The qualitative explanation of the behavior of $\omega_{12}(x)$ as a function of the magnetic field, presented above, was confirmed by a computer calculation (Fig. 4b). It is seen from the presented curves that, just as in the experiment, the shapes of the plots of $\omega_{12}=f(\omega_{10})$ change with increasing magnetic field.

Thus, our investigations of the characteristics of of two-mode He–Ne lasers with orthogonally polarized modes in an axial magnetic field have revealed a number of new effects that add greatly to our concepts concerining the physics of the interaction of modes and on the feasibility of lasers of a new type.

In conclusion, the authors thank M. A. Gubin for useful discussions.

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Stochastic mechanism of excitation of molecules that interact with their own radiation field

P. I. Belobrov, G. P. Berman, G. M. Zaslavskiĭ, and A. P. Slivinskiĭ

L. V. Kirenskiĭ Physics Institute, Siberian Division, USSR Academy of Sciences
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Results are presented of a numerical experiment on the investigation of the interaction of a system of quantum nonlinear oscillators (molecules) with their own radiation field and with an external coherent field of laser radiation. The conditions under which a stochastic regime of excitation of high-lying levels sets in in such a system are obtained. It is shown that in the region of stochasticity of the motion the level population distribution function is substantially altered. An increase of the populations of the high-lying levels takes place simultaneously with generation of a self-consistent radiation field in the system. The character of the level population when several external fields that are resonant to various transitions of the nonlinear oscillator act on the system is investigated in the presence of stochastic instability in the system. The described phenomenon can be regarded as one of the possible mechanisms of stochastic excitation of polyatomic molecules into the region of high-lying levels of the vibrational spectrum.

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1. INTRODUCTION

Much attention is being paid presently to the interaction of coherent laser radiation with multilevel quantum systems. Particular interest attaches to the possibility of selective action of laser radiation on matter, to control of the rates of chemical reactions, and to collisionless dissociation of polyatomic molecules.[1-3] The main difficulties faced by a consistent theoretical interpretation of such results lie in the study of the possible mechanisms that lead to excitation of the molecule to high-lying levels of the vibrational spectrum under conditions of large anharmonicity. The process of vibrational excitation of a molecule was arbitrarily divided in Ref. 4 into two stages, the passage through the low levels and the subsequent excitation of the molecules into the region of large quantum numbers. Resonant character of the interaction was ensured there by a high density of the high-lying vibrational states. A mechanism of rotational compensation of the anharmonicity was proposed in Ref. 5 for low-lying levels. Tunneling in energy space was proposed[6] as a possible mechanism of passage through low-lying levels. Molecule excitation was considered in Ref. 7 as the consequence of the onset of stochastic instability due to the interaction of intermode resonances.

It should also be noted that under certain condition an important factor in the description of the behavior of systems of the indicated type is the allowance for the

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self-consistent radiation field. Jaynes and Cummings[8] considered the self-consistent interaction of a two-level system with its own radiation field and showed that under conditions of the resonance approximation the dynamics is determined by periodic transfer of energy from the two-level system to the radiation field and back. The role of the self-consistent field in the assembly of the interacting particles increases substantially. It was previously shown[9] that under certain conditions a stochastic instability sets in an ensemble of two-level atoms that interact with their own radiation field. An essential condition for the appearance of the stochastic instability is the presence of a threshold value of the constant of the interaction of the atomic system with the radiation field. It is also shown in Ref. 9 that the main consequence of the presence of stochastic instability in this system is the violation of one of the integrals of the motion. This can lead to stochastization of the spectrum of the system.[10] Thus, the presence of a self-consistent radiation field can lead under definite conditions to qualitatively new phenomena connected with the appearance of stochastic properties in the system. Allowance for the stochastic character of the behavior of the system may turn out to be essential in the mechanism whereby the molecules are excited into the region of high-lying states of the vibrational spectrum.

The present paper is devoted to a numerical investigation of the possibility of exciting a multilevel quantum vibrational system by interaction with a self-consistent field in the case when stochastic instability is present. The model chosen for the investigation is a system of N quantum nonlinear oscillators that interact with an external coherent laser-radiation field and with their own radiation field, the latter treated in the classical single-mode approximation.

