The effect of dipole-dipole interactions between atoms in an active medium

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Abstract. Based on the results of numerical modeling, it is shown that dipole-dipole interactions among atoms in the active medium influence strongly the character of the associated superradiation. The main effect is to make the nuclear subsystem behave chaotically. Its strength increases with the atom density, and leads to the suppression of distant collective correlations and superradiation. Near correlations between the atoms are established, causing a confinement effect: a shielding of radiation in the active medium.

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1. Introduction and method

Superradiation (SR) is the cooperative radiation arising in a medium that contains a population inversion of excited states. Originally this effect has been stated for purely quantum systems: i.e. two-level atoms [1]. Experiments have confirmed this prediction [2]. Later work established that this phenomenon also occurs in classical systems [3, 4], and that the phasing effect—the spontaneous origin and strengthening of correlations of originally independent subsystems—underlies it. In the quantum case, these are correlations among phases of electronic states of atoms undergoing radiative transitions; while in the classical regime correlations among phases of oscillations and directions of the electric dipole moments of atoms occur. A full account of the influence on SR of the dipole-dipole interactions among atoms remains incomplete (see references [5, 6, 7]).

The SR theory has been developed in several directions. There exist complementary to each other Schrödinger, Heisenberg and semiclassical approaches. Each of them is applicable to a special area of values of the system parameters. The common methodological lack of these approaches is that the phasing mechanism remains off screen. The mechanism of the transition from casual to a phased state possesses certain spatial, time and statistical behaviors and its nature is not fully clear. The quantum mechanical problem of SR is rather complicated, for example, within the Heisenberg approach it requires to solve a system of nonlinear operational equations. Approximations which are used to simplify this systems have limited and often unclear area of applicability. Classical model of superradiation (CMS), where atoms are substituted by the classical Lorenz oscillators and the electromagnetic field is described by the classical Maxwell equations, allows to answer many difficult questions, in particular, the phasing mechanism. Therefore classical and quantum approaches complement each other. Moreover, radiation produced by pure classical system such as electrons revolved in magnetic field, electron clouds created in wigglers, cathode-ray lamps for microwaves, etc. is also SR.

Let us consider only classical systems. First, phasing leads to the ordering of phases of atoms. Second, according to Earnshaw’s theorem [8, 9], a system of point dipoles cannot maintain a stable static equilibrium configuration. Dipole-dipole interactions cause chaotic behavior that disorders their phases, and hence suppresses SR. SR arrises from a competition between these two opposing effects. This conclusion is inferred from the theory of non-uniform broadening of spectral lines for lasers [10, 11]. Consider now a nonlinear CMS [12, 7], i.e. a system of classical, charged anharmonic oscillators. Maxwell’s equations describe the electromagnetic field. Next, assume that there are sufficient oscillators ($N \gg 1$), and they occupy a small spatial region of length $L$ such that $l \ll L \ll \lambda$, where $l = n^{1/3}$ is the characteristic distance between atoms, and $\lambda$ is the wavelength of the radiation. Each charge has magnitude $e$ and mass $m$, and is located on the ends of springs with stiffness coefficient $k$, at coordinates $r_a + \xi_a (a = 1, 2, ..., N)$, fixed in points $r_a$, where there are also compensating charges $-e$. The equation of motion for the oscillators then takes the form [13]

$$\ddot{\xi}_a + \omega_0^2(1 + \gamma \xi^2_a)\xi_a = -\frac{2e^2 \omega_0^2}{3mc^2} \sum_b \frac{\dot{\xi}_b}{m} \sum_{b \neq a} \nabla_a \times \left( \nabla_a \times \frac{\xi_b (t_{ab})}{r_{ab}} \right).$$

(1)

Here $\nabla_a = \partial / \partial r_a$, $r_{ab} = r_a - r_b$, $t_{ab} = t - r_{ab}/c$ represents the retarded time, $\omega_0 = \sqrt{k/m}$ is the fundamental frequency of the oscillators, and $\gamma$ is the nonlinearity parameter. Substituting the expression

$$\xi_a = b[F_a(t) \exp(-i\omega t) + F^*_a(t) \exp(i\omega t)],$$

(2)
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into equation (1)—where \( b \) represents the characteristic initial amplitude of the oscillations—gives

\[
\dot{F}_a + i\delta(|F_a|^2 - 1)F_a = i\beta \sum_{b \neq a} \nabla \times \left[ \nabla_a \frac{\exp(ikr_{ab})}{r_{ab}} \times F_b(t) \right] - \frac{1}{2}\beta_0 \sum_b F_b. \tag{3}
\]

