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Letter

Super/subradiant second harmonic generation

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Abstract

A scheme for active second harmonics generation is suggested. The system comprises N three-level atoms in ladder configuration, situated into a resonant cavity. The system generates the field whose frequency is twice the frequency of the pumping laser, and the field phase is locked to the phase of the pumping field. It is found that the system can lase in either superradiant or subradiant regime, depending on the number of atoms N. When N passes some critical value the transition from the super to subradiance occurs in a phase-transition-like manner. Stability study of the steady state supports this conclusion. For experimental realization of the super/subradiant second harmonics generation we propose semiconductor quantum well structures, superconducting quantum circuits, and evanescently coupled waveguides in which equally spaced levels relevant to this study exist.

Keywords: superradiant lasing, second harmonic generation, lasers without inversion

(Some figures may appear in colour only in the online journal)

1. Introduction

Traditionally, the second harmonic generation (SHG) is obtained by utilizing the second order ($\chi^{(2)}$) nonlinearity in optical crystals [1, 2]. This demands strong input field intensities and materials with special optical properties. In this paper we report the theoretical development of a novel source for second harmonic generation, free of such demands, which is based on a different physical phenomenon, a field driven super- or sub radiant lasing proposed recently in [3, 4]. A three-level system driven by two lasers, considered in [3], is intrinsically nonlinear with respect to the driving fields [5]. We utilize this nonlinearity to obtain SHG in super/subradiant regimes. Recently developed new semiconductor structures, such as semiconductor quantum wells [6–9], superconducting quantum circuits [10–12], and evanescently coupled waveguides [13, 14] can be designed to have equally spaced levels, which makes them suitable for experimental realization of the super/subradiant SHG. A review of recent studies in nonlinear crystals is given in [15].

The first model of stationary superradiance, a superradiant laser was described by Haake *et al* [16], who considered a model of three-level atoms placed inside a resonant cavity, and pumped with a classical external electromagnetic field. In addition, another 'passive' cavity mode was used to coherently couple one of the non-lasing atomic transition. This 'passive' cavity mode, being adiabatically eliminated, results in nonlinear collective decay. The steady state laser intensity calculated for this model scales as N^2 typical for superradiance. The superradiance obtained in [16] essentially differs from the Dicke superradiance [17] by: (i) it is stationary rather than transient, (ii) the linewidth of the superradiant laser scales as $1/N^2$ which is extremely small compared to the spectral width of Dicke superradiant pulse, (iii) intensity fluctuations of the superradiant laser are essentially squeezed, while those of superfluorescence pulse are close to fluctuations of a coherent state. Since then many theoretical works have been devoted to study this model as well as some other models [18–25].

The key mechanism responsible for stationary superradiance in such lasers is collective nonlinear spontaneous decay of one of the atomic states. Such a cooperative decay

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is provided by incorporating an additional 'passive' resonator to couple the non-lasing atomic transition [16, 18], or to couple two non-lasing transitions in a four-level scheme [21]. Additionally, in order to reach the effect of super-radiance, when the laser intensity scales as N^2, the pumping field strength was taken proportional to N. However, as we have shown in [3], the two aforementioned requirements are not necessary to obtain superradiant lasing. We proposed a model of superradiant laser based on N three-level systems in ladder configuration, driven by two pumping lasers. All spontaneous decay processes are linear, i.e. no correlation in spontaneous emission is introduced. We called this new type of superradiance field driven superradiance since it stems from simultaneous coherent interaction of the atomic system with two driving laser fields. It was found that in the steady state, the generated laser field phase is locked to the relative phase of the two pumping lasers, and at a strong enough pump, the number of photons inside the resonator scales as N^2. It was also found that this system exhibits subradiant behavior manifested in a departure from N^2 scaling, in appropriate conditions.

In terms of nonlinear optics the scheme studied in [3] can be considered as a sum frequency generation, as it produces a field whose frequency is a sum of the frequencies of the two pumping lasers. Our theory is targeted at solid structures, in which all three transitions can be dipole-allowed, such as Ruby [26], semiconductor quantum wells [6–9], superconducting quantum circuits [10–12], and evanescently coupled waveguides [13, 14]. Here we consider a three-level scheme in which both non-lasing transitions have equal resonant frequencies, so they can be pumped by the same laser, and hence the frequency of the generated field is twice the frequency of the pumping laser. An advantage of this scheme is that there is no issues of synchronization and phase matching of different pumping lasers. Within the scope of this article we consider N^2 dependence of the laser intensity as an indication of stationary superradiance, whereas saturation with N as indication of subradiance. Utilizing semiclassical treatment, we solve optical Maxwell–Bloch equations both numerically and, in some particular cases, analytically. It is shown that the second harmonics generated by the system can exhibit both superradiance and subradiance, depending on the number of atoms, which is strong manifestation of collective behavior. The phase of the generated field is locked to that of the pumping laser. We have found that in the vicinity of some critical value of the pump Rabi frequency the system behaves in a phase-transition-like manner, changing from super to subradiant regime.

