

# Stochastic mechanism of generation of optical radiation

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An analysis is made of a resonant interaction of a gas of two-level systems with its own radiation field and with an external amplitude-modulated field. It is shown that an analog of the stochastic Fermi acceleration mechanism may appear in such a gas when certain stochastization conditions are satisfied. In the present case this mechanism leads to significant generation of a radiation field of two-level systems even in the case of relatively low values of the constant representing the interaction of the field with the two-level medium. An analysis is made of the possibility of observing this effect in atomic and molecular gases, and also in crystals containing impurity centers.

## INTRODUCTION

The phenomenon of dynamic chaos in various physical systems is currently attracting much attention.<sup>1-3</sup> One of the topics being investigated is dynamic chaos due to interaction of light with matter.<sup>4</sup> It is meaningful to distinguish two types of nonlinear optical systems manifesting the property of stochasticity: dissipative dynamic systems and Hamiltonian systems. We shall consider dynamic chaos using the Hamiltonian approach, which imposes restrictions on the characteristic times of the dynamics of the system.

A stochastic instability of nonlinear oscillations is frequently a harmful effect which limits the effectiveness of various physical processes.<sup>1-4</sup> However, in some cases such an instability can be used to heat a plasma,<sup>2,3</sup> to exchange energy effectively as a result of interaction of intermode resonances in the process of excitation of polyatomic molecules or in laser photochemistry,<sup>5</sup> etc. Probably the most interesting potential application of dynamic chaos is that based on the stochastic acceleration mechanism first put forward by Fermi in connection with the problem of acceleration of cosmic rays.<sup>6</sup> Zaslavskii and Chirikov demonstrated<sup>7</sup> that this mechanism may be realized in fairly simple nonlinear systems with several degrees of freedom when the conditions for the stochastic instability are satisfied. At present the stochastic acceleration mechanism is regarded as one of the promising methods for the acceleration of particles (see Ref. 8 and the literature cited there). It would therefore be of interest to study an analog of this stochastic acceleration mechanism in nonlinear optical processes.

It was shown in Ref. 9 that when an ensemble of two-level systems interacts with its own radiation field (self-consistent field), fluctuations of the polarization, of the difference between the populations, and of the self-consistent field may become chaotic when the value of the constant representing the interaction between field and matter is sufficiently large so as to correspond to a situation when the approximation of a rotating wave is no longer valid. A numerical experiment<sup>10</sup> was used in a study of the interaction of an ensemble of multilevel systems characterized by a strongly nonequidistant spectrum with a self-consistent radiation field and with an external harmonic field in resonance with the lowest transition in the multilevel systems. It was demonstrated in Ref. 10 that the interaction of the radiation field with such multilevel systems can become chaotic when the

rotating wave approximation is no longer valid and it was also shown that when the conditions are suitable, a self-consistent radiation field may grow in a diffusion-like manner and this may be accompanied by the filling of higher states in multilevel systems (which is an analog of the Fermi stochastic acceleration effect). We demonstrated earlier<sup>11</sup> that in the interaction of an ensemble of two-level systems with a self-consistent field and with an external harmonic field a high value of the constant of the interaction of such systems with the field is not an essential condition for dynamic chaos. A similar conclusion was reached also in Ref. 12 using a different model. It should be pointed out that in all these investigations it was found that dynamic chaos should appear for a nonzero initial population of the upper levels and that chaos should be strong mainly after an initial inversion of two-level or multilevel systems.

In the present paper we shall discuss the interaction of an ensemble of two-level systems (atoms, molecules, impurities in crystals, interband transitions in semiconductors) with its own radiation field and with an external amplitude-modulated field whose carrier frequency is in resonance with a transition of two-level systems. Our main task is to find the conditions for the appearance of an analog of the Fermi stochastic acceleration effect in such a system. This effect results then in significant generation of a self-consistent radiation field when the constant of the interaction between the field and matter is relatively low. We shall find the conditions for a transition from regular to chaotic behavior corresponding to different laws governing modulation of the envelope of the external field. We shall show that the amplitude of the self-consistent field generated under dynamic chaos conditions can exceed the amplitude of the external field and the amplitude of the self-consistent field generated under regular conditions. We shall also demonstrate that chaotic generation of the field is possible when only the lower levels of two-level systems are initially populated. We shall show that unlimited acceleration (in the nondissipative regime) of the self-consistent radiation is possible when two-level systems are subjected to an external field which is a periodic sequence of pulses. Some statistical characteristics of the behavior of two-level systems under advanced chaos conditions will be found. The feasibility of observing these effects in atomic and molecular gases and in crystals with impurity centers will be discussed.

The paper is organized as follows: Sec. 1 gives the deri-

vation of the equations describing the interaction of an ensemble of two-level systems with a self-consistent radiation field and with an external amplitude-modulated field; the criterion of overlap of nonlinear resonances<sup>1-3</sup> is used in Sec. 2 to obtain the conditions for a transition from regular to chaotic behavior for different ratios of the parameters of this system and for different laws of modulation of the envelope of the external field. We shall estimate the maximum amplitudes of the self-consistent field generated in the chaotic regime and find some statistical characteristics of the behavior of two-level systems. Analytic estimates will be confirmed by numerical calculations the results of which are given in Sec. 3. The section headed Conclusions gives the physical parameters which should ensure observation of these effects in gases and in crystals with impurities.