The stochastic excitation of a multilevel molecule is determined by several characteristic times: the time τ_n of nutation of the system under the influence of the external resonant laser field, the time τ_c of decoupling of the phase correlations of the dynamic variables, and the diffusion time τ_D that determines the change of the distribution function of the slow variable in the regime of advanced stochastic motion. The inequalities $\tau_D \gg \tau_n$ and $\tau_D \gg \tau_c$ impose, in view of the finite time of the numerical experiment, restrictions on the choice of the system parameters. Thus, an increase of the anharmonicity constant is preferred in the sense of shortening the time τ_n and accordingly shortening the numerical-experiment time needed for the observation of the characteristic singularities of the stochastic system-motion properties that take place within times $\sim \tau_D$. The anharmonicity parameter chosen by us is therefore higher than used in real experiments on the excitation of polyatomic molecules. The chosen model makes it possible nevertheless to trace the characteristic singularities of the influence of the stochastic component of the motion on the possibility of effective excitation of nonlinear quantum oscillators that interact with their own radiation field. The investigated phenomenon can be regarded as one of the possible mechanisms for effective excitation of polyatomic molecules into the region of high-lying levels of the vibrational spectrum.

2. BASIC EQUATIONS

In accordance with the statements made in the introduction, we write down the single-particle Hamiltonian of a system of interacting multilevel molecules in the form

$$H=H_0+V, \tag{2.1}$$

where H_0 is the unperturbed Hamiltonian of one multilevel molecule and V is the Hamiltonian of the interaction of this molecule with the external coherent field as well as with the self-consistent radiation field.

We write the solution of the Schrödinger equation with the Hamiltonian (2.1) in the form

$$\psi(x,t)=\sum_{n=0}^{M} c_n(t)\varphi_n(x), \tag{2.2}$$

where $M+1=q$ is the number of the vibrational levels of the molecule. The equations for the amplitudes $c_n(t)$ are of the form

$$i\hbar\dot{c}_n=E_nc_n+[E(t)+\mathscr{E}_0\cos\Omega t](d_{n,n+1}c_{n+1}+d_{n,n-1}c_{n-1}), \tag{2.3}$$

E_n is the vibrational spectrum of the molecule:

$$E_n=\hbar\omega\frac{(n+1/2)-\gamma(n+1/2)^2}{1-2\gamma}. \tag{2.4}$$

The form of the spectrum (2.4) is chosen such that the energy of the 0–1 transition be equal to $\hbar\omega$. Expression (2.3) takes into account only transitions between neighboring levels: $d_{n,n+1}=d(n+1)^{1/2}$, $d_{n,n-1}=dn^{1/2}$ (d is the dipole moment of the 0–1 transition), $E(t)$ is the self-consistent field of the radiation, $\mathscr{E}_0\cos\Omega t$ is the external field of the laser radiation, and γ in (2.4) is the anharmonicity parameter.

The equation for the self-consistent field $E(t)$ is of the form

$$\ddot{E}+\nu^2E=-4\pi\dot{P}/V, \tag{2.5}$$

where V is the volume of the system. Equation (2.5) is written in the single-mode approximation. The frequency ν was subsequently chosen equal to the frequency of the 0–1 transition ($\nu=\omega$). The polarization P in (2.5) is given by

$$P=Nd\sum_{n=0}^{M}(c_n^*c_{n+1}d_{n,n+1}+c_n^*c_{n-1}d_{n,n-1}+\text{c.c.}), \tag{2.6}$$

where N is the number of molecules in the volume V.

We represent c_n in (2.4) in the form $c_n=A_n+iB_n$ and introduce the dimensionless variables

$$w_n=\frac{E_n}{\hbar\omega}=\frac{(n+1/2)-\gamma(n+1/2)^2}{1-2\gamma},$$
$$\Lambda^2=16\pi\rho d^2/\hbar\omega,\quad \rho=N/V,\quad \omega t=\tau,\quad \Omega/\omega=\alpha,\quad p=P/N, \tag{2.7}$$
$$\varepsilon_0=\mathscr{E}_0d/\hbar\omega,\quad \mathscr{E}(t)=E(t)d/\hbar\omega.$$

