Effects of interatomic interaction on cooperative relaxation of two-level atoms

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We study effects of direct interatomic interaction on cooperative processes in atom-photon dynamics. Using a model of two-level atoms with Ising-type interaction as an example, it is demonstrated that interparticle interaction combined with atom-field coupling can introduce additional interatomic correlations acting as a phase synchronizing factor. For the case of weakly interacting atoms with $J<\hbar\omega_0$, where J is the interparticle coupling constant and ω_0 is the atomic frequency, dynamical regimes of cooperative relaxation of atoms are analyzed in the Born-Markov approximation both numerically and using the mean field approximation. We show that interparticle correlations induced by the direct interaction result in inhibition of incoherent spontaneous decay leading to the regime of collective pulse relaxation which differs from superradiance in nature. For superradiant transition, the synchronizing effect of interatomic interaction is found to manifest itself in enhancement of superradiance. When the interaction is strong and $J>\hbar\omega_0$, one-particle one-photon transitions are excluded and transition to the regime of multiphoton relaxation occurs. Using a simple model of two atoms in a high-Q single mode cavity we show that such transition is accompanied by Rabi oscillations involving many-atom multiphoton states. Dephasing effects of dipole-dipole interaction and solitonic mechanism of relaxation are discussed.

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I. INTRODUCTION

The phenomenon of superradiance has a long history dating back more than 60 years to the seminal paper by Dicke [1] where the effect was predicted theoretically. Over the past few decades superradiance has been the subject of intense theoretical and experimental studies in a large variety of systems. These include molecular aggregates [2–5], cold atoms [6,7] and Bose-Einstein condensates [8–10], atomic nuclei [11–13], magnetic nanoclusters [14–16], heterostructures [17–19], and many others.

The key process underlying the mechanism of superradiance is phase synchronization of initially independent atoms caused by the coupling with a common environment represented by the electromagnetic field. In order for such a process to occur the phase decoherence time of atoms should be longer than the photon travel time in the sample [20,21].

For samples whose size is smaller than the wavelength of radiation, this condition requires the density of atoms to be sufficiently high. The system-environment (atom-field) coupling also manifests itself as an additional indirect interaction (a sort of the transverse dipole-dipole interaction) which may suppress superradiant transitions depending on the spatial distribution of atoms or the sample geometry [20,21].

On the other hand, when the density of atoms (or, more generally, emitters) is high the direct interparticle interaction starts to play an increasingly important part in determining cooperative behavior of the particles. In particular, this interaction strongly affects the properties of low-dimensional systems. The Mott-insulator quantum phase transition in optically trapped atomic systems [22–25] and in solid structures [26–29], generation of many-particle entangled states or many-particle coherent dynamics, as it is in the case of effectively interacting atoms inside a high quality dissipative cavity [30,31], Bose-Einstein condensate [23,32], and in molecular clusters with strong magnetic [33,34] or Coulomb [35,36] correlations, are examples.

The direct interparticle interaction introduces additional correlations between emitters. These correlations considerably influence the cooperative optical properties of atoms. First, the interaction directly affects the superradiance leading to a number of peculiarities such as changing the order of superradiant phase transitions [37,38]. Recently, the possibility of superradiant relaxation in strongly correlated systems was studied theoretically [15,16]. The experimental results for magnetic molecules of Mn_{12}-acetate type were also reported in Ref. [14]. Earlier, systems of ferroelectric type with strong interparticle interaction were regarded as promising candidates for an active medium of the heat pumping laser [38].

Second, the direct interparticle interaction can play the role of a phase synchronizing factor that may lead to the cooperative behavior which, though it shares many common properties with the superradiance effect, essentially differs from superradiance in nature. The classical example furnishes the spectrum of a P luminescence band in CdS and ZnO where the emission intensity is proportional to the second power of the free exciton number (pumping intensity). In this case the effect is caused by exciton-exciton scattering [39,40]. Recently, such effects were observed in the microcrystalline phase of $CsPbCl_3$ thin films [41].

For interacting atoms, the interaction can drastically change the regime of atom-photon dynamics by inducing (otherwise, excluded) multiphoton transitions [42]. It was shown in Ref. [43] that interatomic interaction can give rise to nonzero multiphoton emission observed with a single-molecule spectroscopy technique as a two-photon cooperative effect for strongly dipole-dipole coupled molecules. Theoretically, this phenomenon was predicted as a large two-

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atom two-photon resonant effect for two atoms inside a high-quality cavity [44].

So, different regimes of radiative decay in correlated atomic systems are mainly governed by the interatomic interaction. By controlling the interaction, radiation properties of such systems can be widely varied, ranging from superradiant transitions to the generation of the Fock state of light. In particular, such control is feasible for the atoms in an optical lattices (see, e.g., [22,45]).

In this work cooperative radiation of interacting atoms coupled to an electromagnetic bath will be of our primary interest. We are aimed to study different relaxation regimes determined by the intensity of interatomic coupling.

The paper is organized as follows. In Sec. II we formulate the model of N two-level atoms with Ising-type interaction and qualitatively discuss various regimes of relaxation by considering realignment of the atomic energy spectrum at different values of the interatomic coupling constant J. There are two limiting cases of weak and strong interaction with $J<\hbar\omega_0$ and $J>\hbar\omega_0$, respectively (ω_0 is the atomic frequency).

We find that, for weakly interacting atoms, Ising interaction would affect dynamical behavior of the system leading to the transition to collective pulse relaxation and enhancement of superradiance. For strong interaction, the regime of multiphoton relaxation is predicted to occur.

Derivation of the master equation for weakly interacting atoms is presented in Sec. III. We show that, for certain atomic configurations, dephasing effects of induced dipole-dipole interaction can be suppressed and dynamics of the atomic system can be described by the simplified master equation.

In Sec. IV, the effects for weakly interacting atoms briefly discussed in Sec. II are investigated in detail. By applying the mean field approximation we obtain the results that agree very well with those calculated by solving the equations for atomic variables numerically.

The regime of multiphoton relaxation that takes place at strong interatomic interaction due to inhibition of one-particle one-photon transitions is described in Sec. V. Finally, in Sec. VI, we draw together the results, discuss the solitonic mechanism of relaxation, and make some concluding remarks. Details on some technical results are relegated to Appendixes A–C.

II. ATOMIC ENERGY SPECTRUM AND REGIMES OF RELAXATION

In order to illustrate a possibility of different relaxational regimes, caused by direct interatomic interaction, we consider the simplest model of a chain of two-level atoms with nearest neighbor Ising-type interaction. Typically, the models of this type are used to describe molecular systems with Coulomb and magnetic interactions such as ferroelectric [46] and magnetic [47–49] clusters, interacting electrons in the tightly binding approximation [28], and interacting atoms in optical lattice [25], where the extended Hubbard model is usually introduced.

The interaction takes into account repulsion of the neighboring atoms so that the system may reveal the correlated many-particle behavior. When the energies of the neighbors are different tunneling transitions between neighboring sites are inelastic.

In addition to the Ising-type direct interaction, the atoms interact with a common electromagnetic field (the atom-field interaction). These two interactions are combined to maintain correlations in the atomic subsystem thus crucially affecting the regimes of cooperative optical transitions.

A. The model

The Hamiltonian of the model atomic chain with the short-range Ising-type interaction can be written in the following form:

$$H = H_A + H_F + H_I, \tag{1}$$

where

$$H_A = \hbar\,\omega_0 \sum_{i=1}^{N} S_i^z - J \sum_{i=1}^{N} S_i^z S_{i+1}^z \tag{2}$$

is the Hamiltonian of the atomic subsystem,

$$H_F = \sum_{\mathbf{k}s} \hbar\,\omega_k a_{\mathbf{k}s}^\dagger a_{\mathbf{k}s} \tag{3}$$

is the Hamiltonian of electromagnetic field, and

$$H_I = -i\hbar \sum_{i=1}^{N} \sum_{\mathbf{k}s} [g_{\mathbf{k}s,i}(S_i^+ + S_i^-)a_{\mathbf{k}s} - \text{H.c.}] \tag{4}$$

is the Hamiltonian of the atom-field interaction and H.c. stands for Hermitian conjugation; ω_0 is the frequency of atomic transition, $J>0$ is the coupling constant of the nearest neighbor interaction, N is the number of atoms, S_i^z is the z-component of the pseudospin operator determining the population of the ith atom, S_i^+ (S_i^-) is the raising (lowering) operator of the pseudospin, $a_{\mathbf{k}s}$ ($a_{\mathbf{k}s}^\dagger$) is the photon annihilation (creation) operator characterized by the wave-number vector $\mathbf{k}$, the frequency ω_k, and the polarization vector $\mathbf{e}_{\mathbf{k}s}$,

$$g_{\mathbf{k}s,i} = \sqrt{\frac{\omega_k}{2\varepsilon_0 \hbar\, v}}(\mathbf{d}_i \cdot \mathbf{e}_{\mathbf{k}s}) e^{i\mathbf{k}\cdot\mathbf{r}_i} \tag{5}$$

is the coupling constant of the dipole interaction between the ith atom and transverse electromagnetic field, $\mathbf{d}_i$ is the effective dipole moment of the ith two-level atom with the vector of spatial coordinates $\mathbf{r}_i$, ε_0 is the vacuum dielectric constant, and v is the volume of field quantization.

The Hamiltonian (1) is distinguished from standard models of a two-level atom interacting with the electromagnetic field by the presence of the interatomic interaction in H_A [see Eq. (2)].

In what follows the dynamics of the collective relaxation of the initially inverted atomic subsystem will be of our primary concern. More specifically, we shall study various dynamical scenarios that occur at different values of the coupling constant J. In the remaining part of this section we first discuss the energy spectrum of the atomic subsystem and qualitatively describe these dynamical regimes.

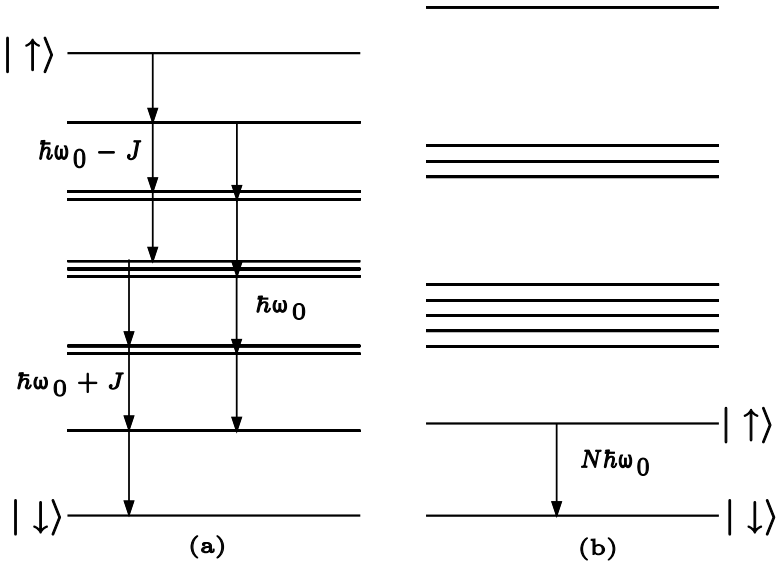

FIG. 1. Energy spectrum of H_A for the chain of six atoms. Two cases are shown: (a) weak interatomic interaction with $J/\hbar\omega_0=0.1$ and (b) strong interatomic interaction with $J/\hbar\omega_0=10$. The state $|\uparrow\rangle=|\uparrow,\uparrow,\ldots,\uparrow\rangle$ ($|\downarrow\rangle=|\downarrow,\downarrow,\ldots,\downarrow\rangle$) indicates that the atoms are all in the excited (ground) state.

