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Structural chaos in reversible spontaneous emission of moving atoms

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Abstract. It is proved analytically and numerically that, under certain conditions, the reversible spontaneous emission of two-level atoms moving in a high-Q resonator and described quantum-classically can be chaotic in the sense of the exponential sensitivity with respect to the initial conditions. The wavelet analysis of the vacuum Rabi oscillations showed that this chaos is structural. The numerical estimates showed that a Rydberg atom maser with a superconducting microwave resonator operating in a strong coupling mode is a promising device for detecting manifestations of the dynamic chaos in the reversible spontaneous emission.

1. Introduction

The spontaneous emission of excited atoms proceeds differently in a free space and an electromagnetic resonator. A continuum of the field modes of the free space vacuum causes the irreversibility of spontaneous emission, which is manifested in the exponential decrease in the probability of finding an atom in the excited state with the exponent containing the Einstein coefficient A [1]. A change in the density of field modes in the resonator or near surfaces results in changes in the rate and spectrum of spontaneous emission and causes the radiative shift of the atomic levels with respect to these characteristics in a free space.

E M Parcell was probably the first to pay attention to this fact in his note published in 1946 [2]. Since then both an increase in the spontaneous emission rate in resonance cavities (see, for example, Refs [3 – 5]) and its suppression in nonresonance cavities (see, for example, Ref. [6]) have been observed for atoms of different types in different frequency regions.

However, in cavities with a comparatively low Q-factor, spontaneous emission does not change qualitatively, being irreversible as in a free space. Owing to efforts of many experimenters, the quality factor Q of cavities, in particular, microcavities was increased to $\sim 10^9$ and more. For such Q-factors, a constant of the interaction of an atom with a vacuum field (the single-atom vacuum Rabi frequency Ω_0) exceeds the spectral width ω_c/Q of the cavity modes, where ω_c is the dominating frequency mode. For $\Omega_0 \gg \omega_c/Q$ (the so-called strong coupling limit in the cavity electrodynamics), the cavity field spectrum exhibits a distinct singularity near the atomic transition frequency, and periodic energy transfer can occur between the atom and the field, i.e., spontaneous emission becomes reversible.

This phenomenon, which is also called vacuum Rabi oscillations, has been observed on microwave transitions of Rydberg atoms in a metal cavity of a centimetre size [7], optical cavities [8], and on excitonic transitions in semiconductor microcavities [9]. Such experiments open up attracting opportunities for controlling spontaneous emission and creating the threshold-free microlasers operating on exciton polaritons in semiconductor microcavities with quantum wells. A brief review of theoretical and experimental papers on spontaneous emission of atoms at rest in cavities was presented in Ref. [10].

This paper is devoted to the theory of reversible spontaneous emission of two-level atoms moving in an ideal cavity with the vacuum field. We proved that vacuum Rabi oscillations described using a combined quantum-classical approach could be chaotic even in a single-mode cavity. This means that, under certain conditions, an ensemble of moving two-level atoms interacts with a single cavity mode in a correlated and coherent way (in the absence of any external energy sources and sources that destroy coherence) and emits and absorbs light chaotically (in the sense of the exponential sensitivity of reversible spontaneous emission to variations in the initial conditions).

The spectrum of such a signal is substantially nonstationary because of the existence of at least three time scales related to the modulation of the vacuum Rabi frequency caused by the spatial inhomogeneity of the cavity mode; energy exchange between atoms and the cavity; and the mismatch between the atomic resonance and the cavity mode. A typical chaotic signal of reversible spontaneous emission consists of the short-lived, high-frequency components closely spaced in time and of the low-frequency components closely spaced in frequency. To reveal the structure of this chaos, we will use the wavelet analysis, which represents a multiscale method that provides good resolution both in frequency and time.

Note that a problem of the dynamic chaos has its own history in quantum electrodynamics. As early as 1964, an aperiodic behaviour of the quantum oscillator model has been found [11]. Later [12], the equivalence was proved between the semiclassical equations for a single-mode two-level laser and the hydrodynamic Lorentz system represent-

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ing a classical model of a strange attractor. The comprehensive literature is available on the problems of instability and dynamic chaos in lasers (see reviews [13, 14]). As for the quantum oscillators with moving particles, the nonlinear dynamics of a single-mode beam maser with a homogeneous field directed along the propagation direction of molecules has been investigated in Ref. [15] using a two-level model. The authors [15] found numerically the regions of bistability, multistability, and chaotic pulsations.