In equation (3) the second derivatives of functions \( F_\alpha(t) \) which vary slowly in comparison with exponents \( \exp(\pm i\omega_0 t) \) are omitted, and a frequency \( \omega = \omega_0 + \delta, \delta = 3\gamma\omega_0b^2/2 \) is chosen. Note, that the case of particles rotating in a magnetic field (important in a practical sense) corresponds \( \delta < 0 \). For a small size system equation (3) can be rewritten as

\[
F_a + i\delta(|F_a|^2 - 1)F_a = i\beta \sum_{b \neq a} \frac{3n_{ab}(n_{ab}F_b) - F_b}{r_{ab}^3} - \frac{1}{2}\beta_0 \sum_b F_b. \tag{4}
\]

Where \( n_{ab} = r_{ab}/r_{ab}, \beta = e^2/(2mc\omega_0), \) and \( \beta_0 = 2e^2\omega_0^3/3mc^3 \). The first term on the right hand side of equation (4) represents the dipole-dipole interaction of the oscillators, while the second term is analogous to a ‘viscosity’ for the radiation in the electromagnetic field. Following Ref. [12], we shall consider one-dimensional oscillators, i.e. that dipoles oscillate along the \( z \) axis, and consequently, that the vectors \( F_a \) are parallel to it: \( F_a = iF_a, \quad i = (1, 0, 0). \) During a given time \( t \) we have \( F_a(t) = \rho_a(t) \exp(i\omega_a(t)) \). Hence, atoms possess a dipole moment that is \( d_a(t) = e\xi_a = eb\rho_a \cos(\omega t + \phi_a) \).

The average radiation intensity of the rapidly oscillating dipoles then is

\[
I = \frac{e^2\omega b^2}{3c^3} \sum_{a,b} |F_a||F_b|\cos(\phi_a - \phi_b). \tag{5}
\]

Thus, equation (4) represents a system of \( N \) oscillators, distributed arbitrarily, that can be solved by numerical means. A similar formalism is described in Ref. [12]: however, dipole-dipole interactions are neglected.

2. Results and discussion

The phasing effect can be described as follows. Consider a complex plane \((x, y) = (\Re(F), \Im(F))\) containing \( N \) points that each represent the state of an individual oscillator, where the distance from the origin is simply the amplitude of oscillation, and the angle is the phase with respect to the fundamental frequency \( \omega_0 \). Points with \( \omega > \omega_0 \) rotate clockwise around the origin; points with \( \omega < \omega_0 \) rotate anticlockwise.

Initially, the points are placed randomly with equal probability phases on a circle of unit radius \( \rho = 1 \). From equation (4), their velocities are

\[
v_a = \omega(\rho_a) \times \rho_a + f + \sum_b d(\rho_a, \rho_b; r_a, r_b). \tag{6}
\]

Here \( \rho_a = (\Re(F_a), \Im(F_a), 0), \) \( v_a = \rho_a, \) \( f = -\beta_0 \sum_a \rho_a^2/2, \) and \( \omega(\rho) = (0, 0, -\delta(\rho^2 - 1)), \) \( d(\rho_a, \rho_b; r_a, r_b). \) The latter dipole-dipole interaction term is not shown in full for reasons of space. Note that the vector \( -f \) is proportional to the total dipole moment of the system \( D = eb\sum_a \rho_a/2; \) and, \( \omega(\rho_a) = 0 \) at \( t = 0 \). Notice also that the sign of \( \gamma \) affects the direction of rotation only: changing it results in a mirror inversion without any other consequences. Points with positive \( \gamma \) rotate clockwise outside the unit circle, and rotate anticlockwise when inside, while the opposite is true when \( \gamma \) is negative. This symmetry, therefore, is exploited by choosing \( \gamma > 0 \).
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Figure 1. Time evolution of the phase distribution of oscillators. The dotted line is a circle with unit radius. The number of oscillators is \( N = 5 \times 10^3 \). The concentration of oscillators \( n = 10^{22} \text{m}^{-3} \) (curve 2 on figure 4).

Figure 2. Time dependence of the radiation intensity for \( N = 5 \times 10^3 \) (all values in arbitrary units).

Having established the basis for the model, we next consider how the system evolves when the density of atoms \( n \) is sufficiently small that dipole-dipole interactions are negligible. Due to the fluctuations of density distribution of the oscillators initial phases \( \varphi_0(0) \), the initial value of the vector \( f \) is not precisely zero. At \( t = 0 \) from equation (6) it follows that

\[
\frac{dD}{dt} = -\frac{D}{\tau_{SR}},
\]

where the characteristic emission time is \( \tau_{SR} = \frac{1}{N\beta_0} \) [1, 5, 6, 7].

