# **Homoclinic chaos in vacuum Rabi oscillations of moving two-level atoms**

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We study analytically and numerically vacuum Rabi oscillations of *N* identical two-level atoms moving through a single-mode lossless cavity. Equations of motion which take into account the atomic quantum correlations are obtained for the quantum mechanical expectation values in the strong-coupling, rotating-wave, pointlike, and Raman-Nath approximations. It is shown that moving atoms may demonstrate an unusual type of spontaneous emission, the chaotic vacuum Rabi oscillations. This manifestation of quantum dynamical chaos in the matter-vacuum interaction is caused by a spatial inhomogeneity of the cavity mode that modulates the vacuum Rabi frequency of moving atoms. For small values of the depth of this modulation we use the Melnikov method and show analytically the presence of homoclinic chaos in this interaction. Transition to global chaos and global phase space stochasticity under conditions of the strong modulation are studied numerically by computing the maximal Lyapunov exponent  $\lambda$  of the atom-field dynamical system as a function of the number and velocity of atoms and detuning. We find a curious structure of the  $\lambda(N)$  dependence reflecting an intermittent route to global chaos. The strength of chaos depends strongly on the initial state preparation of atoms just before injecting into a cavity. It is shown that initially fully inverted atoms, which are in a superfluorescent Dicke state, demonstrate much stronger chaos (under other equal conditions) than the atoms prepared initially in a superradiant state with macroscopic polarization. A maser operating with twolevel Rydberg atoms to be injected into a high-*Q* superconducting microwave cavity seems to be a realistic device for observing some manifestations of the chaotic vacuum Rabi oscillations.

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### **I. INTRODUCTION**

The interaction between matter and vacuum, which is commonly evidenced by spontaneous emission, is one of the most fundamental dynamical interactions in nature. In free space an excited atomic state decays irreversibly because an infinity of vacuum states is available to the radiated photon. It has been predicted that cavity-confined excited atoms may experience an enhancement  $\lceil 1 \rceil$  or an inhibition  $\lceil 2 \rceil$  of spontaneous emission because of a cavity-induced modification in the vacuum-states density. These effects have been demonstrated in a number of experiments in the microwave  $[3,4]$ , infrared  $[5]$ , and optical  $[6,7]$  ranges.

In resonant cavites with one mode close to atomic transition frequency,  $\omega_f \approx \omega_a$ , two distinct regimes of spontaneous emission are realizable. When the cavity damping  $\omega_f/Q$  is large as compared with the vacuum Rabi frequency,  $\omega_f/Q$  $\mathcal{D}_{0}$ , the radiated photon is damped and an excited atomic state decays irreversibly much as it does in free space, through at an enhanced rate. In the strong-coupling regime, corresponding to the opposite condition,  $\omega_f/Q \ll \Omega_0$ , spontaneous emission becomes a reversible, oscillatory process when the atom and the field exchange excitation at the rate  $\Omega_0$ . Such a process with a sample of *N* identical atoms is known under the name ''collective vacuum Rabi oscillations,'' which have been observed both with Rydberg atoms flying through a high- $Q$  microwave cavity  $\lceil 8 \rceil$  and at optical transitions in a high-finesse optical resonator  $[9]$ .

The simple quantum theory neglecting the cavity and atomic damping and the spatial structure of the cavity mode (i.e.,  $\Omega_0$  is assumed to be constant during the interaction) predicts in the rotating-wave approximation a periodic exchange of energy between two-level atoms and a selected cavity mode at the enhanced rate  $\Omega_0\sqrt{N}$  (see, for example, Ref. [10]). It has been demonstrated in numerical experiments by the present authors  $[11]$  that atomic motion through a spatially inhomogeneous high-*Q* mode can change drastically the character of collective vacuum Rabi oscillations. In a recent communication  $[12]$ , one of the authors has shown analytically that the atomic motion produces homoclinic Hamiltonian chaos in vacuum Rabi oscillations. The present paper clarifies the subject by providing a detailed theoretical and numerical treatment of this *unusual type of spontaneous emission*, vacuum Rabi oscillations of identical two-level atoms moving through a single-mode lossless cavity.

Another motivation of the present investigation is the problem of quantum chaos connected with the question of correspondence between classical and quantum dynamics. Experiments with two-level atoms confined in a cavity provide a link between micro-, meso-, and macroscopic physics. Increasing the number of atoms in the cavity, one can force the atom-plus-cavity vacuum system to operate in different regimes, from one in which quantum fluctuations are dominant to one in which the system behaves quasiclassically. Semiclassical dynamical chaos in an ensemble of cavityconfined two-level atoms has been numerically found and investigated by a number of authors  $[13]$ . In particular, we have shown  $\lceil 14 \rceil$  that two-level atoms moving through a spatially varied cavity mode produce, out of the atom-field resonance, homoclinic Hamiltonian chaos *even in the rotatingwave approximation*. However, the semiclassical approximation is known to neglect *all quantum correlations* in the atom-field system. Therefore, the corresponding equations of motion for fully decoupled quantum expectation val-

ues cannot describe vacuum Rabi oscillations at all since the state with fully inverted atoms and a vacuum field is a stationary state in the models of the papers  $[13,14]$ .