2. Problem formulation

Consider a three-level system shown in figure 1. The two transitions $|a\rangle \to |b\rangle$ and $|b\rangle \to |c\rangle$ have closed enough frequencies, i.e. $\omega_{ab} \approx \omega_{bc}$, to the extent that both transitions can be driven by a single pumping laser. The transition $|a\rangle \to |c\rangle$ is in resonance with the laser resonator, therefore the frequency of the generated laser field will be twice the frequency of the driving laser. Relaxation constants γ_{ij} describe the linear spontaneous decay of the corresponding atomic states, where the

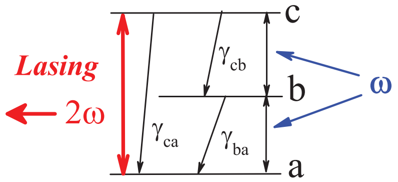

Figure 1. Schematic diagram of three-level system for second harmonics generation. Transitions $|a\rangle \to |b\rangle$ and $|b\rangle \to |c\rangle$ having equal frequency separations, are driven by the resonant field of frequency ω. The frequency of the lasing transition $|a\rangle \to |c\rangle$ equals 2ω.

events of spontaneous emission of different atoms are independent of each other. The interaction Hamiltonian for a single atom is given by

$$H_{\text{int}} = -\hbar[\Omega\, \mathrm{e}^{-\mathrm{i}\varphi}(\sigma_{ab}\mathrm{e}^{-\mathrm{i}\Delta t} + \sigma_{bc}\mathrm{e}^{\mathrm{i}\Delta t}) + g\hat{b}\mathrm{e}^{\mathrm{i}\Delta_{ac}t}\sigma_{ac}] + \text{H.c.} \quad (1)$$

where $\sigma_{ij} = |j\rangle\langle i|$ are the atomic raising/lowering operators, $\Omega = \left|\vec{d}\cdot\vec{E}\right|/\hbar$ is Rabi frequency of the pumping laser, φ is the phase of the pumping field, $\vec{d}$ is the relevant dipole matrix element, $\vec{E}$ is the pumping field amplitude, Δ ($-\Delta$) is the detuning of the pumping field from the atomic transition $|a\rangle \to |b\rangle$ ($|b\rangle \to |c\rangle$), Δ_{ac} is the detuning of the transition $|a\rangle \to |c\rangle$ from the resonator mode frequency, g is the coupling constant and $\hat{b}$ is the photon annihilation operator.

Semiclassical equations of motion for the elements of the atomic density matrix defined by $\rho = \sum \rho_{ij}\sigma_{ij}$ derived from the master equation with Hamiltonian (1) read:

$$\dot{\rho}_{cc} = -(\gamma_{cb} + \gamma_{ca})\rho_{cc} + \mathrm{i}\,\Omega\,(\rho_{cb}\mathrm{e}^{\mathrm{i}\varphi} - \rho_{bc}\mathrm{e}^{-\mathrm{i}\varphi}) + \mathrm{i}g^{*}b\rho_{ca} - \mathrm{i}gb^{\dagger}\rho_{ac} \quad (2)$$

$$\dot{\rho}_{bb} = \gamma_{cb}\rho_{cc} - \gamma_{ba}\rho_{bb} + \mathrm{i}\,\Omega\,(\rho_{bc}\mathrm{e}^{-\mathrm{i}\varphi} - \rho_{cb}\mathrm{e}^{\mathrm{i}\varphi} - \rho_{ab}\mathrm{e}^{-\mathrm{i}\varphi} + \rho_{ba}\mathrm{e}^{\mathrm{i}\varphi}) \quad (3)$$

