Intermittency and dynamical chaos in reversible spontaneous emission

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It is shown that a type of reversible spontaneous emission, the chaotic vacuum Rabi oscillations, may occur in the interaction of two-level atoms strongly coupled with a single cavity mode under a modulation of the atom-field coupling. Such a modulation arises naturally if the atoms move through a cavity in maserlike experiments. The existence of homoclinic chaos in reversible spontaneous emission is proven analytically. Evidence of intermittency associated with the spatial modulation of the vacuum Rabi frequency is shown numerically for the values of parameters that are achievable in present-day experiments with Rydberg atoms moving through a high-Q microwave cavity in the strong-coupling regime.

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It is known that spontaneous emission of excited atoms in a cavity is qualitatively different from spontaneous emission in free space [1]. In the strong-coupling and single-mode regime ($\Omega_0 \gg T_c^{-1}, T_a^{-1}$), it may be even reversible demonstrating the effect of the vacuum Rabi oscillations that has been found experimentally with Rydberg atoms flying slowly through a high-Q microwave cavity [2] and with the usual atoms on optical transitions in a microcavity [3]. The simple theory neglecting the cavity and atomic damping ($T_c, T_a \to \infty$) and the spatial structure of the cavity mode ($\Omega_0 = \mathrm{const}$) predicts in the rotating-wave approximation a periodic exchange of energy between the atoms and a selected cavity mode [4].

Recently, it has been demonstrated numerically [5] that the spatial modulation of the vacuum Rabi frequency of two-level atoms Ω_0 moving through a single-mode cavity can drastically change the character of the vacuum Rabi oscillations. In the present paper we show analytically that a type of reversible spontaneous emission, *the chaotic vacuum Rabi oscillations*, may arise under realistic conditions in the interaction of moving excited two-level atoms with vacuum. The nature of this phenomenon is elucidated with the help of the Melnikov analysis and is proven to be homoclinic. By computing the maximal Lyapunov exponent, we find the ranges of the values of the atom-cavity detuning, of the number and velocity of atoms for which some manifestations of quantum homoclinic chaos in reversible spontaneous emission can be found in real experiments. An intermittent route to chaos is demonstrated in the vacuum Rabi oscillations of the atomic population where the duration of the chaotic state increases with an increasing the velocity of the atoms.

Consider ‘‘a monoenergetic droplet’’ with N identical two-level atoms that moves through a single-mode high-Q cavity along the axis k and is supposed to be so confined that all the atoms ‘‘see’’ the same field when moving. Generally speaking, the cavity field is inhomogeneous spatially, so the vacuum Rabi frequency of the moving atoms should be considered as a time-dependent function $\Omega_0(k) = \Omega_0(v_a t)$, where v_a is the velocity of atoms. For simplicity, we will work in the rotating-wave and Raman-Nath approximations and in the strong-coupling regime neglecting cavity-mode damping and atomic relaxation. The respective Hamiltonian has the form

$$H = \frac{1}{2}\hbar\omega_a \sum_{j=1}^{N} \hat{\sigma}_z^j + \hbar\omega_c\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right) + \hbar\Omega_0(t)\sum_{j=1}^{N}(\hat{a}\hat{\sigma}_+^j + \hat{a}^\dagger\hat{\sigma}_-^j), \tag{1}$$

where the Pauli operators $\hat{\sigma}_{z,\pm}$ and the boson operators $\hat{a}$ and $\hat{a}^\dagger$ describe the atoms and the mode, respectively. The Hamiltonian (1) is known to generate in the Heisenberg picture semiclassical dynamical chaos in the atom-field interaction [6]. However, the semiclassical approximation neglects *all quantum correlations* and, in particular, those that are responsible for spontaneous emission. The intensity radiated by the N-atom system is [7]

$$I(t) = I_1\left\langle \sum_{i=1}^{N}\sum_{j=1}^{N} \hat{\sigma}_+^i \hat{\sigma}_-^j \right\rangle = I_1\frac{N}{2}[z(t)+1] + I_1 N^2 r(t), \tag{2}$$

where I_1 is the intensity of the spontaneous radiation of a single atom. The first term describes the ordinary reversible spontaneous emission proportional to the number of atoms N and to the density of the atomic population inversion z. The second term describes the cooperative spontaneous emission proportional to N^2 [7] and to quantum correlations among the atoms r [8].