In the present paper we go beyond the simple semiclassical factorization intending to take into account the sources of spontaneous emission in the resulting equations of motion in the Heisenberg picture. The simplest way to do this is to take into account quantum correlation between different atoms, which are responsible for cooperative spontaneous emission (see, for example, Refs.  $[15,16]$ ), and to deduce the respective *closed and tractable* set of c-number equations for quantum dynamical expectation values. This is done in Sec. II along with a discussion of the approximations and assumptions involved. In Sec. III we find general exact solutions of the dynamical equations in two integrable limits. The main results of the paper are given in Sec. IV. We introduce first the Melnikov method and obtain analytical predictions for the onset of homoclinic chaos in vacuum Rabi oscillations of moving atoms in the near integrable regime with a weak spatial modulation of the vacuum Rabi frequency. Then we discuss the initial conditions for the atoms and investigate numerically with the help of the maximal Lyapunov exponent  $\lambda$  the transition to global chaos that arises under conditions of strong modulation of  $\Omega_0$ . In Sec. V we give our conclusions.

# **II. SET OF EQUATIONS FOR QUANTUM DYNAMICAL EXPECTATION VALUES WITH ATOMIC CORRELATIONS**

Our model is as simple as posible. It consists of *N* identical two-level atoms, interacting with a single field mode in a perfect cavity, with the following Hamiltonian:

$$
H = \hbar \omega_a R_3 + \hbar \omega_f \left( a^\dagger a + \frac{1}{2} \right) + \hbar \Omega_0(t) (aR_+ + a^\dagger R_-),
$$
\n(1)

where  $\omega_a$  and  $\omega_f$  are the atomic transition frequency and the frequency of the cavity mode, respectively. The operators  $a^{\dagger}$ and *a* are the creation and annihilation operators for the mode under consideration and obey the commutation relation  $[a, a^{\dagger}] = 1$ . The operators  $R_{\pm} = \sum_{j} \sigma_{\pm}^{j}$  are the total atomic dipole operators composed of the raising  $\sigma^j_+$  and lowering  $\sigma^j$  Pauli operators for the individual atoms;  $R_3 = \frac{1}{2} \sum_j \sigma^j_3$ equals the total atomic energy operator apart from factor  $\hbar \omega_a$ . The collective operators obey commutation relations

$$
[R_+, R_-] = 2R_3, [R_3, R_{\pm}] = \pm R_{\pm}.
$$
 (2)

In maser experiments an atom passes along the axis *x* of a closed cavity and consequently experiences a spatial modulation of the coupling coefficient, the single-atom vacuum Rabi frequency  $\Omega_0$ . If we assume its velocity to be constant  $v_a$ , then the effect of motion on the internal dynamics can be included in the usual way  $\Omega_0(t) \rightarrow \Omega_0(x/v_a)$ .

In order to avoid complications, which are not essential to the main scope of this paper, we have made a number of approximations and simplifications.

 $(1)$  The single mode, two-level, and rotating-wave approximations.

~2! The pointlike approximation. In other words, *N* atoms are assumed to be confined to a volume less than the cavitymode wavelength cubed and the atoms inside this volume may be considered as undistinguishable. The pointlike approximation seems to be reasonable in the microwave region with the wavelength of the order of 1 cm.

 $(3)$  The Raman-Nath approximation. We assume that the atoms are injected into a cavity with a velocity high enough to enable us to neglect any change in their kinetic energy. In the microwave region, the recoil energy of atoms accompanying emission of photons is very small  $\lfloor 14 \rfloor$ .

~4! The strong-coupling regime. In this regime, *N* atoms exchange excitation with a cavity field with a period  $T_R$  $=2\pi/\Omega_0\sqrt{N}$  that is much shorter than both the atomic and cavity relaxation times. The present Rydberg atom masers can be operated in this regime, where  $\Omega_0\sqrt{N} \gg \omega_f/Q$  (for a review of Rydberg atom masers see Ref. [17]).

(5) The assumption of partial decorrelation between the atomic and the field degrees of freedom. It means that when deducing c-number equations of motion from operator equations we neglect quantum atom-field correlators higher than second-order ones. This mixed quantum-classical description differs both from a semiclassical one  $[13,14]$ , when one neglects quantum correlators of *all* orders, and from fully quantum description that leads to an infinite hierarchic set of equations for quantum correlators (cumulants).

The problem of dynamical chaos in the quantum system with the Hamiltonian  $(1)$  will be treated in the Heisenberg representation as it is close in its spirit to classical mechanics. Therefore, it is to be desired to derive a tractable closed set of equations of motion for expectation values from the respective operator Heisenberg equations. The simplest way to do this is achieved by writing down the Heisenberg equations for the atomic operators and the field operators, averaging them over an initial quantum state and factorizing all the operator products of the type  $\langle (a \pm a^{\dagger}) R_{\alpha} \rangle$ , where  $\alpha =$  $\pm$ ,3 [13,14]. This simple semiclassical approximation is known to neglect not only the atom-field correlation but the atom-atom correlations as well. The atom-atom correlations occur only through the mediation of the field generated by the atoms (we neglect the dipole-dipole interaction) and are responsible for cooperative spontaneous emission.

