Bonifacio and Lugiato (1975a, b, to be referred to as BL), Banfi and Bonifacio (1974, 1975), Arecchi and Courtens (1970); and see Bonifacio *et al* (1971) for an heuristic theory.

characterized by (iv) a train of pulses in general, with, at fixed geometry, (v) fewer pulses at lower densities, (vi) pulse widths varying as n and intensities as n^2 at intermediate densities, and (vii) widths as $n^{1/2}$ and intensities as n at higher densities very much as the two experiments show. Propagational effects are not ignored and prove to play a significant role.

For simplicity we treat only the case $T_2^* = \infty$ in this paper. We introduce the usual pseudo-spin operators $\sigma_x^{(i)}, \sigma_y^{(i)}, \sigma_z^{(i)}$ for the ith two-level atom. This has resonance frequency ω_s and occupies a site $\boldsymbol{x}_i$ in a finite region V: we assume arbitrarily we can average over the $\boldsymbol{x}_i$ without detriment to the operator commutation relations. In the electric dipole approximation the Hamiltonian H for N atoms coupled to the second-quantized EM field $\boldsymbol{e}(\boldsymbol{x})$ is

$$H = \sum_{\boldsymbol{k},\lambda} \hbar\omega_k a^+_{\boldsymbol{k},\lambda} a_{\boldsymbol{k},\lambda} + \tfrac{1}{2}\hbar\omega_s \sum_{\boldsymbol{x}} \sigma_z(\boldsymbol{x}) - \sum_{\boldsymbol{x}} \boldsymbol{e}(\boldsymbol{x}) \cdot \boldsymbol{p}(\boldsymbol{x}). \tag{1}$$

The dipole density is

$$\hat{\boldsymbol{u}} p(\boldsymbol{x}) = p\hat{\boldsymbol{u}} \sum_{i=1}^{N} \sigma_x^{(i)} \delta(\boldsymbol{x} - \boldsymbol{x}_i)$$

($p\hat{\boldsymbol{u}}$ is the vector matrix element) and $\sigma_z(\boldsymbol{x}) = \Sigma_{i=1}^{N} \sigma_z^{(i)} \delta(\boldsymbol{x} - \boldsymbol{x}_i)$.

Heisenberg's equations yield the operator equations

$$\dot{\boldsymbol{\sigma}}(\boldsymbol{x}, t) = \boldsymbol{\omega} \times \boldsymbol{\sigma}(\boldsymbol{x}, t) \tag{2a}$$

with $\boldsymbol{\omega}(\boldsymbol{x}, t) \equiv (-2\hbar^{-1} p\hat{\boldsymbol{u}} \cdot \boldsymbol{e}(\boldsymbol{x}, t), 0, \omega_s)$ for the atoms and

$$\left(\nabla^2 - c^{-2}\frac{\partial^2}{\partial t^2}\right) e_{\mathrm{SR}}(\boldsymbol{x}, t) = +4\pi c^{-2} \frac{\partial^2 p(\boldsymbol{x}, t)}{\partial t^2} \tag{2b}$$

for the cooperative (super-radiant) field e_{SR}. An additional operator reaction field introduces spontaneous emission and radiative level shifts into the theory and there is also the free field operator, a solution of the homogeneous part of (2*b*) (Saunders *et al* 1975). In this preliminary report we shall focus attention solely on the cooperative field e_{SR}.

A multi-mode theory of SR is obtained through the Fourier transform

$$p(\boldsymbol{x}, t) = n \sum_{\boldsymbol{k}} P(\boldsymbol{k}, t) \cos(\boldsymbol{k} \cdot \boldsymbol{x} - \omega_s t + \phi(\boldsymbol{k}, t)) + n \sum_{\boldsymbol{k}} Q(\boldsymbol{k}, t) \sin(\boldsymbol{k} \cdot \boldsymbol{x} - \omega_s t + \phi(\boldsymbol{k}, t)). \tag{3}$$

The operator envelope and phase functions P, Q and ϕ vary slowly compared with ω_s^{-1}: they are related to the collective operators of Banfi and Bonifacio (1974). We are unable to solve to our satisfaction the multi-mode operator theory for a rod-shaped region V and restrict our precise results and conclusions to the case of the infinite parallel-sided slab $-\tfrac{1}{2}L \leqslant z \leqslant \tfrac{1}{2}L$. We find rigorously that the radiation rate is maximized for resonant modes $|\boldsymbol{k}| = k_s$ travelling *in a direction of maximal length*. These directions are parallel to the surface in the case of the slab. We assume that the result will apply to an arbitrary geometry, however, so that the rate will be maximized for modes parallel to the rod axis in the case of the rod: to this extent the argument justifies Dicke's later emphasis on the 'end fire modes'.