The majority of papers in this field consider the generation of stimulated emission in the presence of relaxation, i.e., the open dissipative systems with a decreasing phase volume. We emphasise here that in this paper we formulated and considered a physical problem of coherent spontaneous emission of excited atoms moving in an ideal cavity, without any external energy pumping. The corresponding system of classical equations of motions is the Hamiltonian (conserving the phase volume) and nonautonomous (because of the inclusion of the spatial structure of the cavity mode) system.

2. Theory of reversible spontaneous emission of two-level atoms moving in an ideal cavity

Consider a single-mode very high-Q cavity. We restrict ourselves to a one-dimensional case and assume that a variation in the electric field strength of a standing wave along the x-axis is described by a spatial function $f(x)$. A monoenergetic cloud of N two-level atoms or molecules, which do not directly interact with each other, is admitted to the cavity with the velocity υ and moves along this axis. The spatial inhomogeneity of the standing wave modulates the interaction energy of the atoms with the selected cavity mode, i.e., the vacuum Rabi frequency becomes time-dependent:

$$\Omega_0 f(x) \to \Omega_0 f(\upsilon t) \equiv \Omega_0(t).$$

The simplest Hamiltonian describing such a situation is the nonstationary operator [16]

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

where ω_{a} and ω_{c} are the frequencies of the atomic transition and cavity mode, respectively; $\hat{\sigma}_{z,\pm}$ is the Pauli operators; $\hat{a}$ and $\hat{a}^{\dagger}$ are the creation and annihilation operators for photons in the selected mode, respectively. Along with the approximations mentioned above, we also used the rotating wave approximation and the Raman–Nath approximation and assumed that the atomic cloud has a small diameter compared to the standing wave wavelength. This allows us to avoid unnecessary complications, keeping in mind our main goal, and to reveal the nature of the appearance of weak chaos in such a simple model of the interaction of atoms with their own radiation field in the absence of any external pump. The emission intensity of atoms is calculated from the expression [17]

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

where I_1 is the emission intensity of an isolated atom. The first term in (2) describes usual spontaneous emission, whose intensity is proportional to the number of atoms and the density z of the population inversion of atoms. The second term describes cooperative spontaneous emission with the intensity that is proportional to the square of the number of atoms and to the expected eigenvalue r of the operator of quantum correlations of the atoms with each other.

Within the framework of the Hamiltonian approach, which is valid for the strong coupling regime, the dynamic equations are found from the Heisenberg equation by averaging the operators over the chosen initial quantum state of the atoms and the mode. It is known that a simple semiclassical averaging, in which all the operator products are decoupled, cannot describe spontaneous emission of completely excited atoms because a combination of the state of atoms with the zero mean dipole moment and the vacuum state of the cavity field represents a stationary state of the corresponding semiclassical system of equations (see, for example, [18]).

To initiate spontaneous emission in semiclassical models, the start fluctuations of the dipole moment and (or) field are required. It is also known [19] that averaging can be performed by retaining atomic quantum correlations, which are produced via the total emission field (it is assumed that atoms do not interact directly with each other!). In this case, the *ad hoc* fluctuations are not required for the description of coherent spontaneous emission. The self-consistent dynamic system of equations for the second-order quantum correlators of the same dimensionality as the semiclassical system, but describing reversible spontaneous emission of moving atoms (see the Appendix), has the form [20]

$$\dot{n} = -\Omega_N(\tau)\upsilon, \quad \dot{z} = 2\Omega_N(\tau)\upsilon, \quad \dot{u} = (\omega - 1)\upsilon,$$
$$\dot{r} = -\Omega_N(\tau)z\upsilon, \dot{\upsilon} = (1-\omega)u - \Omega_N(\tau)\left(\frac{z+1}{N} + 2r + 2nz\right), \tag{3}$$

where z, r, n, u, and υ are the quantum averages of the inversion operator $\hat{z} = N^{-1}\sum_j \hat{\sigma}_z^j$ and of the following bilinear operators:

$$\hat{r} = N^{-2}\sum_{i\neq j}\hat{\sigma}_+^i\hat{\sigma}_-^j, \quad \hat{n} = N^{-1}\hat{a}^{\dagger}\hat{a}, \ \hat{u} = N^{-3/2}\left(\hat{a}\sum_j\hat{\sigma}_+^j + \hat{a}^{\dagger}\sum_j\hat{\sigma}_-^j\right), \quad \hat{=}\mathrm{i}N^{-3/2}\left(\hat{a}^{\dagger}\sum_j\hat{\sigma}_-^j - \hat{a}\sum_j\hat{\sigma}_+^j\right).$$

