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Method of pseudo-coherent states in nonlinear quantum systems

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A new method is proposed for the study of the behavior of wave packets in various quantum systems, including anharmonic ones. It is shown how to approximate such a packet optimally with the aid of a Gaussian. A closed system of ordinary differential equations is obtained for the position and width of this packet. The proposed method is applicable during the time in which the difference between the true solution and the approximating Gaussian solution is small. Wave packets in both one-dimensional and multidimensional systems are considered. It is shown, with a quantum nonlinear string as an example, how to effect the transition from the multidimensional problem to the field problem. The obtained system of ordinary differential equations can be used to set up a numerical experiment on the excitation of molecules. It is shown how to vary the frequency of the field with time at a given anharmonicity, so as to ensure an effective rapid excitation of an oscillator.

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INTRODUCTION

Much attention is being paid recently to the buildup of oscillations in molecules. This question is important for the understanding of the phenomenon of collisionless collective dissociation, of the excitation of molecules by intense light,[1-5] of the excitation of oscillations of the field of surface forces,[6] etc. The behavior of such nonlinear systems in monochromatic fields was considered in a large number of papers (see, e.g., Refs. 7–9).[1)] In Refs. 8 and 9, computer calculations were used to consider both classical and quantum problems dealing with the behavior of a particle in a one-dimensional potential well of non-harmonic type under the influence of an external exciting force. In many of the cases considered in Ref. 8, a curious regularity was observed: despite the relatively strong anharmonicity, the solution in the quantum case was a more or less localized wave packet moving along a trajectory close to classical, but in contrast to the well known coherent state in the harmonic oscillator,[10] the shape of the packet varied with time (the packet pulsated). A situation is possible (and is certainly realized at least during the initial stages of the the excitation), wherein the shape of such localized formations does not deviate noticably from Gaussian. We shall call such wave packets pseudo-coherent states.

In view of the large complexity of the calculation, it is impossible to use directly the method of Refs. 8 and 9 for a numerical experiment on molecules. We propose in this article a method that makes it possible to calculate, in the presence of pseudo-coherent states,[2)] the position and width of a wave packet at each instant of time in both the one-dimensional and miltidimensional cases. Since the method reduces to a solution of a system of ordinary differential equations, it can be used in principle to set up numerical experiments also for multidimensional systems that describe molecules.

When working with pseudo-coherent states we can make use of the following device: we introduce an auxilliary potential $U_1(x,t)$ which, on the one hand, approximates at each instant of time in "optimal" fashion the true potential $U(x,t)$ at the location of the packet [$U_1(x,t)$ can differ quite strongly from $U(x,t)$ in places where there is no packet], and on the other hand greatly simplifies the procedure of solving the Schrödinger equation. We can choose $U_1(x,t)$ to be a potential in the form $\alpha(t)+\beta(t)x+\gamma(t)x^2$.[11] Then, if at the initial instant the packet had a Gaussian form, the solution of the

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Schrödinger equation with $U_1(x,t)$ can be represented in the form

$$\exp\{-A(t)(x-\bar{x}(t))^2+i\hbar^{-1}M\dot{\bar{x}}(t)(x-\bar{x}(t))+B(t)\},$$

where for $A(t), \bar{x}(t)$ and $B(t)$ we can write ordinary differential equations that contain $\alpha(t), \beta(t)$, and $\gamma(t)$. One can regard as "optimal" a choice of coefficients in $U_1(x,t)$ such that both the "center of gravity" and the width of the Gaussian packet coincide at all times with the "center of gravity" and the width of the true solution. In this case it becomes possible (see Sec. 1) to express, with suitable approximation, $\alpha(t), \beta(t)$, and $\gamma(t)$ in terms of $U(x,t), A(t)$, and $\bar{x}(t)$.

In addition to what was done in Ref. 8, i.e., excitation in specified external fields, we have demonstrated[3)] how, by using the obtained equations, it is possible to choose optimal, generally speaking nonmonochromatic exciting signals (see, e.g., Refs. 12 and 13). The frequency of the external signal varies in this case in such a way that its phase coincides at each instant of time with the phase of the wave-packet velocity. We have succeeded, in the numerical experiment, to excite within short times a nonlinear system to energies at which the anharmonicity has already a substantial effect on the spectrum.