The self-consistent field $\mathscr{E}(t)$ is conveniently described in terms of the variables $\vartheta(t)$ and $\chi(t)$:

$$\mathscr{E}(t)=1/2\Lambda\vartheta-1/4\Lambda^2p. \tag{2.8}$$

The system of equations for ϑ and χ is

$$\dot{\vartheta}=\chi,\quad \dot{\chi}=-\vartheta+1/2\Lambda p,\quad p=2\sum_{n=1}^{M}n^{1/2}(A_nA_{n-1}+B_nB_{n-1}). \tag{2.9}$$

These equation take into account the fact that $\nu=\omega$. We rewrite Eqs. (2.3) in terms of the variables A_n and B_n. With allowance for (2.7), we have

$$\dot{A}_n=w_nB_n-(^1/_2\Lambda\vartheta-^1/_4\Lambda^2p+\varepsilon_0\cos\alpha\tau)[(n+1)^{1/2}B_{n+1}+n^{1/2}B_{n-1}],$$
$$\dot{B}_n=-w_nA_n+(^1/_2\Lambda\vartheta-^1/_4\Lambda^2p+\varepsilon_0\cos\alpha\tau)[(n+1)^{1/2}A_{n+1}+n^{1/2}A_{n-1}]. \quad (2.10)$$

The system of equations (2.9) and (2.10) determines fully the dynamics of the investigated model. This system was subsequently solved by numerical means.

We note some of the properties of the system (2.9) and (2.10). In the absence of an external field, $\varepsilon_0=0$, this system has as an integral of the motion

$$Q=W+^1/_2(\vartheta^2+\chi^2+^1/_4\Lambda^2p^2)-^1/_2\Lambda\vartheta p, \quad (2.11)$$

where

$$W=\sum_{n=0}^{M}w_n(A_n^2+B_n^2).$$

In addition, as a result of conservation of the normalization of the wave function (2.2) of an individual molcule, the system (2.9) and (2.10) has also an integral

$$\sum_{n=0}^{M}I_n=1, \quad (2.12)$$

where $I_n=A_n^2+B_n^2$ is the population of the n-th level. We note also that the system (2.9), (2.10) can be expressed in the form of a Hamiltonian-type equivalent system:

$$\dot{A}_m=\frac{1}{2}\frac{\delta\mathcal{H}}{\delta B_m}, \quad \dot{B}_m=-\frac{1}{2}\frac{\delta\mathcal{H}}{\delta A_m}, \quad (2.13)$$
$$\dot{\vartheta}=\delta\mathcal{H}/\delta\chi, \quad \dot{\chi}=-\delta\mathcal{H}/\delta\vartheta,$$

where

$$\mathcal{H}=\sum_{n=0}^{M}w_n(A_n^2+B_n^2)+\frac{1}{2}\left(\vartheta^2+\chi^2+\frac{1}{4}\Lambda^2p^2\right)-\frac{1}{2}\Lambda\vartheta p-p\varepsilon_0\cos\alpha\tau. \quad (2.14)$$

The polarization p in (2.14) is assumed expressed in terms of the variables A_n and B_n in accord with (2.9).

The system of equations (2.9) and (2.10) [or (2.13)] was considered in Ref. 3 for the particular case $M=1$ (a gas of two-level molecules) and $\mathcal{E}_0=0$. As noted in the Introduction, the results of Ref. 9 show that under certain conditions a stochastic instability develops in the system and the motion becomes random. It becomes possible then to describe the behavior of the system by statistical methods.

3. RESULTS OF NUMERICAL ANALYSIS OF THE EQUATIONS OF MOTION

To ascertain the characteristic singularities of the appearance of stochastic properties in the system under consideration, the equations of motion (2.9) and (2.10) were numerically analyzed with a computer. It is known that one of the criteria for the presence of a stochastic-motion component in a system is local instability of the trajectories in phase space. We have chosen to characterize the distance between two trajectories in phase space by the quantity

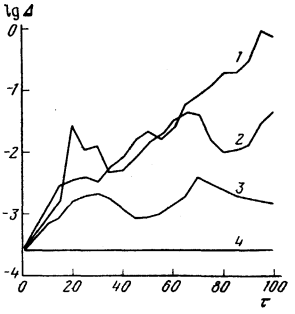

FIG. 1. Distance between initially close trajectories vs. the time at $\Lambda=2.4$ (curve 1), $\Lambda=0.4$ (curve 2), $\Lambda=0.01$ (curve 3), and $\Lambda=0$ (curve 4); $q=8$, $\varepsilon_0=0.1$, $\gamma=0.05$, $I_0(0)=0.2$, $I_1(0)=0.8$, $I_2(0)=I_3(0)=\ldots=I_7(0)=0$.