The averaging was performed over the factorised initial quantum state of atoms and the field mode

$$|\psi(0)\rangle = |\psi_N(0)\rangle_{\mathrm{a}}|0\rangle_{\mathrm{c}}\,. \tag{4}$$

The derivatives in the system (3) are taken with respect to the dimensionless time $\tau = \omega_{\mathrm{a}}t$, while the collective vacuum Rabi frequency $\Omega_N(\tau) = \Omega_0(\tau)\sqrt{N}/\omega_{\mathrm{a}}$ and the normalised mismatch $\omega = \omega_{\mathrm{c}}/\omega_{\mathrm{a}}$ represent the dimensionless controlling parameters. The unitary property of the atomic evolution and conservation of the total energy of the atomic-field system in the case of neglecting any relaxation during the interaction (i.e., it is assumed that the flight time of atoms through the cavity is far shorter than the times of atomic relaxation and the field decay) results in two conservation laws

$$z^2 + 4r = 4N^{-2}R(R+1), \quad z + 2n = S. \tag{5}$$

Here, R is the cooperative number that numbers the atomic

$|R, M\rangle$ Dicke states; $M = Nz/2$ changes in such a way that $|M| \leqslant R \leqslant N/2$ and is proportional to the energy of atoms.

It follows from Eqns (3) that, unlike the semiclassical theory, a state with completely excited atoms injected into a cavity with a vacuum field, i.e., the initial state of the system of five equations (3) ($z_0 = 1$, $n_0 = r_0 = u_0 = v_0 = 0$) is not equilibrium in our model. Therefore, even when photons in the resonance mode, atomic correlations, and polarisation of atoms are absent at the initial moment, all these quantities begin to oscillate with time because of the presence of the term $(z+1)/N$.

Therefore, weak quantum oscillations $\sim 1/N$ represent a source of spontaneous emission in our model. As time passes, the quantum oscillations increase generally. Because our model neglects the third-order and higher-order quantum correlators, the range of its application is restricted by the times at which quantum corrections of this order can be neglected. While this time scale is comparatively great for regular vacuum Rabi oscillations, $\tau_{\rm q}^{\rm reg} \approx N$, it substantially decreases in the chaotic regime: $\tau_{\rm q}^{\rm ch} \simeq \lambda^{-1} \ln N$ [21, 22], where λ is the maximum Lyapunov index.

One can easily show that when $\Omega_0 = \text{const}$ (i.e., when atoms are at rest or flying in the direction along which the cavity field can be considered homogeneous), the system (3) acquires the additional integral of motion $C = 2\Omega_N u - (\omega - 1)z$ and is integrable in quadratures. The exact solution for the atomic inversion density has the form

$$z(\tau) = z_1 + (z_2 - z_1) \times \mathrm{sn}^2\left\{\left[\frac{1}{2}(z_3 - z_1)\right]^{1/2} \Omega_N(\tau - T); \frac{z_2 - z_1}{z_3 - z_1}\right\}, \qquad (6)$$

where

$$T = \frac{1}{\Omega_N \sqrt{2}} \int_{z_0}^{z_1} \frac{\mathrm{d}z}{[(z - z_1)(z - z_2)(z - z_3)]^{1/2}}; \qquad (7)$$

Here $z_{1,2,3}$ are the roots of a cubic algebraic equation, which appears upon inversion of the elliptic integral, and z_0 is the initial value of z. Solutions for other variables can be readily found using the integrals of motion of the system.

It follows from the form of equations (3) that they are integrable in the case of the exact resonance for arbitrary modulation $f(\tau)$ of the vacuum Rabi frequency, $\Omega_N(\tau) = \Omega_N f(\tau)$, because the variable u becomes constant for $\omega = 1$. In the limit of the exact resonance, exact solutions can be found from the corresponding solutions for $\Omega_0 = \text{const}$ using the substitution $\tau \to \int f(\tau')d\tau'$. Therefore, vacuum Rabi oscillations of the resonance atoms moving through a high-Q cavity with an arbitrary spatial mode configuration are regular. This fact can be used to verify numerical calculations. Prants et al. showed [18] by the Mel'nikov method [23] that in the presence of an arbitrarily small degree of modulation of the Rabi frequency in the semiclassical atomic-field system with moving nonresonance atoms, the so-called transverse intersections of the stable and unstable manifolds of a hyperbolic singularity of this system appear. A similar analysis of our model (3) showed that the Mel'nikov function in the first order of the perturbation theory in a small modulation parameter $\varepsilon \ll \Omega_N$ ($\Omega_N(\tau) = \Omega_N + \varepsilon \sin(b\omega\tau)$ and $b = v_{\rm a}/c$ is the ratio of the velocity of atoms to the speed of light in vacuum) has the form