In the Heisenberg picture, the main problem is to translate the respective operator equations into a tractable closed set of c-number equations that are able to describe adequately a physical situation considered. With the aim to describe the reversible spontaneous emission we go beyond the simple semiclassical factorization and take into consideration the following bilinear operators: $\hat{r} = N^{-2}\Sigma_i'\Sigma_j \hat{\sigma}_+^i\hat{\sigma}_-^j$ (the prime denotes summation over different atoms), $\hat{n} = N^{-1}\hat{a}^\dagger\hat{a}$, $\hat{u} = N^{-3/2}(\hat{a}\Sigma_j\hat{\sigma}_+^j + \hat{a}^\dagger\Sigma_j\hat{\sigma}_-^j)$, $\hat{v} = iN^{-3/2}(\hat{a}^\dagger\Sigma_j\hat{\sigma}_-^j - \hat{a}\Sigma_j\hat{\sigma}_+^j)$, and the population inversion operator $\hat{z} = N^{-1}\Sigma_j\hat{\sigma}_z^j$. It can be shown that *a low-dimensional closed set of equations* for the quantum correlators of the second order n, r, u, and v and for z over an initial quantum state that is supposed to be factorized into a product of the collective initial atomic state just before entering the cavity and vacuum field state can be derived

$$\dot{n} = -\Omega_N(\tau)v, \quad \dot{z} = 2\Omega_N(\tau)v, \quad \dot{u} = (\omega-1)v, \tag{3}$$

$$\dot{r} = -\Omega_N(\tau)zv, \quad \dot{v} = (1-\omega)u - \Omega_N(\tau)\left(\frac{z+1}{N} + 2r + 2nz\right).$$

When deducing Eq. (3), we have neglected the quantum correlations beyond the second order; for example, $\langle\hat{a}\hat{a}^\dagger\rangle\langle\hat{\sigma}_z\rangle$ is written in place $\langle\hat{a}\hat{a}^\dagger\hat{\sigma}_z\rangle$.

The derivatives in Eqs. (3) are taken with respect to $\tau = \omega_a t$. The time-dependent, $\Omega_N(\tau) = \Omega_0(\tau)\sqrt{N}/\omega_a$, and time-independent, $\omega = \omega_c/\omega_a$, control parameters are the dimensionless collective vacuum Rabi frequency and the dimensionless detuning, respectively. The dynamical system (3) possesses two integrals of motion

$$z^2 + 4r = 4N^{-2}R(R+1), \quad z + 2n = W, \tag{4}$$

resulting from the unitarity of atomic evolution and conservation of the total energy of the atom-field system in a lossless cavity, respectively. Here R is the cooperation number which labels the atomic Dicke states $|R,M\rangle$ [7], and $M = Nz/2$ is proportional to the energy stored by the atoms and is such that $|M| \leqslant R \leqslant N/2$.

If there are initially no correlations among the atoms, polarization, and cavity photons, we still have on the right-hand side of the last equation in the set (3) the term $z+1$ which equals twice the density of the atoms in the excited state. Namely, this term is the source of the spontaneous emission. It drives v, which in turn drives the other variables in Eq. (3), creating the atom-atom correlations, polarization, and cavity photons. The situation differs strongly from the semiclassical model [6,9] where the respective initial state with zero polarization and cavity vacuum is an equilibrium state. Our model accounts for small quantum corrections $\simeq 1/N$ that are responsible for spontaneous emission. During the evolution, quantum corrections grow. Let us estimate the time scale of validity of the factorization of the third-order correlators involved. As is known [10], the scale of quantum-classical correspondence depends strongly on the type of motion. For regular motion, the time scale has a power dependence on N. For chaotic motion, a ''distance'' between initially adjacent trajectories can grow exponentially, $d(\tau) \simeq d_0 \exp(\lambda\tau)$, where λ is the maximal Lyapunov exponent. Initially we have $d_0 \simeq 1/N \ll 1$, and from the condition $d(\tau) \ll 1$ one can estimate the time scale of validity of our dynamical equations (3) in the chaotic regime to be $\tau_q \ll \lambda^{-1}\ln N$.