B. Weak interatomic interaction, $J<\hbar\omega_0$

The transition energy of an atom depends on the states of its neighbors as a result of interatomic interaction. This effect can be described as renormalization of the transition frequency of the ith atom $\omega_0\rightarrow\tilde{\omega}_i$ which can also be readily seen from the mean field expression for the Hamiltonian H_A:

$$H_A \sim \sum_i \hbar \left[\omega_0 - \frac{J}{2\hbar}(\langle S_{i-1}^z\rangle + \langle S_{i+1}^z\rangle) \right] S_i^z = \sum_i \hbar\,\tilde{\omega}_i S_i^z.$$

Sufficiently weak interaction, $J\ll\hbar\omega_0$, results in inhomogeneous broadening of the radiation spectrum. When the coupling constant of interatomic interaction J increases and $J<\hbar\omega_0$, the spectrum of the atomic subsystem undergoes more pronounced rearrangement. As is shown in Fig. 1, the spectrum is no longer equidistant, but its structure is identical to that for noninteracting atoms with $J=0$. In particular, the latter implies that the state with all the atoms excited $|\uparrow\rangle=|\uparrow,\uparrow,\ldots,\uparrow\rangle$ (spin up represents the excited state of the atom) gives the level of highest energy and the level of lowest energy corresponds to the case where atoms are all in the ground states $|\downarrow\rangle=|\downarrow,\downarrow,\ldots,\downarrow\rangle$.

Similar to the case of noninteracting atoms, the dynamics of radiative decay is dominated by one-particle one-photon transitions and can be described using the Born-Markov approximation.

A cooperative relaxation of the system from an excited state can be realized by different schemes of transitions. It is a possibility to have a generation in the two mode regime with the frequencies $\omega=\omega_0\pm J/\hbar$ [see Fig. 1(a)] arising due to renormalization of the atomic frequency $\tilde{\omega}_i$ that governs the intensity of radiation.

It is well-known [21] that the intensity of atom radiation I is proportional to the fourth power of the transition frequency $I\propto\omega_0^4|\mathbf{d}|^2$, where $\mathbf{d}$ is the transverse dipole moment of the atomic transition. For N weakly interacting atoms, the intensity I is defined by the renormalized frequency and can be represented by a sum of the coherent and the incoherent parts, $I=I_{\mathrm{coh}}+I_{\mathrm{incoh}}$, where $I_{\mathrm{coh}}\propto\Sigma_{i\neq j}\tilde{\omega}_i^4|\mathbf{d}_i\mathbf{d}_j^*|\propto\tilde{\omega}^4N^2$ and $I_{\mathrm{incoh}}\propto\Sigma_i\tilde{\omega}_i^4\mathbf{d}_i^2\propto\tilde{\omega}^4N$.

Dependence of the renormalized frequency on the states of neighboring atoms introduces additional correlations into the incoherent part of radiation. Such correlations may result in a cooperative radiation of fundamentally different origin than superradiance described by the coherent part I_{coh}. The effect of superradiance, for its part, can be enhanced by the Ising-type interaction which may act as a phase synchronizing factor.

Figure 1(a) shows that, when the atomic subsystem is not completely inverted, an excited state may relax through a series of transitions whose frequencies are identically equal to the resonant frequency ω_0. This regime of relaxation resembles the solitonic mechanism where the process of emission is connected with inelastic motion of defects (Bloch walls) induced by a short-range interaction. We shall discuss this point at greater length in Sec. VI.

C. Strong interatomic interaction, $J>\hbar\omega_0$

In the case of strong interaction with $J>\hbar\omega_0$, arrangement of the energy level significantly differs from that of noninteracting atoms. Now the state $|\uparrow\rangle$ does not give the level of highest energy and $E_N<E_{N-1}$, where E_j is the energy of the state with j excited atoms. Such realignment indicates that many-atom multiphoton transitions will dominate the process of relaxation.

Figure 1(b) illustrates that, in the limiting case with $J\gg\hbar\omega_0$, the states $|\uparrow\rangle$ and $|\downarrow\rangle$ give two lowest lying energy levels. They are separated from the other excited states by the gap of the width $\sim J$. Relaxation of fully inverted state (transition from $|\uparrow\rangle$ to $|\downarrow\rangle$) can be achieved through a N-photon process.

At this stage it should be stressed that an accurate description of the interaction between the system and the field in the presence of strong interatomic correlations requires careful consideration of interplay between many-atom multiphoton dipole and multipole transitions. In a sense, our system can be regarded as an intermediate case between a collection of

two-level atoms independently interacting with the field and a single many-level system.

A good example provides the spin-phonon interaction in magnetic molecules $Mn_{12}O_{12}$ of the multipole form $H_I \propto (S^+)^4 a_k + (S^-)^4 a_k^\dagger$ whose symmetry is consistent with the anisotropy energy [50]. Similarly, the spin superradiance in atomic nuclei can be dominated by multipole transitions [13].

For the multipole superradiance [51], the exponent of the N-dependence of the radiation intensity peak (N is the number of emitters) can be greater than two, $I \propto N^\alpha$ with $\alpha > 2$. This is the case for the cooperative spin-phonon relaxation in $Mn_{12}O_{12}$ where a rough estimate gives $I \propto \langle (S^+)^4 (S^-)^4 \rangle \propto N^8$.

The standard approach to the dissipative dynamics of atoms relies on the Lindblad equation for the reduced density matrix of the atomic subsystem derived by eliminating boson variables from the master equation for the density operator of the atom-field system. This master equation is obtained using the Born-Markov approximation [52,53] that neglects multiphoton transitions and thus is applicable only for weakly interacting atoms at $J < \hbar\omega_0$. In the subsequent section we discuss the derivation procedure for the Lindblad equation for the density matrix of weakly interacting atoms.

III. MASTER EQUATION FOR WEAKLY INTERACTING ATOMS

In this section we derive and discuss the master equation for the reduced atomic density operator ρ for the case of weak interaction at $J < \hbar\omega_0$. We show that the direct interaction renormalizes both the induced dipole-dipole interaction and the damping operator in the Lindblad equation. In addition, we find that, under certain conditions, the destructive effect of the dipole-dipole interaction on superradiance is suppressed.

A. Derivation of master equation

We begin by applying the standard procedure of elimination of field variables in the Born-Markov approximation [52] to the equation for the density matrix of the combined field-atom system, ρ_{AF}, written in the representation of interaction

$$i\hbar \frac{d}{dt}\tilde{\rho}_{AF}(t) = [V(t), \tilde{\rho}_{AF}(t)], \tag{6}$$

where

$$\tilde{\rho}_{AF}(t) = e^{i(H_A+H_F)t/\hbar}\rho_{AF}e^{-i(H_A+H_F)t/\hbar},$$

$$\begin{aligned} V(t) &\equiv e^{i(H_A+H_F)t/\hbar}H_I e^{-i(H_A+H_F)t/\hbar} \\ &= -i\hbar\sum_{i=1}^{N}\sum_{\mathbf{k}s}[g_{\mathbf{k}s,i}(S_i^+ e^{i(\omega_0\hat{\Gamma}_i-\omega_k)t} + S_i^- e^{-i(\omega_0\hat{\Gamma}_i+\omega_k)t})a_{\mathbf{k}s} \\ &\quad - \text{H.c.}], \end{aligned} \tag{7}$$

$$\hat{\Gamma}_i = 1 - \beta(S_{i+1}^z + S_{i-1}^z), \quad \beta = J/(\hbar\omega_0) \tag{8}$$

and the expression for the operator (7) is derived using the cyclic boundary conditions.

As it was pointed out in Sec. II, for the ith atom, the energy of transition depends on the state of the neighboring atoms. Alignment of the energy spectrum of the atomic subsystem and, as a result, the character of atom-field interaction are determined by the value of the ratio $\beta = J/\hbar\omega_0$. In Eq. (7) the frequency of the dipole transition is effectively described by the operator (8) $\omega_0\hat{\Gamma}_i$ entering the Hamiltonian of interaction V. When $J > \hbar\omega_0$ and $\beta > 1$, ordering of the energy levels changes and certain transitions appear to be excluded.

In this case, for example, the energy E_N of the "monodomain" state $|\uparrow,\uparrow,\ldots,\uparrow\rangle$ is below the energy E_{N-1} of the state where only one atom is in the ground state $|\downarrow,\uparrow,\uparrow,\ldots,\uparrow\rangle$. The result is that relaxation of the completely inverted state $|\uparrow,\uparrow,\ldots,\uparrow\rangle$ cannot occur as a one-photon process. This can be seen from the expression for the probability of the transition from the state $|\uparrow,\uparrow,\ldots,\uparrow\rangle|0\rangle$ to the one-photon state $|\downarrow,\uparrow,\uparrow,\ldots,\uparrow\rangle|1_{\mathbf{k}s}\rangle$

$$\begin{aligned} W^{(1)} &= \lim_{t\to\infty}\frac{d}{dt}\frac{1}{\hbar^2}\sum_{\mathbf{k}s} q(\mathbf{k}s) \\ &\quad \times \left|\int_0^t d\tau \langle\downarrow,\uparrow,\uparrow,\ldots,\uparrow|\langle 1_{\mathbf{k}s}|V(\tau)|0\rangle|\uparrow,\uparrow,\ldots,\uparrow\rangle\right|^2 \\ &= \lim_{t\to\infty}\frac{d}{dt}\frac{2d^2}{3\pi^2\varepsilon_0\hbar c^3}\int_0^\infty d\omega\omega^3\frac{\sin^2\{[\omega_0(1-\beta)-\omega]t/2\}}{[\omega_0(1-\beta)-\omega]^2} \\ &\to 0, \quad J > \hbar\omega_0, \end{aligned} \tag{9}$$

where $q(\mathbf{k}s)$ is the density of the electromagnetic field modes. The interaction renormalizes the frequency of an atom transition $\omega_0 \to \omega_0(1-\beta)$. At $\beta > 1$, the one-particle one-photon transition becomes nonresonant and its probability $W^{(1)}$ goes to zero.