The indicated transition can be directly generalized to a multidimensional problem. In this case U_1 must be represented in the form

$$\alpha(t)+\sum_i \beta_i(t)x_i+\sum_{ij}\gamma_{ij}(t)x_ix_j.$$

On the other hand, the transition from multidimensional problems to field problems is less trivial: the field equations must be renormalized. What is remarkable here is that the renormalization can be effected in the equations in general form, without going over to concrete problems.

1. ONE-DIMENSIONAL NONLINEAR OSCILLATOR

Let the Hamiltonian of the system in question be

$$\hat{H}=\hat{p}^2/2M+U(x,t), \tag{1}$$

where $U(x,t)$ is a potential with weak anharmonicity $\lambda\Phi(x)$:

$$U(x,t)=\alpha_0+\beta_0 x+{}^1/_2 M\omega_0^2x^2+\lambda\Phi(x). \tag{2}$$

We represent the exact solution of the Schrödinger equation with Hamiltonian (1) in the form

$$\Psi_{ex}(x,t)=\Psi_{ap}(x,t)+\varphi(x,t), \tag{3}$$

where

$$\Psi_{ap}(x,t)=\exp\{-A(t)y^2+iC(t)y+B(t)\}, \tag{4}$$
$$y=x-\bar{x}(t),\quad A(t)=A_1(t)+iA_2(t),\quad B(t)=B_1(t)+iB_2(t),$$

$A_1(t), A_2(t), B_1(t), B_2(t), \bar{x}(t), C(t)$ are real functions of the time. We stipulate furthermore that the parameters of the function $\Psi_{ap}(x,t)$ satisfy the conditions

$$\bar{x}(t)=\int x|\Psi_{ex}|^2dy,\quad Y(t)=\frac{1}{4A_1(t)}=\int(x-\bar{x}(t))^2|\Psi_{ex}|^2dy. \tag{5}$$

Let, in addition, $\Psi_{ap}(x,t)$ be a solution of a Schrödinger equation with a real potential. It is easy to show that the following relations are then valid:

$$C(t)=\frac{M\dot{\bar{x}}(t)}{\hbar},\quad \dot{A}_1=\frac{4\hbar}{M}A_1A_2,\quad B_1=\frac{1}{4}\ln\frac{2A_1}{\pi}; \tag{6}$$

$$\int(|\varphi|^2+\Psi_{ap}\varphi^*+\varphi\Psi_{ap}^*)dy=0,$$
$$\int(|\varphi|^2+\Psi_{ap}\varphi^*+\varphi\Psi_{ap}^*)y\,dy=0,\quad \int(|\varphi|^2+\Psi_{ap}\varphi^*+\varphi\Psi_{ap}^*)y^2dy=0. \tag{7}$$

The function $\Psi_{ap}(x,t)$ yields the first two moments of the exact solution $\Psi_{ex}(x,t)$. It is easy to show that the quantity $\langle Y+\bar{x}^2(t)\rangle$ averaged over the fast time oscillations yields, accurate to terms proportional to λ, the energy of the investigated system, namely

$$E(t)\approx M\omega_0^2\langle Y+\bar{x}^2(t)\rangle.$$

Differentiating (5) twice with respect to time and using the Ehrenfest equations, we obtain

$$\int\left(\varphi^*\frac{\partial\Psi_{ap}}{\partial y}-\varphi\frac{\partial\Psi_{ap}^*}{\partial y}\right)dy=-\int\varphi^*\frac{\partial\varphi}{\partial y}dy,$$
$$\int y\left[\varphi^*\frac{\partial\Psi_{ap}}{\partial y}-\varphi\frac{\partial\Psi_{ap}^*}{\partial y}\right]dy=-\frac{1}{2}\int y\left[\varphi^*\frac{\partial\varphi}{\partial y}-\varphi\frac{\partial\varphi^*}{\partial y}\right]dy, \tag{8}$$
$$\int[\varphi^*\Psi_{ap}-\varphi\Psi_{ap}^*]dy=0$$