$$\Delta^2(\tau)=\sum_{n=0}^{M}\{(\bar{A}_n-A_n)^2+(\bar{B}_n-B_n)^2\}+\frac{(\bar{\vartheta}-\vartheta)^2+(\bar{\chi}-\chi)^2}{\vartheta^2+\chi^2}, \quad (3.1)$$

where $\bar{A}_n$, $\bar{B}_n$, $\bar{\vartheta}$, $\bar{\chi}$ and A_n, B_n, ϑ, χ pertain respectively to two different trajectories in phase space. The local instability corresponds to an exponential growth, with time, of two trajectories that are close to each other at the initial instant of time.

As shown in Ref. 9, for a gas of two-level molecules that interact with their own radiation field, the dimension of the region of stochasticity of the motion depends substantially on the interaction constant Λ [see (2.7)]. At $\Lambda\ll1$ the size of the stochasticity region is exponentially small, $\sim\exp(-1/\Lambda)$. Complete stochastization of the motion takes place in such a system at $\Lambda\lesssim\Lambda_{cr}\approx1$. It is of interest to determine Λ_{cr} for the case of multilevel molecules. Our numerical experiments show that Λ_{cr} decreases with increasing number of levels. For example, at $M=7$ (eight-level molecule) we have $\Lambda_{cr}\approx0.4$. It should be noted that local instability takes place also at lower values of Λ, but the rate of its development slows down in this case (Fig. 1) and vanishes at $\Lambda=0$ (in the absence of the self-consistent field). The rate of development of local instability in the stochasticity region ($\Lambda>\Lambda_{cr}$) depends essentially on the character of the initial population of the levels, namely, it increases with increasing initial population of the upper levels, i.e., with increasing initial energy of the molecule (Fig. 2). Comparison of curves 2 and 4 on Fig. 2 shows a substantial increase of the rate of development of the local instability (at $\Lambda>\Lambda_{cr}$) when the high-lying levels are initially populated. At the same time, in the stochasticity region, the rate of development of the local instability depends little on the number of levels if their initial populations are equal (see curves 3 and 4 of Fig. 2). In addition, the rate of development of the local instability is practically independent of the

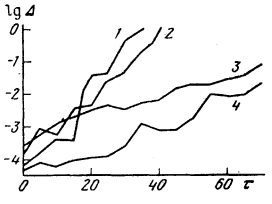

FIG. 2. Time dependence of the distance between initially close trajectories: curve 1—$q=2$, $I_0(0)=10^{-4}$, $I_1(0)=0.9999$; curve 2—$q=3$, $I_0(0)=0$, $I_1(0)=0.8$, $I_2(0)=0.2$; curve 3—$q=8$, $I_0(0)=0.2$, $I_1(0)=0.8$; curve 4—$q=3$, $I_0(0)=0.2$, $I_1(0)=0.8$. $\varepsilon_0=0$, $\gamma=0.05$; $\Lambda=2.4$.

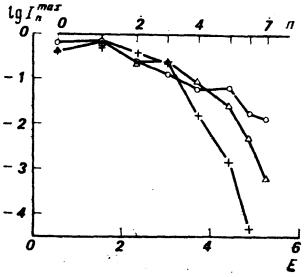

FIG. 3. Dependence of the maximum population on the number of the level. The abscissas are the level number (upper scale) and the level energy in $\hbar\omega$ units (lower scale). $\Lambda = 2.4$ (○), $\Lambda = 1$ (△), $\Lambda = 0.4$ (+); $q = 8$, $\varepsilon_0 = 0.1$.

presence in the system of an external field at resonance with the 0–1 transition (see curve 1 of Fig. 1, and curve 3 of Fig. 2). In the presence of local instability in the system, the correlations of the phases $\varphi_n = \tan^{-1}(B_n/A_n)$ of the complex amplitudes c_n of the level populations become decoupled. The phases φ_n then become random functions of the time and the molecule excitation process can be described in the diffusion approximation. The stochastization of the motion is analogous in this case to the phenomenon of the Fermi stochastic acceleration, considered in Ref. 11. It is known that in the Fermi stochastic acceleration regime the energy of the accelerated particle increases with time in accord with the diffusion law. The physical counterpart in our system is the excitation of the molecule to high-lying levels. Figure 3 shows the dependence of the distribution of the maximal populations $\log I_n^{\max}$ on the number of the level. With increasing Λ, the population of the upper levels increases and at $\Lambda = 2.4$ it exceeds 1% for the eighth level.