## 1. PRINCIPAL EQUATIONS

We shall consider a sample in the form of a gas of two-level systems with a transition frequency  $\omega_0$  enclosed in a single-mode ring cavity with a natural frequency  $\omega$ . We shall seek the field and polarization of the two-level medium inside the cavity in the form

$$\begin{aligned} E_s(z, t) &= E_1(z, t) \cos(\omega t - kz) + E_2(z, t) \sin(\omega t - kz), \quad (1) \\ P(z, t) &= \rho d [u(z, t) \cos(\omega t - kz) - v(z, t) \sin(\omega t - kz)], \\ \omega &= ck, \end{aligned} \quad (2)$$

where  $d$  is the magnitude of a matrix element of a dipole transition;  $\rho$  is the density of the two-level medium. We shall assume that an external amplitude-modulated field injected into the cavity in the  $z = 0$  plane can be described by

$$E_e(t) = E_0 F(t) \cos(\omega t), \quad (3)$$

where  $F(t)$  is a given slowly varying function which we shall assume to be a periodic function of time. The field inside the resonator satisfies the following boundary conditions<sup>13</sup>:

$$\begin{aligned} E_1(z=0, t) &= E_1(z=l, t) + E_0 F(t), \\ E_2(z=0, t) &= E_2(z=l, t), \end{aligned} \quad (4)$$

where  $l$  is the characteristic size of the medium containing two-level systems. The interaction of these systems with the electromagnetic field will be considered using a semiclassical approach, describing the two-level systems in terms of quantum mechanics and the electromagnetic field using the classical Maxwell equations.<sup>14</sup> The corresponding equations deduced from the self-consistent system of the Maxwell-Bloch equations subject to the boundary conditions of Eq. (4), are

$$\begin{aligned} \dot{\bar{u}} &= \Delta \bar{v} + \bar{w} \bar{\epsilon}_2, \\ \dot{\bar{v}} &= -\Delta \bar{u} + \bar{w} \bar{\epsilon}_1, \\ \dot{\bar{w}} &= -\bar{u} \bar{\epsilon}_2 - \bar{v} \bar{\epsilon}_1, \\ \dot{\bar{\epsilon}}_1 &= \omega_c^2 \bar{v} + GF(t), \\ \dot{\bar{\epsilon}}_2 &= \omega_c^2 \bar{u}; \quad G = c\epsilon_0/l, \end{aligned} \quad (5)$$

where  $\epsilon_{0,1,2} = dE_{0,1,2}/\hbar$ ,  $\Delta = \omega_0 - \omega$ ;  $\omega_c = (2\pi\rho d^2\omega_0/\hbar)^{1/2}$  is the cooperative frequency of Ref. 15; a dot above a symbol denotes differentiation with respect to  $t$ . The variables  $u(z, t)$  and  $v(z, t)$  are related to the polariza-

tion of the two-level medium in accordance with Eq. (2) and the variable  $w$  is expressed in terms of the amplitudes of the populations of the upper ( $a_j$ ) and lower ( $b_j$ ) levels of the  $j$ th two-level system as follows:

$$w(z, t) = \frac{1}{N_s} \sum_{j \in \Delta V}^{N_s} (|a_j|^2 - |b_j|^2),$$

where  $\Delta V = (\Delta z)\pi r^2$  is a physically infinitesimally small volume;  $z$  is the coordinate of the center of a layer of thickness  $\Delta z \ll \lambda$  ( $\lambda = 2\pi/k$  is the wavelength of the radiation);  $r$  is the characteristic radius of the sample containing a gas of two-level systems;  $N_s$  is the number of such systems in  $\Delta V$  ( $N_s \gg 1$ ). A bar above dynamic variables in the system (5) denotes spatial averaging defines as follows:

$$\bar{A}(t) = \frac{1}{l} \int_0^l dz A(z, t),$$

where  $A(z, t)$  is one of the functions  $u, v, w$ , or  $\epsilon_{1,2}$ . The system (5) is derived by separation of variables corresponding to the mean field approximation<sup>16,17</sup>

$$\overline{AB} \rightarrow \bar{A}\bar{B}. \quad (6)$$

The condition of validity of Eq. (6) will be discussed later. The system of equations (5) admits the following conservation law (we shall drop the bar above the dynamic variables):

$$u^2(t) + v^2(t) + w^2(t) = 1. \quad (7)$$

If  $\epsilon_0 = 0$  the system (5) describes nonlinear cooperative fluctuations: it describes a periodic process of energy transfer from the two-level medium and vice versa.<sup>15,18</sup> Such fluctuations have been observed experimentally.<sup>19,20</sup>

The system (5) can be simplified in the case of an exact resonance ( $\Delta = 0$ ). In fact, in this case we can show that if  $u(0) = \epsilon_2(0) = 0$ , then  $\epsilon_2(t) = u(t) = 0$  for any value of  $t$ . Then the first and last equations drop out of the system (5). In future, whenever possible, we shall drop the index 1 of the field component  $\epsilon_1$ .

We shall introduce the following variables satisfying the law of conservation of Eq. (7):

$$v(t) = -\sin x(t), \quad w(t) = -\cos x(t). \quad (8)$$

In terms of the variables defined by Eq. (8) the system of equations (5) with  $\Delta = 0$  becomes

$$\ddot{x} + \omega_c^2 \sin x = GF(t). \quad (9)$$

The variable  $\dot{x}(t)$  is related to the self-consistent field by  $\dot{x}(t) = \epsilon(t)$ . Equation (9) describes a physical pendulum acted upon by an external force  $F(t)$ . An analysis of the dynamics of the system described by Eqs. (5) and (9) will be made in the next section.