In order to take into account the sources of spontaneous emission in the Heisenberg representation one should go beyond the simple semiclassical factorization. Let us introduce operators normalized to the number *N* of atoms

$$
A = \frac{a}{\sqrt{N}}, \quad A^{\dagger} = \frac{a^{\dagger}}{\sqrt{N}}, \quad S_{\alpha} = \frac{1}{N} R_{\alpha} \tag{3}
$$

with the commutation relations

$$
[A^{\dagger}, A] = \frac{1}{N}, \ [S_+, S_-] = \frac{2}{N} S_3, \ [S_{\pm}, S_3] = \pm \frac{1}{N} S_{\pm},
$$
  

$$
\alpha = \pm, 3,
$$
 (4)

and consider the following set of the bilinear products of the operators

*d*

*d*

$$
A^{\dagger}A
$$
,  $S_{+}S_{-}$ ,  $U=AS_{+}+A^{\dagger}S_{-}$ ,  $V=i(A^{\dagger}S_{-}-AS_{+})$ . (5)

The Heisenberg equations for the density of the atomic inversion  $S_3$  and the operators  $(5)$  are easily derived from the Hamiltonian (1)

$$
\frac{d}{dt}(A^{\dagger}A) = -\Omega_0(t)\sqrt{N}V,
$$
\n
$$
\frac{d}{dt}S_3 = \Omega_0(t)\sqrt{N}V,
$$
\n
$$
\frac{d}{dt}U = (\omega_f - \omega_a)V,
$$
\n(6)\n
$$
\frac{d}{dt}(S_+S_-) = 2i\Omega_0(t)\sqrt{N}(S_+AS_3 - A^{\dagger}S_3S_-),
$$

$$
\frac{d}{dt}V = -(\omega_f - \omega_a)U - 2\Omega_0(t)\sqrt{N}(S_+S_- + 2A^{\dagger}AS_3).
$$

At  $t=0$ , just before injecting into a cavity, the atoms and field are assumed to be uncorrelated. The initial quantum state, over which we shall average Eqs.  $(6)$ , is the following product state:

$$
|\psi(0)\rangle = |\psi_a(0)\rangle \otimes |\psi_f(0)\rangle. \tag{7}
$$

The equations of motion are obtained by taking the expectation values for the Heisenberg operators with the initial quantum state  $(7)$ . The nonlinearity of the two last equations in the set  $[Eq. (6)]$  brings second- and third-order correlations into the equations for the first moments. Our task is to ''save'' those second-order atomic correlations which are responsible for the cooperative spontaneous emission and to derive a closed and tractable dynamical system. When averaging the polarization operator we separate the term representing correlations between different atoms [16]  $\mathcal{R}$  $=N^{-2}\langle \sum_{i \neq j} \sigma^i + \sigma^j \rangle$ , where the sum is over all pairs of different atoms. Thus

$$
\langle S_{+} S_{-} \rangle = \frac{1}{N^2} \left( \left\langle \sum_{j=1}^{N} \sigma_{+}^{j} \sigma_{-}^{j} + \sum_{i \neq j=1}^{N} \sigma_{+}^{i} \sigma_{-}^{j} \right\rangle \right)
$$

$$
= \frac{1}{2N} + \frac{1}{N} \langle S_{3} \rangle + \langle \mathcal{R} \rangle, \tag{8}
$$

where we have used the following property of the Pauli operators  $\sigma_+ \sigma_- = \frac{1}{2}(I + \sigma_z)$  with *I* being the identity operator. When averaging the products of three operators in the last two equations of Eq.  $(6)$  we factorize quantum correlators of third order  $\langle S_+AS_3\rangle$ ,  $\langle A^{\dagger}S_3S_-\rangle$ , and  $\langle A^{\dagger}AS_3\rangle$  into products of second-order and first-order ones (see the fifth assumption in our list of approximations involved). Using the known properties of the Pauli operators,  $\sigma_z^i \sigma_-^i = -\sigma_-^i$  and  $\sigma_+^i \sigma_z^i$  $=-\sigma^i_+$ , and the assumption of decorrelation between polarization and inversion of different atoms,  $\langle \sigma_z^i \sigma_z^j \rangle$  $= \langle \sigma_z^i \rangle \langle \sigma_z^{-i} \rangle$  and  $\langle \sigma_+^i \sigma_z^i \rangle = \langle \sigma_+^i \rangle \langle \sigma_z^{-i} \rangle$ , we obtain

$$
\langle S_+AS_3 \rangle = \langle AS_+ \rangle \left( \langle S_3 \rangle - \frac{1}{2N} \right),
$$
  

$$
\langle A^{\dagger} S_3 S_- \rangle = \langle A^{\dagger} S_- \rangle \left( \langle S_3 \rangle - \frac{1}{2N} \right),
$$
  

$$
\langle A^{\dagger}AS_3 \rangle = \langle A^{\dagger}A \rangle \langle S_3 \rangle.
$$
 (9)