We have investigated the question of the existence and of the properties of the distribution function of the populations of a multilevel molecule in the advanced stochastic motion regime corresponding to $\Delta(\tau) \sim 1$. A typical the distribution function is shown in Fig. 4 for the case of an eight-level system. The angle brackets denote time averaging. It follows from our experiments that the presence of stochastic instability leads to a relatively rapid establishment of a stationary distribution function and affects its form significantly. The most appreciable change in the population distribution function occurs in this case in the region of the high-lying levels.

In the course of the numerical experiment it was observed that in the region $\Lambda > \Lambda_{cr}$ the excitation of the

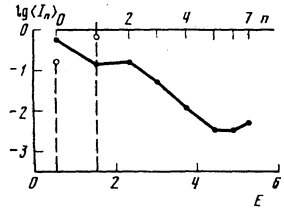

FIG. 4. Distribution function of level populations. $q = 8$; $\varepsilon_0 = 0.1$; $\gamma = 0.05$; $\Lambda = 2.4$. The points correspond to the time-averaged populations on the corresponding levels. The dashed lines show the initial populations.

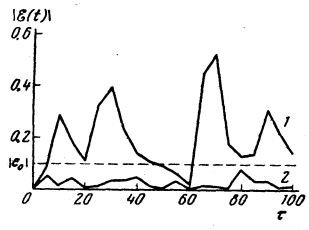

FIG. 5. Time dependence of the envelope of the self-consistent field in the presence of stochastic instability. The dashed line shows the amplitude of the external field that is at resonance with the 0–1 transition. Curve 1—$\Lambda = 2.4$; 2—$\Lambda = 0.4$; $q = 8$, $\varepsilon_0 = 0.1$, $\gamma = 0.05$.

molecule into the region of the high-lying level is always accompanied by an increase of the self-consistent radiation field. The maximum values of $\mathscr{E}(t)$ greatly exceed in this case the external-field amplitude ε_0 (curve 1 in Fig. 5). In the case when the amplitude of the self-consistent field in the system is not large ($\mathscr{E}_{\max}(t) \leq \varepsilon_0$, curve 2, Fig. 5), the upper levels (as seen from Fig. 3, the curve with the points marked +) are barely excited, although local instability is present in the system (see curve 2 of Fig. 1). It should be noted in this connection that the characteristic values of the constant Λ, at which the excitation of the molecule to the high-lying levels turns out to be substantial, lie in a range of value exceeding the critical Λ_{cr} determined from the criterion for the local instability. Thus, both the presence of local instability in the system and generation of a self-consistent radiation field are essential for the excitation of a molecule into the region of high-lying levels.

It is known that increasing the number of resonant harmonics of the external field lowers the thresholds of the transition of the system into the stochastic motion regime. It is therefore of interest to investigate the conditions of stochastic excitation of a molecule in the case when there are several external laser-radiation fields that are at resonance with the transitions between the various levels. To study this question in the numerical experiment, a system with twenty levels was chosen. A comparative analysis was made for two cases: 1) the external field ε_0 is resonant to the 0–1 transition; 2) two external fields are resonant to the transition 0–1 (ε_0) and to the transition 3–4 (ε_3). In the case when two resonant field act simultaneously on the system

$$\varepsilon(t) = \varepsilon_0 \cos\tau + \varepsilon_3 \cos[(1-8\gamma)/(1-2\gamma)]\tau$$

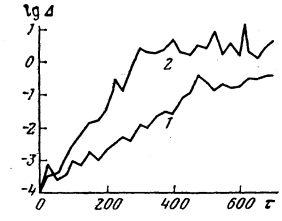

FIG. 6. Time dependence of the distance between initially close trajectories. Curve 1—external field at resonance with 0–1 transition; 2—external field at resonance with transitions 0–1 and 3–4. $q = 20$, $\varepsilon_0 = 0.04$, $\gamma = 0.02$, $\Lambda = 0.4$; $I_0(0) = 0.057$, $I_1(0) = 0.908$, $I_2(0) = 0$, $I_3(0) = 0.004$, $I_4(0) = 0.031$, $I_5(0) = I_6(0) = \ldots = I_{19}(0) = 0$.