$$M(\tau_0) = \frac{2\pi(1-\omega)(b\omega)^2}{\Omega_N^3 \sinh[b\omega\pi/(z_3 - z_1)^{1/2}\Omega_N]} \cos(b\omega\tau_0). \qquad (8)$$

This function characterises the distance (with sign) between the above-mentioned perturbed manifolds at the moment τ_0 along the direction of the normal to the unperturbed homoclinic surface (see the description of homoclinic structures, for example, in Ref. [24]).

It follows from the Mel'nikov function (8) that in the absence of the exact resonance ($\omega \neq 1$) it has the infinite set of simple zeroes over the variable τ_0. Intersections of the stable and unstable manifolds in the infinite set of homoclinic points form a complex homoclinic structure, which generates in the vicinity of these points the transformation of the phase volume of the Smale horseshoe type and results in the Hamiltonian chaos in the reversible spontaneous emission of moving atoms even in the rotating wave approximation and at an arbitrarily small degree of modulation ε of the vacuum Rabi frequency. In the semiclassical limit, a small stochastic layer is formed in the vicinity of the separatrix of the unperturbed system, which expands with increasing ε [18].

3. Wavelet analysis of vacuum Rabi oscillations

Let us analyse numerically the nonlinear dynamics of the atomic-field system (3) in the presence of strong modulation, which is chosen for definiteness in the form $\Omega_N(\tau) = \Omega_N \sin(\omega b\tau)$ (i.e., the spatial structure of a standing wave in the cavity is described by a simple sine). It is assumed that the atoms entering the cavity are prepared (say, by means of a laser π-pulse) in the completely excited state $|\psi_N(0)\rangle_{\rm a} = |N/2, N/2\rangle$, and the cavity field is in the vacuum state. In terms of the variables of our model, such a factorised state is described by the 5-vector with co-ordinates $z_0 = 1, n_0 = 0, r_0 = 0, u_0 = 0, v_0 = 0$.

Because in the experiments with moving atoms, excitation of atoms is usually detected at the cavity exit, to illustrate the signals of reversible spontaneous emission, we present in Figs 1a–c the dependence $z(\tau)$ for $\Omega_N = 1$, $N = 10^6$, and the mismatch $\omega = 1.5$ for different velocities b of the atoms and Fig. 1d, the dependence $z(\tau)$ for $\Omega_N = 1$, and $b = 0.1$ for the exact resonance $\omega = 1$.

Note first of all that the dependence $z(\tau)$ exhibits two specific features, namely, a delay of the first superradiance pulse during which the interatomic quantum correlation is established, and a characteristic periodic structure of the regular signal caused by spatial modulation of the vacuum Rabi frequency. The dimensionless period of this modulation is $\tau_{\rm m} = \pi/b\omega$, and its numerical estimates yield the values ~ 2090, ~ 209 and ~ 31.4 for the cases in Figs 1a, b, and d, respectively. These values well agree with the corresponding periods in Fig. 1.

The maximum Lyapunov index, which characterises the dynamic chaos of a nonlinear system, was calculated to be $\lambda \approx 0$ (within the error of numerical calculations) for Figs 1a, b, and d, which, as is known, suggests the quasi(periodic) type of the corresponding dynamics. The signal in Fig. 1 not only appears as chaotic but it is indeed chaotic because the corresponding Lyapunov index is positive and equals ~ 0.1.