For the slab it now appears reasonable to approximate (3) by

$$p(\boldsymbol{x}, t)=n[P^{(+)}(t)\cos(k_s z-\omega_s t+\phi^{(+)}(t))+P^{(-)}(t)\cos(k_s z+\omega_s t+\phi^{(-)}(t))$$
$$+Q^{(+)}(t)\sin(k_s z-\omega_s t+\phi^{(+)}(t))+Q^{(-)}(t)\sin(k_s z+\omega_s t+\phi^{(-)}(t))] \qquad (4)$$

in which the two modes are both resonant and *normal* to the slab surface.

This *ansatz* proves to be consistent with (2*a*) only if the cooperative field e_{SR} takes the form

$$e_{SR}(\boldsymbol{x}, t)=\hbar p^{-1}[\epsilon^{(+)}(t)\cos(k_s z-\omega_s t+\phi^{(+)}(t))+\epsilon^{(-)}(t)\cos(k_s z+\omega_s t+\phi^{(-)}(t))] \qquad (5)$$

in which envelopes and phases depend only on t. The forms (4) and (5) must therefore consistently ignore propagational effects. In practice we find from (4) and (2*b*) by a careful analysis of the chirp (cf Hassan 1976) that the envelopes and phases in (5) do depend on z. This result is not peculiar to the slab and it is hard to avoid the conclusion that it also applies to the rod.

Instead of (4) and (5) we must therefore assume that

$$p(\boldsymbol{x}, t)=n[P^{(+)}(z, t)\cos\Phi^{(+)}(z, t)+P^{(-)}(z, t)\cos\Phi^{(-)}(z, t)+Q^{(+)}(z, t)\sin\Phi^{(+)}(z, t)$$
$$+Q^{(-)}(z, t)\sin\Phi^{(-)}(z, t)] \qquad (6a)$$
$$e_{SR}(\boldsymbol{x}, t)=\hbar p^{-1}[\epsilon^{(+)}(z, t)\cos\Phi^{(+)}(z, t)+\epsilon^{(-)}(z, t)\cos\Phi^{(-)}(z, t)]$$

where

$$\Phi^{(\pm)}(z, t)=\omega_s t\mp k_s z+\phi^{(\pm)}(z, t). \qquad (6b)$$

In the slowly varying envelope and phase approximation (2*b*) reduces to

$$c^{-1}\frac{\partial\epsilon^{(\pm)}(z, t)}{\partial t}\pm\frac{\partial\epsilon^{(\pm)}(z, t)}{\partial z}=\alpha p^{(\pm)}(z, t)$$
$$\epsilon^{(\pm)}(z, t)\left(c^{-1}\frac{\partial\phi^{(\pm)}(z, t)}{\partial t}\pm\frac{\partial\phi^{(\pm)}(z, t)}{\partial z}\right)=\alpha Q^{(\pm)}(z, t); \qquad (7a)$$
$$\alpha=2\pi np^2k_s\hbar^{-1}.$$

The operator Bloch equation (2*a*) reduces to

$$\frac{\partial P^{(\pm)}(z, t)}{\partial t}=-\tfrac{1}{2}\Gamma_0 P^{(\pm)}(z, t)+\epsilon^{(\pm)}(z, t)N(z, t) \qquad \Gamma_0\equiv\tfrac{4}{3}p^2k_s^3\hbar^{-1};$$
$$\frac{\partial Q^{(\pm)}(z, t)}{\partial t}=-\tfrac{1}{2}\Gamma_0 Q^{(\pm)}(z, t)-\frac{\partial\phi^{(\pm)}(z, t)}{\partial t}P^{(\pm)}(z, t); \qquad (7b)$$
$$\frac{\partial N(z, t)}{\partial t}=-\Gamma_0(1+N(z, t))-[\epsilon^{(+)}(z, t)P^{(+)}(z, t)+\epsilon^{(-)}(z, t)P^{(-)}(z, t)]$$