There exist, at least, two integrable versions of the five-dimensional set of the nonlinear equations (3). In the ''homogeneous'' limit, a spatial inhomogeneity of the cavity mode is neglected, and the vacuum Rabi frequency Ω_0 is supposed to be a constant. This is the case, for example, with motionless atoms in the pointlike approximation. The extra integral $C = 2\Omega_N u - (\omega-1)z$ helps to integrate Eqs. (3) in terms of Jacobian elliptic functions. The exact solution for the density of the atomic inversion looks as

$$z(\tau) = z_1 + (z_2 - z_1)\mathrm{sn}^2\left[\sqrt{\frac{1}{2}(z_3 - z_1)}\,\Omega_N(\tau-\tau_0); \frac{z_2 - z_1}{z_3 - z_1}\right], \tag{5}$$

where

$$\tau_0 = \frac{1}{\Omega_N\sqrt{2}}\int_{z_0}^{z_1}\frac{dz}{\sqrt{(z-z_1)(z-z_2)(z-z_3)}}, \tag{6}$$

and $z_{1,2,3}$ are the roots of an algebraic cubic equation that arises when inverting the elliptic integral. The exact solutions for the other variables are $2n = W - z$, $4r = 4N^{-2}R(R+1) - z^2$, $2\Omega_N u = C + (\omega-1)z$, $2\Omega_N v = \dot{z}$. It should be mentioned that solutions for the related simple semiclassical version (without any quantum correlations) in terms of Jacobian elliptic functions have been known for a long time [4,11].

In the resonance limit, when the frequency of the cavity mode coincides exactly with the atomic transition frequency, i.e., $\omega = 1$, the set (3) is integrable for any kind of modulation $f(\tau)$ of the vacuum Rabi frequency $\Omega_N(\tau) \equiv \Omega_N f(\tau)$ due to a reduction of the system's dimension (the variable u becomes a constant). In this integrable limit, exact solutions are obtained from the respective solutions of the ''homogeneous'' limit when transforming to the new ''time'' $\tau \to \int f(\tau')d\tau'$. Thus, the resonant two-level atoms when moving through a lossless single-mode cavity will experience a periodically modulated exchange of energy with the cavity field *regardless of the spatial structure of the cavity mode along the propagation axis*. This analytic result will be used in checking the results of our numerical simulations of the nonintegrable system (3).

In order to show what happens when the nonresonant two-level atoms move through a spatially varying cavity field we will use the perturbative Melnikov method [12] that enables us to detect transversal intersections between perturbed stable and unstable manifolds of a hyperbolic fixed point. Suppose the atoms move through a cavity in a direction along which the depth of the spatial modulation of the vacuum Rabi frequency may be considered as small as compared with its amplitude value $\Omega_N(\tau) = \Omega_N + \epsilon\sin(b\omega\tau)$, $\epsilon \ll \Omega_N$, where $b = v_a/c$ is the dimensionless velocity of the atoms and c is the velocity of light in vacuum. The signed distance between the perturbed stable and unstable manifolds of the hyperbolic fixed point at a time moment τ_0 along the normal $\mathbf{n}$ to an unperturbed homoclinic surface is proportional to $\epsilon M(\tau_0) + O(\epsilon^2)$. Here

$$M(\tau_0) = \int_{-\infty}^{\infty}\mathbf{n}(\mathbf{s})\cdot\mathbf{G}(\mathbf{s})d\tau \tag{7}$$