As the collective Rabi frequency Ω_N increases, i.e., the number N of atoms in a cluster and (or) the monatomic vacuum Rabi frequency Ω_N increases, chaos is revealed for more and more slower moving atoms. The maximum Lyapunov

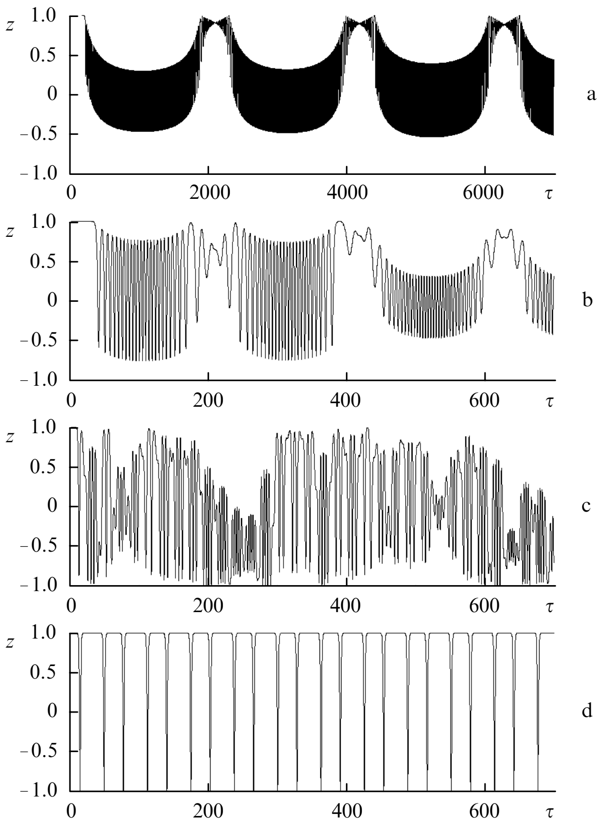

Figure 1. Inversion density oscillations of 10^6 moving atoms with the collective Rabi frequency $\Omega_N = 1$ for the velocity $b = 0.001$ (a), 0.01 (b) and 0.1 (c) and$\omega = 1.5$ (a – c) and 1 (the exact resonance, d).

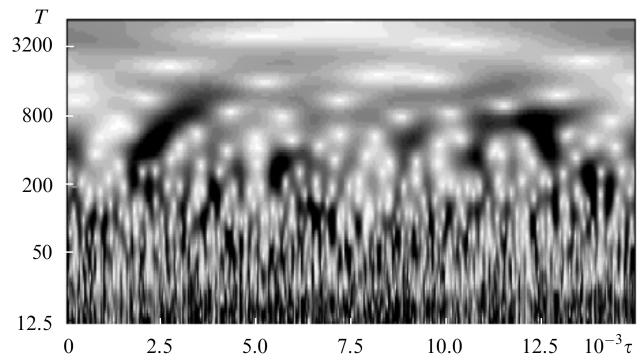

Figure 2. Wavelet transform of the chaotic signal of vacuum Rabi oscillations shown in Fig. 1c.

index calculated for $\Omega_N = 8.5$ amounts to ~ 0.5 for the velocity of atoms $\upsilon_a \simeq 1.5{\cdot}10^5$ m s^{-1} ($b = 5{\cdot}10^{-4}$) and $\omega = 0.9$.

However, the temporal type of chaos can be established neither from the form of chaotic Rabi oscillations nor from their Fourier spectrum. To determine the temporal type of chaos, we performed the wavelet transform of the corresponding signal $z(\tau)$ shown in Fig. 1c:

$$W(\alpha, \beta) = \int_{-\infty}^{\infty} z(\tau)\phi^*_{\alpha\beta}(\tau)d\tau \,, \tag{9}$$

where $\phi_{\alpha\beta}(\tau) = \alpha^{-1}\phi((\tau - \beta)/\alpha)$; $\phi = \exp(\mathrm{i}k_0\tau)\exp(-\tau^2/2)$ is the basis Morlet wavelet; α is a scaling coefficient; β is the displacement parameter; and k_0 is the fitting parameter.

The obtained two-dimensional matrix of numbers can be represented as a two-dimensional pattern with axes α and β in which the absolute values of W are shown by black half-tones (Fig. 2). The axes α and β correspond to the frequency and time scales of the signal, respectively. Each point α_0, β_0 at the picture is a convolution of $z(\tau)$ with the basis wavelet ϕ displaced by β_0 and extended by a factor of α_0. Therefore, $W(\alpha,\beta)$ contains information both on the time and frequency properties of the signal, which allows one to analyse the signal in more detail than with the help of the Fourier analysis (see, for example, [25]).