(The last equality ensures a minimum deviation of the phase of $B_2(t)$ from the phase of the exact solution $\Psi_{ex}(x,t)$,

$$M\ddot{\bar{x}}(t)=-\frac{1}{M}\int|\Psi_{ap}|^2\frac{\partial U}{\partial y}dy-\int\frac{\partial U}{\partial y}(\varphi\Psi_{ap}^*+\varphi^*\Psi_{ap}+|\varphi|^2)dy,$$
$$\dot{Y}=-\frac{2}{M}\int y\frac{\partial U}{\partial y}|\Psi_{ap}|^2dy+\frac{\dot{Y}^2}{2Y}+\frac{\hbar^2}{2M^2Y}$$
$$-\frac{2\hbar^2}{M^2}\int y\frac{\partial U}{\partial y}[\varphi^*\Psi_{ap}+\varphi\Psi_{ap}^*+|\varphi|^2]dy+\frac{2\hbar^2}{M^2}I. \tag{9}$$
$$I=\left(\frac{M^2}{\hbar^2}\dot{\bar{x}}^2-\frac{1}{2Y}\right)\int|\varphi|^2dy+\left[\frac{1}{4Y^2}+\frac{M^2}{4\hbar^2}\frac{\dot{Y}^2}{Y^2}\right]\int y^2|\varphi|^2dy$$
$$-\frac{M^2\dot{\bar{x}}\dot{Y}}{\hbar^2Y}\int y|\varphi|^2dy+\int\varphi^*\frac{\partial^2\varphi}{\partial y^2}dy+\frac{2M\dot{\bar{x}}i}{\hbar}\int\varphi^*\frac{\partial\varphi}{\partial y}dy$$
$$+\frac{iM\dot{Y}}{2\hbar Y}\int y\left[\varphi^*\frac{\partial\varphi}{\partial y}-\varphi\frac{\partial\varphi^*}{\partial y}\right]dy.$$

For the correction $\varphi(y,t)=\chi(y,t)\Psi_{ap}(y,t)$ we have the equation

$$i\hbar\dot{\chi}=-\frac{\hbar^2}{2M}\chi''+\frac{2\hbar^2A}{M}y\chi'+\Delta U(1+\chi), \tag{10}$$

where

$$\Delta U=U(y,t)-\alpha-\beta y-\gamma y^2,$$
$$\alpha=-\hbar\dot{B}_2+M\dot{\bar{x}}^2/2-\hbar^2/4MY,\quad \beta=-M\ddot{\bar{x}}, \tag{11}$$
$$\gamma=M[\hbar^2/2M^2Y+\dot{Y}^2/2Y-\ddot{Y}]/(4Y).$$

Considering only those exact solutions of the Schrödinger equation which can be represented at the initial instant in the form of a Gaussian function, and recognizing that the contribution from $\varphi(x,t)$ to (9) is small in terms of λ^2, we obtain a system of equations that is closed relative to $\bar{x}(t)$ and $Y(t)$:

$$M\ddot{\bar{x}}=-\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\frac{\partial^{2n-1}U}{\partial x^{2n-1}}\Big|_{x=\bar{x}(t)}Y^{n-1},$$
$$M\ddot{Y}=\frac{M}{2}\frac{\dot{Y}^2}{Y}+\frac{\hbar^2}{2MY}-2\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\frac{\partial^{2n}U}{\partial x^{2n}}\Big|_{x=\bar{x}(t)}Y^n. \tag{12}$$

It can be shown, using relations (7) and (8), that Eq. (12) in first order in λ yields the minimum of the norm of the increment $\varphi(y,t)$. In the case of a linear oscillator the system (12) yields exact solutions for the Hamiltonian $\hat{H}$ in the form of Gaussian packets. On the other hand, if the potential $U(x,t)$ is anharmonic, the nonlinear quantum corrections to the packet motion are automatically taken into account in (12). It is important that (12) yields not a quasiclassical but a quantum description of the evolution of the system, with a single limitation: the deviation of the exact solution from the approximate one must be small.

