Resonant excitation of an anharmonic quantum-mechanical oscillator

N. N Rozanov and V. A. Smirnov

(Submitted 4 January 1983; resubmitted 3 October 1983)
Zh. Eksp. Teor. Fiz. **86**, 1180–1192 (April 1984)

Wave functions for the coherent states of an anharmonic oscillator subjected to external resonant excitation are constructed and investigated. The equation of motion of the center of gravity of the wave function, which is the quantum-mechanical analog of the Duffing equation, is derived. Solutions of the equation of motion and the corresponding expansions of the wave functions over the stationary states are obtained for a monochromatic external force. Periodic self-oscillations in the absence of the external force are found, and their period is in agreement with numerical analyses published in the literature. The conditions for the excitation of high vibrational states are determined, and the possibility of hysteresis in the excitation of the anharmonic oscillator is demonstrated.

Hysteresis effects accompanying the application of laser radiation to various macroscopic objects are currently under intensive investigation (this is the so-called "optical bistability"; see, for example, Refs. 1 and 2). There is also undoubted interest in the possibility of hysteresis in elementary quantum-mechanical systems, including the anharmonic quantum-mechanical oscillator (AQO). Although hysteresis in the resonant excitation of the classical anharmonic oscillator is well known,[3,4] hysteresis in AQO has remained an open question. The analogy with the classical oscillator in the quasiclassical limit ($\hbar \to 0$) has been noted[5] but, when the higher-order terms in $\hbar$ were taken into account in Ref. 6, it was concluded that the quantum-mechanical oscillator with a definite phase differed from the classical oscillator in that it could occupy only a single state. We shall show that the linearity of the time-dependent Schrödinger equation (for the wave function) does not, in general, prevent the possibility of hysteresis, since we are dealing with a partial differential equation. Actually, solutions of the nonlinear ordinary differential equations describing classical hysteresis (the Duffing equation) can be compared with solutions of the suitably constructed linear partial differential equation.[7] By analogy with the stochastic behavior of the anharmonic classical oscillator when the amplitude of the external force exceeds a certain critical value,[8,9] the AQO model enables us to investigate the possibility of stochasticity in the quantum-mechanical region.[10–18]

We note that the harmonic quantum-mechanical oscillator serves as the basis of the theory of a wide range of physical phenomena. When nonlinear phenomena are described, the anharmonicity of the oscillator must be taken into account.[14,15] A review of papers on the application of the AQO model to resonant excitation of molecules by intense laser radiation is given in Ref. 16. A quantum-mechanical oscillator is essentially a multilevel system. The spectrum of the harmonic oscillator has a degenerate structure. The exact solution of the problem of interaction between a harmonic oscillator and an arbitrary external force is given in Ref. 17. This degeneracy is removed in the case of the AQO, but a large number of multiphoton transitions becomes possible. The problem is much more complicated and, at present, solutions are available only for the limiting cases of weak (perturbation theory[15,18]) and strong (semiclassical approximation[19–21,5]) fields.

A promising approach to a more complete solution of the above problem is to use the coherent states of the harmonic oscillator, introduced by Schrödinger.[17,22] These states correspond to wave packets whose center of gravity moves on a classical trajectory of the oscillator. This ensures the closest correspondence between the quantum-mechanical and classical descriptions. The coherent discrete states differ from the Glauber states used in Refs. 6, 23, and 24. The Glauber formalism is convenient in the description of the excitation of AQO with allowance for fluctuations. However, because of the overpopulation of the set of these states, special steps must be taken to regularize the density matrix.[25]

As in Refs. 6 and 16, we shall not take into account the finite lifetime of the states, i.e., our analysis will be confined, for example, to radiative lifetimes Γ^{-1}. We note that inclusion of relaxation in the anharmonic classical oscillator produces only a slight distortion of its resonance curve, i.e., the dependence of the steady-state amplitude on the frequency of the external force.[3,4] Damping plays an important role in forced steady-state oscillations (of frequency equal to that of the external force, ω_f) and, eventually, stops the free oscillations. However, when free oscillations are initially absent, a sufficiently slow variation (scanning) of the frequency or amplitude of the external field ensures that the relative amplitude of free oscillations is exponentially small even in the absence of damping, i.e., it is $\sim \exp(-\pi\Delta\omega/2\gamma)$, where γ is the rate of scanning, $\Delta\omega$ is the frequency detuning, and $\gamma \ll \Delta\omega$. In this sense, relaxation (damping) does not introduce any fundamental modification to the problem of bistability. On the contrary, even low-level noise produces the metastability of the two states of the classical oscillator that are present in the absence of noise.[26,27] Noise gives rise to a finite probability of transfer between these two states so that, in the statistical description, a single distribution function is established and is independent of the initial conditions. In this sense, there is neither hysteresis nor bistability, and the two phenomena can be observed only within time intervals that are shorter than the time necessary to leave the metastable state. Steady states of the AQO[1)] subjected to a mono-

 0038-5646/84/040689-07$04.00

chromatic external force and a noise component are discussed in Refs. 23 and 24, where it is concluded that bistability is possible only over restricted intervals of time. Since the radiative lifetime of IR oscillations is sufficiently long,[28] ($\Gamma^{-1} \sim 10^{-8}$ s), the foregoing discussion justifies the physical formulation of the AQO excitation problem without taking relaxation into account.

In this paper, we shall use the coherent states of the harmonic oscillator and the methods of the theory of nonlinear oscillations[3,29] to find the solutions of the Schrödinger equation for the AQO. We shall derive the equation of motion for the center of gravity of the wave packet (quantum-mechanical analog of the Duffing equation[3,29]). A brief report of these results was given in Ref. 30. The derivation of the quantum-mechanical Duffing equation given here is augmented by an analysis of effects that are the analogs of parametric resonance. They could give rise to the destruction of the coherent states and, in fact, restrict the possibility of their excitation. Solutions of the equation of motion and the corresponding expansions over the stationary states are obtained for the case of a monochromatic external force. The conditions for the excitation of high vibrational states of the AQO and the hysteresis phenomena occurring for slow frequency scanning are determined. The essentially quantum-mechanical excitation, whose description is qualitatively different from that obtained in the semiclassical approximation, is investigated.