We shall conclude the present section by considering the criterion of validity of the mean field approximation described by Eq. (6).

The approximation of Eq. (6) is correct if the envelopes of  $u, v, w$ , and  $\epsilon_{1,2}$  vary little along the coordinate in a distance equal to the characteristic length  $l$ . We shall introduce a quantity  $\Omega_{\max} = \{\omega_c, G^{1/2}, \Omega\}$ , where  $\Omega$  is the characteristic frequency of variation of the function  $F(t)$ . The criterion

of validity of Eq. (6) can then be written in the form

$$\Omega_{\max} l / c < 1. \quad (10)$$

## 2. STOCHASTICITY CRITERION

We shall now consider possible behavior of the solutions of Eq. (9) for different ratios between the parameters  $G$  and  $\omega_c$  and for different forms of the functions  $F(t)$ . We shall represent the function  $F(t)$  in the form of a Fourier series

$$F(t) = f_0 + \sum_{n=1}^{\infty} [f_n^c \cos(n\Omega t) + f_n^s \sin(n\Omega t)]. \quad (11)$$

In this section we shall consider the dynamics of the pendulum described by Eq. (9) under the action of a force  $F(t)$  containing one harmonic ( $F(t) = \sin \Omega t$ ) or two or more harmonics, and we shall describe the influence of the zeroth harmonic  $f_0$ .

The motion of this pendulum in the absence of any perturbation ( $G = 0$ ) is periodic and in the phase plane it has two singularities: elliptic with the coordinates  $\dot{x} = 0$  and  $x = 2\pi n$  ( $n = 0, \pm 1, \pm 2, \dots$ ) corresponding to the initial populations of the lower levels of two-level systems ( $v = 0, w = -1, \varepsilon = 0$ ) and hyperbolic with the coordinates  $\dot{x} = 0$  and  $x = \pi(n + 1)$  ( $n = 0, \pm 1, \dots$ ) corresponding to complete filling of the upper levels of two-level systems ( $v = 0, w = 1, \varepsilon = 0$ ). The separatrix of the pendulum (representing a special path in the phase plane separating vibrational and rotational motion and passing through hyperbolic points) corresponds to complete transfer of energy from the atoms in the system to the field and back again.

The behavior of the solutions of Eq. (9) in the  $G \neq 0$  case depends strongly on the number of harmonics in the spectrum  $F(t)$  and on the value of the parameter  $G/\Omega^2$ . We shall analyze the case when  $F(t)$  contains one harmonic.

1) We shall assume that  $G/\Omega^2 < 1$ , so that when  $G/\omega_c^2 \lesssim 1$ , the action of an external force on the pendulum can be regarded as a perturbation resulting in nonlinear resonances between the harmonics of the natural frequency of the nonlinear motion of the pendulum and the frequency  $\Omega$  of the external force.<sup>1-3</sup> If  $\Omega > \omega_c$ , an overlap of the nonlinear resonances in the vicinity of the separatrix creates a narrow stochastic layer and the rest of the phase space is filled mainly by periodic paths.<sup>1</sup> If  $\Omega \lesssim \omega_c$  a wide stochastic layer forms in the vicinity of the separatrix and it fills a major part of the phase space, with the exception of the vicinity of an elliptic point.<sup>1</sup> In this case the width of the stochastic layer in the rotational part of the phase space is of the order of the size of the vibrational part.<sup>1</sup> Therefore, the maximum self-consistent field which can be generated under stochastic condition when the modulation law is  $F(t) = \sin \Omega t$ , amounts to  $|\varepsilon_{\max}| \sim 4\omega_c$ .

2) It is pointed out in Ref. 21 that very different behavior is possible if  $G/\Omega^2 > 1$ . In this case it is convenient to replace  $x(t)$  with a new variable  $\psi(t) = x(t) + G\Omega^{-2} \sin \Omega t$ . The equation describing the behavior of the pendulum in this case is

$$\ddot{\psi} + \omega_c^2 \sin \left[ \psi - \frac{G}{\Omega^2} \sin \Omega t \right] = 0. \quad (12)$$

An equation of the (12) type is encountered in studies of the

dynamics of a problem of passage across a nonlinear resonance.<sup>2,22</sup> Adopting an expansion

$$\begin{aligned} & \exp \left[ i \left( \psi - \frac{G}{\Omega^2} \sin \Omega t \right) \right] \\ &= \sum_{m=-\infty}^{\infty} (-1)^m J_m \left( \frac{G}{\Omega^2} \right) \exp i(\psi + m\Omega t), \end{aligned} \quad (13)$$

we find that Eq. (12) admits nonlinear resonances of the  $\psi = m\Omega$  type ( $m = \pm 1, \pm 2, \dots$ ). The condition for overlap of these nonlinear resonances is<sup>2,22</sup>

$$\begin{aligned} K \gg 1, \quad K &= \frac{2\omega_c}{\Omega} \left[ \left| J_m \left( \frac{G}{\Omega^2} \right) \right|^{1/2} + \left| J_{m-1} \left( \frac{G}{\Omega^2} \right) \right|^{1/2} \right] \\ &\rightarrow (23\omega_c^2) / (G^{1/2}\Omega); \quad G\Omega^{-2} \gg 1. \end{aligned} \quad (14)$$