We now indicate a criterion for the applicability of (12). In first order we obtain the following equation for χ:

$$i\hbar\dot{\chi}=-\frac{\hbar^2}{2M}\chi''+\frac{\hbar^2}{2M}y\left(\frac{1}{Y}-\frac{iM\dot{Y}}{Y}\right)\chi'+\Delta U(y,t).$$

Let $\Delta U(y,t)$ be a polynomial in y of degree N. Then, taking (11) and (12) into account we have, choosing $\dot{B}_2(t)$ in suitable fashion,

$$\Delta U=\sum_{n=3}^{N}Z_n(t)H_n\left(\frac{y}{(2Y)^{1/2}}\right),$$

where $H_n(y)$ is a Hermite polynomial. The coefficients $\chi_n(t)$ in the expression

$$\chi(y,t)=\sum_{n=3}^{N}\chi_n(t)H_n\left(\frac{y}{(2Y)^{1/2}}\right)\exp\left\{-\frac{n\hbar i}{2M}\int_0^t\frac{d\tau}{Y}\right\}$$

then take the form

$$\chi_n(t)=-\frac{1}{i\hbar}\int_0^t Z_n(\tau)\exp\left\{\frac{n\hbar i}{2M}\int_0^\tau\frac{d\tau_1}{Y}\right\}d\tau. \tag{13}$$

Solving (12), we obtain the dependence of $\bar{x}(t)$ and $Y(t)$ on the time. We can then easily determine $\chi(y,t)$ from (13). On the other hand, the system (12) can be easily solved for a concrete form of the potential $U(x,t)$ with a computer (see Sec. 3).

The criterion for the applicability of the proposed description is satisfaction of the inequality

$$\int_{-\infty}^{+\infty}|\chi|^2|\Psi_n|^2\,dx\ll 1. \tag{14}$$

2. EQUATIONS IN SLOW VARIABLES

For a linear oscillator, Eqs. (12) take the form

$$M\ddot{\bar{x}}=-\beta_0(t)-M\omega_0^2\bar{x},\quad \dot{Y}=\tfrac{1}{2}\dot{Y}^2/Y+\hbar^2/2M^2Y-2\omega_0^2Y. \tag{15}$$

These equations can be solved exactly. In particular,

$$Y=(p^2+p_0^2)^{1/2}+p\sin\left\{2\omega_0 t+\arcsin\frac{Y(0)-(p^2+p_0^2)^{1/2}}{p}\right\}, \tag{16}$$

where

$$p_0=\frac{\hbar}{2M\omega_0},\quad (p^2+p_0^2)^{1/2}=\left(\frac{M\dot{Y}^2(0)}{2Y(0)}+2M\omega_0^2Y(0)+\frac{\hbar^2}{2MY(0)}\right)\frac{1}{4M\omega_0^2};$$

at $p=0$ we obtain the well known coherent states.

For a nonlinear oscillator, we represent $\bar{x}(t)$ and $Y(t)$ in the form

$$\begin{aligned}\bar{x}&=a(t)\cos(\omega_1 t+\theta(t))+\lambda u_1+\dots,\\ Y(t)&=q(t)+p(t)\cos(2\omega_1 t+\eta(t))+\lambda p_1+\dots.\end{aligned} \tag{17}$$

For concreteness, we consider anharmonicity of the form

$$\lambda\Phi(x)=\tfrac{1}{4}\lambda x^4.$$

By the averaging method[14] we obtain for the slow variables $\eta(t)$, $\theta(t)$, $a(t)$, $q(t)$, and $p(t)$, in first-order approximation,

$$\begin{aligned}\dot{a}&=\frac{f_0}{2M\omega_0}\sin(\delta-\theta)+\frac{3\lambda ap}{4M\omega_0}\sin(2\theta-\eta),\\ \dot{p}&=-\frac{3}{4}\frac{\lambda a^2q}{M\omega_0}\sin(2\theta-\eta),\\ \dot{\theta}&=-\frac{f_0\cos(\delta-\theta)}{2M\omega_0 a}+\frac{3\lambda a^2}{8M\omega_0}+\frac{3\lambda q}{2M\omega_0}+\frac{3\lambda p}{4M\omega_0}\cos(2\theta-\eta)+\omega_0-\omega_1,\\ \dot{\eta}&=\frac{3}{2}\frac{\lambda a^2}{M\omega_0}+\frac{9\lambda q}{2M\omega_0}+\frac{3}{4}\frac{\lambda a^2q}{M\omega_0 p}\cos(2\theta-\eta)+2(\omega_0-\omega_1),\\ q&=(p^2+p_0^2)^{1/2},\end{aligned} \tag{18}$$

where

$$p_0=\hbar/2M\omega_0,\quad \beta_0(t)=-f_0\cos(\omega_1 t+\delta)=f(t).$$

The system (18) is easier to investigate than (12).