1. DUFFING'S QUANTUM-MECHANICAL EQUATION

Consider the Schrödinger equation ($\hbar = m = 1$)

$$i\frac{\partial\Psi}{\partial t}+\frac{1}{2}\frac{\partial^2\Psi}{\partial x^2}-\frac{1}{2}\omega^2(t)x^2\Psi+f(t)x\Psi-U_{\rm nl}(x)\Psi=0, \tag{1.1}$$

where $U_{\rm nl}=\alpha x^3+\beta x^4$) is the anharmonic component of the potential and $f(t)$ is the external force with characteristic frequency ω_f, which is close to the frequency ω of the oscillator, so that

$$|\omega^2(t)/\omega_f^2-1|\ll 1. \tag{1.2}$$

For the zero-order approximation, we shall take the coherent states of the free harmonic oscillator[17]

$$\Psi_n^{(0)}=\Phi_n(x_1)\exp[i(\dot\eta^{(0)}x_1-\varepsilon_n t+\sigma_n^{(0)}(t))], \quad n=0,1,2,\ldots, \tag{1.3}$$

where

$$\Phi_n(x_1)=\left[\frac{1}{2^n n!}\left(\frac{\omega_f}{\pi}\right)^{1/2}\right]^{1/2}\exp\left(-\frac{1}{2}\omega_f x_1^2\right)H_n(\omega_f^{1/2}x_1),$$
$$\varepsilon_n=(n+1/2)\omega_f,\ x_1=x-\eta^{(0)}(t), \tag{1.4}$$
$$\ddot\eta^{(0)}+\omega_f^2\eta^{(0)}=0,\ \dot\sigma^{(0)}=1/2(\dot\eta^{(0)2}-\omega_f^2\eta^{(0)2}),$$

and H_n are Hermite polynomials. The center of gravity of the wave packet corresponding to the solutions given by (1.4) moves on the classical trajectory of the harmonic oscillator with arbitrary oscillation amplitude (because of the linearity of the oscillator). Transforming in (1.1) to the variables $x_1 = x - \eta(t)$, t, we shall seek the solution in the form

$$\Psi_n=\tilde\Phi_n(x_1,\ t)\ \exp[a(t)x_1-i\varepsilon_n t+i\sigma(t)]. \tag{1.5}$$

The function $a(t)$ will be determined below. We shall need the following recurrence relations in our subsequent analysis:

$$x\Phi_n=\frac{1}{(2\omega_f)^{1/2}}[n^{1/2}\Phi_{n-1}+(n+1)^{1/2}\Phi_{n+1}],$$
$$x^2\Phi_n=\frac{1}{2\omega_f}\{[n(n-1)]^{1/2}\Phi_{n-2}+(2n+1)\Phi_n+[(n+1)(n+2)]^{1/2}\Phi_{n+2}\}, \tag{1.6}$$
$$x^3\Phi_n=\frac{3(n+1/2)}{2\omega_f}x\Phi_n-\frac{3}{(2\omega_f)^2}\frac{d\Phi_n}{dx}+\frac{1}{(2\omega_f)^{3/2}}$$
$$\times\{[n(n-1)(n-2)]^{1/2}\Phi_{n-2}+[(n+1)(n+2)(n+3)]^{1/2}\Phi_{n+3}\},$$
$$x^4\Phi_n=\frac{1}{(2\omega_f)^4}\{[n(n-1)(n-2)(n-3)]^{1/2}\Phi_{n-4}+(4n-2)[n(n-1)]^{1/2}\Phi_{n-2}$$
$$+3(2n^2+2n+1)\Phi_n+(4n+6)[(n+1)(n+2)]^{1/2}\Phi_{n+2}+[(n+1)(n+2)(n+3)(n+4)]^{1/2}\Phi_{n+4}\}.$$

These will be used in the first approximation in the small parameters proportional to the anharmonic coefficient α, β, the force $f(t)$, and the detuning $\omega^2(t)-\omega_f^2$, to show that

$$i\frac{\partial\tilde\Phi_n}{\partial t}+\frac{1}{2}\frac{\partial^2\tilde\Phi_n}{\partial x_1^2}+\left[-i\dot\eta+(\alpha+4\beta\eta)\frac{3}{(2\omega_f)^2}+a\right]\frac{\partial\tilde\Phi_n}{\partial x_1}$$
$$-\frac{1}{2}\Omega^2(t)x_1^2\tilde\Phi_n+\varepsilon_n\tilde\Phi_n+X(t)x_1\tilde\Phi_n+Y(t)\tilde\Phi_n=Z, \tag{1.7}$$
$$\Omega^2(t)=\omega^2(t)+6\alpha\eta+12\beta\eta^2,$$

where

$$X(t)=f(t)+i\dot a-\omega^2(t)\eta-(\alpha+4\beta\eta)\frac{3(n+1/2)}{2\omega_f}-3\alpha\eta^2-4\beta\eta^3, \tag{1.8}$$
$$Y(t)=-\dot\sigma+\frac{1}{2}a^2-ia\dot\eta+\frac{1}{2}\omega^2(t)\eta^2+f(t)\eta$$
$$-\frac{3\beta}{(2\omega_f)^2}(2n^2+2n+1)-\alpha\eta^3-\beta\eta^4,$$
$$Z=\frac{\beta}{(2\omega_f)^2}\{[n(n-1)(n-2)(n-3)]^{1/2}\Phi_{n-4}+(4n-2)[n(n-1)]^{1/2}\Phi_{n-2}$$
$$+(4n+6)[(n+1)(n+2)]^{1/2}\Phi_{n+2}+[(n+1)(n+2)(n+3)(n+4)]^{1/2}\Phi_{n+4}\}$$
$$+\frac{\alpha+4\beta\eta}{(2\omega_f)^{3/2}}\{[n(n-1)(n-2)]^{1/2}\Phi_{n-3}+[(n+1)(n+2)(n+3)]^{1/2}\Phi_{n+3}\}.$$