The value $\beta = 1$ separates the two different regimes of atomic relaxation. For $\beta < 1$, the atomic decay is governed by the one-photon process and the multiphoton relaxation prevails at $\beta > 1$.

It should be noted that, for $\beta > 1$, the change of sign in the eigenvalues of the operator $\hat{\Gamma}_i$ introduces new resonant terms of the form $S_i^+ a_{\mathbf{k}s}^\dagger$. However, as it was discussed in Sec. II C, this case is additionally complicated by multipole transitions that need to be taken into consideration.

When $J < \hbar\omega_0$ and $\beta < 1$, the eigenvalues of the operator $\hat{\Gamma}_i$ are all positive. The Ising-type interaction just renormalizes the transition frequencies inducing additional correlations in the atomic subsystem.

Dynamics of the atomic relaxation is now governed by one-photon processes and, in the remaining part of this section, we restrict ourselves to this case of weakly interacting atoms with $\beta < 1$.

We can now use the Born approximation to derive the equation for the reduced density matrix of the atomic subsystem [52], $\rho = \mathrm{Tr}_F\{\rho_{AF}\}$. For simplicity, we consider the zero-temperature case where only vacuum fluctuations of the electromagnetic field are taken into account. So, the density operator ρ_{AF} can be written in the following form:

$$\rho_{AF}(t) = |0\rangle\langle 0| \otimes \rho(t). \tag{10}$$

We additionally assume that the wavelength of radiation is much longer than the size of the system and use the Markov approximation [20]. Omitting the technical details that can be found in Appendix A, the resulting equation is given by

$$\frac{d}{dt}\tilde{\rho}(t) = \eta\sum_{i,j}\int_0^\infty d\omega\omega^3 F_{ij}(\omega) \times\left\{\left[R_j(t),\tilde{\rho}(t)S_i^+(t)\left(\pi\delta(\omega-\omega_0\hat{\Gamma}_i)+\mathrm{P}\frac{i}{\omega-\omega_0\hat{\Gamma}_i}\right)\right] + \left(R_j(t),\tilde{\rho}(t)S_i^-(t)\mathrm{P}\frac{i}{\omega+\omega_0\hat{\Gamma}_i}\right)+\mathrm{H.c.}\right\}, \tag{11}$$

where

$$S_i^\pm(t) \equiv e^{i(H_A+H_F)t/\hbar}S_i^\pm e^{-i(H_A+H_F)t/\hbar} = S_i^\pm e^{\pm i\omega_0\hat{\Gamma}_i t},$$

$$\tilde{\rho}(t) = e^{i(H_A+H_F)t/\hbar}\rho e^{-i(H_A+H_F)t/\hbar},$$

$$R_i(t) = S_i^+(t) + S_i^-(t), \tag{12}$$

and

$$F_{ij}(\omega) = \frac{3}{2}\left[[1-(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]\frac{\sin kr_{ij}}{kr_{ij}} + [1-3(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]\left(\frac{\cos kr_{ij}}{(kr_{ij})^2}-\frac{\sin kr_{ij}}{(kr_{ij})^3}\right)\right], \tag{13}$$

$k=\omega/c$, $r_{ij}\equiv|\mathbf{r}_{ij}|=|\mathbf{r}_i-\mathbf{r}_j|$ is the separation distance between the ith and jth atoms, we assume $\mathbf{d}_i=\mathbf{d}_j\equiv\mathbf{d}$, $\bar{\mathbf{r}}_{ij}\equiv\mathbf{r}_{ij}/|\mathbf{r}_{ij}|$, $\bar{\mathbf{d}}\equiv\mathbf{d}/|\mathbf{d}|$, and

$$\eta = \frac{d^2}{6\varepsilon_0\hbar\,\pi^2c^3}. \tag{14}$$

For brevity, in Eq. (11) we used operator functions defined on eigenvalues of $\hat{\Gamma}_i$, so that if $\hat{\Gamma}_i|\psi\rangle=\Gamma_i|\psi\rangle$ then, e.g., $\delta(\omega-\omega_0\hat{\Gamma}_i)|\psi\rangle=|\psi\rangle\delta(\omega-\omega_0\Gamma_i)$. It should be stressed once again that Eq. (11) is valid only for the case of weakly interacting atoms, when $J<\hbar\omega_0$ and the Born-Markov approximation is applicable.

Principal values of the integrals that enter the right-hand side of the master equation (11) define the dipole-dipole interatomic interaction (see Appendix A) and the Bethe-part of the Lamb shift, which depend on the direct interparticle interaction. Equation (11) can be further simplified under the condition that the characteristic wavelength of radiation λ is much longer than the size of the sample.

It is well-known [20] that in this case the dipole-dipole interaction is of quasistatic character and is independent of the frequency of atomic transition. Since the direct interaction in Eq. (7) just renormalizes the atomic frequencies $\omega_0\rightarrow\tilde{\omega}_i=\omega_0\hat{\Gamma}_i$, the asymptotic form of the dipole-dipole interaction in the limit $r_{ij}/\lambda\ll1$ must be identical to that for the system of noninteracting atoms with $J=0$. At the same time the relaxation terms proportional to $\tilde{\omega}_i^3$ essentially depend on the interatomic interaction.

Mathematically, Eq. (A6) in the limit $r_{ij}/\lambda\ll1$ gives the relation

$$\mathrm{P}\int_0^\infty d\omega\omega^3\frac{F_{ij}(\omega)}{\omega\pm\omega_0\hat{\Gamma}_i}\sim-\frac{3}{4}\frac{\pi}{(r_{ij}/c)^3}[1-3(\mathbf{d}\cdot\hat{\mathbf{r}}_{ij})^2],\quad i\neq j. \tag{15}$$

With $F_{ij}(\omega)\approx1$ and the Lamb shift disregarded Eq. (11) gives the master equation in the simplified form

$$\frac{d}{dt}\tilde{\rho}(t) = \gamma_0\sum_{i,j}[R_j(t),\tilde{\rho}(t)\hat{\Gamma}_i^3S_i^+(t)] + i\sum_{i\neq j}\frac{\Omega_{ij}}{2}[R_j(t),\tilde{\rho}(t)R_i(t)]+\mathrm{H.c.}, \tag{16}$$

where $\gamma_0=\pi\eta\omega_0^3$ is one-half the spontaneous decay rate for an isolated atom and

$$\Omega_{ij} = -\pi\eta\frac{3}{2}\frac{1-3(\mathbf{d}\cdot\hat{\mathbf{r}}_{ij})^2}{(r_{ij}/c)^3} \tag{17}$$

is the constant of the quasistatic dipole-dipole interaction [20].

The first term on the right-hand side of Eq. (16) describes renormalization of the damping term by the interatomic interaction. After neglecting the rapidly oscillating terms (the rotating wave approximation) in Eq. (16) and going back to the Schrödinger representation, we derive the master equation for the case of weakly interacting atoms, $\beta<1$, in the final form:

$$\frac{d}{dt}\rho(t) = -\frac{i}{\hbar}[H_A,\rho(t)] + i\sum_{i\neq j}\Omega_{ij}[S_i^+S_j^-,\rho(t)] + \gamma_0\sum_{i,j}([S_j^-,\rho(t)\hat{\Gamma}_i^3S_i^+]+[S_i^-\hat{\Gamma}_i^3\rho(t),S_j^+]). \tag{18}$$

Equation (18) describes dynamics of the Ising-like interacting atoms coupled to an electromagnetic bath. The key feature of this equation is that only the damping terms are renormalized by the direct interatomic interaction at $r_{ij}\ll\lambda$.

B. Effects of dipole-dipole interaction

It is known (see, e.g., reviews [20,21]) that the dipole-dipole interaction may destroy the cooperative radiation due to the frequency chirping effect. This effect is caused by the dynamical shift of energy levels that results in a rapid loss of phase synchronization with the characteristic time of phase decoherence much shorter than the rate of relaxation [20].

Influence of the dipole-dipole interaction on the dynamics of the atomic subsystem is strongly affected by the spatial configuration of atoms and the angular distribution of dipole moments. It turns out that, for certain configurations, dephasing induced by the dipole-dipole interaction is inhibited and the cooperative radiation takes place [20,21].

In particular, the latter is the case when atoms are all in an identical environment and the system is invariant under permutations of atoms, $\Omega_{ij}=\Omega_{i'j'}=\Omega$. These conditions are met for a two atom system or an atomic ring [20].

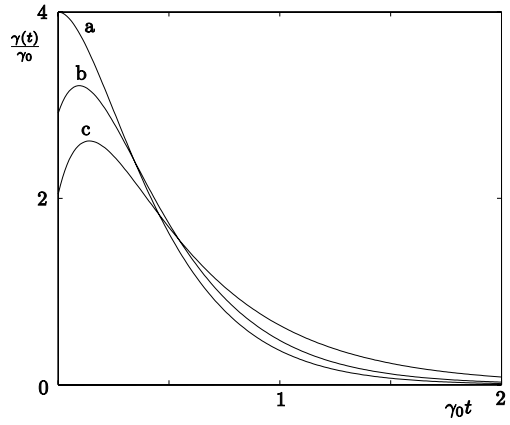

FIG. 2. Relaxation rate $\gamma(t)$ (19) for two interacting two-level atoms at various values of the ratio $\beta \equiv J/\hbar\omega_0$: (a) $\beta=0$, (b) $\beta=0.1$, and (c) $\beta=0.2$.

For disordered atomic configuration (atomic cloud), the superradiance effect depends on the sample shape. It can occur for pencil-shaped patterns as a result of suppression of the destructive effect of the dipole-dipole interaction [21].

In our case, when the interatomic spacings are much smaller than the wavelength ($r_{ij} \ll \lambda$), the form of the dipole-dipole interaction (17) is the same for both interacting and noninteracting atoms. Symmetry of the renormalized damping terms is also identical to the symmetry of the system of noninteracting atoms with $J=0$. So, atomic configurations suppressing the dephasing effect of the dipole-dipole interaction are the same for both cases: $J=0$ and $J\neq 0$.

For the simplest case of two atomic configurations, this point can be illustrated explicitly. Similar to the case of two noninteracting atoms considered in Ref. [54], the master equation (18) for two atoms with $J\neq 0$ is exactly solvable. The exact solution described in Appendix B shows that relaxation is independent of the dipole-dipole interaction.

The analytical results presented in Appendix B can also be used to demonstrate the effect of additional correlations induced by the interaction. For this purpose, we consider the expression for the relaxation rate of initially excited atoms

$$\gamma(t) \equiv -\frac{d}{dt}\mathrm{Tr}\{\rho(S_1^z + S_2^z)\} = 4me^{-4mt}\left[1 + \frac{n}{n-m}(1 - e^{-4(n-m)t})\right], \tag{19}$$

where $n=\gamma_0(1+\beta/2)^3$ and $m=\gamma_0(1-\beta/2)^3$.