Consequently from equation (6), the system responds by moving in a direction opposite to the dipole moment \( D \), with a collective net velocity \( f \). The system at time ~ \( \tau_{SR} \) is displaced a distance ~ \( D(0)/(Ne) \) (see figure 1b). The resulting displacement moves half of the points outside the unit circle (\( \rho > 1 \)), and the other half inside (\( \rho > 1 \)). Hence, points outside the circle will move in clockwise orbits, while those within circulate the opposite way. After an interval \( t \sim 10\tau_{SR} \), the net motion results in a bunching of points on the inside of the circle (figure 1b), thus the atoms emit most of their stored energy in a sharp pulse of coherent radiation (figure 2). For two-level atoms, the characteristic delay time \( t_0 = \tau_{SR} \log N \) given in [1] is consistent with this. The bunch subsequently develops into a spiral-shaped distribution.
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Figure 3. Time evolution of the phase distribution of oscillators in systems with a strong dipole-dipole interaction. The dotted line is a circle with unit radius. Figure a and b corresponds to concentration of oscillators $n = 8 \times 10^{22} \text{m}^{-3}$ (curve 4 on figure 4). Figure c corresponds to $n = 1.8 \times 10^{23} \text{m}^{-3}$ (curve 6 in figure 4).

Figure 4. Intensity of radiation (arbitrary units) for systems with different oscillator concentrations $n \ (10^{22} \text{m}^{-3})$: 0.083, 1.0, 2.3, 8.0, 12.13, 18.38, 27.86, for 1–7 respectively.

At high density $n$, dipole-dipole interactions have a significant effect. Figure 3 shows the outcome of equation (4) for large $n$; the initial conditions are the same as described previously. Notice that the points on the phase plane now move in a more chaotic manner than before. When $n$ is high, dipole-dipole interactions among adjacent oscillators are strong and this leads to incoherence. However, SR is not entirely suppressed. In spite of the chaotic behavior of dipole-dipole interaction, the initial total dipole moment results in bunching of points, and correspondently in the SR pulse (figure 3a,b). On figure 3c, where the concentration of oscillators was doubled, dipole-dipole interaction suppress the bunching.

High density systems are also complicated by collective effects. Localized groups of
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resonant atoms induce antiphase dipole moments among their neighbors. This preserves coherence while screening SR [7].

The SR delay \( t_0 \) and peak intensity \( I_{\text{max}} \) also depend on \( n \): increasing \( n \) makes \( t_0 \) longer, and \( I_{\text{max}} \) smaller (see figures 4, 5, and 6). This is a consequence of the effect of coherence on the collective interactions among the dipoles, which becomes weaker with increasing \( n \).

Unlike classical systems, quantum systems do not behave chaotically. The intensity varies smoothly with time as described by the following formula [1].

\[
I(t) = \frac{\hbar \omega_0}{4\mu \tau_N} (\mu N + 1)^2 \sech^2 \left( \frac{t - t_0}{2\tau_N} \right),
\]

where \( \mu \) represents the form-factor of the oscillators’ mutual position, and \( \tau_N = 1/\beta_0 \) is the characteristic emission time. This curve is plotted in figure 5 to illustrate the difference between the classical and quantum cases. When \( N \) is large, at \( t = t_0 \), equation 7 suggests \( I_{\text{max}} \sim N^2 \). However, the CMS predicts that the exponent \( \alpha = \frac{\log(I_{\text{max}})}{\log(N)} \) rises to a peak value that is less than two, then declines as \( N \) increases (see figure 7). Experimental observations of SR in semiconductors exhibit similar behavior [14].

The results are consistent with Ref. [7]. Localized, dynamic metastable states are formed when the atom density \( n \) is sufficiently large. Each oscillator perturbs the motion of its nearest neighbors such that their relative phase differs by \( \pi \). Hence, in effect each oscillator appears to be screened in a manner analogous to Debye shielding. This leads to confinement of electromagnetic fields in the active medium.

3. Conclusions

This study examines the phenomenon of superradiation for systems of classical nonlinear charged oscillators. The results of our numerical simulations show that after a characteristic
Figure 6. Dependence of a maximum of radiation intensity (arbitrary units) on oscillator density $n$ (in $10^{22} m^{-3}$).

Figure 7. Dependencies on the number of oscillators $N$, of (a) the ratio $\log_{10}(I_{\text{max}})/\log_{10}(N)$; and (b) the peak radiation intensity.
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delay time $t_0$, a peak in radiated power occurs, which subsequently decays in a chaotic, oscillatory manner, superimposed on a sech$^2(t - t_0)$ background. SR is also suppressed progressively with increasing oscillator density $n$. This behavior is ultimately a consequence of collective dipole-dipole interactions. These both induce incoherence among the oscillators, and cause a screening effect.

Within localized regions, the individual dipoles possess correlated moments. Dipoles separated by sufficient large distances are nearly uncorrelated. As $n$ increases, the system breaks up into more of these regions. Each region emits SR impulses independently, resulting in the chaotic decay described above.

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