To ensure that the term including $\partial\Phi_n/\partial x_1$ vanishes in (1.7), we shall require that

$$a(t)=i\dot\eta-\frac{3}{(2\omega_f)^2}(\alpha+4\beta n). \tag{1.9}$$

Terms including $Y(t)$ are eliminated by suitably choosing $\sigma(t)$. The quantity $X(t)$ is of the first order of small quantities, but it cannot be taken into acocunt within the framework of standard perturbation theory because it contains resonant terms. We are therefore forced to set $X(t) = 0$, which gives an equation of motion for $\eta(t)$. The quantity Z contains nonresonant terms, namely, a small admixture of the states

$\Phi_{n\pm2,\,n\pm3,\,n\pm4}$. These can be taken into account in the usual way of perturbation theory,[22] but do not contribute to the equation of motion. The solution of the remaining Schrödinger equation for the harmonic oscillator with effective frequency $\Omega(t)$ [i.e., of the form (1.1) with $U_{nl}=0$] is given in Ref. 17. We note that our analysis is valid if the functions $\tilde{\Phi}_n$ are not very different from the unperturbed functions Φ_n. This condition requires further justification in the region of parametric resonance.[17] This is provided in Sec. 3 and in the Appendix.

The contribution to the equation of motion in the first approximation in α leads to a small shift and small oscillations in $\eta(t)$. The contribution of the second order in α can be effectively taken into account by changing the coefficient β. We shall therefore substitute $\alpha=0$ and $\omega^2(t)=\omega_0^2=\text{const}$ in (1.8). The equation of motion then assumes the form

$$\ddot{\eta}+\tilde{\omega}_n^2\eta+4\beta\eta^3+i\frac{\rho}{\omega_0}\dot{\eta}=f(t), \tag{1.10}$$

where

$$\rho=\frac{3\beta}{\omega_0}\left(=\frac{3\hbar\beta}{m^2\omega_0}\right), \quad \tilde{\omega}_n^2=\omega_0^2+2\rho\left(n+\frac{1}{2}\right). \tag{1.11}$$

The classical Duffing equation is obtained from (1.10) when $\rho\to0$ ($\hbar\to0$). According to the "quantum-mechanical Duffing equation" (1.10), the trajectory of the center of gravity of the wave packet is complex. Quantum-mechanical corrections are relatively small when the amplitude of the oscillations $\eta(t)$ is much greater than the characteristic width of the wave packet, $\sim\omega_0^{-1/2}$. More detailed analysis of (1.10) is given in the next section.

2. SOLUTION OF THE EQUATION OF MOTION FOR A MONOCHROMATIC EXTERNAL FORCE

Suppose that the external force is monochromatic:

$$f(t)=A\cos(\omega_f t). \tag{2.1}$$

To elucidate the role of the "quantum-mechanical" term in (1.10), consider, to begin with, the case of small oscillations:

$$\beta\eta^2(t)\ll\rho. \tag{2.2}$$

We can then neglect the nonlinear term in (1.10), which enables us to write its general solution in the form

$$\eta(t)=C^{(+)}e^{i\omega^{(+)}t}+C^{(-)}e^{-i\omega^{(-)}t}+\eta^{(B)}(t), \tag{2.3}$$

where

$$\eta^{(B)}(t)={}^1\!/_2(pe^{i\omega_f t}+qe^{-i\omega_f t}) \tag{2.4}$$

and

$$p=\frac{A}{\tilde{\omega}_n^2-\omega_f^2-\rho}, \quad q=\frac{A}{\tilde{\omega}_n^2-\omega_f^2+\rho},$$

$$\omega^{(\pm)}=(\tilde{\omega}_n^2\pm\rho)^{1/2}, \quad C^{(\pm)}=\text{const}. \tag{2.5}$$

In the absence of the external force ($A=0$), Eq. (2.3) shows that there are natural oscillations with frequencies $\pm\omega^{(\pm)}$ and amplitudes $C^{(\pm)}$, which are determined by the initial conditions. Inclusion of relaxation (finite lifetime) should lead to the damping of the natural oscillations. The forced oscillations (2.4) are characterized by two oscillation amplitudes p and q at frequencies $\pm\omega_f$. The physical meaning of these amplitudes is different. Thus, for $n=0$, the time dependence of the square of the modulus of the wave function is ($C^{(\pm)}=0$)

$$|\Psi_0|^2\sim\exp\left[-\omega(x-q\cos(\omega_f t))^2\right]. \tag{2.6}$$

In the above case, the amplitude of the motion of the center of gravity of the wave packet is equal to q, and the average value of the coordinate is $\bar{x}=q\cos(\omega_f t)$, so that p does not appear in the time dependence of the wave function.

We must now find the "forced" periodic solutions of (1.10) without assuming that (2.2) is satisfied. Taking $\eta(t)$ in the form given by (2.4), and substituting it into the resonance approximation usually employed for the Duffing equation,[3,4,23] we obtain

$$[(u+d)^2-\rho^2]u=v\ , \tag{2.7}$$

where

$$u=3|\beta|pq,\ v=3|\beta|A^2,\ d=\text{sign}(\beta)(\tilde{\omega}_n^2-\omega_f^2). \tag{2.8}$$

The amplitudes p and q can be uniquely expressed in terms of u:

$$p=\frac{A}{(\tilde{\omega}_n^2-\omega_f^2+3\beta pq)-\rho}=\frac{A\,\text{sign}(\beta)}{(d+u)-|\rho|}$$
$$q=\frac{A}{(\tilde{\omega}_n^2-\omega_f^2+3\beta pq)+\rho}=\frac{A\,\text{sign}(\beta)}{(d+u)+|\rho|}. \tag{2.9}$$

The cubic equation (2.7) for u has for all values of the parameters three solutions of which at least one must be real. It can be shown that complex roots correspond to unstable solutions of (1.10). The necessary condition for the stability of solutions with real u is

$$(d+u)(d+3u)\geqslant0. \tag{2.10}$$

"Inversion" of (2.7) yields

$$d=-u\pm(v/u+\rho^2)^{1/2} \tag{2.11}$$

and this shows that the real values of u occur in the following two intervals: $-\infty<u<-v/\rho^2$ and $0<u<\infty$. Figure 1 shows the resonance curve corresponding to (2.7), i.e., the frequency dependence of u. Broken curves show the solutions that do not satisfy the stability condition (2.10).