In Fig. 2 the relaxation rate (19) is plotted as a function of time at various values of β. It is seen that the interaction noticeably influences the character of relaxation even at small values of β. By contrast to the case of noninteracting atoms with $\beta=0$, the system features collective decay with a pronounced emitted pulse arising as a result of additional correlations induced by Ising-type interaction.

Now it is our primary task to examine how the cooperative relaxation is influenced by the interatomic interaction. So, in what follows we shall restrict our study to the atomic configurations where the frequency chirping effect can be disregarded. For such configurations, the master equation (18) can be considerably simplified by neglecting the terms describing the dipole-dipole interaction.

IV. COOPERATIVE RADIATION OF WEAKLY INTERACTING ATOMS

From the master equation (18) without the dipole-dipole interaction, $\Omega_{ij}=0$, it is straightforward to deduce a system of equations for averages of the atomic variables $\langle S_n^z\rangle = \mathrm{Tr}_A\{\rho S_n^z\}$ and $\langle S_n^\pm\rangle = \mathrm{Tr}_A\{\rho S_n^\pm\}$. The result is given by

$$\frac{d}{dt}\langle S_n^z\rangle = -\gamma_0 \sum_i \langle \hat{\Gamma}_i^3 S_i^+ S_n^- + S_n^+ S_i^- \hat{\Gamma}_i^3\rangle, \tag{20a}$$

$$\frac{d}{dt}\langle S_n^+\rangle = i\omega_0\langle S_n^+ \hat{\Gamma}_n\rangle + 2\gamma_0 \sum_i \langle \hat{\Gamma}_i^3 S_i^+ S_n^z\rangle, \tag{20b}$$

$$\frac{d}{dt}\langle S_n^-\rangle = -i\omega_0\langle S_n^- \hat{\Gamma}_n\rangle + 2\gamma_0 \sum_i \langle S_n^z S_i^- \hat{\Gamma}_i^3\rangle, \tag{20c}$$

where $\gamma_0=\pi\eta\omega_0$ is one-half the spontaneous decay rate for an isolated atom.

The relaxation rate of the atomic subsystem

$$\gamma(t) = -\sum_n \frac{d\langle S_n^z\rangle}{dt} = \gamma_0 \sum_{n,i} \langle \hat{\Gamma}_i^3 S_i^+ S_n^- + S_n^+ S_i^- \hat{\Gamma}_i^3\rangle \tag{21}$$

is of primary importance in our subsequent analysis. The parameter $\gamma(t)$ is defined in the right-hand side of Eq. (20a).

By contrast to the case of noninteracting atoms, there is no intimate connection between the relaxation rate $\gamma(t)$ and the total intensity of radiation $I(t)$ which is proportional to $\Sigma_{i,n}\langle S_n^+ S_i^- \hat{\Gamma}_i^4 + S_i^+ \hat{\Gamma}_i^4 S_n^-\rangle$. The reason is that the interatomic interaction renormalizes the frequency of the dipole transition $\tilde{\omega} \propto \omega_0 \hat{\Gamma}$, whereas $\dot{S}^z \propto \tilde{\omega}^3$ and $I \propto \tilde{\omega}^4$.

The right-hand side of Eq. (21) can be conveniently rewritten as a sum of the coherent ($i\neq n$) and incoherent ($i=n$) parts

$$\frac{d}{dt}\langle S_n^z\rangle = -\gamma_0\langle(1+2S_n^z)\hat{\Gamma}_n^3\rangle - \gamma_0 \sum_{i\neq n} \langle \hat{\Gamma}_i^3 S_i^+ S_n^- + S_n^+ S_i^- \hat{\Gamma}_i^3\rangle, \tag{22}$$

where we used the identities $S_i^+ S_i^- = \frac{1}{2} + S_i^z$ and $[S_n^z, \hat{\Gamma}_n]=0$.

In order to decouple correlations in Eq. (20), we use the Bloch representation for the wave functions of two-level atoms $|\Phi\rangle$ [55]

$$|\Phi\rangle = \prod_{j=1}^{N} |\theta_j, \varphi_j\rangle,$$

$$|\theta_j, \varphi_j\rangle = \sin\frac{\theta_j}{2} e^{-i\varphi_j/2}|\uparrow\rangle_j + \cos\frac{\theta_j}{2} e^{i\varphi_j/2}|\downarrow\rangle_j \tag{23}$$

and obtain the closed system of equations for the averages of atomic variables $\langle S_j^z\rangle = -\frac{1}{2}\cos\theta_j$, $\langle S_j^\pm\rangle = \frac{1}{2}\sin\theta_j e^{\pm i\varphi_j}$ of the following form:

$$\frac{d}{d\tau}\langle S_n^z\rangle = -(1+2\langle S_n^z\rangle)\langle\hat{\Gamma}_n^3\rangle - (\langle S_{n+1}^+\rangle\langle S_n^-\rangle + \langle S_n^+\rangle\langle S_{n+1}^-\rangle)\langle\hat{K}_{n+1}\rangle$$
$$-(\langle S_{n-1}^+\rangle\langle S_n^-\rangle + \langle S_n^+\rangle\langle S_{n-1}^-\rangle)\langle\hat{K}_{n-1}\rangle$$
$$-\sum_{i\neq n,n\pm1}\langle\hat{\Gamma}_i^3\rangle(\langle S_n^+\rangle\langle S_i^-\rangle + \langle S_i^+\rangle\langle S_n^-\rangle),$$

$$\frac{d}{d\tau}\langle S_n^\pm\rangle = (\pm i\alpha\langle\hat{\Gamma}_n\rangle - \langle\hat{\Gamma}_n^3\rangle)\langle S_n^\pm\rangle + 2\langle\hat{E}_{n+1}\rangle\langle S_{n+1}^\pm\rangle + 2\langle\hat{E}_{n-1}\rangle$$
$$\times\langle S_{n-1}^\pm\rangle + 2\sum_{i\neq n,n\pm1}\langle S_n^z\rangle\langle S_i^\pm\rangle\langle\hat{\Gamma}_i^3\rangle, \quad (24)$$

where $\tau=\gamma_0 t$, $\alpha=\omega_0/\gamma_0\gg 1$, and

$$\langle\hat{\Gamma}_i^3\rangle = 1-\beta(3+\beta^2)(\langle S_{n-1}^z\rangle + \langle S_{n+1}^z\rangle) + \frac{3\beta^2}{2}(1+4\langle S_{n-1}^z\rangle\langle S_{n+1}^z\rangle), \quad (25)$$

$$\langle\hat{K}_{n\pm1}\rangle = 1+\beta(3+3\beta+\beta^2)\left(\frac{1}{2}-\langle S_{n\pm2}^z\rangle\right), \quad (26)$$

$$\langle\hat{E}_{n\pm1}\rangle = \langle\hat{\Gamma}_{n\pm1}^3 S_n^z\rangle = \langle S_n^z\rangle - \beta(3+\beta^2)\left(\frac{1}{4}+\langle S_{n\pm2}^z\rangle\langle S_n^z\rangle\right) + \frac{3\beta^2}{2}(1+\langle S_{n\pm2}^z\rangle). \quad (27)$$

There is a rapidly oscillating term $i\alpha\langle\hat{\Gamma}_n\rangle\langle S_n^\pm\rangle$ in the system of equations (24). In contrast to the case of noninteracting atoms with $\hat{\Gamma}_i\equiv 1$, this term cannot be removed by the phase shift $\langle S_z^\pm\rangle\to e^{\pm i\alpha\tau}\langle S_z^\pm\rangle$. In Appendix C we apply the method of multitime scales [56,57] to eliminate the rapidly oscillating terms and deduce the following equations for slowly varying atomic amplitudes:

$$\sigma_n^z(\tau) = \overline{[\langle S_n^z\rangle(\tau,\tau')]}_{\tau'}, \quad (28)$$

$$\sigma_n^\pm(\tau) = \overline{\left[\langle S_n^\pm\rangle(\tau,\tau')\exp\left(\mp i\int_0^{\tau'}dt''\langle\hat{\Gamma}_n\rangle(\tau,t'')\right)\right]}_{\tau'},$$

where $\overline{(\cdots)}_{\tau'}$ denotes averaging over the time $\tau'=\alpha\tau$ of fast motion,

$$\frac{d}{d\tau}\sigma_n^z = -(1+2\sigma_n^z)\Gamma_n^3 - (\sigma_{n+1}^+\sigma_n^- w_{n+1,n} + \sigma_n^+\sigma_{n+1}^- w_{n,n+1})K_{n+1}$$
$$-(\sigma_{n-1}^+\sigma_n^- w_{n-1,n} + \sigma_n^+\sigma_{n-1}^- w_{n,n-1})K_{n-1}$$
$$-\sum_{i\neq n,n\pm1}\Gamma_i^3(\sigma_n^+\sigma_i^- w_{n,i} + \sigma_i^+\sigma_n^- w_{i,n}),$$

$$\frac{d}{d\tau}\sigma_n^+ = -\sigma_n^+\Gamma_n^3 + 2E_{n+1}\sigma_{n+1}^+ w_{n+1,n} + 2E_{n-1}\sigma_{n-1}^+ w_{n-1,n} + 2\sum_{i\neq n,n\pm1}\Gamma_i^3\sigma_i^+\sigma_n^z w_{i,n},$$

$$\sigma_i^- = (\sigma_i^+)^*, \quad (29)$$

where

$$w_{i,j} = -i\frac{e^{i2\pi(\Gamma_i-\Gamma_j)}-1}{2\pi(\Gamma_i-\Gamma_j)}$$

and functions

$$\Gamma_n = 1-\beta(\sigma_{n+1}^z+\sigma_{n-1}^z), \quad (30)$$

$$\Gamma_n^3 = 1-\beta(3+\beta^2)(\sigma_{n+1}^z+\sigma_{n-1}^z) + \frac{3}{2}\beta^2(1+4\sigma_{n+1}^z\sigma_{n-1}^z), \quad (31)$$

$$K_{n\pm1} = 1+\beta(3+3\beta+\beta^2)\left(\frac{1}{2}-\sigma_{n\pm2}^z\right), \quad (32)$$

$$E_{n\pm1} = \sigma_n^z - \frac{1}{4}\beta(3+\beta^2)(1+4\sigma_{n\pm2}^z\sigma_n^z) + \frac{3}{2}\beta^2(1+\sigma_{n\pm2}^z) \quad (33)$$

describe correlations due to the direct interatomic interaction.