3. RESULTS OF COMPUTER CALCULATIONS

The systems of ordinary differential equations (12) and (18), obtained in the preceding sections can be used to formulate a numerical experiment on the excitation of oscillations of a molecule by laser radiation. It must be borne in mind that for polyatomic molecules at high excitation energies, where the multimode character is already significant, Eqs. (12) and (18) no longer work. During the initial stages, however, so long as only one mode is excited and so long as the criterion (14) is valid, it is possible to use these equations to describe the excitation of the molecule.

Using (12) and (18), we can choose an optimal modulation of the laser radiation, such that the effective build-up of the oscillations of the molecule turns out to be possible at much lower powers than at present. One of the methods of such a choice is the following. We substitute $f(t)$ in (18) in the form $f(t)=-f_0\sin(\omega_0 t+\theta)$. The external field at any instant of time performs then positive work on the molecule, inasmuch as in this case the force has the same phase as the volocity. Determining

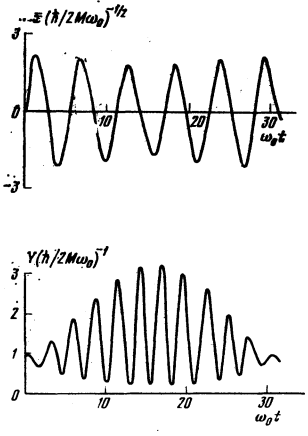

FIG. 1. Free motion of oscillator. Solution of Eqs. (12). Fast variables, $\lambda_0=0.03$.

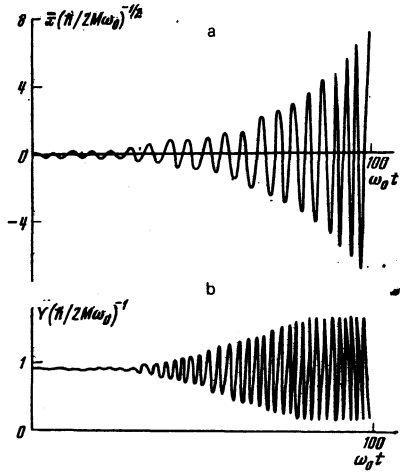

FIG. 2. Excitation of oscillator by a driving force. Solution of Eqs. (12). Fast variables, $f = 0.1\dot{X}$, $\lambda_0 = 0.1$.

in self-consistent fashion $f(t)$ in the course of the solution (18), we obtain the explicit form of the exciting signal.

For the concrete form of the potential $U(x, t)$

$$U = M\omega_0^2 x^2/2 + \lambda x^4/4 - fx \tag{19}$$

we present examples of the solutions of Eqs. (12) and (18), obtained with the aid of PDP and WANG computers. The following cases were considered:

1. Free motion of the system ($f = 0$, see Fig. 1).

2. Evolution of the system under the influence of an external force $f(t) = \alpha\bar{x}(t)$, $\alpha > 0$ [we solved Eqs. (12), see Fig. 2].