It is clear from (2.8) that in the case $\beta<0$, which is

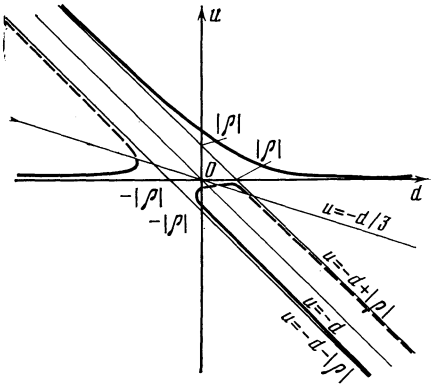

FIG. 1. Resonance curve of the anharmonic quantum-mechanical oscillator.

characteristic for molecular oscillations, the quantity d increases with increasing frequency ω_f of the exciting radiation. The quantum-mechanical case, where the Rabi frequency is much lower than the anharmonic shift $A\omega^{1/2}/|\rho| \ll 1$, is of particular interest. Let us rewrite (2.7) in the form

$$\frac{u}{|\rho|}\left(\frac{u+d}{|\rho|}-1\right)\left(\frac{u+d}{|\rho|}+1\right)=\frac{A^2\omega}{\rho^2} \tag{2.12}$$

The point $d=-|\rho|$ in Fig. 1 corresponds to resonance at the frequency of the $n\to n+1$ transition. In the immediate neighborhood of this point, there is only one real solution (right-hand external branch of the resonance curve)

$$u_1+d-|\rho| \approx \frac{A^2\omega}{4|\rho|}, \quad p=\frac{4\rho}{A\omega}, \quad q=\frac{A}{2\rho}. \tag{2.13}$$

Two other solutions

$$u_{2,3}+d+|\rho|=\frac{d+|\rho|}{2}\pm\left[\left(\frac{d+|\rho|}{2}\right)^2-\frac{A^2\omega}{2}\right]^{1/2} \tag{2.14}$$

correspond to the outer $d<-|\rho|$ and inner $d>-|\rho|$ branches of the resonance curve. These solutions exist when the detuning from the frequency of the $n\to n+1$ transition is not too small: $|\omega_f-(\tilde{\omega}_n-|\rho|/2\omega)| \geqslant A(\omega/2)^{1/2}$. At the branch points of the resonance curve,

$$p=-A/2\rho, \quad q=\pm\text{sign}\,(\beta)\,(2/\omega)^{1/2}. \tag{2.15}$$

The point $d=|\rho|$ corresponds to resonance at the frequency of the $n\to n-1$ transition. There are three solutions in the neighborhood of this point. For the solution corresponding to the lower branch of the inner curve,

$$u_1+d+|\rho| \approx \frac{A^2\omega}{4|\rho|}, \quad p=-\frac{A}{2\rho}, \quad q=\frac{4\rho}{A\omega}. \tag{2.16}$$

For the other two solutions (external and internal branches), we have, correspondingly,

$$u_{2,3}+d-|\rho|=\frac{d-|\rho|}{2}\pm\left[\left(\frac{d-|\rho|}{2}\right)^2+\frac{A^2\omega}{2}\right]^{1/2}, \quad p=\pm\text{sign}\,(\beta)\left(\frac{2}{\omega}\right)^{1/2}, \quad q=\frac{A}{2\rho}. \tag{2.17}$$

Thus, the effective rise in the amplitude q [which, in accordance with (2.6) and Sec. 3, determines the excitation of the oscillator] occurs at low frequencies along the left-hand outer branch of the resonance curve, as the excitation frequency is approached. For this type of frequency scanning $\omega^{1/2}q \sim 1$ at the top of the curve. We note that q is then practically independent of the amplitude of the external force. On the right-hand branch, on the other hand, the amplitude p increases as the top of the resonance curve is approached, and q is always small ($\sim A/2\rho$).

As in the case of small oscillations (2.2), Eq. (1.10) allows the existence of self-oscillations, i.e., oscillations of the center of the packet that are not damped out after the external force is turned off (in the absence of relaxation). They can be found by replacing (2.4) with

$$\eta=\tfrac{1}{2}[P(\tau)e^{i\tau}+Q(\tau)e^{-i\tau}], \quad \tau=\tilde{\omega}_n t. \tag{2.18}$$

Assuming that P and Q change little when τ is increased by 2ρ, and using the method of slowly varying amplitudes (or the method of two-scale expansions,[25] which is equivalent to it in the lowest-order approximation), we obtain

$$2i\frac{dP}{d\tau}+3\frac{\beta}{\tilde{\omega}_n^2}P^2Q-\frac{\rho}{\tilde{\omega}_n^2}P=0, \quad 2i\frac{dQ}{d\tau}-3\frac{\beta}{\tilde{\omega}_n^2}PQ^2-\frac{\rho}{\tilde{\omega}_n^2}Q=0, \tag{2.19}$$

and hence

$$P(\tau)Q(\tau)=P(0)Q(0)\exp\left(-i\frac{\rho}{\tilde{\omega}_n^2}\tau\right). \tag{2.20}$$

Substituting the last result in (2.19), we obtain

$$P(\tau)=P(0)\exp\left\{-\frac{1}{2}\tilde{\omega}_n P(0)Q(0)\times\left[\exp\left(-i\frac{\rho\tau}{\tilde{\omega}_n^2}\right)-1\right]-\frac{i\rho}{2\tilde{\omega}_n^2}\tau\right\} \tag{2.21}$$

$$Q(\tau)=Q(0)\exp\left\{\frac{1}{2}\tilde{\omega}_n P(0)Q(0)\times\left[\exp\left(-i\frac{\rho\tau}{\tilde{\omega}_n^2}\right)-1\right]-\frac{i\rho}{2\tilde{\omega}_n^2}\tau\right\},$$

or, when $P(0)Q(0)$ is small,

$$P(\tau)=P(0)\exp\left(-i\frac{\rho\tau}{2\tilde{\omega}_n^2}\right)\times\left\{1-\frac{1}{2}\tilde{\omega}_n P(0)Q(0)\left[\exp\left(-i\frac{\rho\tau}{\tilde{\omega}_n^2}\right)-1\right]\right\}, \tag{2.22}$$

$$Q(\tau)=Q(0)\exp\left(-i\frac{\rho\tau}{2\tilde{\omega}_n^2}\right)\times\left\{1+\frac{1}{2}\tilde{\omega}_n P(0)Q(0)\left[\exp\left(-i\frac{\rho\tau}{\tilde{\omega}_n^2}\right)-1\right]\right\}.$$