$$\dot{\rho}_{bc} = -(\frac{\gamma_{ca} + \gamma_{cb} + \gamma_{ba}}{2} - \mathrm{i}\Delta)\rho_{bc} - \mathrm{i}\,\Omega\,[(\rho_{cc} - \rho_{bb})\mathrm{e}^{\mathrm{i}\varphi} + \rho_{ac}\mathrm{e}^{-\mathrm{i}\varphi}] + \mathrm{i}g^{*}b^{\dagger}\rho_{ba} \quad (4)$$

$$\dot{\rho}_{ac} = -\frac{\gamma_{ca} + \gamma_{cb}}{2}\rho_{ac} - \mathrm{i}g^{*}b^{\dagger}(\rho_{cc} - \rho_{aa}) + \mathrm{i}\,\Omega\,\mathrm{e}^{\mathrm{i}\varphi}(\rho_{ab} - \rho_{bc}) \quad (5)$$

$$\dot{\rho}_{ab} = -(\frac{\gamma_{ba}}{2} + \mathrm{i}\Delta)\rho_{ab} - \mathrm{i}\,\Omega\,[(\rho_{bb} - \rho_{aa})\mathrm{e}^{\mathrm{i}\varphi} - \rho_{ac}\mathrm{e}^{-\mathrm{i}\varphi}] - \mathrm{i}g^{*}b^{\dagger}\rho_{cb} \quad (6)$$

$$\dot{b} = -\kappa b + \mathrm{i}\sum_{j}^{N} g^{*}\rho_{ca}^{j} \quad (7)$$

where N is the number of atoms.

This is a standard set of optical Maxwell–Bloch equations where irreversible processes of spontaneous emission and resonator losses are governed by linear terms with atomic relaxation constants γ_{ij} and the field decay rate κ in the cavity. In semiclassical approximation the field operators can be replaced with c-numbers, so we put $b = \sqrt{n}\,\mathrm{e}^{\mathrm{i}\phi}$, where n is the number of photons in the lasing mode and ϕ is the laser field phase. No assumption is made regarding the initial atomic cooperativity such as nonlinear collective relaxation imposed by the presence of a 'passive' resonator, as in [16, 18, 21].

3. Results and discussion

We have solved (2)–(7) both numerically and, in the particular case of resonance, analytically using proper approximations. For the sake of simplicity, we take all three atomic relaxation rates equal, i.e. $\gamma_{ij} = \gamma$, and using $g \ll \Omega$, we obtain at resonance $\Delta_{ac} = \Delta = 0$ the following approximate expressions for the photon number n:

$$n_1 \approx \frac{4g^2\gamma^4\Omega^4}{\kappa^2(\gamma^4 + 12\gamma^2\Omega^2 + 12\,\Omega^4)^2}N^2, \tag{8}$$

$$n_2 \approx \frac{4\,\Omega^2 - 3\gamma^2}{4g^2}. \tag{9}$$

Expression (8) is valid at $N \ll N_c$, while expression (9) is valid for $N > N_c$, where

$$N_c \approx \frac{3\kappa\,\Omega^2}{\gamma g^2} \tag{10}$$

is a critical number of atoms that separates the superradiant regime from the subradiant one, as will be seen in the following. The field phase $\phi \approx 2\varphi + \pi/2$ for $N < N_c$ and $\phi \approx 2\varphi + \mathrm{ArcTan}[\gamma/\Omega]$ for $N > N_c$. In figure 2 the number of photons and the field phase are plotted as a function of the number of atoms. One can see that approximate analytical expressions (8) and (9) describe quite well the system behavior, except the vicinity of the critical point $N \sim N_c$, where it differs from the numerical solution. It can also be observed that the number of photons increases gradually with the number of atoms N, without any kinks or other threshold-like sharp changes. This may be interpreted as a thresholdless lasing [27–30].

When N approaches its critical value N_c defined by (10), both the number of photons and the field phase abruptly change their values, which indicates to the presence of a phase transition. At $N \ll N_c$ the photon number scales as N^2 in agreement with (8)—a signature of superradiance. At $N = N_c$ the photon number n sharply increases by several orders of magnitude, and at $N > N_c$ the photon number asymptotically approaches its limit value $(\Omega/\gamma)^2$ defined by (9). When $N \gg N_c$ the number of pumping photons is less than the number of atoms and a further increase in N does not affect the lasing process. This situation resembles a scenario described by Eberly [31] where subradiant spontaneous emission was considered in a system of N atoms excited by a single photon. We call this regime, in which the number of photon does not depend on N subradiant. It is interesting to follow the reverse transition from subradiant to superradiant regime by varying the pumping Rabi frequency Ω at fixed number of atoms N, as depicted in figure 3, where the number of photons is plotted as a function of Ω.