Fig. 2 shows the result of the wavelet transform of the chaotic signal $z(\tau)$ for $\Omega_N = 1$, $b = 0.1$ and $\omega = 1.5$. One can see that the Hamiltonian chaos in the reversible spontaneous emission of atoms moving in a high-Q cavity is transient in the sense that irregular oscillations occur during a random time interval and then transform to regular oscillations, which in turn transform to irregular oscillations, etc. Fig. 2 shows a distinct quasi-regular structure of the high-frequency components of the signal, with chaotic low-frequency components that appear and disappear against its background (black spots in Fig. 2).

The appearance and destruction in the reversible spontaneous emission of moving atoms is caused by the coexistence of vacuum Rabi oscillations (doubly periodic in the resonance limit) and the periodic modulation of the coupling coefficient of atoms with the cavity mode. When the periods of these processes become comparable in the absence of the resonance (i.e., at a sufficiently great velocity of atoms), the quasi-regular structure is changed by chaos even at comparatively short time scales. For low velocities of atoms or in the case of the exact resonance (for any velocities), the frequency-time structure of the reversible spontaneous emission is regular.

Fig. 3 shows an example of such a structure which represents the wavelet transform of the signal $z(\tau)$ (shown in Fig. 1d) in the case of the exact resonance between atoms and the cavity for the same values of the other parameters of the system as in Fig. 2. The regular Rabi oscillations appear under resonance conditions due to the conservation of the energy of interaction between atoms and the cavity mode, which in turn is explained by the appearance of the additional integral of motion C in the system in this limit. The regularity the high-frequency and low-frequency components of the signal is distinctly observed in its wavelet transform.

4. Some numerical estimates

In principle, our model describes the interaction between any two-level moving objects with a single emission mode. However, the validity range of the model pointed out at the beginning of the Section 2 of this paper cannot be self-consistent for any such objects and any range of the electromagnetic waves. The Rydberg atoms moving in a high- microwave cavity appear to be a suitable system for the observation of manifestations of quantum chaos in reversible spontaneous emission. The Rydberg atom maser can operate in the regime for which the assumptions we used in deriving basic Eqns (3) are valid. Below, we present our estimates of the parameters of atoms and a cavity, which we made for the experimental setup in the Paris Ecole Polytechnique [26].

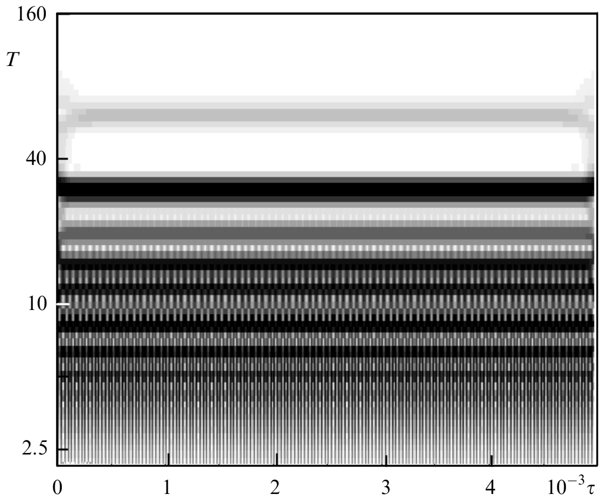

Figure 3. Wavelet transform of the regular signal of vacuum Rabi oscillations shown in Fig. 1d.

The frequency of transition between the neighbouring circular Rydberg levels of the rubidium atom with principal quantum numbers 50 and 51 is $\omega_a/2\pi = 51.099$ GHz and the electric transition dipole moment is $d = 1250$ D. The relaxation of excitation of these levels with maximum quantum numbers of the angular and magnetic momenta occurs only via a microwave transition to the nearest lower-lying circular state, so that the characteristic relaxation time is very long (~ 30 ms). Because of a large distance between the excited electron and a nucleus even a moderate electric field is sufficient for ionisation of these atoms. This fact is used for highly sensitive and selective detection of the states of atoms emerging out the cavity.

The Q-factor of the niobium superconducting cavity at a temperature of 1 K achieves $\sim 10^9 - 10^{10}$, which corresponds to the relaxation time $\sim 10 - 100$ ms of the cavity itself. The typical monatomic vacuum Rabi frequency Ω_0 equals $\sim 10^5 - 10^6$ rad s^{-1}. For such parameters of the atoms and cavity, all the conditions of the very strong coupling regime ($\Omega_0 \gg \omega_c/Q$) and of the Hamiltonian dynamics (at least for a few thousands of periods of collective vacuum Rabi oscillations) can be assumed valid.