A. Order parameter and superradiance transition

It is known [21] that relaxation of the atomic subsystem essentially depends on the number of atoms N. For noninteracting atoms ($J=0$ and $\hat{\Gamma}_i\equiv 1$), the first term on the right-hand side of Eq. (22) describes processes of spontaneous incoherent decay.

Transition to the regime of superradiant relaxation, where correlations are caused by interaction with the electromagnetic field in the vacuum state, occurs when the term describing the coherent part in Eq. (22) (a sum with $i\neq n$) starts to play the dominating role. Such a transition can be conveniently described using the order parameter of the form (see, e.g., [58]):

$$C = \lim_{T\to\infty}\frac{1}{T}\int_0^T dt\frac{\gamma_{\rm coh}(t)}{\gamma_{\rm incoh}(t)} = \lim_{T\to\infty}\frac{1}{T}\int_0^T dt\frac{\sum_j^N\sum_{i\neq j}^N\langle\hat{\Gamma}_i^3 S_i^+ S_j^- + S_j^+ S_i^-\hat{\Gamma}_i^3\rangle(t)}{\sum_i^N\langle S_i^+ S_i^-\hat{\Gamma}_i^3 + \hat{\Gamma}_i^3 S_i^+ S_i^-\rangle(t)} \quad (34)$$

and takes place if the number of atoms N exceeds its critical value N_c [21].

The presence of the factor $\hat{\Gamma}_i=1-\beta(S_{i+1}^z+S_{i-1}^z)$ in Eq. (34) indicates that the direct interaction introduces additional correlations into both the coherent and the incoherent parts of radiation. It is the effect of such correlations on the incoherent part that is responsible for the regime of pulse radiation [39,40] discussed in Sec. I.

In Fig. 3 we present the results for N dependence of the order parameter

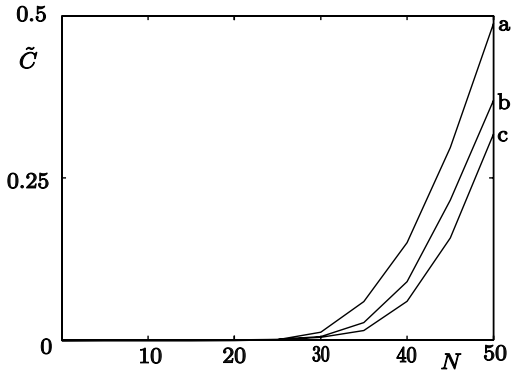

FIG. 3. Order parameter $\tilde{C}$ defined in Eq. (35) as a function of the number of atoms N. The transition to the superradiant regime is shown for three different values of the ratio $\beta=J/\hbar\omega_0$: (a) $\beta=0$, (b) $\beta=0.5$, and (c) $\beta=0.9$.

$$C \approx \tilde{C} = \lim_{T\to\infty} \frac{1}{T} \int_0^T dt \frac{\sum_j^N \sum_{i\neq j}^N (\Gamma_i^3 \sigma_i^+ \sigma_j^- + \sigma_j^+ \sigma_i^- \Gamma_i^3)}{\sum_i^N (1 + 2\sigma_i^z)\Gamma_i^3} \tag{35}$$

computed by solving the system (29) numerically at different values of the interaction parameter $\beta=J/\hbar\omega_0$. The curves show that the critical number of atoms and the supperradiance transition are almost insensible to the interatomic interaction. So, similar to the case of noninteracting atoms, we have two qualitatively different regimes of relaxation: the incoherent regime at $N<N_c$ and the regime of superradiance at $N>N_c$.

B. Collective pulse relaxation

Let us consider the decay of an initially inverted system of interacting atoms at subcritical number of the atoms $N<N_c$ in more detail. Dependence of the relaxation rate on time calculated by solving Eq. (29) with the initial conditions $\langle S_n^z\rangle(0)=1/2$ is shown in Fig. 4.

Referring to Fig. 4, it is seen that, in the absence of interaction ($J=0$ and $\hat{\Gamma}_i \equiv 1$), relaxation occurs as an incoherent spontaneous decay of excited atoms. This regime is characterized by a monotonic exponential decrease of the relaxation rate.

When the interaction parameter β increases, spontaneous processes are suppressed and the time dependence of the relaxation rate reveals a nonmonotonic behavior with a pronounced peak (see Fig. 4). This is the regime of collective pulse relaxation induced by the direct interatomic interaction.

Collective pulse relaxation can be easily described using the mean field approximation for decoupling of correlations. To this end we assume that, for a small number of atoms, the relaxation rate (22) is predominantly determined by the first term (incoherent part) describing incoherent mechanism of decay. So, Eq. (22) takes the simplified form

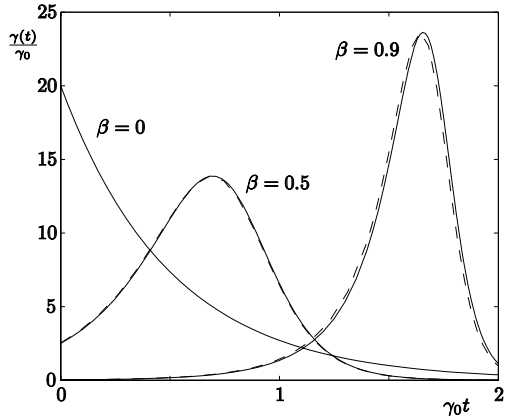

FIG. 4. Relaxation rate $\gamma(t)$ defined in Eq. (21) as a function of time for ten interacting atoms at different values of the interaction parameter $\beta=J/\hbar\omega_0$. Solid lines represent the numerical results computed by solving the system (18). The dashed line is computed from the solution of Eq. (39).

$$\frac{d}{dt}\langle S_n^z\rangle \approx -\gamma_0\langle(1+2S_n^z)\hat{\Gamma}_n^3\rangle \approx -\gamma_0(1+2\langle S_n^z\rangle)\langle\hat{\Gamma}_n^3\rangle, \tag{36}$$

where

$$\langle\hat{\Gamma}_n^3\rangle = 1 - \beta(3+\beta^2)(\langle S_{n+1}^z\rangle + \langle S_{n-1}^z\rangle) + \frac{3}{2}\beta^2(1+4\langle S_{n+1}^z\rangle\langle S_{n-1}^z\rangle). \tag{37}$$

As opposed to the case of noninteracting atoms where Eqs. (36) with $\beta=0$ are one-particle and describe spontaneous independent atomic decay, at $\beta\neq 0$ the interparticle interaction enters Eqs. (36) thus describing a collective atomic dynamics.

We can now substitute the ansatz

$$\langle S_n^z\rangle = \langle\tilde{S}^z\rangle + \delta\langle\tilde{S}_n^z\rangle, \tag{38}$$

assuming that $\langle\tilde{S}^z\rangle \gg \delta\langle\tilde{S}_n^z\rangle$, into Eq. (36) and use the relation to derive the equation governing the dynamics of the atomic subsystem

$$\frac{d\langle\tilde{S}^z\rangle}{dt} = -\gamma_0(1+2\langle\tilde{S}^z\rangle)\langle\Gamma^3\rangle, \quad \langle\tilde{S}^z\rangle(0) = \frac{1}{2}, \tag{39}$$

where

$$\langle\Gamma^3\rangle \approx 1 - 2\beta(3+\beta^2)\langle\tilde{S}^z\rangle + \frac{3}{2}\beta^2(1+4\langle\tilde{S}^z\rangle^2). \tag{40}$$

It can be shown that the mean field solution $\langle\tilde{S}^z\rangle(t)$ is a steplike function of time and is stable at $|\langle\tilde{S}^z\rangle| \gg |\delta\langle\tilde{S}_n^z\rangle|$. The curves calculated in the mean field approximation for the relaxation rate

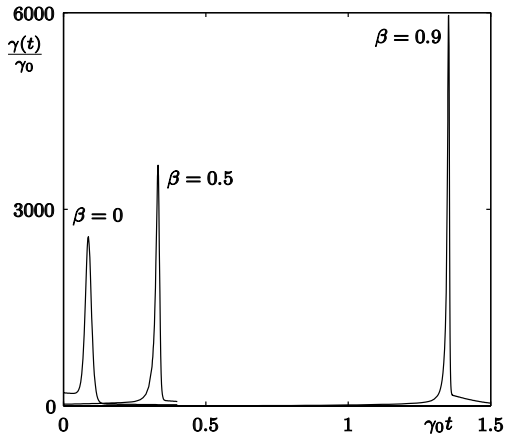

FIG. 5. Relaxation rate $\gamma(t)$ defined in Eq. (21) as a function of time for 100 interacting atoms at different values of the interaction parameter $\beta=J/\hbar\omega_0$ ($2\gamma_0$ is the spontaneous decay rate for an isolated atom). The curves are computed by solving the system (18) numerically.

$$\gamma(t) = -\sum_n \frac{d\langle S_n^z\rangle}{dt} \approx -N\frac{d\langle \tilde{S}^z\rangle}{dt}$$

are shown in Fig. 4 as dashed lines. It can be seen that the results are in excellent agreement with the data of numerical analysis.

It should be noted that the peak intensity is proportional to the number of atoms N which is a consequence of a short-range character of interatomic interaction. By contrast, for the regime of superradiance, the intensity is typically proportional to N^2.

So, we have shown that the direct Ising-type interaction has a synchronizing effect on the system behavior. The result is that the regime of incoherent spontaneous decay changes to the regime of collective pulse relaxation characterized by an increase in the radiation time. For initially inverted atomic configuration, the eigenvalue of the operator $\hat{\Gamma}_i$ goes to zero as the parameter β approaches unity. It follows that, in the limiting case with $\beta\to 1$, the relaxation rate γ of one-atom one-photon transitions Eq. (21) vanishes and, as a result, the radiation time increases.

C. Enhancement of superradiance

As it was previously discussed in Sec. IV A, the effect of superradiance dominates the regime of relaxation of the atomic subsystem at supercritical values of the number of atoms, $N>N_c$. Figure 5 presents the time dependence of the relaxation rate for $N=100$ at various values of the interaction parameter β.

At $\beta=0$, the peak with the intensity proportional to N^2 corresponds to the superradiance effect in the system of noninteracting atoms. In this system interatomic correlations are solely due to interaction between atoms and electromagnetic field.

Synchronizing effect of the Ising-type interaction characterized by the parameter β manifests itself in enhancement of superradiance and an increase in the peak intensity.

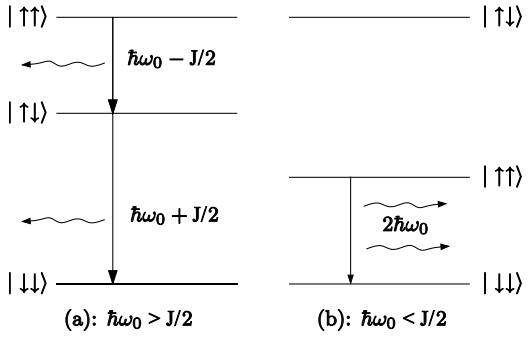

FIG. 6. Energy spectrum of two interacting atoms with $H_A=\hbar\omega_0(S_1^z+S_2^z)-JS_1^zS_2^z$.