in which the damping arises from the reaction field and $N(z, t)\equiv\sigma_z(\boldsymbol{x}, t)$. Solution of these operator equations is intractable at the present time. We have therefore replaced them by the equivalent c-number equations and integrated them numerically. A consistent treatment of the initiating spontaneous emission cannot be given this way and we have set $\Gamma_0 \equiv 0$ and assumed the initial condition†

$$N(z,0) = \begin{cases} 1-\delta & |z| < \frac{1}{2}L \\ 0 & |z| > \frac{1}{2}L \end{cases}, \tag{8}$$

with $\delta < 10^{-3}$. Skribanowitz *et al* (1973) adopted comparable equations and conditions empirically and restricted study to only one of the two oppositely directed wave trains $\epsilon^{(+)}$ and $\epsilon^{(-)}$ travelling along the direction of L.

Numerical results (figure 1) for $\tau_R \sim 5$ ns ($n = 9{\cdot}4 \times 10^{11}$ molecules cm^{-3}) and data for the HF gas experiment ($L = 100$ cm, $\lambda = 84$ μm, $p = 6{\cdot}7 \times 10^{-19}$) agree well with the observations. Pulse intensities are proportional to $\tau_R^{-2}(\propto n^2)$ and only the delays (roughly of the order of $\tau_R \ln \delta$ and determined weakly by (8)) are too short. Plainly the fields (inside the slab (shown in figure 1(*a*)) rapidly become spatially inhomogeneous; but the two oppositely directed wave trains interfere only during the emission of the later ringing pulses and justify the assumption of Skribanowitz *et al* that these trains evolve separately.

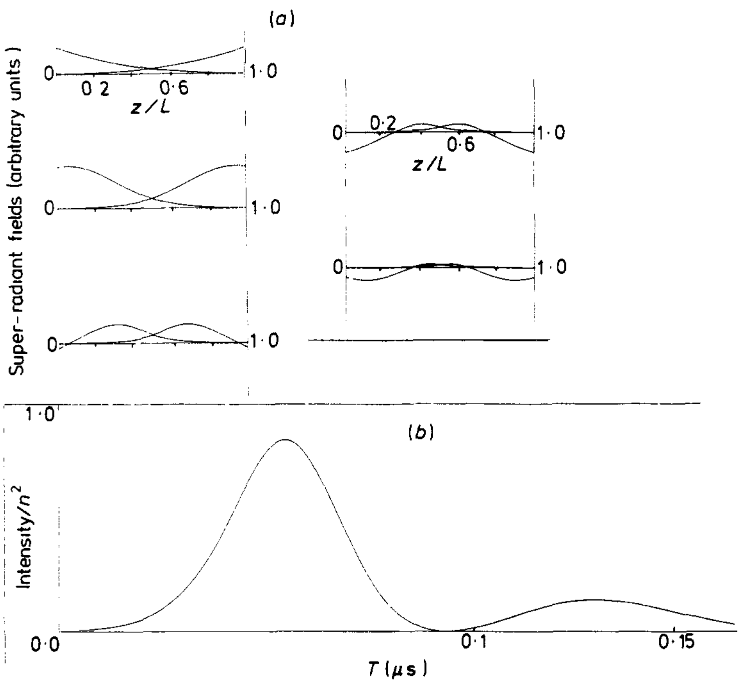

Figure 1. (*a*) Fields inside $|z| \leq \frac{1}{2}L$ and (*b*) output intensity for $\tau_R = 5$ ns, $\lambda = 84$ μm, $L = 100$ cm, $n = 9{\cdot}4 \times 10^{11}$ molecules cm^{-3}, $p = 6{\cdot}7 \times 10^{-19}$.

For shorter $\tau_R \sim 5 \times 10^{-10}$–$5 \times 10^{-11}$ s ($n \sim 10^{13}$–10^{14} molecules cm^{-3}) figure 2 shows that delays and pulse widths become roughly proportional to $\sqrt{\tau_R}$ and intensities to τ_R^{-1}: oppositely directed wave trains now interfere strongly throughout the motion. Such densities were outside the range of the experiments on HF; but equivalent

† We assume the phased initial condition $P(z, 0) = [1-(1-\delta)^2]^{1/2}$ for the dipoles.