With the help of Eqs.  $(8)$  and  $(9)$  we can now derive the closed *c*-number equations of motion from the operator equations  $(6)$ 

$$
\dot{n} = -\Omega_N(\tau)v,
$$
  
\n
$$
\dot{z} = 2\Omega_N(\tau)v,
$$
  
\n
$$
\dot{u} = (\omega - 1)v,
$$
  
\n
$$
\dot{r} = -\Omega_N(\tau)zv,
$$
  
\n
$$
\dot{v} = (1 - \omega)u - \Omega_N(\tau)\left(\frac{z + 1}{N} + 2r + 2nz\right),
$$
\n(10)

where dot denotes differentiation with respect to the dimensionless time  $\tau = \omega_a t$ . The time-dependent and timeindependent coupling coefficients are the dimensionless collective vacuum Rabi frequency and the dimensionless detuning, respectively

$$
\Omega_N(\tau) = \frac{\Omega_0(\tau)\sqrt{N}}{\omega_a}, \quad \omega = \frac{\omega_f}{\omega_a}.
$$
 (11)

The classical variables  $n=\langle A^\dagger A\rangle$  and  $z=2\langle S_3\rangle$  are the density of photons in a cavity and the density of the atomic inversion, respectively. The variables  $u = \langle U \rangle$  and  $v = \langle V \rangle$ are the atom-field correlators of second order describing the dressed atoms. The quantity  $r=\langle \mathcal{R} \rangle$  represents correlations among different atoms. If there are initially no correlations, polarization, and photons, we still have on the right side of the last equation in the set  $(10)$  the term  $z+1$  which equals twice the density of the atoms in the excited state. Namely, this term is the source of spontaneous emission. It drives *v*, which in turn drives the other variables in our atom-field dynamical system  $(10)$ , creating atom-atom correlations, polarization, and cavity photons.

Two integrals of motion can be found by inspection from the system  $(10)$ . The first one

$$
W = z + 2n \tag{12}
$$

reflects a conservation of energy in a lossless cavity. The other one

$$
S = z^2 + 4r \tag{13}
$$

results from the unitarity of atomic evolution. The value of the constant *S* will be found in Sec. IV in terms of the Dicke cooperation number and the number of atoms.

## **III. THE INTEGRABLE LIMITS OF THE EQUATIONS OF MOTION**

If a spatial inhomogeneity of the cavity mode can be neglected (for example, with motionless atoms in the pointlike approximation), our model  $(10)$  is integrable. It becomes clear after finding the third integral of motion if one assumes the vacuum Rabi frequency  $\Omega_0$  to be constant. This extra integral

$$
C = 2\Omega_N u - (\omega - 1)z\tag{14}
$$

reflects a conservation of the interaction energy between motionless atoms and a cavity field that is a consequence of the rotating-wave approximation made.

With the help of the three integrals *W*, *S*, and *C* it is easy to show that the density of the atomic inversion satisfies the following second-order nonlinear differential equation:

$$
\ddot{z} = 3\Omega_N^2 z^2 - \left[ (\omega - 1)^2 + 2\Omega_N^2 \left( W + \frac{1}{N} \right) \right] z - (\omega - 1)C
$$

$$
-\Omega_N^2 \left( S + \frac{2}{N} \right),\tag{15}
$$

$$
\dot{z}(\tau=0) = 2\Omega_N v_0, \ \ z(\tau=0) = z_0.
$$

The energy integral for Eq.  $(15)$  has the form

$$
E = \frac{1}{2}\dot{z}^2 + F,\tag{16}
$$

where

$$
F = -\Omega_N^2 z^3 + \left[ \frac{(\omega - 1)^2}{2} + \Omega_N^2 \left( W + \frac{1}{N} \right) \right] z^2
$$

$$
- \left[ (\omega - 1)C + \Omega_N^2 \left( S + \frac{2}{N} \right) \right] z \tag{17}
$$

is the ''potential energy.'' By inverting the elliptic integral of the first kind

$$
d\tau = \pm \frac{dz}{\sqrt{2(E - F)}},\tag{18}
$$

it is easy to find the solution for the density of the atomic inversion in terms of the elliptic Jacobian function

$$
z(\tau) = z_1 + (z_2 - z_1) \operatorname{sn}^2 \left[ \sqrt{\frac{1}{2} (z_3 - z_1)} \Omega_N(\tau - \tau_1); \frac{z_2 - z_1}{z_3 - z_1} \right],
$$
\n(19)

where

$$
\tau_1 = \frac{1}{\Omega_N \sqrt{2}} \int_{z_0}^{z_1} \frac{dz}{\sqrt{(z - z_1)(z - z_2)(z - z_3)}},\tag{20}
$$

and  $z_{1,2,3}$  are the roots of the algebraic cubic equation

$$
E - F = 0.\t(21)
$$

The periodic exchange of energy between initially excited atoms and cavity vacuum field to be described by Eq.  $(19)$  is known as regular vacuum Rabi oscillations. The exact solutions for the other variables are found after solving Eq.  $(15)$ 

$$
2n = W - z,
$$
  
\n
$$
4r = S - z^2,
$$
 (22)  
\n
$$
2\Omega_N u = C + (\omega - 1)z,
$$
  
\n
$$
2\Omega_N v = \dot{z}.
$$

There exists another nontrivial integrable limit of the equations of motion  $(10)$  when the frequency of the cavity mode  $\omega_f$  coincides exactly with the atomic transition frequency  $\omega_a$ , i.e., if  $\omega=1$ . The atom-field system with  $\omega=1$ 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 five-dimensional problem  $(10)$  to the four-dimensional one (the variable  $u$  becomes a constant). In this integrable limit, the general exact solution is obtained by putting  $\omega=1$  in the formulas  $(15)$ – $(22)$  and transforming to the new "time"  $\tau$  $\rightarrow \int_0^{\tau} f(\tau') d\tau'$ . The resonant two-level atoms when moving through a lossless single-mode cavity will experience a periodic exchange of energy with the cavity field *regardless of the spatial structure of the cavity mode along the propagation axis*. In the next section we will show what happens with nonresonant two-level atoms moving in a spatially varying field.