The amplitudes P and Q obtained in this way are periodic functions with period

$$T=4\pi\tilde{\omega}_n^2/|\rho|. \tag{2.23}$$

3. DISCUSSION OF RESULTS

There is considerable interest in the degree of excitation of the AQO in a coherent state. This is characterized by the distribution of the population of stationary states, which is determined by the coefficients of the expansion of the coherent state in terms of the wave functions of stationary states. For the harmonic oscillator, this expansion is

$$\psi_n=\sum_k a_{nk}(t)\,\Phi_k(x)\,e^{-i\varepsilon_k t}, \tag{3.1}$$

$$a_{nk}=\frac{1}{(2^n n!\,2^k k!)^{1/2}}\times\exp\left[i(\varepsilon_k-\varepsilon_n)t+i\sigma(t)-\frac{\omega}{4}\left(\eta^2+\frac{\dot{\eta}^2}{\omega^2}\right)-i\frac{\eta\dot{\eta}}{2}\right]\times\begin{cases}2^n k!\left[-\frac{\omega^{1/2}}{2}\left(\eta-i\frac{\dot{\eta}}{\omega}\right)\right]^{n-k}L_k^{n-k}\left[\frac{\omega}{2}\left(\eta^2+\frac{\dot{\eta}^2}{\omega^2}\right)\right], & n\geqslant k\\ 2^k n!\left[\frac{\omega^{1/2}}{2}\left(\eta+i\frac{\dot{\eta}}{\omega}\right)\right]^{k-n}L_n^{k-n}\left[\frac{\omega}{2}\left(\eta^2+\frac{\dot{\eta}^2}{\omega^2}\right)\right], & n\leqslant k,\end{cases}$$

where L_n^m are Laguerre polynomials.

The expansion coefficients of the normal coherent state ($n=0$) have the simplest form: to within quantities $\sim(\omega-\omega_f)/\omega$ and a constant normalizing factor, the coefficients (3.1) can then be written in the form

$$|a_{0k}|=\frac{1}{(k!)^{1/2}}\left[\left(\frac{\omega}{2}\right)^{1/2}q\right]^k e^{-\omega q^2/4}, \tag{3.2}$$

where q is the amplitude of the oscillations of the center of the wave packet, given by (2.4). We note that, in this particular approximation, the expansion coefficients are independent of the amplitude p. We have already shown in Sec. 2 [Eq. (2.6)] that p does not appear in the time dependence of the wave function of the zeroth coherent states. We can verify with the aid of (3.1) that, when n is not too high, the amplitude q determines mainly the coefficients with $k>n$, and p determines the coefficients with $k<n$, i.e., q characterizes the excitation and p the deactivation of the stationary states. The main assumption that was used in obtaining the AQO wave functions was that the spatial structure of these functions was very similar and, consequently, so were the coefficients of the expansion of these functions in terms of the stationary states in the case of the harmonic oscillator. In the Appendix, we shall examine the distortion of the spatial dependence of the wave functions due to anharmonism, and will give the conditions for the validity of the analysis. For simplicity, we shall confine our attention to the normal coherent state $n=0$. To ensure that the admixture of the stationary state for $n=2$, which appears as a result of anharmonism, is less than the corresponding expansion coefficient (3.2), we must have

$$b(t),\ c/\omega,\quad d\ll\omega^2q^2. \tag{3.3}$$

The most stringent restriction ensuing from these conditions is that $b_1(t)\ll\omega^2q^2$. From this, it follows [see (8.5)] that, at resonance ($\omega_f\approx\omega$), we must have, even for small amplitudes,

$$c/\omega,\quad d\gg\beta/\omega^2. \tag{3.4}$$

This means that, when the detuning is small, the coherent states of the AQO differ from the harmonic oscillator in that they do not become pure stationary states, which complicates their excitation by monochromatic radiation. When the exciting frequency is scanned in a region well away from resonance, $b_1(t)$ is quite small for large detuning. Hence, in the limit as $q, p\to0$, the coherent states become stationary states whose quantum numbers are equal to the number n of coherent states. By scanning the frequency ω_f between the low-frequency region and detunings of the order of the anharmonic shift $|\omega-\omega_f|\sim\beta/\omega^2$, we can excite the oscillator. It is then essential that (3.3) and (3.4) be satisfied, i.e., the excitation amplitude q must be sufficiently large. In this frequency range, $q\sim A\omega/\beta$ [see (2.13)]. We now introduce the Rabi frequency of the exciting radiation $\nu_P=A/(2\omega)^{1/2}$ and the anharmonic shift $\Delta_a=\rho/\omega$, and write this condition in the form

$$\nu_P\gg\Delta_a(\Delta_a/\omega)^{1/2}. \tag{3.5}$$

We note that the well-known condition for the excitation of multilevel systems, obtained within the framework of the two-level approach, has the form $\nu_P>\Delta_a$. The criterion for the excitation of the anharmonic oscillator differs from this by the presence of the factor $(\Delta_a/\omega)^{1/2}\ll1$. Thus, when $(\Delta_a/\omega)^{1/2}\ll\nu_P/\Delta_a\ll1$, the AQO does not reduce to the two-level system even when $|\omega_f-\omega|\sim\nu_P$.