3. Evolution of the system under the influence of the external force

$$f(t) = -f_0 \sin(\omega_0 t + \theta(t)), \quad f_0 > 0$$

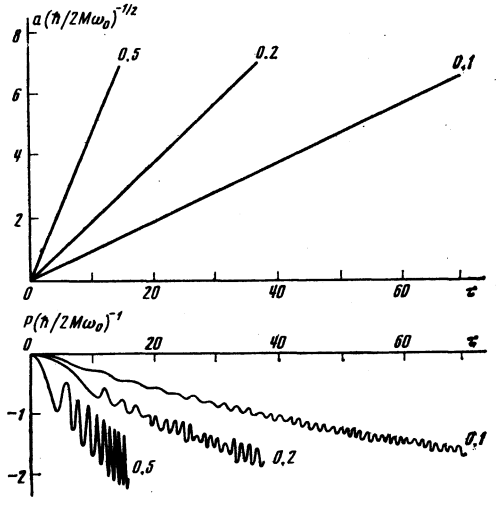

FIG. 3. Excitation by a driving force $f = -f_0 \sin(\omega_0 t + \theta)$; $f_{00} = 0.1$; 0.2; 0.5. Solution of Eqs. (18). Slow variables. The dimensionless quantities are here

$$f_{00} = \frac{f_0}{6\lambda}\left(\frac{2M\omega_0}{\hbar}\right)^{3/2}, \quad \lambda_0 = \frac{\lambda\hbar}{2M^2\omega_0^3}, \quad \tau = 3\lambda_0\omega_0 t\,.$$

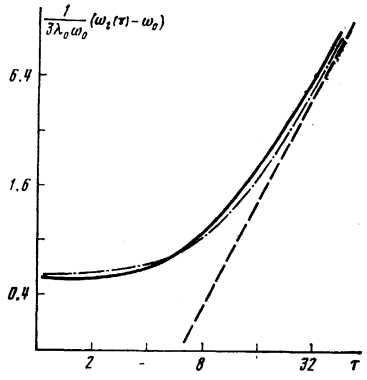

FIG. 4. Dependence of the frequency of the induced force on the time. The same quantities are used as in Fig. 3; $f_{00} = 0.02$.

at $\bar{x}(t) \approx -a\omega_0 \sin(\omega_0 t + \theta(t))$ [we solved Eqs. (18), see Fig. 3].

The results of the solution of the equations are shown in the figures. As seen from Figs. 2 and 3, the exciting signal causes rather fast excitation of the system. Figure 4 shows a plot of $\omega_f(t)$ at optimal excitation (solid line). For comparison, the same figure shows a plot of $\omega_f(t)$ at optimal classical excitation (dashed line). In addition, also for comparison, the figure shows a plot of $\omega_f(t)$ obtained neglecting the change of the width of the packet (dash–dot).

Choosing the anharmonicity constant for a concrete molecule and using the function plotted in Fig. 4, we can determine the required temporal variation of the laser frequency in the optimal regime of excitation of this molecule. The intensity of the laser radiation field is then

$$E = 1.2\frac{\lambda}{e}\left(\frac{\hbar}{2M\omega_0}\right)^{3/2}, \quad f_0 = eE. \tag{20}$$

As already mentioned in the Introduction, the method considered here can be generalized also to multidimensional problems (see Sec. 5). This makes it possible in principle to organize a numerical experiment also when a large number of modes is excited. The philosophy behind the search for the optimal exciting signal remains the same as before.

4. THE EFFECTIVE HAMILTONIAN

Equations (12) can be obtained also in another way. Namely, we can obtain the previously derived equations by averaging the Hamiltonian (1) over a pseudo-coherent state and introducing the necessary canonical variables. In fact, after integration and after simple transformations we obtain

$$H_{\text{eff}} = \frac{\hbar^2}{8q} + \frac{2p^2 q}{M} + \frac{P^2}{2M} + U(\bar{x}, t) + \sum_{n=1}^{\infty} \frac{1}{2n!!}\frac{\partial^{2n} U}{\partial x^{2n}} q^n. \tag{21}$$

The canonical variables were chosen in the following manner:

$$x = \bar{x}(t), \quad P = M\dot{\bar{x}}(t), \quad q = Y, \quad p = -\hbar A_2. \tag{22}$$

(The one-dimensional quantum problem goes over in this case into a two-dimensional classical problem). Taking the variational derivatives of H_{eff} with respect to the canonical variables and using the Hamilton equations, we then obtain exactly Eqs. (12). It is thus clear

that by a purely "classical" method we can obtain from (21) the equations (12) for the pseudo-coherent states, by using the Hamilton equations.