### **IV. CHAOTIC VACUUM RABI OSCILLATIONS**

#### **A. The onset of homoclinic chaos**

We have shown in Sec. III that both in the limit of the nonhomogeneous resonant interaction  $(\omega=1)$  and in the limit of the homogeneous nonresonant interaction  $(\Omega_N)$  $=$ const) the vacuum Rabi oscillations are periodic. In particular, the atomic inversion,  $z(\tau)$ , is goverened by the nonlinear oscillator equation  $(15)$  whose phase plane is divided into regions of bounded and unbounded motion by a separatrix loop corresponding to a homoclinic orbit

$$
z(\tau) = z_1 + (z_2 - z_1) \tanh^2 \left[ \sqrt{\frac{1}{2} (z_3 - z_1)} \Omega_N(\tau - \tau_0) \right],
$$
\n(23)

where  $\tau_0$  is the time parametrizing this orbit and the roots  $z_{1,2}$  can found from Eqs. (17) and (21) with  $z_2 = z_3$  and the given initial conditions. In this section we use the Melnikov method  $[18]$  to prove a homoclinic structure in the vicinity of the unperturbed separatrix  $(23)$  that is produced out of resonance ( $\omega \neq 1$ ) even under an extremely small spatial modulation of the vacuum Rabi frequency  $\Omega_N(\tau)$  of moving atoms. It leads to a replacement of the separatrix of the unperturbed system by a stochastic layer in the perturbed one. The modulation is assumed to be equal to

$$
\Omega_N(\tau) = \Omega_N + \epsilon \sin b \omega \tau,\tag{24}
$$

where  $\epsilon$  is sufficiently small as compared with  $\Omega_N$ , and  $b\omega$ is the dimensionless modulation frequency with  $b = v_a/c$  be $\overline{1}$ 

ing the ratio of velocity of atoms to the velocity of light. Stable and unstable homoclinic manifolds of a hyperbolic fixed point coinciding in the unperturbed system ( $\epsilon=0$ ) begin to intersect transversally in the perturbed system with the parametric modulation  $(24)$ . The signed distance between the perturbed stable and unstable manifolds at  $\tau_0$  along a normal **n** to the unperturbed homoclinic manifold is proportional to  $\epsilon M(\tau_0)+O(\epsilon^2)$ , where the Melnikov function is given by  $\lceil 18 \rceil$ 

$$
M(\tau_0) = \int_{-\infty}^{\infty} \mathbf{n} \cdot \mathbf{G} d\tau.
$$
 (25)

The perturbation part of the vector field **G** has the form

$$
\mathbf{G} = \left( -v \sin b \omega \tau, 2v \sin b \omega \tau, 0, -zv \sin b \omega \tau, -\left( \frac{z+1}{N} + 2r + 2nz \right) \sin b \omega \tau \right)^{T}.
$$
 (26)

The Melnikov distance is nonzero only in the direction **n**  $=(0,1-\omega,2\Omega_N,0,0)^T$ . After evaluating the scalar product  $\mathbf{n} \cdot \mathbf{G}$  on the separatrix (23) and carrying out the integration by parts we obtain the final result

$$
M(\tau_0) = \frac{2\,\pi(1-\omega)(b\,\omega)^2}{\Omega_N^3 \sinh(b\,\omega\,\pi/\sqrt{z_3 - z_1}\Omega_N)} \cos b\,\omega\,\tau_0. \tag{27}
$$

The Melnikov function has, out of resonance, simple zeros as a function of  $\tau_0$  implying transverse intersections between stable and unstable manifolds in an infinite variety of homoclinic points. These intersections produce in the vicinity of each homoclinic point a transformation of the type of the Smale horseshoe  $[19]$ . In other words, the initial volume element in the system's phase space stretches along the unstable manifold, shrinks along the stable manifold, and folds. It results in the local instability of almost all trajectories starting inside this homoclinic structure where Kolmogorov-Arnol'd-Moser (KAM) tori cannot exist because of changing the topology of trajectories  $[20]$ . Thus the transverse intersections proven analytically above in the absence of resonance lead to the onset of homoclinic chaos in the vacuum Rabi oscillations of moving atoms.