When Eq. (14) is obeyed, a stochastic modulation layer appears in the phase space and its width is<sup>2,22</sup>

$$|\varepsilon_{\max}| \sim G/\Omega. \quad (15)$$

The width of this modulation layer determines the maximum amplitude of the self-consistent field generated when  $G/\Omega^2 > 1$  and the condition for chaos [Eq. (14)] is satisfied. An important feature is that in this case we can expect stochastic excitation of the self-consistent field when only the lower levels of the two-level systems are initially populated:  $w(0) = -1, v(0) = 0, \varepsilon(0) = 0$ . The diffusion rate  $D$  of the self-consistent field  $\varepsilon$  inside a stochastic modulation layer depends strongly on the value of the parameter  $V = G/\omega_c^2$ . If  $V \gtrsim 1$ , the diffusion is rapid:  $D \propto (\omega_c^2 \Omega) / V$ , whereas if  $V \ll 1$ , the diffusion is slow:  $D \propto \omega_c^2 \Omega (V \ln V)^2$  (Ref. 22). In the limit  $\Omega \rightarrow 0$  the width of the stochastic modulation layer increases in accordance with Eq. (15), but then the rate of diffusion inside the layer falls proportionally to  $\Omega$ .

We shall now analyze the case when the external force of Eq. (11) contains two or more harmonics (we shall confine ourselves to the specific case when all the harmonics are characterized by  $f_n^c = 0$ ) and when the condition  $G/\Omega^2 < 1$  is satisfied. Then, the condition for the appearance of chaos in Eq. (9), valid in any part of the phase space, can be obtained if we go over rigorously from canonic variables ( $x, \varepsilon$ ) to the action-angle variables and then apply the criterion of overlap of nonlinear resonances. However, in view of the complex functional relationship between ( $x, \varepsilon$ ) and the action-angle variables, it is more convenient to carry out an approximate analysis of the conditions for the appearance of chaos in three parts of the phase space: in the vicinity of an elliptic point, in the vicinity of the separatrix, and in the region corresponding to rotational motion. In the vicinity of an elliptic point the Chirikov parameter is

$$K_1 \approx (Gf|x(0)|)^{1/2} / \Omega, \quad (16)$$

where  $f$  denotes the characteristic average value of the non-vanishing harmonics  $f_n$  ( $n > 0$ ) of the force described by Eq. (11). If  $\varepsilon(0) = 0$  and  $x(0) \ll 1$ , we find that

$$w(0) = -\cos x(0) \approx -1 + x^2(0)/2$$

and the criterion of the appearance of chaos near the elliptic point  $K_1 \gtrsim 1$  becomes

$$[Gf(2\delta w)^{1/2}]^{1/2} / \Omega \gtrsim 1, \quad \delta w = w(0) + 1, \quad |\delta w| \ll |w(0)|. \quad (17)$$

It is clear from Eq. (17) that even when the initial excitation of the system is weak so that  $|\delta w| \ll 1$ , the stochasticity criterion can be readily satisfied.

In the vicinity of the separatrix (in the vibrational and rotational regions), we always have a stochastic layer.<sup>1-3</sup> When the conditions<sup>1</sup>

$$G/\omega_c^2 \ll 1, \quad \Omega/\omega_c \ll 1 \quad (18)$$

are satisfied, the thickness of this stochastic layer becomes of the order of  $\sim 4\omega_c$ , which is the width of the region inside the separatrix.

The parameter of overlap of resonances in the rotational region of the phase space ( $|\varepsilon| \gg \omega_c$ ) is

$$K_3 \approx [(8Gf)^{1/2} \omega_c] / \varepsilon \Omega. \quad (19)$$

Since for  $G/\Omega^2 < 1$  and  $|\varepsilon|/\omega_c \gg 1$ , we have  $K_3 < 1$ , it follows that in this part of the phase space the transition to chaotic dynamics is impossible. The above analysis demonstrates that an increase in the number of harmonics in the spectrum  $F(t)$  compared with the case when  $F(t) = \sin \Omega t$  or  $G/\Omega^2 < 1$  makes possible stochastic generation of the radiation field when the initial excitation of the two-level systems is very weak, but it does not change significantly the maximum amplitude of the self-consistent field produced by chaotic generation.

We shall assume now that  $G/\Omega^2 > 1$ . For simplicity, we shall consider the case when  $F(t)$  contains just two harmonics:

$$F(t) = G_1 \sin \Omega_1 t + G_2 \sin \Omega_2 t, \quad G_{1,2}/\Omega_{1,2}^2 > 1, \quad (20)$$

where  $G_1 = Gf_n$ ,  $G_2 = Gf_m$ ,  $\Omega_1 = n\Omega$ ,  $\Omega_2 = m\Omega$ ,  $n \neq m$ . A new feature compared with  $F(t) = \sin \Omega t$ , and  $G/\Omega^2 > 1$  is the feasibility of formation of a wider stochastic region because of overlap of the individual stochastic modulation layers. The equation describing the dynamics of a pendulum of Eq. (9) under the influence of an external force of Eq. (20) is

$$\ddot{\psi} + \omega_c^2 \sin \left[ \psi - \frac{G_1}{\Omega_1^2} \sin \Omega_1 t - \frac{G_2}{\Omega_2^2} \sin \Omega_2 t \right] = 0, \quad (21)$$

where

$$\psi(t) = G_1 \Omega_1^{-2} \sin(\Omega_1 t) + G_2 \Omega_2^{-2} \sin(\Omega_2 t).$$