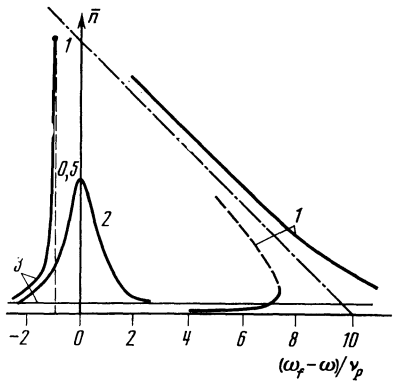

FIG. 2. Population of excited states of the anharmonic quantum-mechanical oscillator as a function of excitation frequency: $\Delta_a/\omega=0.01$, $\nu_P/\Delta_a=0.1$; 1—classical oscillator, 2—excitation of the AQO averaged over fluctuations, 3—excitation of the AQO over time intervals shorter than the life of the metastable states.

Condition (3.5) ensures a Poisson distribution (3.2) over the stationary levels for all scanning frequencies. The maximum value of q is reached at the top of the resonance curve, near the point $d=-|\rho|$ in Fig. 1. It is given by $q=(2/\omega)^{1/2}$. The amplitudes of the stationary states in the normal coherent state are

$$|a_{0k}|=e^{-1/4}/(k!)^{1/2}. \tag{3.6}$$

When the exciting frequency ω_f is scanned in the region of high frequencies, the amplitude p is found to rise but q remains small. As noted in Sec. 2, the normal coherent state does not then have an admixture of excited stationary states. It can be verified that, even in this case, it is essential to satisfy (3.5). Thus, coherent states with the same quantum number n are characterized by different degrees of excitation that are determined by past history.

Figure 2 shows the population $\bar{n}$ of the excited states of the anharmonic oscillator in the normal coherent state as a function of the frequency ω_f. Curve 1 describes the resonant excitation of the classical anharmonic oscillator. The value of $\bar{n}$ for this oscillator is given by $\bar{n}=E/\hbar\omega$ ($E=1/2m\omega^2\eta_0^2$ is the oscillator excitation energy). Curve 2 describes the excitation of the AQO, averaged over the quantum oscillations,[23,24] in the limit of infinitely small damping. We recall that this case occurs when the damping time of the metastable state is still shorter than the time of operation of the external force. Curve 3 corresponds to the case where damping can be neglected, which we assume in this paper. It corresponds to the left-hand outer branch of the resonance curve shown in Fig. 1 ($\bar{n}=1/2\omega q^2$). As noted above, no appreciable excitation of the oscillator occurs along the right-hand outer branch. For the indicated parameter values, $\bar{n}=0.025$ on this branch in this part of the spectrum. It is important to note that, in our case, bistability is essentially quantum-mechanical in character ($\nu_P\ll\Delta_0$). In contrast to the classical

case, excitation occurs on the left-hand branch of the resonance curve ($\omega_f < \tilde{\omega}_n$ if $\beta < 0$). None of the quasiclassical approximations that are admissible for $\nu_P > \Delta_0$ can then be used.[6,23,24]

The above results show that the anharmonic oscillator is an essentially multilevel system and that many-photon processes play an important role in the evolution of its coherent states. In our approach, we start with the coherent states of the harmonic oscillator and the basic many-photon resonances are taken into account even in the early approximations. These are, above all, the allowed many-photon transitions in the harmonic oscillator for which the number of photons is equal to the difference between the quantum numbers of the stationary states between which the transition occurs. The exciting frequency ω_f for such transitions is then bounded by the transition frequency ω_{01}. When $\beta < 0$, so that the natural frequencies of the AQO decrease with increasing quantum number, this region is $\omega_f < \omega_{01}$. Hence, it follows that, when the exciting frequency is scanned in the region $\omega_f > \omega_{01}$, there should be a situation approaching the two-level scheme in which the many-photon resonances do not appear. Let us consider the $n = 1$ coherent state. According to (31), the amplitudes of the stationary oscillations are

$$|a_{1k}| = \frac{N}{(2^k k!)^{1/2}} \omega^{1/2} p, \quad k=0,$$
$$|a_{1k}| = \frac{N}{(2^k k!)^{1/2}} \cdot 4(\omega q^2)^{k-1} L_1^{k-1}\left(\frac{\omega}{2} pq\right), \quad k \geqslant 1, \tag{3.7}$$

where N is a normalizing factor. Near the point $d = |\rho|$ on the resonance curve for this state, which corresponds to the $1 \to 0$ transition, we have from (2.9) and (2.17)

$$p \approx 2A\{d - |\rho| \pm [(d-|\rho|)^2 + 2\omega A^2]^{1/2}\}^{-1}, \quad q \approx A/2\rho, \tag{3.8}$$

where the signs $\pm$ correspond to the outer and inner branches of the resonance curve, respectively (Fig. 1). When $\nu_P \ll \Delta_a$, we have $\omega q^2 \ll \omega pq \ll 1$ near the resonance, and only the two levels[2)] with $k = 0$ and 1 are appreciably populated. Substituting $d - |\rho| \approx 2\omega_{01}(\omega_{01} - \omega_f)$, we can readily verify tht (3.7) and (3.8) describe the amplitudes of coherent states of the two-level system.[22] The outer branch corresponds to the coherent state in which the population of the excited state increases with increasing ω_f, whereas the inner branch shows a decrease. We note that, well away from the resonance ($\omega_f > \omega_{01}$), the inner branch of the resonance curve of the $n = 1$ coherence state, like the outer branch of the $n = 0$ state, corresponds to the same normal stationary state of the oscillator. There is a considerable difference between the intensities that are necessary for the excitation of these states near resonance. Whereas the excitation of the $n = 0$ coherent state in the neighborhood of the ω_{01} resonance must obey condition (3.5) (the amplitude is $\omega^{1/2} p \approx \Delta_a / \nu_P \gg 1$), the state corresponding to the two-level system is formed at low intensities $\omega^{1/2} p \approx 1$.