The effect of enhancement can be analyzed in the mean field approximation by applying the procedure described in the previous section to the coherent part (the second term) of the relation (22). So, the equation for the average of atomic population $\langle\tilde{S}^z\rangle$ is now given by

$$\frac{d\langle\tilde{S}^z\rangle}{dt} \approx -\gamma_0 N\left(\frac{3}{2} - 2\langle\tilde{S}^z\rangle^2\right)\langle\Gamma^3\rangle, \tag{41}$$

where $\langle\Gamma^3\rangle$ is given in Eq. (40) and the identity (the conservation law for the pseudo-spin) $\frac{1}{2}(S^+S^-+S^-S^+)+(S^z)^2=\frac{3}{4}$ is used.

The peak of the relaxation rate $\gamma(t)$ is at $\langle\tilde{S}^z\rangle\approx 0$ and Eq. (41) combined with the relation (40) provide an estimate for the gain factor $\langle\Gamma^3\rangle|_{\langle\tilde{S}^z\rangle=0}\propto(1+\frac{3}{2}\beta^2)$. As for the case of $N<N_c$, the interatomic interaction delays the time of radiation.

V. REGIME OF MULTIPHOTON RELAXATION

It is known that interaction between atoms at interatomic spacings smaller than the wavelength may bring about multiphoton processes in the atomic system [42,43]. At weak interaction with $J<\hbar\omega_0$, probability of multiphoton transitions is much smaller than that of single-photon transitions. So, the first order of the perturbative expansion over the atom-field interaction can be used to describe dynamical behavior of the system. At strong interaction with $J>\hbar\omega_0$, single-photon processes are nonresonant and multiphoton transitions start to play an increasingly important role. This is the so-called regime of multiphoton relaxation.

In order to illustrate how this regime may occur we qualitatively consider a model system of two two-level atoms with the Hamiltonian $H_A=\hbar\omega_0(S_1^z+S_2^z)-JS_1^zS_2^z$. Its energy spectrum is schematically represented in Fig. 6.

If the Hamiltonian of atom-field interaction has the form of a sum $H_{int}=H_+e^{i\omega t}+H_-e^{-i\omega t}$, where H_+ (H_-) describes photon emission (absorption), then the second order perturbative expression for the probability of two-photon transition $|0\rangle|\uparrow\uparrow\rangle\to|2\rangle|\downarrow\downarrow\rangle$ is given by

$$W^{(2)} = \frac{2\pi}{\hbar^2}\left|\frac{\langle\downarrow\downarrow|H_+|\uparrow\downarrow\rangle\langle\uparrow\downarrow|H_+|\uparrow\uparrow\rangle}{\hbar\omega_0(1-\beta/2)-\hbar\omega} + \frac{\langle\downarrow\downarrow|H_+|\downarrow\uparrow\rangle\langle\downarrow\uparrow|H_+|\uparrow\uparrow\rangle}{\hbar\omega_0(1-\beta/2)-\hbar\omega}\right|^2 \delta(2\omega_0-2\omega). \tag{42}$$

For strong interaction with $J > \hbar\omega_0$, the energy level of one-particle excited states, $|\uparrow\downarrow\rangle$ and $|\downarrow\uparrow\rangle$, is higher than the energy of the state $|\uparrow\uparrow\rangle$ [see Fig. 6(b)], so that one-photon relaxation of the two-particle excited state is excluded by the energy conservation law. As a consequence, relaxation occurs as a two-photon process.

Probability of multiphoton transitions for N excited atoms in free space is small because its magnitude is determined by the Nth order term of perturbative expansion over the atom-field coupling constant $g_{\mathbf{k}s,i}$. So, we arrive at the conclusion that the effect of interatomic interaction can be the formation of long living excited states in the system.

This conclusion, however, is not strictly valid if the density of electromagnetic modes has a singularity near the energy of atomic transition, $\hbar\omega_0$. An example is a high quality resonant cavity. In this case, dynamical behavior of the atom-field system is characterized by Rabi oscillations involving many-particle and multiphoton states [44].

We demonstrate this effect for the two atomic system embedded into a high-Q single-mode resonant cavity. The Hamiltonian of the model is

$$H = \hbar\,\omega_0 a^\dagger a + \hbar\,\omega_0(S_1^z + S_2^z) - JS_1^zS_2^z + \hbar\, g[a(S_1^+ + S_2^+) + a^\dagger(S_1^- + S_2^-)], \tag{43}$$

where a the $(a^\dagger)$ is the photon annihilation (creation) operator for the cavity mode, and g is the coupling constant of the interaction between atoms and the cavity mode.

The model (43) is exactly solvable and the wave function for the system initially prepared in the state $|\psi(0)\rangle = |\uparrow\uparrow, n\rangle$, where n is the number of photons, can be written in the explicit form:

$$|\psi(t)\rangle = e^{-i\omega_0(n+1)t}\Bigg\{\Bigg[\frac{n+2}{2n+3}e^{iJ't} + g^2\frac{n+1}{D}\left(\frac{e^{iDt}}{D-J'} + \frac{e^{-iDt}}{D+J'}\right)\Bigg]|\uparrow\uparrow, n\rangle + \sqrt{(n+1)(n+2)}\Bigg[\frac{g^2}{D}\left(\frac{e^{iDt}}{D-J'} + \frac{e^{-iDt}}{D+J'}\right) - \frac{e^{iJ't}}{2n+3}\Bigg] \times|\downarrow\downarrow, n+2\rangle - i\frac{g\sqrt{n+1}}{D}\sin Dt(|\downarrow\uparrow, n+1\rangle + |\uparrow\downarrow, n+1\rangle)\Bigg\}, \tag{44}$$

where

$$J' = J/4\hbar, \quad D = \sqrt{(J')^2 + 2g^2(2n+3)}.$$

In the absence of the interatomic interaction $(J=0)$, the amplitudes of one- and two-photon processes are of the same order of magnitude. But, at strong interaction with $J \gg \hbar g\sqrt{n}$, the two-particle two-photon amplitudes dominate and the wave function (44) can be approximated as follows:

$$|\psi(t)\rangle \approx e^{-i[\omega_0(n+1)-J'-\Delta]t}\Bigg\{\Bigg[\frac{n+1}{2n+3}e^{i\Delta t} + \frac{n+2}{2n+3}e^{-i\Delta t}\Bigg]|\uparrow\uparrow, n\rangle + 2i\frac{\sqrt{(n+1)(n+2)}}{2n+3}\sin\Delta t|\downarrow\downarrow, n+2\rangle\Bigg\}, \tag{45}$$

where

$$\Delta = \frac{g^2}{2J'}(2n+3),$$

and for $n \gg 1$

$$|\psi(t)\rangle \approx e^{-i(\omega_0 n - J')t}(\cos\Delta t|\uparrow\uparrow, n\rangle + i\sin\Delta t|\downarrow\downarrow, n+2\rangle).$$

This means the build up of two-atomic two-photon Rabi oscillations and can be used to generate a Schrödinger catlike entangled atom-field state.

Thus interparticle interaction may give rise to the regime of many-particle multiphoton dynamics. When interaction is strong as compared with the energy of atomic transition, inhibition of one-photon processes in the system is accompanied by transition from the regime of cooperative pulse radiation (superradiance) to the generation of the Fock state of light. In a cavity, multiphoton dynamical effects come into play under the condition $J \gg \hbar g\sqrt{n}$, so that the level of intermediate energy is essentially shifted away from one-photon resonance (see Fig. 6).

VI. DISCUSSION AND CONCLUSIONS

We studied effects of direct interatomic interaction on collective processes in atom-photon dynamics using, as an example, a simple model of two-level atoms with Ising interaction of ferromagnetic type. We have found that this interaction influences radiation processes of the atomic ensemble acting as an additional synchronizing factor.

For weakly interacting atoms at $J < \hbar\omega_0$, we have shown that interatomic interaction results in inhibition of incoherent spontaneous decay of atoms and dynamical behavior of the system is governed by the regime of collective pulse relaxation. This regime, though it bears a resemblance to superradiance, has nothing to do with the effect of phase synchronization induced by fluctuations of the electromagnetic field. For example, in solid state structures, collective pulse relaxation is caused by inelastic exciton-exciton scattering and is characterized by quadratic dependence of the radiation peak on the number of particles [39–41].

We have also found that interaction induced synchronization enhances superradiance. In the presence of interparticle interaction collective pulse radiation and enhanced superradiance are both characterized by an increase in the delay time of emission.

At the end of Sec. II B, we have pointed out that when the excited state is not fully inverted its relaxation can be determined by transitions with frequencies equal to the atomic resonant frequency ω_0. If we consider nonexcited atoms of the initial state as defects of the atomic chain, such a relaxation scheme can be referred to as the solitonic mechanism of relaxation. The solitonic mechanism implies that in the

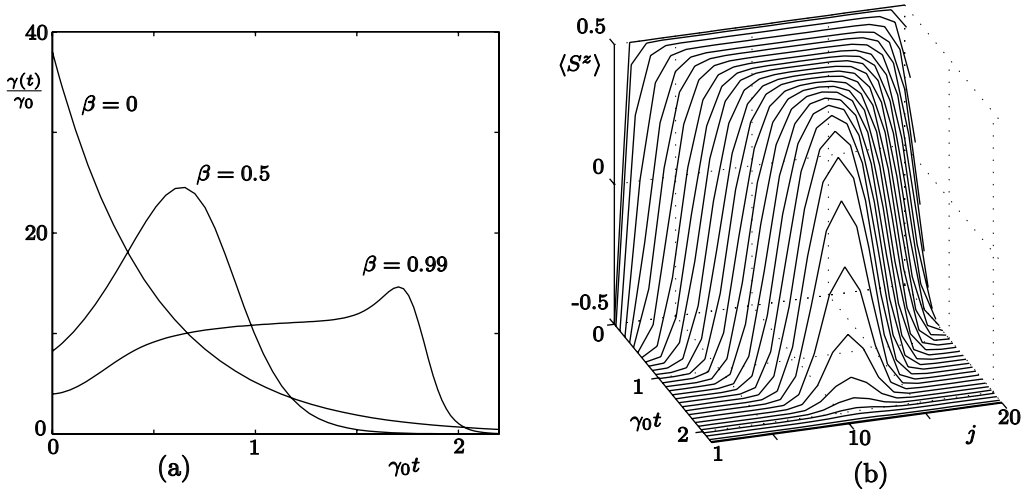

FIG. 7. (a) Relaxation rate $\gamma(t)$ as a function of time at different values of the interaction parameter β. (b) Averaged population of ith atom $\langle S_j^z \rangle$ as a function of time and atomic number i at β=0.99. The curves are computed by numerically solving Eq. (39) for a ring of 20 atoms. One atom is initially in the ground state representing a point defect in the atomic ring. At sufficiently strong interaction, relaxation of the atomic ring clearly demonstrates the solitonic mechanism when atoms undergo transition to the ground state consecutively one after another.

course of relaxation atoms undergo transition to the ground state successively one after another. So, such behavior can be interpreted as a defect motion. More generally, since the system of equations (29) governing dynamics of atoms at $J < \hbar\omega_0$ is similar in structure to the Volterra system and the Toda lattice [61], it might be expected that the system possesses solitonlike solutions.