#### **B. Initial conditions**

The theory developed and the solutions obtained in the preceding sections are valid with arbitrary initial conditions. We have only assumed atoms and field to be initially in the product state  $(7)$ . Because we are mainly interested in cooperative spontaneous emission, we shall adopt the vacuum state of the initial cavity field

$$
|\psi_f(0)\rangle = |0\rangle. \tag{28}
$$

In the pointlike approximation, all the atoms experience identical conditions and must be treated as undistinguishable particles. The collective states of a *N*-atom system are the Dicke states  $[21]$   $|R,M\rangle$  which are labeled by the cooperation number *R* and the transition number  $M = Nz/2$ 

$$
\mathbf{R}^2|R,M\rangle = R(R+1)|R,M\rangle, R_3|R,M\rangle = M|R,M\rangle, \tag{29}
$$

$$
R_{\pm}|R,M\rangle=[(R\mp M)(R\pm M+1)]^{1/2}|R,M\pm 1\rangle,
$$

where

$$
\mathbf{R}^2 = \frac{1}{2} (R_+ R_- + R_- R_+) + R_3^2, \quad R = \frac{N}{2},
$$
  

$$
\frac{N}{2} - 1, \dots, 1, \quad 0 \quad \left(\frac{1}{2}\right), \quad M = -R, \quad -R + 1, \dots, R - 1, \quad R. \tag{30}
$$

Averaging the operator  $R_+R_- = N^2S_+S_-$  in Eq. (8) over the Dicke states  $|R, M\rangle$  and using the operator identity (30) we can write down the atomic integral of motion  $(13)$  in terms of the cooperation number *R* and the number of atoms *N*

$$
S = \frac{4}{N^2}R(R+1) - \frac{2}{N}.
$$
 (31)

Those Dicke states, which are symmetric under atom exchange, possess the maximal cooperation number  $R = N/2$ and are easily realized with Rydberg atoms in cavities  $[17]$ . In these states  $|N/2,M\rangle$  the excitation is symmetrically shared among all atoms, and the collective *N*-atom system has only  $N+1$  nondegenerate equidistant energy levels. Being prepared is one of the symmetric Dicke states, the system will make transitions, only into other symmetric states. It is easy to show that  $S=1$  for the atoms initially prepared in a symmetric Dicke state.

Computer simulations in this paper will be done with two different initial symmetric Dicke states. Adopting the terminology of the theory of supperradiance (see Refs.  $[21,15]$ ), we name the state

$$
|\psi_a(0)\rangle_1 = \left|\frac{N}{2},0\right\rangle
$$
 (32)

with strongly correlated atoms  $(r_0=1/4)$  and a macroscopic electric dipole ( $z_0=0$ ) as a superradiant state. The radiation is due to this dipole. Within the accuracy  $O(1/N)$  the superradiant state  $(32)$  can be shown [15] to be coincide with a coherent atomic state prepared from the ground state by a classical  $\pi/2$  — pulse. The state with fully excited and uncorrelated atoms

$$
|\psi_a(0)\rangle_2 = \left|\frac{N}{2}, \frac{N}{2}\right\rangle
$$
 (33)

is known as a superfluorescent atomic state. The state with all the atoms on the upper level is realized, for example, by preparing the atoms in the ground state and then radiating the *N*-atom system with a classical  $\pi$  — pulse. In the superfluorescent state  $r=0$  and no macroscopic polarization is present in the system  $(z=1)$ . Nevertheless, the atoms begin to radiate spontaneously creating atomic correlations, a macroscopic dipole, and photons in a cavity. Other quasiclassical coherent atomic states, which may be represented as a linear combination of the symmetric Dicke states, are prepared from the ground state by a classical  $2|\gamma|$  — pulse with  $\pi/2$  $\leq$  2 $\gamma$   $\leq$   $\pi$  [15].

#### **C. Transition to global chaos**

We proved analytically the presence of homoclinic chaos in vacuum Rabi oscillations in the near integrable regime where the depth of spatial modulation of the vacuum Rabi frequency of moving atoms  $[Eq. (24)]$  is small as compared with its amplitude value. In Hamiltonian systems, the stochastic layer in the vicinity of an unperturbed separatrix exists under any arbitrary small perturbation strength  $[20,22]$ . Due to intersections of stable and unstable homoclinic manifolds invariant KAM tori cannot survive in such a layer. As the perturbation strength increases, the chaotic dynamics, typically, must cover extended regions of phase space.

In order to study transition to global chaos we will assume atoms moving through a cavity in a direction *x* along which the spatial variation of a cavity mode is described by the function

$$
f(x) = \sin \frac{p \pi x}{L_c}.
$$
 (34)

It corresponds to a  $TE<sub>p</sub>$  mode in a rectangular cavity with  $L<sub>c</sub>$ being the cavity length and  $p+1$  being the number of nodes in the cavity. Then the vacuum Rabi frequency becomes the time-periodic function

$$
\Omega_N(\tau) = \Omega_N \sin b \,\omega \,\tau,\tag{35}
$$

where the normalized cavity frequency (the dimensionless detuning) is  $\omega = p \pi c / L_c \omega_a$ .