is the Melnikov function. The integral should be evaluated along the separatrix $\mathbf{s}$ that can be easily found in an explicit form $\mathbf{s}:(n_s, z_s, u_s, r_s, v_s)$ from the integrable ''homogeneous'' version of Eqs. (3). After calculating the scalar product with the normal vector $\mathbf{n}(\mathbf{s}) = [0, 1-\omega, 2\Omega_N, 0, 0]^T$ and the $O(\epsilon)$ perturbation part of the vector field on the separatrix $\mathbf{G}(\mathbf{s}) = [-v_s\sin(b\omega\tau), 2v_s\sin(b\omega\tau), 0, -z_s v_s\sin(b\omega\tau), -(z_s/N + 1/N + 2r_s + 2n_s z_s)\sin(b\omega\tau)]^T$ and carrying out the integration we find

$$M(\tau_0)=\frac{2\pi(1-\omega)(b\omega)^2}{\Omega_N^3 \mathrm{sh}(b\omega\pi/\sqrt{z_3-z_1}\Omega_N)}\cos(b\omega\tau_0). \quad (8)$$

It is clear from Eq. (8) that out of resonance, $\omega\neq 1$, the Melnikov integral has simple zeroes as a function of τ_0, implying transversal intersections of the stable and unstable manifolds in an infinite variety of homoclinic points (for a review of homoclinic chaos in Hamiltonian systems see, e.g., [13]). Therefore, we may conclude from the above analysis that horseshoe chaos becomes possible in the interaction of moving two-level atoms with a cavity vacuum field even in the rotating-wave approximation and under a small modulation.

In order to confirm numerically the onset of chaos in reversible spontaneous emission we have calculated the maximal Lyapunov exponent λ in the Eqs. (3) assuming modulation of the vacuum Rabi frequency in the form $\Omega_N(\tau)=\Omega_N\sin(\omega b\tau)$. The results depend strongly on the initial atomic state. With initially fully inverted atoms (the so-called superfluorescent state $|N/2,N/2\rangle$ [14]), λ may reach the values of the order of 0.5 at $\Omega_N\simeq 8.5$ and $\omega\simeq 0.9$ even if the atoms move comparatively slowly ($b\simeq 5\times 10^{-4}$), whereas λ is negligibly small at the same values of the control parameters but with the atoms prepared initially in the superradiant state $|N/2,0\rangle$. We have observed windows, i.e., parameter intervals in which the Rabi oscillations are periodic ($\lambda\approx 0$), that are punctuated by intervals in which the oscillations are chaotic with positive values of λ. This effect of intermittency is especially prominent with initially fully inverted atoms, $|N/2,N/2\rangle$, when the windows have been found very close to almost every parameter value that leads to chaos.

The maximal Lyapunov exponent cannot, of course, be measured directly in a real experiment. What is measured is the radiated intensity that can be written in terms of the atomic variables as Eq. (2). Both the atomic population inversion z and the quantum correlation among the atoms r should display chaotic behavior in a range of the values of the system's control parameters, v_a, ω, and Ω_N, for which the respective maximal Lyapunov exponent is positive. To illustrate the structural chaos and intermittency in the reversible spontaneous emission that could be found in a real experiment, we show in Fig. 1 the oscillations of z with $N=10^6$ initially fully inverted atoms at the fixed values of the control parameters $\Omega_N=1$ and $\omega=1.5$ but with different values of the atomic velocity: (a) $b=0.001$, (b) $b=0.01$, and (c) $b=0.1$. As is seen from the figure, the duration of the chaotic phase of the oscillations increases with an increasing the velocity of the atoms. Part d of this figure shows for a comparison the evolution of the density of the atomic population at exact resonance ($\omega=1$) when the atom-cavity system was shown to be integrable. As expected, the oscillations are regular at exact resonance.