A great wavelength (~ 1 cm) and a low recoil energy of the atoms, which accompanies the emission of microwave photons, provide the validity of point models of Dicke and Raman – Nath, respectively. In the chaotic regime of reversible spontaneous emission, the time scale of the quantum-classical correspondence decreases logarithmically, and its estimate $t_q^{ch} \simeq (\omega_a \lambda)^{-1} \ln N$ depending on the number of atoms and the Laypunov index can vary from a few tens to a few hundreds of periods of collective vacuum Rabi oscillations.

5. Conclusions

Thus, we showed analytically and numerically the possibility of the appearance of the structural dynamic chaos in the reversible spontaneous emission of two-level atoms moving inside a single-mode ideal cavity. We emphasise that this was done within the framework of a model with the mixed quantum-classical dynamics. Therefore, the validity of this result is limited by a time interval of the quantum-classical correspondence. The observation of manifestations of a classical chaos in a such fundamental process of the interaction of a matter with vacuum as spontaneous emission in real experiments would shed additional light on the problem of quantum-classical correspondence.

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Appendix

Let us introduce new operators $\hat{A} = \hat{a}/\sqrt{N}$ and $\hat{A}^\dagger = \hat{a}^\dagger/\sqrt{N}$, $\hat{S}_\rho = \hat{\sigma}_\rho/N$ with the commutation relations $[\hat{A}^\dagger, \hat{A}] = 1/N[\hat{S}_+, \hat{S}_-] = 2\hat{S}_3/N[\hat{S}_\pm, \hat{S}_3] = \mp\hat{S}_\pm/N (\rho = \pm, 3)$, which disappear in the macroscopic limit $N \to \infty$. The Heisenberg equations for the bilinear combinations of new operators $\hat{A}^\dagger\hat{A}$, $\hat{S}_+\hat{S}_-$, $\hat{u} = \hat{A}\hat{S}_+ + \hat{A}^\dagger\hat{S}_-$, $\hat{v} = \mathrm{i}(\hat{A}^\dagger\hat{S}_- - \hat{A}\hat{S}_+)$ and the atomic inversion density $\hat{S}_3$ have the form

$$\frac{\mathrm{d}}{\mathrm{d}t}(\hat{A}^\dagger\hat{A}) = -\Omega_0(t)\sqrt{N}\hat{};\quad \frac{d}{\mathrm{d}t}\hat{S}_3 = \Omega_0(t)\sqrt{N}\hat{};$$

$$\frac{\mathrm{d}}{\mathrm{d}t}(\hat{S}_+\hat{S}_-) = 2\mathrm{i}\Omega_0(t)\sqrt{N}(\hat{S}_+\hat{A}\hat{S}_3 - \hat{A}^\dagger\hat{S}_3\hat{S}_-), \tag{A.1}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\hat{u} = (\omega_c - \omega_a)\hat{};\quad \frac{\mathrm{d}}{\mathrm{d}t}\hat{=} - (\omega_c - \omega_a)\hat{u}$$

$$- 2\Omega_0(t)\sqrt{N}(\hat{S}_+\hat{S}_- + 2\hat{A}^\dagger\hat{A}\hat{S}_3).$$

By averaging the polarisation operator, we will separate the term that represents the quantum correlations of different atoms [27] $r = N^{-2}\langle \sum_{i\neq j} \hat{\sigma}_+^i \hat{\sigma}_-^j \rangle$ with summation over all the pairs of different atoms

$$\langle \hat{S}_+\hat{S}_- \rangle = \frac{1}{N^2}\left\langle \sum_{j=1}^{N} \hat{\sigma}_+^j \hat{\sigma}_-^j + \sum_{i\neq j=1}^{N} \hat{\sigma}_+^i \hat{\sigma}_-^j \right\rangle$$

$$= \frac{1}{2N} + \frac{1}{N}\langle \hat{S}_3 \rangle + \langle \hat{r} \rangle. \tag{A.2}$$

By neglecting in the averaging all the quantum correlators of the order higher than the second one, we obtain the closed five-dimensional system of equations (3) for average values $n = \langle \hat{A}^\dagger\hat{A} \rangle$, $z = 2\langle \hat{S}_3 \rangle$, $u = \langle \hat{u} \rangle$, $v = \langle \hat{v} \rangle$ and $r = \langle \hat{r} \rangle$.

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