5. SYSTEM OF SEVERAL PARTICLES WITH NONLINEAR INTERACTION

Let the Hamiltonian of the system in question be of the form

$$\hat{H}=-\frac{\hbar^2}{2}\sum_{k=1}^{N_0}\frac{1}{M_k}\frac{\partial^2}{\partial x_k^2}+U(x_1,\ldots,x_{N_0};t).$$

We stipulate beforehand that we shall consider for simplicity a system consisting of unlike particles. The wave function of the pseudo-coherent state is of the form

$$\Psi_{ap}=\exp\{-A_{nm}y_ny_m+i\hbar^{-1}M_n\dot{\bar{x}}_ny_n+B\},$$
$$B_1=-\tfrac{1}{4}\ln(2\pi)^{N_0}\det[\lambda_{nm}],$$
$$\lambda_{nm}=\tfrac{1}{4}A^{-1}_{nm(1)};\quad A_{nm}=A_{mn}=A_{mn(1)}+iA_{mn(2)};\quad \bar{x}_n=\langle x_n\rangle_x, \tag{23}$$
$$A_{nm(1)}=2\hbar^2\sum_{k=1}^{N_0}\left[A_{nk(1)}\frac{1}{M_k}A_{km(2)}+A_{nk(2)}\frac{1}{M_k}A_{km(1)}\right].$$

Generalizing (21) to the multiparticle case, we obtain

$$H_{\text{eff}}=\langle\hat{H}\rangle_x=\sum_{k=1}^{N_0}\frac{P_k^2}{2M_k}+U(x_1,\ldots,x_{N_0};t)+\frac{\hbar^2}{8}\sum_{k=1}^{N_0}\frac{q_{kk}^{-1}}{M_k}$$
$$+2\sum_{k=1}^{N_0}\frac{p_{km}p_{kn}q_{nm}}{M_k}+\sum_{i_1,i_2=1}^{N_0}\sum\frac{1}{2}\frac{\partial^2U}{\partial x_{i_1}\partial x_{i_2}}q_{i_1i_2}$$
$$+\sum_{n=2}^{\infty}\frac{1}{2n!!}\frac{\partial^{2n}U}{\partial x_{i_1}\ldots\partial x_{i_{2n}}}q_{i_1i_2}\ldots q_{i_{2n-1}i_{2n}}, \tag{24}$$

where

$$q_{nm}=\lambda_{nm},\quad p_{nm}=-\hbar A_{nm(2)},\quad x_n=\bar{x}_n,\quad P_n=M\dot{\bar{x}}_n. \tag{25}$$

Varying (24) with respect to the canonical variables (25), we obtain a system of equations for the pseudo-coherent multiparticle state

$$M\ddot{\bar{x}}_i=-\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\frac{\partial^{2n-1}U}{\partial x_i\partial x_{k_1}\ldots\partial x_{k_{2n-2}}}\lambda_{k_1k_2}\ldots\lambda_{k_{2n-3}k_{2n-2}},$$
$$\hbar\dot{A}_{ij(2)}=-2\hbar^2\sum_{k=1}^{N_0}A_{ik(1)}\frac{1}{M_k}A_{kj(1)}+2\hbar^2\sum_{k=1}^{N_0}A_{ik(2)}\frac{1}{M_k}A_{kj(2)}$$
$$+\frac{1}{2}\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\frac{\partial^{2n}U}{\partial x_i\partial x_j\partial x_{k_1}\ldots\partial x_{k_{2n-2}}}\lambda_{k_1k_2}\ldots\lambda_{k_{2n-3}k_{2n-2}}, \tag{26}$$
$$\hbar\dot{A}_{nl(2)}\frac{\lambda_{lm}}{M_m}+\hbar A_{nl(2)}\frac{\dot{\lambda}_{lm}}{M_n}=-\frac{1}{2}\frac{d}{dt}\lambda_{nm}.$$

In the derivation of (26) we must use the relation $d\lambda^{-1}_{kk}/d\lambda_{ij}=-\lambda^{-1}_{ki}\lambda^{-1}_{kj}$ and assume in the variation that λ_{ij} is independent of λ_{ji} and $A_{ij(2)}$ is independent of $A_{ji(2)}$. The solution of the system (26) yields the complete information on the time variation of the approximate solution of the Schrödinger equation—the multiparticle wave function (23).