Using the expansion of Eq. (13), we find that Eq. (21) admits the possibility of nonlinear resonances of the type  $\psi = m_1 \Omega_1 + m_2 \Omega_2$  ( $m_1, m_2 = \pm 1, \pm 2, \dots$ ). The condition of overlap of nonlinear resonances is now

$$K_{1,2} \gg 1, \quad K_{1,2} = \frac{13\omega_c^2}{(G_1 G_2)^{1/2}} \left\{ \frac{\Omega_2/\Omega_1}{\Omega_1/\Omega_2}, \quad \frac{G_{1,2}}{\Omega_{1,2}^2} \gg 1. \right. \quad (22)$$

In Eq. (22) we no longer have the transition in the limit to Eq. (14) even when  $m_2 = 0$  and this is due to the condition  $G_{1,2} > \Omega_{1,2}^2$ . When the conditions of Eq. (22) are obeyed, a chaotic region forms in the phase space and the width of this region determines the maximum amplitude of the self-consistent field:

$$|\varepsilon_{\max}| \sim G_1/\Omega_1 + G_2/\Omega_2. \quad (23)$$

We can see from Eq. (23) that the presence of two harmonics in  $F(t)$  on condition that  $G_{1,2}/\Omega_{1,2}^2 > 1$  increases the

maximum amplitude of the self-consistent field  $|\varepsilon_{\max}|$  approximately twofold compared with the case when the perturbation  $F(t)$  contains just one harmonic [see Eq. (15)].

In the subsequent numerical analysis of the system (5) we shall consider also the case when  $F(t)$  is a periodic sequence (train) of pulses. The spectrum  $F(t)$  then also has the zeroth Fourier harmonic  $f_0$ . Therefore, we shall consider the influence of  $f_0$  on the dynamics of Eq. (9). The effect of the zeroth harmonic reduces essentially to deformation of the pendulum potential  $U_0(x) = -\omega_c^2 \cos x$ , resulting in the loss of translational invariance:

$$U(x) \neq U(x+2\pi), \quad U(x) \rightarrow -\infty \quad \text{when } x \rightarrow \infty, \quad (24)$$

where  $U(x) = U_0(x) - f_0 x$ . Now, if stochastization of fluctuations in a potential well formed by  $U(x)$  occurs under the influence of the nonzerth harmonics of  $F(t)$  and the energy of the system (of the self-consistent radiation field) growing in accordance with the diffusion law reaches the value of the energy near the separatrix, unlimited (in the nondissipative approximation) growth of the self-consistent radiation field is possible. Such unlimited growth of the self-consistent field can occur for any, no matter how small, value of the zeroth harmonic  $f_0 \ll 1$ . It should be pointed out that this effect of a strong increase in the velocity of motion of a nonlinear system [in our case the field  $\varepsilon(t)$  performs the role of this velocity] as a result of stochastization of nonlinear fluctuations is fairly common and can be observed in other physical systems with the potential exhibiting the property described by Eq. (24).

We shall end this section by deriving some statistical characteristics of the behavior of two-level systems under advanced chaos conditions when  $G/\Omega^2 > 1$ . For simplicity, we shall consider only the case  $F(t) = \sin \Omega t$  (the case of many harmonics can be considered similarly). The behavior of the system is then deduced from Eq. (12) and the variables  $v(t)$  and  $w(t)$  describing the two-level system are related to  $\psi(t)$  in accordance with the expressions

$$\begin{aligned} v(t) &= -\sin[\psi - G\Omega^{-2} \sin(\Omega t)], \\ w(t) &= -\cos[\psi - G\Omega^{-2} \sin(\Omega t)]. \end{aligned} \quad (25)$$

All the calculations will now be carried out for  $v(t)$  and the results of  $w(t)$  will be fully analogous. It is known from Refs. 1, 3, and 22 that when the stochasticity criterion of Eq. (14) is obeyed,  $\psi(t)$  is a random variable which can be described satisfactorily by a function distributed uniformly in an interval  $[0, 2\pi]$ . Using this fact, we find that the distribution function  $f(v)$  and the first moments of the random process  $v(t)$  are of the form

$$\langle v \rangle = 0, \quad \langle v^2 \rangle = 1/2; \quad f(v) = \pi^{-1} [1 - v^2]^{-1/2}. \quad (26)$$

We shall now consider the behavior of another important characteristic of the random process  $v(t)$ , which is the correlation function

$$R(\tau) = \langle v(t+\tau)v(t) \rangle - \langle v(t) \rangle^2. \quad (27)$$

It is known from Ref. 1 that when the overlap criterion of Eq. (14) is satisfied so that  $K \gg 1$ , the correlation function of the process  $\psi(t)$  behaves as follows:

$$\left\langle \frac{\cos}{\sin} [\psi(t+\tau) - \psi(t)] \right\rangle \sim \exp\left(-\frac{\tau}{t_c}\right), \quad (28)$$

where the time for decoupling the correlations amounts to  $t_c = 2T/\ln K$ , where  $T = 2\pi/\Omega$ . Using Eq. (28), we can show that

$$R(\tau) \sim \Phi(\tau) \exp(-\tau/t_c), \quad (29)$$

where  $\Phi(\tau)$  is an oscillatory function of time. According to the Wigner-Khinchin theorem,<sup>23</sup> the width of the spectrum of the process  $v(t)$  is  $\sim 1/t_c$  and, therefore, the system under consideration exhibits a line broadening mechanism associated with the appearance of the chaotic instability. The contribution to the line width made by the broadening mechanism associated with dynamic chaos is the largest, since we are considering the dynamics of the interaction of the field with the two-level systems over intervals shorter than all the other characteristic field and matter relaxation times.