The origin of dynamical chaos is in extremal sensitive to initial conditions which is characterized by the Lyapunov exponents

$$
\lambda_i = \lim_{\tau \to \infty} \lambda_i(\tau), \quad \lambda_i(\tau) = \lim_{\Delta_i(0) \to 0} \frac{1}{\tau} \ln \frac{\Delta_i(\tau)}{\Delta_i(0)},\tag{36}
$$

where  $\Delta(\tau)$  is the distance between two initially adjacent trajectories at time  $\tau$ , which may be specified as the Euclidean distance between two phase-space points. Due to two conserved quantities  $(12)$  and  $(13)$ , the motion of the fivedimensional atom-field system with moving atoms  $(10)$  is restricted on a three-dimensional hypersurface and characterized by three Lyapunov numbers ( $\lambda_i$ ,  $i=1,2,3$ ). The volume of a given element of the phase space of our conservative system should be invariant, i.e.,  $\lambda_1 + \lambda_2 + \lambda_3 = 0$ . Therefore, we have positive, negative, and zero Lyapunov exponents. If  $\lambda_i$  < 0, then the volume element shrinks in the corresponding direction, and if  $\lambda_i$ >0, then it expands exponentially in that direction. One has the linear growth if  $\lambda_i=0$ . A chaotic flow arises when the initial volume element stretches, shrinks, and folds (for a review of dynamical chaos in Hamiltonian systems see Refs.  $[20,22,23]$ . Because of the global confinement in phase space of the atom-field system, the local exponential divergence of trajectories, which produces a local stretching, is accompanied by folding. Repeated stretching and folding produces homoclinic chaos in cooperative emission and absorption by moving two-level atoms. For chaotic motion, the maximal Lyapunov exponent is a measure of the rate of divergence of phase trajectories. For a wide class of dynamical systems (see Refs. [22,23]),  $\lambda$  has been shown to be of the order of the reciprocal of a mixing time  $\tau_c$ , the time scale of decay of a correlation function  $C(t)$  $\sim \exp(-t/\tau_c)$ . It means that the mixing property of chaos follows from the property of local dynamical instability.

The maximal Lyapunov exponent  $\lambda$  gives us the quantitative criterion of dynamical chaos. The motion is said to be chaotic when  $\lambda > 0$ . We have computed the maximal Lyapunov exponents of the atom-field system  $(10)$  under the perturbation in the form  $(35)$  with the following initial conditions: no photon present and the atoms strongly correlated at  $\tau=0$ 

$$
|\psi(0)\rangle_1=|0\rangle\otimes\left|\frac{N}{2},0\right\rangle
$$
:  $(n_0=z_0=u_0=v_0=0, r_0=\frac{1}{4}),$  (37)

and no photons present and the atoms uncorrelated at  $\tau=0$ 

$$
|\psi(0)\rangle_2 = |0\rangle \otimes \left|\frac{N}{2}, \frac{N}{2}\right\rangle: (n_0 = r_0 = u_0 = v_0 = 0, z_0 = 1).
$$
\n(38)

As was shown in the preceding subsection, both atomic states can be prepared from the ground atomic state by a classical pulse of the appropiate area just before injecting the atoms into a cavity.

Numerical experiments have been carried out for the dynamical system  $(10)$  with those values of parameters that may be considered as achievable, in principle, with a Rydberg atom maser, a device operating with Rydberg two-level atoms moving through a high-*Q* superconducting microwave cavity  $[3,8,17]$ . This device can be really operated in the regime when all the assumptions adopted in our model (see Sec. II) may be considered as valid. In the strong-coupling regime, the one-photon vacuum Rabi frequency of a single atom may reach  $\Omega_0 \approx 10^6$  rad/s [17]. The period of collective vacuum Rabi oscillations  $2\pi/\Omega_0\sqrt{N}$  is much shorter than the lifetimes of Rydberg states and microwave photons  $(2.10^{-2} \text{ s})$  in a few centimeter size cavity with  $Q \approx 10^{9}$ . It implies the Hamiltonian approach, strong-coupling limit, pointlike, and Raman-Nath approximations adopted to be valid. Typical transition frequencies between working Rydberg states are in the range  $\omega_a \approx 10^{10} - 10^{11}$  rad/s. The dimensionless single-atom and collective vacuum Rabi frequencies with the given value of  $\Omega_0$  are estimated to be in the ranges  $\Omega = \Omega_0 / \omega_a \approx 10^{-5} - 10^{-4}$  and  $\Omega_N = \Omega_0 \sqrt{N}/\omega_a$  $\approx$  1–10 (with *N*=10<sup>10</sup>), respectively.

First of all, we compute the maximal Lyapunov exponent  $\lambda$  as a function of the number of atoms *N*. The numerical simulations are run for the nonresonant case,  $\omega$ =0.9, with the dimensionless single-atom vacuum Rabi frequency  $\Omega$  $=10^{-5}$  and the dimensionless velocity of atoms  $b=0.01$  and 0.001. Figure 1 demonstrates the dependence of the maximal Lyapunov exponent on the common logarithm of the number of atoms for the superradiant initial condition  $(37)$ . There is a curious structure of the dependence  $\lambda(\log_{10}N)$  in the beginning of transition to global chaos. Figure  $1(b)$  shows enlargement of this structure. Quite regular peaks on a fine scale



FIG. 1. (a) Dependence of the maximal Lyapunov exponent  $\lambda$ on the number of atoms *N* on a logarithmic scale for the superradiant initial atomic state  $|N/2,0\rangle$ : $\Omega = 10^{-5}$ ,  $\omega = 0.9$ ,  $b = 0.01$ ; (b) enlargement of the initial portion of this dependence. All variables and parameters are in dimensionless units:  $\Omega = \Omega_0 / \omega_a$ ,  $\omega$  $= \omega_f/\omega_a$ , and  $b = v_a/c$ .

reflect an intermittent route to chaos and a possible cascade of bifurcations when changing the number of atoms in an atomic ''droplet'' to be injected into a cavity. Under the superradiant initial condition and with given Rabi frequency and the detuning, vacuum Rabi oscillations becomes chaotic when *N* reaches  $\simeq 10^9$  atoms.