The analysis given above has demonstrated the existence of coherent states of the AQO in a monochromatic external field. However, detailed description of the excitation of coherent states and transitions between different branches of the resonance curve turns out to be too complicated and probably cannot be obtained within the framework of the purely analytical approach. There are several papers[31-33] in which numerical methods are used to investigate, in the absence of the external force ($f = 0$), the evolution of the AQO state formed initially as a coherent state of the harmonic oscillator. In our approach, this corresponds to the self-oscillations considered at the end of Sec. 2. Brickmann and Russegger[31] have determined, in particular, the time t_k after which the packet returns to its initial state for a number of model potentials. According to equation (2.23) in that paper, the self-oscillation period is

$$T = 2\left[\frac{\omega_0^2}{\rho} - 2\left(n + \frac{1}{2}\right)\right]\frac{2\pi}{\omega_0}, \quad \beta < 0. \tag{3.9}$$

We have carried out a comparison for the smallest anharmonicity parameter $\varepsilon = 0.1$ used in Ref. 31 because, for higher values, the continuous spectrum, which we have not examined here, begins to play an important role. It then turns out that $t_K = (45 \pm 5) \cdot 2\pi/\omega_0$. Equation (3.9) yields $T = 40 \cdot 2\pi/\omega_0$ for the corresponding anharmonicity parameters, which is in good agreement with numerical experiment.

The above effects accompany the excitation of coherent states and can be observed in simple molecules exposed to coherent radiation.

Table I lists the parameters of well-known diatomic molecules, namely, the frequency ω, the anharmonic shift $\Delta_a = \omega_e x_e$, the minimum value of the Rabi frequency calculated from (3.5), and the laser power density corresponding to this Rabi frequency. It is clear from the table that the lowest power density necessary for the observation of effective resonant excitation by atomic molecules and of excitation hysteresis is 2 GW/cm^2 for SO and 10 GW/cm^2 for CO. These values are attainable when modern laser beams are suitably focused down.

APPENDIX

In Eq. (1.7) in the main text, the square of the effective frequency, $\Omega^2(t)$, contains the term $12\beta\eta^2$. It is clear from (2.4) that this term includes oscillations of frequency $2\omega_f$.

TABLE I.

	CO	HBr	HCl	HF	HI	NO	SO
ω, cm^{-1}	2170	2650	2990	4138	2310	1904	1124
Δ_a, cm^{-1}	13.46	45.2	52.05	90	39.7	13.91	6.12
ν_P, cm^{-1}	1.1	5.9	6.9	13.3	5.2	1.2	0.45
P, GW/cm^2	10	350	480	2700	270	15	2

This means that, for small detuning $|\omega_f - \omega| < 3\beta q^2/2\omega$, we can have parameteric resonance-type phenomena that lead to distortions of the wave function. We shall show below that these distortions do not modify the equation of motion although they impose specific limitations on the possibility of resonant excitation of coherent states.

We shall seek the solution of (1.1) in the form

$$\Psi_n = \exp[-i\sigma(t) - i\varepsilon_n t + a(t)x_1 + b(t)x_1^2 + cx_1^4]\,\tilde{\Phi}_n(x_1 + dx_1^3). \tag{A1}$$

For $b = c = d = 0$, this expression becomes identical with (1.5). Substituting this Ψ_n into the Schrödinger equation (1.1), we obtain an equation of the form of (1.7) with $X(t)$ and $Y(t)$ retaining their form. The choice of the wave function (A1) leaves the equation of motion (1.10) unaltered. Let us divide the coefficient $b(t)$ into two parts, namely, $b = b_0 + b_1(t)$, where b_0 is a constant, and let us write out the terms of the equation relates the states with $\Delta n = \pm 2$ (analog of the expression $1/2\Omega^2 x_1^2 \Phi_n$):

$$\begin{aligned}&[(i\dot{b}_1 + 4b_0b_1 + 2b_1^2 - 6\beta\eta^2)x_1^2 + 8b_1cx_1^4]\,\tilde{\Phi}_n \\ &+ 2b_1x_1\tilde{\Phi}_n' + [(6c + 2b_0^2 - {}^1/_2\omega^2)x_1^2 - \beta x_1^4 \\ &+ 8c^2x_1^6]\,\tilde{\Phi}_n + (2b_0x_1 + 4cx_1^3)\,\tilde{\Phi}_n' + {}^1/_2\tilde{\Phi}_n''.\end{aligned} \tag{A2}$$

It can be verified that the resonant term $6\beta\eta^2$ will not cancel out when $b_0 = c = d = 0$. Moreover, the constant shift of the effective frequency $\Omega(t)$ by the amount $\sim b_0$ leads, as a result of the mixing of the states $\Phi_{n\pm 2}$, to the appearance of resonant terms in higher orders of perturbation theory, which are of the same order as the term to be compensated. Equating to zero the coefficients in the expansion for the time-independent part of (A2), and assuming that $b/\omega, c/\omega^2, d/\omega \ll 1$, we obtain two equations for the constants b_0, c and d:

$$c = \frac{(2n+1)}{4}\omega d, \qquad b_0 = \frac{d}{4}[9 - 4n(n+1)]. \tag{A3}$$

To cancel out the time-dependent resonant terms, we substitute

$$b_1 = b_1^{(+)} e^{2i\omega_f t} + b_1^{(-)} e^{-2i\omega_f t}. \tag{A4}$$

By analogy with the foregoing, we find that

$$\begin{aligned} b_1^{(+)} &= \frac{3\beta}{2} p^2 \left[\omega - \omega_f + (4n-2)\left(\frac{c}{\omega} + d\right)\right]^{-1}, \\ b_1^{(-)} &= \frac{3\beta}{2} q^2 \left[\omega - \omega_f + (4n+6)\left(\frac{c}{\omega} - d\right)\right]^{-1}. \end{aligned} \tag{A5}$$

Thus, the wave function given by (A1) is determined to within one of the constant b_0, c, d, related by (A3). In the above derivation, we made the assumption that

$$b_0/\omega, c/\omega^2, d/\omega \ll 1, b_1/\omega \ll 1. \tag{A6}$$

It is clear from (A5) that the last condition is the most important in the case of resonant excitation, when $|\omega - \omega_f| < |\beta|/\omega^2$. The quantities ωp^2 or ωq^2 (see Sec. 2) can then be ~ 1. For condition (A6) to be satisfied in this case as well, we must ensure that $d \gg |\beta|/\omega^2$.

[1)]A nonlinear Fabry-Perot interferometer is investigated in Refs. 23 and 24. Its Hamiltonian is identical with the AQO Hamiltonian. Noise is ignored in Ref. 24, and the analysis is confined to the semiclassical approximation in which hysteresis has the usual form.[3,4]

[2)]In coherent states with $n > 1$, all the lowest states 0, 1, ..., n are populated in the neighborhood of the $n \to n-1$ resonance, i.e., the system does not reduce to a two-level system.