### 3. RESULTS OF NUMERICAL CALCULATION

We checked the analytic estimates obtained in Sec. 2 by a numerical investigation of the system of equations (5). In this investigation we considered the laws governing modulation of the external field, when  $F(t)$  contains one harmonic or two harmonics [Eq. (20)], and also when  $F(t)$  represents a periodic sequence of rectangular pulses with the duration of a single pulse  $T_0$  and an interval between the pulses  $T - T_0$ . In the latter case the expressions for the coefficients in the Fourier expansion of Eq. (11) are

$$f_0 = \frac{T_0}{T}, \quad f_n^s = 0, \quad f_n^c = \frac{2T_0}{T} \frac{\sin(n\pi T_0/T)}{\pi T_0/T},$$

$$\Omega = \frac{2\pi}{T}, \quad n \geq 1. \quad (30)$$

The precision of these calculations was monitored by checking that the conservation law (7) was satisfied. We found that Eq. (7) was satisfied to within a few tenths of a percent. The difference between chaotic and regular paths in the dynamic system (5) was found by calculating not only the paths, but also the Fourier spectrum and the logarithm of the separation between two paths closest at the initial moment

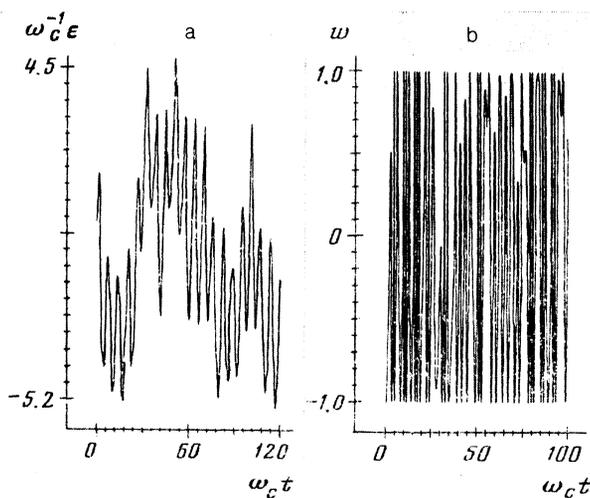


FIG. 1. Self-consistent field  $\epsilon(t)$  (a) and the difference between the populations  $w(t)$  (b) in the case of chaos:  $G/\omega_c^2 = 2$ ,  $\Omega/\omega_c = 1$ ,  $u(0) = v(0) = \epsilon_{1,2}(0) = 0$ ,  $w(0) = -1$ , and  $F(t) = \sin \Omega t$ .

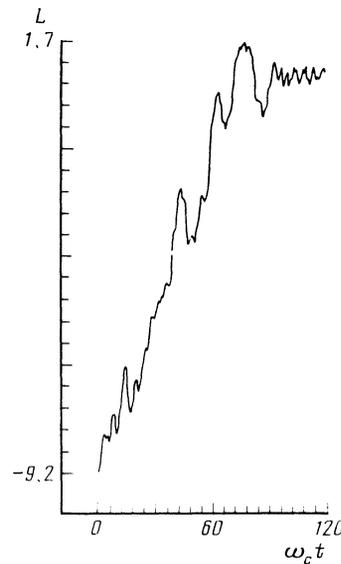


FIG. 2. Local instability; the initial conditions and the parameters are the same as in Fig. 1.

$$L(t) = \ln \left[ (u(t) - u'(t))^2 + (v - v')^2 + (w - w')^2 + \omega_c^{-2} (\epsilon_1 - \epsilon_1')^2 + \omega_c^{-2} (\epsilon_2 - \epsilon_2')^2 \right]^{1/2}, \quad (31)$$

where a prime is used for a path with similar initial conditions. The results of these numerical calculations are presented in Figs. 1–6. The paths characterized by chaotic behavior (Figs. 1a and 1b) exhibit a local instability (Fig. 2) and a wide Fourier spectrum (Fig. 4a). Regular paths (Figs. 3a and 3b) are characterized by the absence of a local instability and by a discrete Fourier spectrum (Fig. 4b).