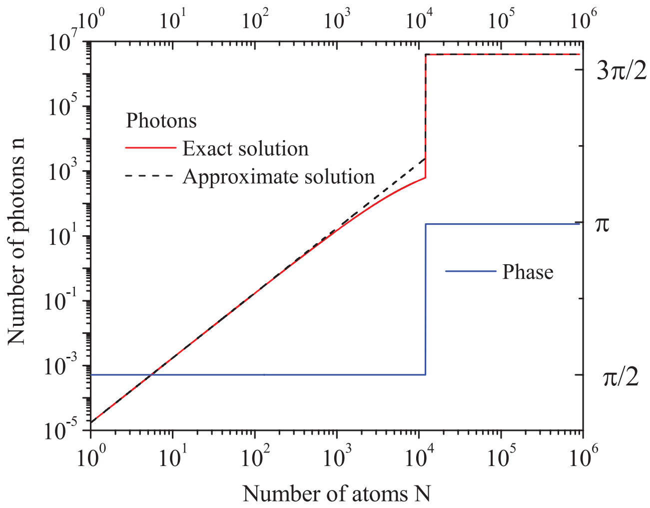

Figure 2. The number of photons and the phase of the generated field as a function of the number of atoms, as calculated from (2)–(7). Dashed lines are plotted using approximate expressions 8 and 9 for superradiant and subradiant regimes, respectively. Parameter values: $\Omega = 10, N = 10\,000, \gamma = 1, g = 0.01, \kappa = 0.001$.

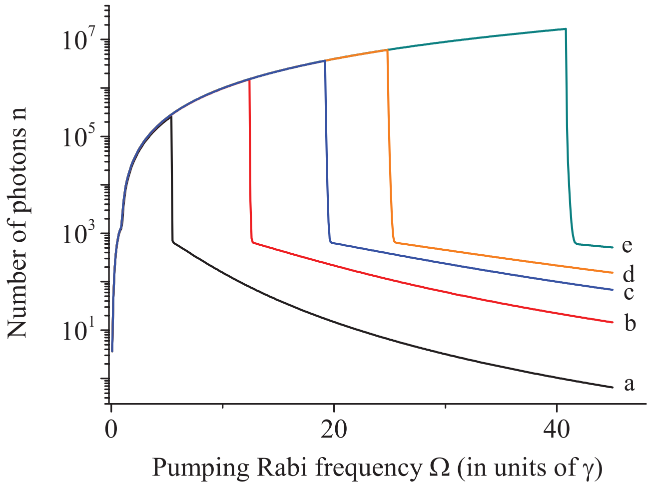

Figure 3. The number of photons as a function of pumping Rabi frequency at various values of the number of atoms N: (a) 1000, (b) 5000, (c) 12 000, (d) 20 000, (e) 50 000. Parameter values: $\gamma = 1, g = 0.01, \kappa = 0.001$.

There exists some critical value of the pumping Rabi frequency $\Omega_c \approx \sqrt{\gamma g^2 N/3k}$ which separates the two regimes of lasing. If we associate Ω with a number n_P of pumping photons and compare it with the number of atoms N, then the ratio between the number of pumping photons and N will define in which regime the laser works. Increasing Ω results in gradual increase of the number of photons, as long as $\Omega < \Omega_c$. When the pumping Rabi frequency reaches its critical value, i.e. at $\Omega = \Omega_c$, the number of photons drops several orders of magnitude and then slowly decreases with Ω. To get more physical insight to this behavior, we show in figure 4 the dependence of atomic populations on Ω. In subradiant regime $\Omega < \Omega_c$ the number of pumping photons n_P is less than N, and the larger is n_P, the more atoms are involved into the lasing process giving rise to growing laser intensity. When n_P becomes larger than N, strong pumping field results in sinchronization of fast Rabi