Figure 7 presents the numerical results for the excited state with one initially nonexcited atom. The number of atoms is small, $N < N_c$, and the relaxation rate is computed from the mean field equation (39).

Referring to Fig. 7, it is seen that relaxation of the atomic ring can be described as motion of the defect (Bloch wall). When the interaction parameter β approaches the limit of strong interaction, the relaxation rate $\gamma(t)$ assumes the kinklike form and is determined by defect velocity. The peak is caused by collective pulse relaxation indicating that the retardation time of emission is too short for the defect to travel through the ring.

However, it should be noted that we have neglected the dipole-dipole interaction by considering permutationally invariant atomic configurations, the approximation that is not valid in the presence of a defect in the system. This interaction may have a destructive effect on the solitonic mechanism of relaxation.

By contrast to the case of weakly interacting atoms where dynamics of the atomic subsystem is governed by one-particle one-photon transitions, at strong interatomic interaction with $J \gg \hbar\omega_0$, these transitions appear to be excluded. In this case multiphoton transitions will determine relaxation of the excited atomic subsystem. This is what we called the regime of multiphoton relaxation.

The regime is characterized by transition from generation of superradiant pulse to generation of the Fock quantum state of light. We have used a simple model of two atoms in a high-Q single mode cavity to show that such transition is accompanied by Rabi oscillations involving many-atom multiphoton states. In other words, it means generating a many-particle entangled atom-field state [59].

Interestingly, transition to multiphoton dynamics is analogous to the Mott-insulator quantum phase transition in optically trapped atomic systems where a Fock state characterizing the number of localized atoms is formed [22,45]. In our case, the energy of atomic transition $\hbar\omega_0$ and the interatomic coupling J play the role of kinetic and potential energy, respectively. So, the transition to generation of the Fock state of light takes place when the potential energy J becomes greater than $\hbar\omega_0$.

In lattice atomic systems with inelastic tunneling transitions between neighboring wells, the Mott or Peierls transitions and the transition to multiphoton relaxation can be related to each other. We illustrate such a possibility by the simple example of a one-dimensional periodic chain of potential wells, Fig. 8.

In the half-filling case, when there are half as many atoms as there are wells and the energy of repulsion between neighboring particles J is much larger than the hopping energy of tunneling between neighboring wells ε_t, the ground state for identical wells should be degenerate in energy, which corresponds to the atoms localized in either odd or even wells, see Fig. 8(a). Such ordering of particles is similar to the Wigner crystal or the charge density waves observed in low-dimensional conductors [63,64]. The possibility of the density wave Mott-insulator phase for atoms embedded into an optical lattice was also discussed in Ref. [25]. These ordered states are separated from the excited ones by the gap of width $\sim J$.

Local tunneling transitions may lift this degeneracy of the ground state. In this case, at $J > \varepsilon_t$, the ground state can be described, at least for a short chain, as a many-particle entangled state of the Schrödinger cat type, i.e., $\approx \frac{1}{\sqrt{2}}(|\uparrow\uparrow\cdots\uparrow\rangle \pm |\downarrow\downarrow\cdots\downarrow\rangle)$ (the so-called GHZ state [60]), associated with many-particle tunneling oscillations between odd and even wells (see, e.g., Ref. [35]), which is similar to

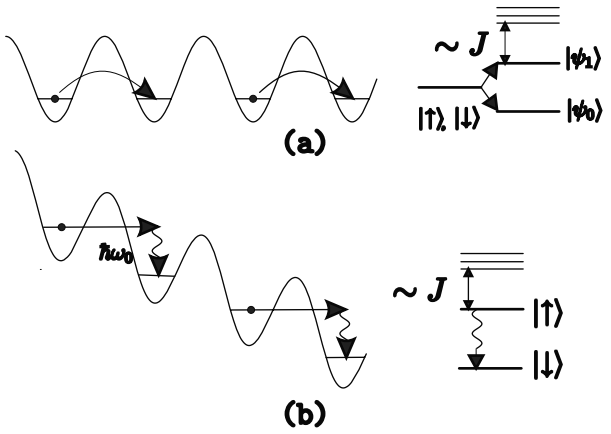

FIG. 8. (a) Interacting particles embedded into potential lattice in the case of half-filling. Ground state is double degenerate corresponding to the particles localized in either odd or even wells ($|\uparrow\rangle=|\uparrow\uparrow\cdots\uparrow\rangle$ and $|\downarrow\rangle=|\downarrow\downarrow\cdots\downarrow\rangle$). Tunneling transitions lift degeneracy so that the two lowest levels correspond to the many-particle entangled states $|\psi_{0,1}\rangle\approx\frac{1}{\sqrt{2}}(|\uparrow\rangle\pm|\downarrow\rangle)$. Excited states are separated by the gap of the width $\propto J$. (b) For energetically nonequivalent wells, tunneling transitions are inelastic. For $J\gg\hbar\omega_0$, they can be accompanied by multiphoton processes.

the tunneling creep of charge density waves [64].

If the wells are energetically inequivalent, Fig. 8(b), the tunneling many-particle transitions from the excited state $|\uparrow\rangle=|\uparrow\uparrow\cdots\uparrow\rangle$ are inelastic and can be accompanied by multiphoton processes or the cooperative dynamic regimes discussed above.

But our model is oversimplified and, strictly speaking, cannot be applied to inelastic tunneling transitions of atoms between energetically different wells. The Hamiltonian of atoms H_A need to be modified to take into account overlapping of the particle wave function in the neighboring well. This requires additional terms proportional to S^x. Symmetry of interaction between particles and field E also has to be of more complicated form $H_{int}\propto E(d_0+d_xS^x+d_zS^z)$. Another limitation of our model is neglecting multipole transitions that, in the case of strong interaction, may compete with multiphoton transitions [13,50].

Among important omissions in this paper are spectral characteristics of the radiation. In our case there are only two modes with the frequencies $\hbar\omega_0\pm J$. We also have not discussed superradiant and subradiant Dicke states. These have the standard form because the Ising-type interparticle interaction makes the atomic spectrum nonequidistant but does not affect symmetry of these states.

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APPENDIX A: THE BORN-MARKOV APPROXIMATION

In this section we concentrate on the case of weakly interacting atoms with $J<\hbar\omega_0$ and describe the dissipative dynamics of the atomic subsystem using the Born approximation. To this end we apply to the master equation (6) the standard technique of elimination of bosonic variables [52].

For simplicity, we consider the zero temperature case and choose an initial state with no correlations between the atomic subsystem and the vacuum field. So, the density operator can be taken in the factorized form (10).

Substituting Eq. (10) into Eq. (6) and taking the trace over the field variables, we obtain

$$\frac{d}{dt}\tilde{\rho}(t)=\eta\sum_{i,j}\int_0^t d\tau\int_0^\infty d\omega\omega^3F_{ij}(\omega)\times(e^{i\omega\tau}([R_j(t),\tilde{\rho}(t-\tau)S_i^+e^{i\omega_0\hat{\Gamma}_i(t-\tau)}]+[R_j(t),\tilde{\rho}(t-\tau)S_i^-e^{-i\omega_0\hat{\Gamma}_i(t-\tau)}])+\text{H.c.}),\quad\text{(A1)}$$

where the sum over $\mathbf{k}s$ is transformed into the integral form [53]

$$\sum_{\mathbf{k}s}\rightarrow\frac{V}{(2\pi c)^3}\int_0^\infty d\omega\omega^2\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\varphi\sum_s,\quad\text{(A2)}$$

and the functions $R_j(t)$, $F_{ij}(\omega)$, $\tilde{\rho}(t)$, and η are described by Eqs. (12)–(14).

The next step is to employ the Markov approximation. This approximation implies that a reservoir relaxation time is much shorter than a time scale of the atomic subsystem evolution, so that $\tilde{\rho}(t-\tau)$ can be replaced by $\tilde{\rho}(t)$ in the right-hand side of Eq. (A1) and the upper limit of the integral can be extended to infinity.

For $J<\hbar\omega_0$, the eigenvalues of the operators $\hat{\Gamma}_i$ are all positive. Using the relation

$$\lim_{t\rightarrow\infty}\int_0^t d\tau e^{\pm ix\tau}=\pi\delta(x)\pm\text{P}\frac{i}{x},\quad\text{(A3)}$$

where the symbol P stands for the Cauchy principal value of the integral, we can perform the integral in Eq. (A1) and derive Eq. (11). We have used the operator functions defined on the eigenvalues of $\hat{\Gamma}_i$ so that if $\hat{\Gamma}_i|\psi\rangle=\Gamma_i|\psi\rangle$ then, e.g., $\delta(\omega-\omega_0\Gamma_i)|\psi\rangle=\delta(\omega-\omega_0\hat{\Gamma}_i)|\psi\rangle$.