The results of numerical calculations carried out for the case when  $F(t) = \sin \Omega t$  demonstrate that stochastic excitation of the self-consistent field in the case of initial population of the lowest levels of the two-level systems [ $w(0) = -1$ ,  $\epsilon_1(0) = \epsilon_2(0) = 0$ ] is possible only if  $\Omega \lesssim \omega_c$

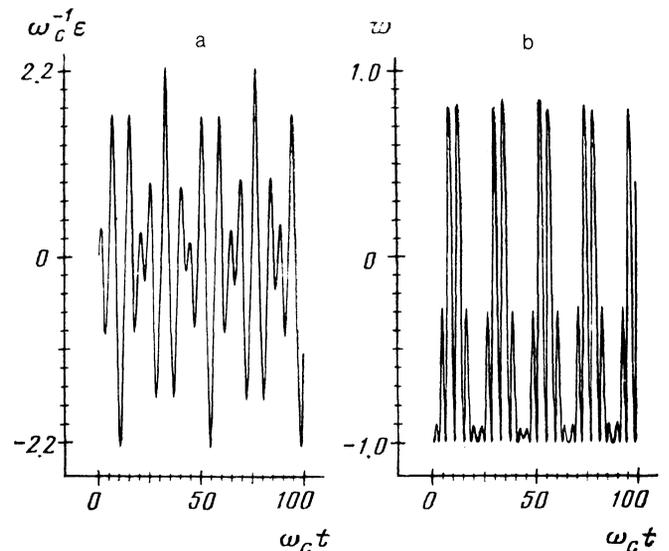


FIG. 3. Self-consistent field  $\epsilon(t)$  (a) and the difference between the populations  $w(t)$  (b) in the case of regular motion:  $G/\omega_c^2 = 0.5$ ,  $\Omega/\omega_c = 1$ ,  $u(0) = v(0) = \epsilon_{1,2}(0) = 0$ ,  $w(0) = -1$ ,  $F(t) = \sin \Omega t$ .

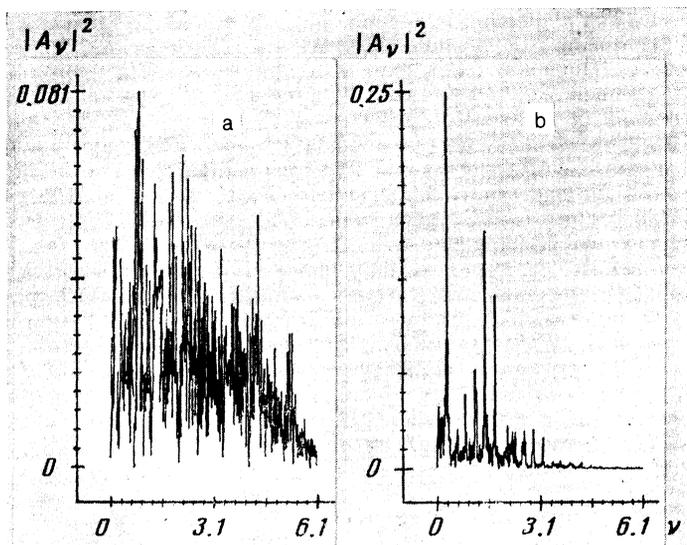


FIG. 4. Fourier spectrum of the process  $w(t)$  in the case of chaotic (a) and regular (b) motion. The initial conditions and the parameters are the same as in Figs. 1 and 3, respectively. The frequency  $\nu$  is measured in units of  $\omega_c$ .

and  $G \gtrsim G_{cr}$  (if  $\Omega \approx 1$ , we have  $G_{cr} \approx 0.7$ , whereas for  $\Omega = 0.1$ , we find that  $G_{cr} \approx 0.9$ ). The maximum amplitudes of the self-consistent field generated under these conditions are in good agreement with the theoretical estimates obtained in Sec. 2. Moreover, we investigated numerically the influence of a finite detuning  $\Delta \neq 0$  on the chaotic dynamics of the system. If  $\Delta \lesssim \omega_c$ , the size of the chaotic region in the phase space changes only slightly compared with the case when  $\Delta = 0$ . If  $\Delta > \omega_c$ , the size of the chaotic region becomes narrower and for  $\Delta \gg \omega_c$  only a narrow stochastic layer remains near the separatrix ( $w = 1, \varepsilon_1 = \varepsilon_2 = 0$ ).

A numerical investigation carried out using  $F(t)$  described by Eq. (20) subject to the conditions  $G_{1,2}/\Omega_{1,2}^2 \lesssim 1$ ,  $G_1/\Omega_1^2 \gg 1$ ,  $G_2/\Omega_2^2 \lesssim 1$ , and  $G_{1,2}/\Omega_{1,2}^2 \gg 1$  demonstrated that chaotic generation of the field is possible when only the lower levels of the two-level systems are populated initially [ $w(0) = -1$ ]; then,  $|\varepsilon_{\max}|$  is described satisfactorily by the

theoretical estimates given by Eqs. (15) and (23).

Figure 5 demonstrates the dynamics of growth of a self-consistent radiation field when an ensemble of two-level systems is subjected to a periodic sequence of pulses [Eqs. (11) and (30)]. When the conditions for chaos are satisfied, diffusion growth of the field from  $\varepsilon = 0$  to  $\varepsilon \sim 2\omega_c$  is observed (pendulum separatrix) and this is followed by an almost linear regular unlimited growth. It should be noted that such a time dependence is exhibited also by the differences between the populations and by the polarization of the two-level systems: chaotic fluctuations become fast regular ones. An increase in the pulse repetition time  $T$  increases, other conditions being equal, the time during which chaotic fluctuations  $\varepsilon(t)$ ,  $v(t)$ , and  $w(t)$  take place and this is associated with the reduction in the amplitude of the zeroth harmonic  $f_0 = T_0/T$  and with a corresponding reduction in deforma-