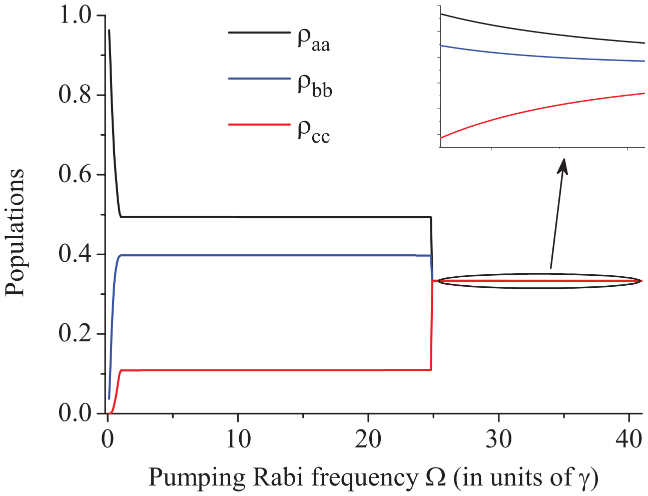

Figure 4. Atomic populations as a function of pumping Rabi frequency at $N = 20\,000$. The inset shows convergence of atomic populations to 1/3. Parameter values: $N = 10\,000, \gamma = 1, g = 0.01, \kappa = 0.001$.

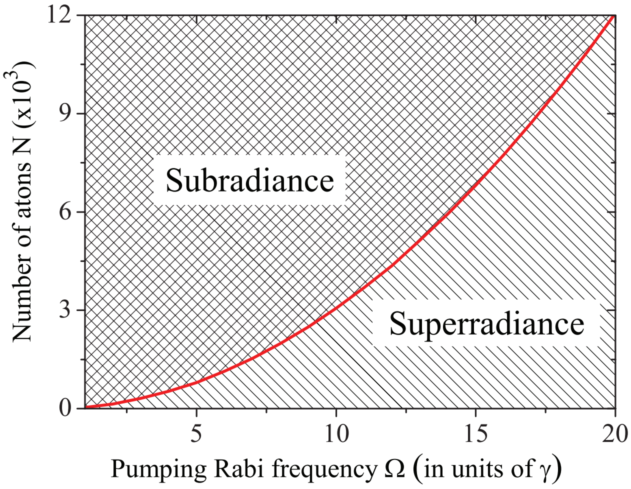

Figure 5. Phase diagram N versus Ω. Solid line (red online), corresponding to the critical number of atoms N_c defined by (10), separates two regions of parameters, corresponding to different regimes: superradiant below the line, and subradiant above the line. Parameter values: $\gamma = 1, g = 0.01, \kappa = 0.001$.

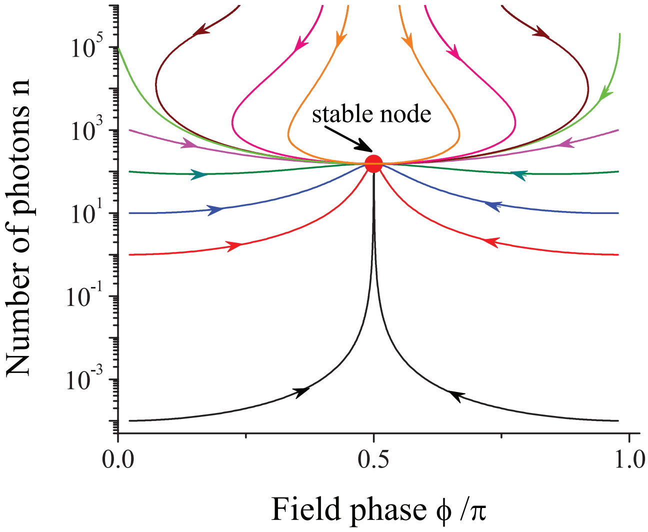

Figure 6. Superradiant regime: phase portrait of $n(t)$ versus $\phi(t)$. Each line represents a phase trajectory for a specific set of initial conditions $\{n(0), \phi(0)\}$. Red dot marks a single stationary state of the system (stable node) attracting all phase trajectories, independently on the initial conditions. Parameter values: $\Omega = 10, N = 1000, \gamma = 1, g = 0.01, \kappa = 0.001$.