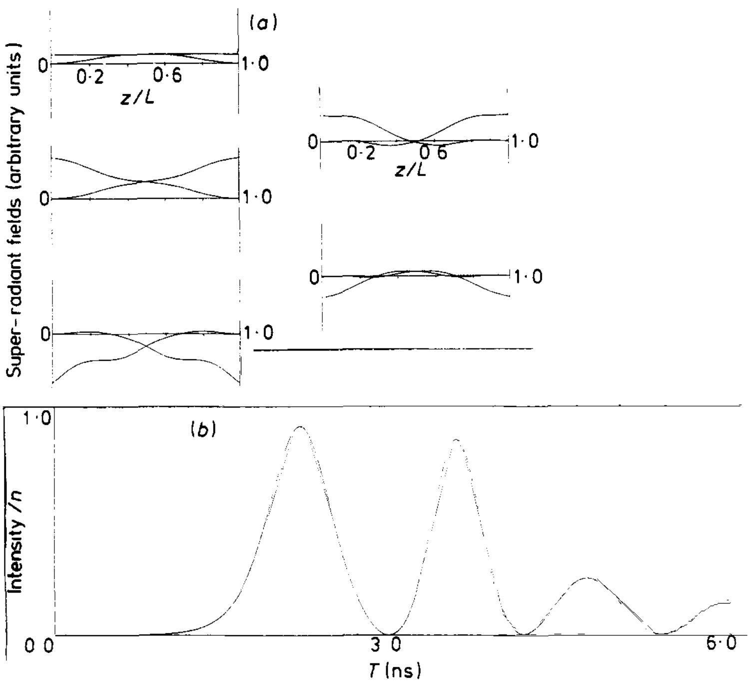

Figure 2. (*a*) Field inside $|z| \leq \frac{1}{2}L$ and (*b*) output intensity for $\tau_R \sim 50$ ps. $\lambda = 84\ \mu$m, $L = 100$ cm, $n = 9 \times 10^{13}$ molecules cm^{-3}, $p = 6{\cdot}7 \times 10^{-19}$.

densities and these behaviours are reported by Gross *et al* (1976) on Na. The data of figures 1 and 2 particularly exemplify points (vi) and (vii) above: other numerical data to be published elsewhere show that the number of ringing pulses decreases with n (compare figure 1 with 2), that if $T_2^* < \infty$, $\tau_R \ll T_2^*$ is a condition for SR, and that ringing pulses are suppressed for $\tau_R \lesssim T_2^*$.

Equations (7) reduce to the usual c-number resonant sharp-line self-induced transparency equations when

$$N(z, t) = N^{(+)}(z, t) + N^{(-)}(z, t) \tag{9}$$

and the $(\pm)$ qualities do not overlap. If $\epsilon^{(\pm)}(z, t) \equiv \partial\sigma^{(\pm)}(z, t)/\partial t$, $\tau \equiv (\alpha c)^{1/2} t$, $\xi = (\alpha c)^{1/2}(t - 2z/c)$, equations (7) then reduce to the pairs of sine–Gordon equations $\sigma_{\xi\xi}^{(\pm)} - \sigma_{\tau\tau}^{(\pm)} = -\sin \sigma^{(\pm)}$ with the unusual boundary conditions $\partial \to 0(\xi \to -\infty)$, $\to \pi(\xi \to +\infty)$. These equations contrast with the spatially homogeneous damped-pendulum equation

$$\sigma_{tt} + k(\alpha c)^{-1/2}\sigma_t - \sin \sigma = 0$$

in which $\sigma = \sigma(t)$ obtained by Bonifacio and Lugiato (1975a, b). Note that $k^{-1} \equiv 2Lc^{-1}$ appears neither in these sine–Gordon equations nor in (7a) whilst BL's semi-classical equations reduce to (7) when, but only when, $k = 0$. This is because we have not attempted to quantize new modes inside the occupied region V. Instead, the effect of the boundary of V occurs naturally through the c-number Green function of (2b), or else in the numerical integration of (7) we follow the field across the boundary of V and compute the fields inside and outside V (see figures 1 and 2). The region outside V is not treated simply as an energy sink therefore and no damping terms k need to appear in (7a) in consequence.

Note also in contrast with BL that the initial condition (8) is not spatially homogeneous and the equations (7) are not independent of z. Propagational effects are therefore of considerable importance as the evolutions of the fields in the two figures show. Since k does not appear in (7) the PSR of BL is not a possible consequence of the Hamiltonian (1). It is interesting to note however that any effective conductivity damping of sufficient magnitude in (7a) opens up the possibility of this particular type of radiation.

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