Figure 2(a) demonstrates the dependence  $\lambda(\log_{10}N)$  with initially fully inverted atoms [see Eq.  $(38)$ ] at the same fixed values of the control parameters as in Fig. 1. It should be stressed that with the superfluorescent atoms the strength of chaos measured by the value of  $\lambda$  at a fixed value of *N* is almost ten times more than that with the superradiant atoms, and chaos begins to show up with  $N \approx 10^8$  atoms. As a result of extremal instability of the system with initially fully inverted atoms, the respective dependence  $\lambda(log_{10}N)$  is a highly irregular one. In Fig. 2(b) the dependence of  $\lambda$  on the number of atoms is computed at  $b=0.001$ , i.e., at  $v_a=3$  $\times 10^5$  m/s.



FIG. 2. Dependence  $\lambda(N)$  on a logarithmic scale for the superfluorescent initial atomic state  $|N/2,N/2\rangle$ :  $\Omega = 10^{-5}$ ,  $\omega = 0.9$ . (a) *b*  $=0.01$ ; (b)  $b=0.001$ . All variables and parameters are in the same dimensionless units as in Fig. 1.

Behavior of the maximal Lyapunov exponent near the atom-cavity resonance is computed at  $\Omega_N = 8.5$ ,  $N = 10^{10}$ , and  $v_a = 3 \times 10^5$  m/s( $b = 0.001$ ) with initially fully inverted atoms and shown in Fig. 3. At exact resonance, the atomfield dynamics was analytically shown in Sec. III to be regular. In this case  $\lambda$  should be equal to zero as one can see in the figure at  $\omega=1$ . Nearby the exact resonance, Fig. 3 shows very strong chaos with the values of  $\lambda$  of the order of unity. Dependence of  $\lambda$  on the velocity of initially fully inverted atoms has been computed at  $\Omega_N = 8.4$ ,  $N = 10^{10}$ , and  $\omega$  $=0.9$ . A fragment of this rather irregular dependence on a logarithmic scale in the velocity range  $v_a = 3 \times 10^4 - 3$  $\times 10^5$  m/s is shown in Fig. 4. Weak chaos ( $\lambda \approx 0.01$ ) begins to show up with the velocity of atoms of the order of  $v_a$  $\approx$ 3×10<sup>4</sup> m/s.

It should be stressed that chaos arises in the adiabatic regime of the parametric vacuum Rabi oscillations,  $b\omega$  $\mathcal{L}_{N}$ , where the frequency of the spatial modulation is much less than the collective vacuum Rabi frequency, that is,



FIG. 3. Dependence of  $\lambda$  on the normalized cavity frequency (dimensionless detuning) near the atom-cavity resonance for the superfluorescent initial atomic state :  $\Omega_N = 8.5$ , $N = 10^{10}$ , and  $b = 0.001$ . All variables and parameters are in the same dimensionless units as in Fig. 1 with  $\Omega_N = \Omega_0 \sqrt{N/\omega_a}$ .

the frequency of the energy exchange between the collection of *N* identical atoms and a cavity mode.

### **V. CONCLUSION**

Under appropriate conditions, cooperative spontaneous emission from moving two-level atoms becomes a reversible process. In the present work we have shown that the simplest model of this process may demonstrate a very complicated dynamics which is chaotic in the sense of extremal sensitivity to initial atomic state. The basic results of our study are as follows.

The dynamical equations for quantum expectation values that take into account quantum atomic correlations (responsible for the cooperative spontaneous emission) are derived. The equations of motion are shown to be integrable in the limit of exact resonance between moving atoms and a spatially inhomogeneous cavity mode and in the limit of a spatially homogeneous field interacting with nonresonant atoms. The respective vacuum Rabi oscillations may be considered as periodic in these limits.



FIG. 4. Dependence of  $\lambda$  on the velocity of atoms (in m/s units) on a logarithmic scale for the superfluorescent initial atomic state :  $\Omega_N$ =8.4, *N* = 10<sup>10</sup>, and  $\omega$ =0.9.

In general, moving nonresonant atoms may demonstrate a type of reversible spontaneous emission, chaotic vacuum Rabi oscillations, which are shown to be of a homoclinic nature. This process depends strongly on the initial preparation of atoms just before injecting into a cavity. Under the other equal conditions, chaos is much stronger with atoms to be prepared in the fully inverted state  $|N/2,N/2\rangle$  than with those prepared initially in the superradiant state  $|N/2,0\rangle$ . The numerical experiments computing maximal Lyapunov exponents predict the values of the control parameters (the detuning, the velocity, and the number of atoms) for which it

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would be worthwhile to search for the chaotic vacuum Rabi oscillations in real experiments.

Rydberg atoms in superconducting microwave cavities are a well-suited system to observe manifestations of quantum chaos in the vacuum Rabi oscillations. This device can be actually operated in the regime when all the assumptions adopted in our model may be considered as valid.

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