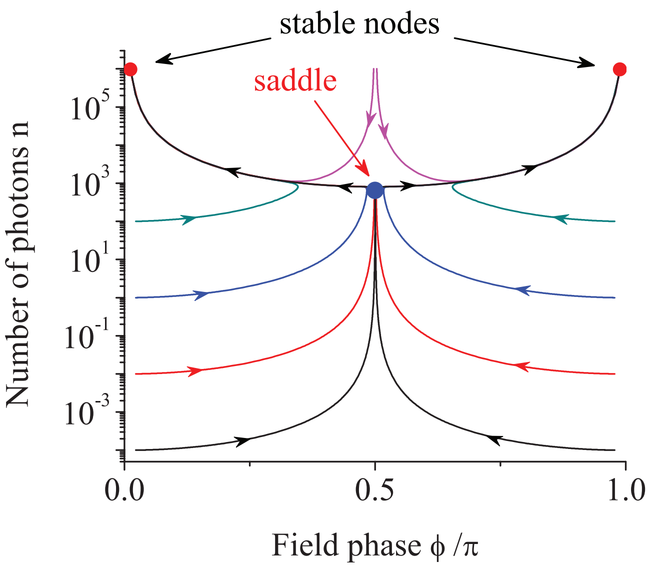

Figure 7. Subradiant regime: phase portrait of $n(t)$ versus $\phi(t)$. Each line represents a phase trajectory for a specific set of initial conditions $\{n(0), \phi(0)\}$. Blue dot marks the unstable stationary state of the system (saddle point) that repels all phase trajectories, independently on the initial conditions. There are two stable nodes (marked with red dots): the left one attracts phase trajectories with $\phi(0) < \pi/2$, and the right one that attracts phase trajectories with $\phi(0) > \pi/2$. If the initial value $\phi(0)$ equals exactly $\pi/2$, the field phase remains constant, i.e. $\phi(t) = \pi/2$, however, this state is unstable, and any small phase deviation will destroy it. Parameter values: $\Omega = 10, N = 4000, \gamma = 1, g = 0.01, \kappa = 0.001$.

oscillations on $|a\rangle \rightarrow |b\rangle$ and $|b\rangle \rightarrow |c\rangle$ transitions, the three atomic states are equally populated and the most energy is in the atoms.

Figure 5 shows a 'phase diagram' of the system where the solid line (red online), $N_c \propto \Omega^2$ according to (10), separates two different areas of parameters corresponding to superradiant (above the line) and subradiant (below the line) regimes.

Analysis of time behavior and the stability of the steady state solutions of (2)–(7) shows that at $N < N_c$ the superradiant solution is stable, while the subradiant one is unstable. At $N > N_c$ the situation is opposite: the subradiant solution becomes stable and the superradiant one becomes unstable. Thus the critical point $N = N_c$ is the instability point, which gives another indication to the presence of phase transition at this point. Figures 6 and 7 illustrate different stability properties of the super and subradiant regimes. In these figures each line represents a parametric plot in coordinates $\{\phi(t), n(t)\}$ for a specific initial condition. Phase portrait of the system at $N < N_c$ presented in figure 6 shows that in superradiant regime the steady state solution does not depend on the initial conditions, i.e. there exists a single stationary point/attractor which is a stable node. In this regime the system gradually approaches its steady state that is stable.

Essentially different behavior is seen in figure 7 showing phase portrait in the subradiant regime, in which $N > N_c$. The node at $\phi = \pi/2$ has lost its stability and transformed into a saddle point that repels slowly approaching phase trajectories, which then quickly reach one of the two stable steady states. As distinct from the superradiant regime, now these stationary states, being still independent on the initial photon number $n(0)$, depend upon the initial field phase $\phi(0)$. Phase trajectories with initial condition $\phi(0) < \pi/2$ reach the left stable node, while the phase trajectories with initial condition $\phi(0) > \pi/2$ reach the right stable node. Stability analysis of the subradiant regime shows that first the field reaches the long-lived metastable state, and then, at some critical time, grows abruptly to stable steady state. In theory of phase transitions [32] such a behavior is known as a phenomenon of critical slowing down.

4. Conclusions

We have shown that a three-level ladder scheme with equally separated levels can generate a second harmonics of the pumping laser. The intensity of the generated field is nonlinear with respect to that of the pumping one: depending on the ratio between the number of atoms and the intensity of the pumping laser the SHG can operate in either superradiant or subradiant regime. Transition from one regime to another occurs abruptly, in phase-transition-like manner. In subradiant regime the steady state is also established in phase-transition-like manner, via critical slowing down. The laser field phase is locked to the phase of the driving laser. Details of the time evolution of this system will be published elsewhere.

For experimental realization of the described super/subradiant SHG we propose semiconductor quantum well structures [6–9], superconducting quantum circuits [10–12], and evanescently coupled waveguides [13, 14], in which equally spaced levels relevant to this study exist.

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