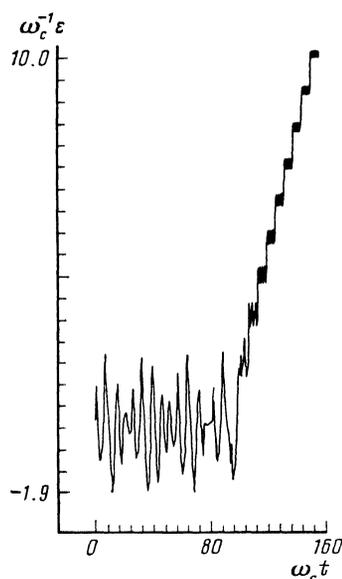


FIG. 5. Self-consistent field  $\varepsilon(t)$  in the case when  $F(t)$  is a periodic sequence of rectangular pulses:  $G/\omega_c^2 = 2$ ,  $\omega_c T = 6.28$ ,  $\omega_c T_0 = 0.5$ ,  $u(0) = v(0) = 0$ ,  $w(0) = -0.99$ , and  $v(0) = [1 - w(0)^2]^{1/2}$ .

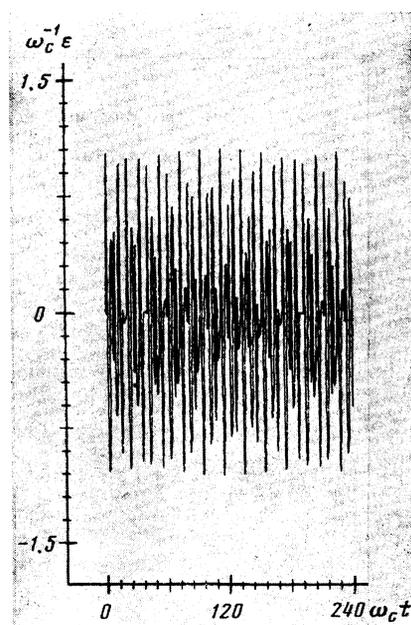


FIG. 6. Field  $\varepsilon(t)$  in the case of regular motion; the data are as in Fig. 5, except for  $\omega_c T = 4$ .

tion of the potential  $U(x) = -\omega_c^2 \cos x - f_0 x$ . When the condition for chaos is not satisfied, regular fluctuations are observed and the maximum amplitude of the self-consistent field does not exceed  $|\varepsilon| \sim 2\omega_c$  (Fig. 6).

## CONCLUSIONS

Our results thus demonstrate that in the case of a resonant interaction of a gas of two-level systems with an amplitude-modulated external field and with an intrinsic radiation field we can expect stochastization of fluctuations of the difference between the populations and of the polarization of the two-level systems accompanied by a diffusion-like growth of the self-consistent field. The conditions for stochastization and maximum amplitude of the self-consistent field  $|\varepsilon_{\max}|$  depend strongly on the spectrum of modulation of the external field and on the ratio  $G/\Omega^2$ . When the stochastization conditions are satisfied, we can expect excitation of the self-consistent field with  $|\varepsilon_{\max}|$  exceeding the amplitude of the external field and significant growth of the self-consistent field may occur even when only the lower levels of the two-level systems are initially populated. When a gas of two-level systems is subjected to a periodic sequence of pulses and the stochastization conditions are satisfied, the self-consistent field grows in two stages: 1) diffusion growth to  $\sim 2\omega_c$ ; 2) unlimited (in the nondissipative approximation) regular growth. Under advanced chaos conditions the difference between the populations and the polarization are random processes and the spectra of these processes are broadened by an amount of the order of the modulation frequency  $\Omega$ .

These effects can be observed clearly when laser radiation interacts with atomic and molecular gases characterized by a relaxation time  $T_2 \sim 10^{-7} - 10^{-9}$  s and  $d \sim 10^{-18}$  cgs units,  $l \sim 10 - 0.1$  cm,  $\rho \sim 10^{12} - 10^{16}$  cm $^{-3}$ ,  $\omega_c$ ,  $\varepsilon_0$ ,  $1/T$ ,  $1/T_0 \sim 10^9 - 10^{11}$  s $^{-1}$  or in crystals with impurity centers under conditions close to those needed for the observation of optical superradiance<sup>24,25</sup>:  $d \sim 10^{-20}$  cgs units,  $\rho \sim 10^{16} - 10^{18}$  cm $^{-3}$ ,  $l \sim 1 - 0.001$  cm,  $T_2 \sim 10^{-7} - 10^{-10}$  s,  $\omega_c$ ,  $\varepsilon_0$ ,  $1/T$ ,  $1/T_0 \sim 10^9 - 10^{11}$  s $^{-1}$ . Weak attenuation of the field in the resonator (with the attenuation time  $\tau_p \sim 10^{-7} - 10^{-10}$  s) can be achieved if the transmission coefficient of the mirrors is  $\sim 1 - 0.1\%$ .

When an amplitude-modulated field interacts with two-level systems, dynamic chaos can appear in principle also in the microwave range due to Rydberg transitions of atoms and molecules in experiments of the type described in Refs. 20 and 26. In this case the density  $\rho$  can be quite low because of the large values of the dipole moments of the transitions and of the relaxation times of the Rydberg states.

We shall conclude by noting one of the interesting generalizations of the problem considered above: chaotic dynamics of the interaction of an amplitude-modulated field with an ensemble of multilevel systems allowing for the co-

operative effects. The stochastic Fermi acceleration effect may be manifested by multilevel systems both by the growth of the self-consistent radiation field and by filling of the upper levels of the multilevel systems.

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