[1]V. N. Lugovoi, Kvantovaya Elektron. (Moscow) **6**, 2053 (1979) [Sov. J. Quantum Electron. **9**, 1207 (1979)].
[2]N. N. Roganov, Zh. Eksp. Teor. Fiz. **80**, 96 (1981) [Sov. Phys. JETP **53**, 47 (1981)].
[3]N. N. Bogolyubov and Yu. A. Mitropol'skiĭ, Asimptoticheskie metody v teoriu nelineĭnykh kolebaniĭ (Asymptotic Methods in the Theory of Nonlinear Oscillations), Nauka, Moscow, 1974.
[4]L. D. Landau and E. M. Lifshitz, Mekhanika (Mechanics), Nauka, Moscow, 1965, p. 113 (English translation by Pergamon Press).
[5]P. A. Braun, Teor. Mat. Fiz. **37**, 355 (1978).
[6]V. N. Sazonov, Teor. Mat. Fiz. **31**, 107 (1977).
[7]V. I. Klyatskin, Stokhasticheskie uravneniya i volny v sluchaĭnoneodnorodnykh sredakh (Stochastic Equations and Waves in Randomly Inhomogeneous Media), Nauka, Moscow, 1980, p. 76.
[8]A. D. Morozov, Dif. Uravneniya **12**, 241 (1976).
[9]B. A. Huberman and J. P. Crutchfield, Phys. Rev. Lett. **43**, 1743 (1979).
[10]E. V. Shuryak, Zh. Eksp. Teor. Fiz. **71**, 2039 (1976) [Sov. Phys. JETP **44**, 1070 (1976)].
[11]G. P. Berman and G. M. Zaslavsky, Physica A **91**, 450 (1978).
[12]B. V. Chirikov, F. M. Izrailev, and D. L. Shepelyansky, Transient Stochasticity in Quantum Mechanics, Preprint 80-210 IYaF SO AN SSSR, Novosibirsk, 1980.
[13]G. P. Berman, G. M. Zaslavskiĭ, and A. R. Kolovskiĭ, Zh. Eksp. Teor. Fiz. **81**, 506 (1981) [Sov. Phys. JETP **34**, 272 (1981)].
[14]G. A. Askar'yan, Zh. Eksp. Teor. Fiz. **46**, 403 (1964) [Sov. Phys. JETP **19**, 273 (1964)]; **48**, 666 (1965) [**21**, 439 (1966)].
[15]F. V. Bunkin, R. V. Karapetyan, and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. **47**, 216 (1964) [Sov. Phys. JETP **20**, 145 (1965)].
[16]V. N. Bagratashvili, V. S. Letokhov, A. A. Makarov, and E. A. Ryabov, Mnogofotonnye protsessy v molekulakh v infrakrasnom lazernom pole. Itogi nauki i tekhniki. Fizika atoma i molekuly. Optika. Magnitnyĭ rezonans (Many-Photon Processes in Molecules in an Infrared Laser Field. Advances in Science and Technology, Physics of the Atom and the Molecule, Optics, Magnetic Resonance, VINITI, Moscow, 1981, Vol. 2, Chap. 4).
[17]A. I. Baz', Ya. B. Zel'dovich, and A. M. Perelomov, Rasseyanie, reaktsii i raspady v nerelyativistskoĭ kvantovoĭ mekhanike (Scattering Reactions and Decays in Nonrelativistic Quantum Mechanics), Nauka, Moscow, 1971, Chapt. 6 [Wiley, 1969].
[18]D. M. Larsen and N. Bloembergen, Opt. Commun. **17**, 254 (1976).
[19]M. V. Kuz'min and V. N. Sazonov, Zh. Eksp. Teor. Fiz. **73**, 422 (1977) [Sov. Phys. JETP **46**, 220 (1977)].
[20]M. K. Kuz'min, Kvantovaya Elektron. (Moscow) **5**, 789 (1978) [*sic*].
[21]V. N. Sazonov, Teor. Mat. Fiz. **35**, 361 (1978).
[22]L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika (Quantum Mechanics), Nauka, Moscow, 1974, p. 95 (English translation by Pergamon).
[23]P. D. Drummond, C. W. Gardiner, and D. F. Walls, Phys. Rev. A **24**, 914 (1981).
[24]P. D. Drummond and D. F. Walls, J. Phys. A **13**, 725 (1980).
[25]P. D. Drummond and C. W. Gardiner, J. Phys. A **13**, 2353 (1980).
[26]A. D. Venttsel' and M. I. Freĭdkin, Fluktuatsii v dinamicheskikh sistemakh pod deĭstviem malykh sluchaĭnykh vozmushcheniĭ (Fluctuations in Dynamic Systems Under the Influence of Small Random Perturbations), Nauka, Moscow, 1979, Chap. 4.
[27]M. I. Dykman and M. A. Krivoglaz, Zh. Eksp. Teor. Fiz. **77**, 60 (1979) [Sov. Phys. JETP **50**, 30 (1979)].
[28]G. Herzberg, Spectra and Structure of Diatomic Molecules, Van Nostrand [Russian translation, IL, Moscow, 1949, p. 297].
[29]I. G. Malkin, Nekotorye zadachi teorii nelineĭnykh kolebaniĭ (Some Problems in the Theory of Nonlinear Oscillations), Gostekhizdat, Moscow, 1956, p. 32.
[30]N. N. Rozanov and V. A. Smirnov, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 504 (1981) [JETP Lett. **33**, 488 (1981)]; Opt. Spectrosk. **53**, 912 (1982) [Opt. Spectrosc. (USSR) **53**, 544 (1982)].
[31]J. Brickman and P. Russegger, J. Chem. Phys. **75**, 5744 (1981).
[32]P. F. Endres and K. K. Wieson, J. Chem. Phys. **46**, 425 (1967).
[33]P. Gutschic, M. M. Nieto, and L. M. Simmens Jr., Phys. Lett. A **76**, 15 (1980).

Translated by S. Chomet