This result can also be obtained directly by representing the terms $\exp(\pm i\omega_0\hat{\Gamma}_i\tau)$ that enter Eq. (A1) as follows:

$$e^{\pm i\omega_0\hat{\Gamma}_i\tau}=(1-\hat{U}_i)e^{\pm i\omega_0\tau}+\frac{1}{2}(\hat{U}_i+\hat{\Omega}_i)e^{\pm i\omega_0(1-\beta)\tau}+\frac{1}{2}(\hat{U}_i-\hat{\Omega}_i)e^{\pm i\omega_0(1+\beta)\tau},\quad\text{(A4)}$$

where

$$\hat{U}_i=\frac{1}{2}(1+4S_{i+1}^zS_{i-1}^z),\quad\hat{\Omega}_i=S_{i+1}^z+S_{i-1}^z.\quad\text{(A5)}$$

The principal values of the integrals in Eq. (11) with $i\neq j$ can be estimated as

$$\mathrm{P}\int_0^\infty d\omega\omega^3 \frac{F_{ij}(\omega)}{\omega+\omega_0\hat{\Gamma}_i} = \frac{3}{2}(\omega_0\hat{\Gamma}_i)^3\left\{\left[\frac{1-(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2}{k_0\hat{\Gamma}_i r_{ij}} - \frac{1-3(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2}{(k_0\hat{\Gamma}_i r_{ij})^3}\right]A(k_0\hat{\Gamma}_i r_{ij}) + \frac{[1-3(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]B(k_0\hat{\Gamma}_i r_{ij}) - [1-(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]}{(k_0\hat{\Gamma}_i r_{ij})^2}\right\}, \tag{A6a}$$

$$\mathrm{P}\int_0^\infty d\omega\omega^3 \frac{F_{ij}(\omega)}{\omega-\omega_0\hat{\Gamma}_i} = \frac{3\pi}{2}(\omega_0\hat{\Gamma}_i)^3\left\{[1-(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]\frac{\cos(k_0\hat{\Gamma}_i r_{ij})}{k_0\hat{\Gamma}_i r_{ij}} - [1-3(\bar{\mathbf{d}}\cdot\bar{\mathbf{r}}_{ij})^2]\left[\frac{\sin(k_0\hat{\Gamma}_i r_{ij})}{(k_0\hat{\Gamma}_i r_{ij})^2} + \frac{\cos(k_0\hat{\Gamma}_i r_{ij})}{(k_0\hat{\Gamma}_i r_{ij})^3}\right]\right\} - \mathrm{P}\int_0^\infty d\omega\omega^3 \frac{F_{ij}(\omega)}{\omega+\omega_0\hat{\Gamma}_i}, \tag{A6b}$$

where $k_0=\omega_0/c$,

$$A(x) = \sin(x)\mathrm{ci}(x) - \cos(x)\mathrm{si}(x),$$

$$B(x) = \sin(x)\mathrm{si}(x) + \cos(x)\mathrm{ci}(x), \quad x>0,$$

and $\mathrm{si}(x)=\int_\infty^x \frac{\sin t}{t}dt$ and $\mathrm{ci}(x)=\int_\infty^x \frac{\cos t}{t}dt$ [62].

In the limit $r_{ij}\omega/c\to 1$, Eq. (A6) assumes the asymptotical form (15) coincident with the standard expression for the dipole-dipole interaction that does not depend on the coupling constant of Ising interaction J.

APPENDIX B: TWO INTERACTING ATOMS: EXACT SOLUTION OF THE MASTER EQUATION

For the case of two noninteracting atoms with $J=0$, the exact solution of the master equation (18) was previously obtained in Ref. [54]. It was shown that the decay of initially excited atoms demonstrates the superradiant regime and the dipole-dipole interaction does not influence the cooperative behavior of atoms.

In this section we show that the master equation (18) for two Ising-like interacting atoms, $J\neq 0$, can be solved along similar lines. The Hamiltonian of two interacting atoms is given by

$$H_A = \hbar\,\omega_0(S_1^z + S_2^z) - JS_1^zS_2^z.$$

In the basis of atomic states $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$ the density operator ρ can be written in the matrix form

$$\rho = \begin{pmatrix} \rho_{11} & 0 & 0 & 0 \\ 0 & \rho_{22} & \rho_{23} & 0 \\ 0 & \rho_{32} & \rho_{33} & 0 \\ 0 & 0 & 0 & \rho_{44} \end{pmatrix} \tag{B1}$$

and Eq. (18) reduces to the system

$$\dot{\rho}_{11} = -4m\rho_{11},$$

$$\dot{x}_0 = 4m\rho_{11} - 2n(x_0+x_1),$$

$$\dot{x}_1 = 4m\rho_{11} - 2n(x_1+x_0),$$

$$\dot{x}_2 = -2nx_2 + i2\Omega x_3,$$

$$\dot{x}_3 = i2\Omega x_2 - 2nx_3,$$

$$\dot{\rho}_{44} = 2n(x_0+x_1), \tag{B2}$$

where $x_0=\rho_{22}+\rho_{33}$, $x_1=\rho_{23}+\rho_{32}$, $x_2=\rho_{23}-\rho_{32}$, $x_3=\rho_{22}-\rho_{33}$, $n=\gamma_0(1+\beta/2)^3$, $m=\gamma_0(1-\beta/2)^3$, and $\Omega_{12}=\Omega_{21}=\Omega$ is the constant of the dipole-dipole interaction.

For the initial conditions $\rho_{11}(0)=1$, $\rho_{22}(0)=\rho_{23}(0)=\rho_{32}(0)=\rho_{33}(0)=\rho_{44}(0)=0$ describing two initially excited atoms, it is easy to write down the solution of Eq. (B2)

$$\rho_{11} = e^{-4mt},$$

$$x_0 = x_1 = \frac{m}{n-m}(e^{-4mt} - e^{-4nt}),$$

$$\rho_{44} = 1 + \frac{1}{m-n}(ne^{-4mt} - me^{-4nt}), \tag{B3}$$

and also $x_2=x_3=0$.

It is seen that similar to the case of noninteracting atoms the solution (B3) and the decay rate of the atomic subsystem

$$\gamma(t) \equiv -\frac{d}{dt}\,\mathrm{Tr}\{\rho(S_1^z + S_2^z)\} = \dot{\rho}_{44} - \dot{\rho}_{11} \tag{B4}$$

are both independent of the constant of the dipole-dipole interaction Ω.

APPENDIX C: ELIMINATION OF RAPIDLY OSCILLATING VARIABLES

In order to eliminate from Eq. (24) the rapidly oscillating terms $i\alpha\langle\hat{\Gamma}_n\rangle\langle S_n^\pm\rangle$ we apply the method of multitime scales [56,57] representing the atomic variables $\langle\vec{S}_n\rangle$ as functions of two time scales $\langle\vec{S}_n\rangle(\tau,\tau')$, where $\tau'=\alpha\tau$ $(\alpha\gg 1)$ is a characteristic time of fast motion. We use the following power series expansion of $\langle S_n^\pm\rangle$ and $\langle S_n^z\rangle$ over the small parameter α^{-1}

$$\langle S_n^{\pm}\rangle(\tau,\tau') = \delta_n^{\pm}(\tau')\tilde{\sigma}_n^{\pm}(\tau,\tau') = \delta_n^{\pm}(\tau')\left(\sigma_n^{\pm}(\tau) + \frac{1}{\alpha}\sigma_{n,1}^{\pm}(\tau)\gamma_n^{\pm}(\tau') + \cdots\right), \tag{C1a}$$

$$\langle S_n^{z}\rangle(\tau,\tau') = \tilde{\sigma}_n^{z}(\tau,\tau') = \sigma_n^{z}(\tau) + \frac{1}{\alpha}\sigma_{n,1}^{z}(\tau)\gamma_n^{z}(\tau') + \cdots, \tag{C1b}$$

where $\gamma_n^{\pm}(\tau')$, $\gamma_n^{z}(\tau')$, and $\delta_n^{\pm}(\tau')$ are rapidly oscillating periodic functions, so that

$$\overline{(\gamma_n^{\pm})}_{\tau'} \equiv \frac{1}{2\pi}\int_0^{2\pi}\gamma_n^{\pm}(\tau')d\tau' = 0.$$

Taking into account that

$$\frac{d}{d\tau} = \frac{\partial}{\partial\tau} + \alpha\frac{\partial}{\partial\tau'} \tag{C2}$$

and substituting the expansion (C1a) into Eq. (24), we deduce the following relation for $\tilde{\sigma}_n^{+}$:

$$\left(\frac{\partial\tilde{\sigma}_n^{+}}{\partial\tau} + \alpha\frac{\partial\tilde{\sigma}_n^{+}}{\partial\tau'}\right)\delta_n^{+} + \alpha\tilde{\sigma}_n^{+}\frac{\partial\delta_n^{+}}{\partial\tau'} = (i\alpha\langle\hat{\Gamma}_n\rangle - \langle\hat{\Gamma}_n^{3}\rangle)\tilde{\sigma}_n^{+}\delta_n^{+} + 2(\langle\hat{E}_{n+1}\rangle\tilde{\sigma}_{n+1}^{+}\delta_{n+1}^{+} + \langle\hat{E}_{n-1}\rangle\tilde{\sigma}_{n-1}^{+}\delta_{n-1}^{+}) + 2\sum_{i\neq n,n\pm1}\langle\hat{S}_n^{z}\rangle\langle\hat{\Gamma}_i^{3}\rangle\tilde{\sigma}_i^{+}\delta_i^{+}. \tag{C3}$$

In order to eliminate the imaginary term in the right-hand side of Eq. (C3) we choose $\delta_n^{+}(\tau')$ in the form

$$\delta_n^{+}(\tau') = \exp\left(i\int_0^{\tau'}\langle\hat{\Gamma}_n\rangle(\tau,\xi)d\xi\right) \approx e^{i\Gamma_n\tau'},$$

where Γ_n is the zero order term of the expansion for $\langle\hat{\Gamma}_n\rangle \times(\tau,\tau')$ given by Eq. (30), to yield the equation

$$\frac{\partial\tilde{\sigma}_n^{+}}{\partial\tau} + \alpha\frac{\partial\tilde{\sigma}_n^{+}}{\partial\tau'} = -\langle\hat{\Gamma}_n^{3}\rangle\tilde{\sigma}_n^{+} + 2(\langle\hat{E}_{n+1}\rangle\tilde{\sigma}_{n+1}^{+}e^{i(\Gamma_{n+1}-\Gamma_n)\tau'} + \langle\hat{E}_{n-1}\rangle\tilde{\sigma}_{n-1}^{+}e^{i(\Gamma_{n-1}-\Gamma_n)\tau'}) + 2\sum_{i\neq n,n\pm1}\langle\hat{S}_n^{z}\rangle\langle\hat{\Gamma}_i^{3}\rangle\tilde{\sigma}_i^{+}e^{i(\Gamma_i-\Gamma_n)\tau'}. \tag{C4}$$

Averaging Eq. (C4) over τ' and retaining only the lowest order of the correlations, we have

$$\frac{d\sigma_n^{+}}{d\tau} = -\Gamma_n^{3}\sigma_n^{+} + 2(E_{n+1}\sigma_{n+1}^{+}w_{n+1,n} + E_{n-1}\sigma_{n-1}^{+}w_{n-1,n}) + 2\sum_{i\neq n,n\pm1}\Gamma_i^{3}\sigma_i^{+}\sigma_n^{z}w_{i,n}, \tag{C5}$$

where $w_{i,j}$ is given by

$$w_{i,j} = \frac{e^{i2\pi(\Gamma_i-\Gamma_j)} - 1}{i2\pi(\Gamma_i - \Gamma_j)}.$$

Equation (C5) is the second equation of the system (29). The complex conjugate of Eq. (C5) gives the equation for σ_n^{-}. Along the same line averaging the equation for $\langle S_n^{z}\rangle$ from Eq. (24) provides the last equation of Eq. (29).

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