Two peculiarities of the parametric vacuum Rabi oscillations are seen distinctly in the figure: (i) the delay time during which quantum correlations among the atoms build up and (ii) a characteristic periodic structure of the dependence $z(\tau)$ that is due to a spatial modulation of the vacuum Rabi frequency. The dimensionless half-period of the modulation

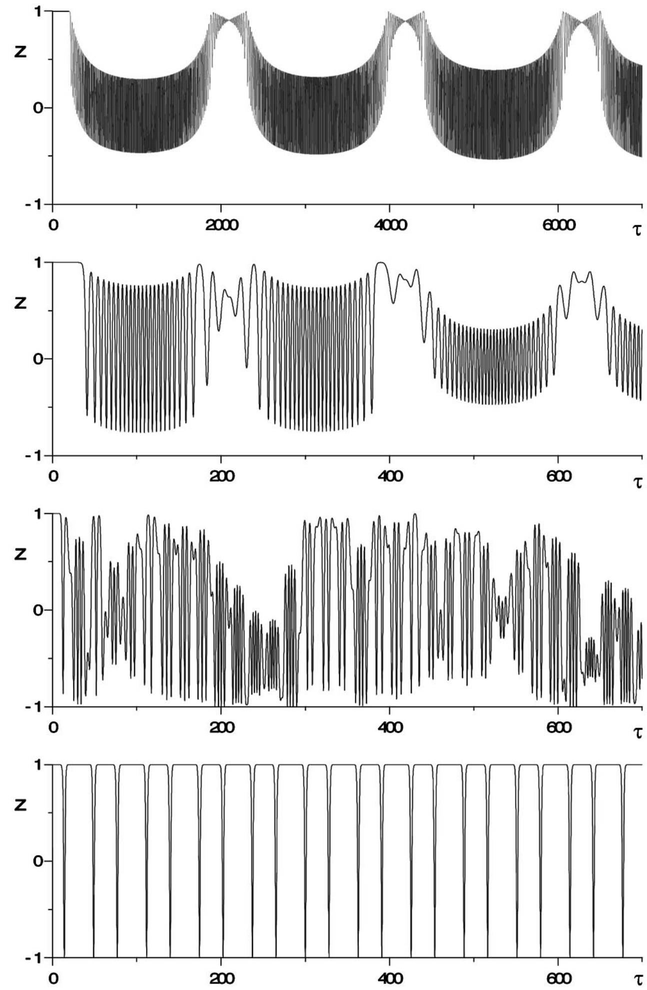

FIG. 1. Dependence $z(\tau)$ with $N=10^6$ initially fully inverted atoms at $\Omega_N=1$: (a) $b=0.001$, $\omega=1.5$, (b) $b=0.01$, $\omega=1.5$, (c) $b=0.1$, $\omega=1.5$, and (d) $b=0.1$, $\omega=1$, exact resonance.

is equal to $\pi/b\omega$ and may be estimated to be $\simeq 2000$ with $b=0.001$ and $\simeq 200$ with $b=0.01$.

Rydberg atoms in a high-Q microwave cavity [2] seem to be a physical system that is well suited to observe the parametric chaotic vacuum Rabi oscillations and is adequate to the theory presented. This device can be really operated in the regime when all the assumptions adopted in our model may be considered as valid. In a high-Q superconducting microcavity, the single-atom vacuum Rabi frequency Ω_0 may reach 10^5-10^6 rad/s. Therefore, the period of the collective vacuum Rabi oscillations $T_R=2\pi/\Omega_0\sqrt{N}$ is much smaller than the lifetimes of the circular Rydberg states $\simeq 10^{-2}$ s and of the microwave photons $\simeq 10^{-1}-10^{-2}$ s in a superconducting microwave cavity with $Q\simeq 10^9$ [2], implying the Hamiltonian approach adopted and the strong-coupling limit to be valid. A comparatively long wavelength $\simeq 1$ cm implies the pointlike approximation to be valid. The recoil energy of atoms accompanying the emission of microwave photons can be estimated to be very small [6] (the Raman-Nath approximation). The time-scale of quantum-classical correspondence $t_q=\omega_a^{-1}\tau_q$ in the chaotic regime of reversible spontaneous emission from Rydberg atoms moving through a high-Q microwave cavity may reach $100-1000T_R$ with $N=10^{10}$ atoms.

In conclusion, we showed analytically and numerically that the intermittency and dynamical chaos of a homoclinic nature may occur in the interaction of a very simple quantum system with vacuum. It should be possible to observe some manifestations of this kind of reversible spontaneous emission in experiments with Rydberg atoms moving through a high-Q microwave cavity.

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