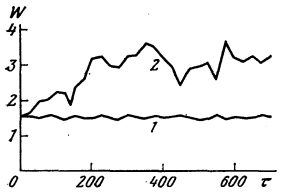

FIG. 7. Dependence of the molecule energy on the time. Curve $1-\varepsilon_0=0.04$, $\varepsilon_3=0$; $2-\varepsilon_0=0.04$, $\varepsilon_3=0.04$. The remaining conditions are indicated in the legend of Fig. 6.

the rate of development of the local instability increases substantially and the transition of the system into the stochastic regime of motion occurs within shorter times (Fig. 6).

An important circumstance is the substantial increase of the molecule energy W in the second case, on account of the energy of the external field (curve 2, Fig. 7). A study of the time evolution of the level-population distribution shows that in the case when two resonant fields act on the system the number of excited high-lying levels is larger than under the action of one resonant field. As seen from Fig. 8, in the former case seven levels are excited, and in the latter thirteen. The level-population distribution function becomes stationary in the second case within much shorter times than in the first case. Thus, turning on additional sources of coherent laser radiation that are at resonance with transitions of the vibrational spectrum of the molecule increases the rate of stochastization of the system, increases the average molecule energy at the expense of the external field, and causes substantial excitation of the high-lying levels.

4. CONCLUSION

Notice should be taken of two substantial circumstances connected with allowance for the self-consistent radiation field of interacting multilevel systems. First, allowance for the self-consistent radiation field at $\Lambda > \Lambda_{cr}$ leads to a qualitatively new effect—development of stochastic instability in the system. The latter decouples the phase correlations of the dynamic variables, so that the motion can be described by statistical methods. This leads to establishment of a stationary distribution function of the level population of the system. This function differs substantially from the behavior of the populations in the case when there is no stochastic instability in the system ($\Lambda < \Lambda_{cr}$), when the motion is mainly quasiperiodic. Effective excitation of the high-lying levels of the molecule occurs in the region of stochastic instability of the motion ($\Lambda > \Lambda_{cr}$) simultaneously with the generation, in the system, of a self-consistent radiation field with characteristic amplitudes that exceed the amplitude of the external field.

Second, the presence of a resonant laser-radiation field acting simultaneously on several transitions increases the rate of development of the local instability and increases the molecule energy. A stationary distribution function of the level populations is established

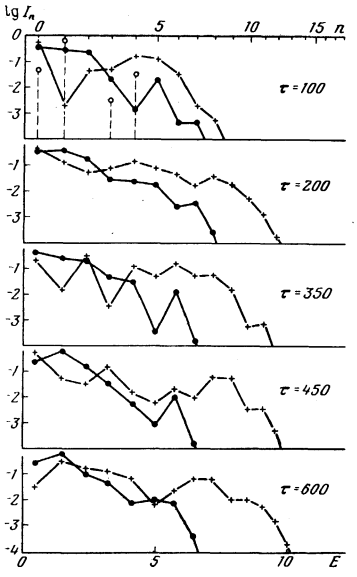

FIG. 8. Time dependence of the level populations in the presence of one resonant field (●) $\varepsilon_0=0.04$ and two resonant fields (+): $\varepsilon_0=0.04$ and $\varepsilon_3=0.04$. The remaining conditions are indicated in the legend of Fig. 6.

in this case much more rapidly, and substantial excitation of the high-lying level takes place. The considered stochastic excitation of multilevel nonlinear quantum oscillators interacting with their own radiation field and with an external coherent radiation field can be one of the possible mechanisms of effectively exciting polyatomic molecules into the region of high-lying levels of the vibrational spectrum. We note that allowance for the multimode character of the self-consistent radiation field and for the fact that real molecules are three-dimensional can substantially lower the critical value Λ_{cr} of the interaction constant at which the stochastic motion regime develops in the system.

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