The derivation of the criterion in the multidimensional case is quite cumbersome because of the tensor character of the transformations, but its idea does not differ in principle from that of the one-dimensional case.

The choice of the canonical variables (25) is governed principally by convenience considerations. In particular, from the theorems on the simultaneous diagonalization of two quadratic forms it follows (since λ_{ij} is a positive definite quadratic form) that there exists a transformation, generally speaking time-dependent, which yields a set of canonical variables out of $4N_0$ quantities.

If the explicit time dependence in H_{eff} is caused only by the external field, then that part of (24) which does not depend explicitly on the time yields the eigenenergy of the system in a definite pseudo-coherent state. This energy consists of three parts: 1) the energy of the "classical" motion (the first two terms), 2) the quantum "internal" energy in the quasicoherent state (third, fourth, and fifth terms of the sum), 3) the "interaction" energy of these "degrees of freedom," due to both the quantum and the nonlinear character of the system. For a harmonic oscillator, the "internal" and "classical" degrees of freedom are uncoupled. The pseudo-coherent state corresponding to the minimum of the "internal" energy, is a well known coherent state of a quadratic system.

We have considered above the averaging of the Hamiltonian over a pseudo-coherent state. We shall show now that for a Hermitian operator $\hat{Q}(\hat{p},\hat{x})$ whose anharmonicity parameters remain small with respect to $\hat{p}$ and $\hat{x}$, also satisfies accurate to second order in λ the relation

$$\langle O(p,x)\rangle_{ex}=\langle\hat{O}\rangle_{ap} \tag{27}$$

In fact

$$\int\Psi_{ex}^{*}\hat{O}\Psi_{ex}dx\approx\int\Psi_{ap}^{*}\hat{O}\Psi_{ap}dx+\int[\varphi^{*}\hat{O}\Psi_{ap}+\Psi_{ap}^{*}\hat{O}\varphi]dx.$$

The Hermitian operator $\hat{O}(\hat{p},\hat{x})$ which depends analytically on the variables p and x, can always be expanded, accurate to second-order terms inclusive:

$$\hat{O}(p,\hat{x})=O(M\dot{\bar{x}},\bar{x})+\left(\frac{\partial\hat{O}}{\partial p}\right)_{p=\bar{p}}\dot{y}+\left(\frac{\partial\hat{O}}{\partial x}\right)_{x=\bar{x}}y$$
$$+\left(\frac{\partial^2\hat{O}}{\partial p^2}\right)_{\hat{p}=\bar{p}}\dot{y}^2+\left(\frac{\partial^2\hat{O}}{\partial x^2}\right)_{x=\bar{x}}y^2+\left(\frac{\partial^2\hat{O}}{\partial p\partial x}\right)_{\hat{p}=\bar{p}\,x=\bar{x}}(y\dot{y}+\dot{y}y)+\ldots.$$

Using Eqs. (7) and (8), we can easily verify the validity of (27). The generalization to the multidimensional case is automatic.

We assume now that there exists an "ensemble" of pseudo-coherent states and that we are interested in the mean value of the operator $\hat{O}$ over this "ensemble." Obviously, this mean value can be expressed in terms of a sort of "distribution function" F, which determines the density of the probability that the canonical variables (22) take on definite values. In the case when the "ensemble" is so constructed that the pseudo-coherent states contained in it do not decay, we can write for F the equation

$$\frac{\partial F}{\partial t}+\frac{\partial F}{\partial x_i}\frac{\partial H_{\text{eff}}}{\partial p_i}-\frac{\partial F}{\partial p_i}\frac{\partial H_{\text{eff}}}{\partial x_i}+\frac{\partial F}{\partial q_{ij}}\frac{\partial H_{\text{eff}}}{\partial p_{ij}}-\frac{\partial F}{\partial p_{ij}}\frac{\partial H_{\text{eff}}}{\partial q_{ij}}=0.$$

6. NONLINEAR QUANTUM STRING

We extend the method to include a system with an infinite number of particles. Namely, using the results presented above, we obtain equations that describe the

pseudo-coherent states of a nonlinear quantum string in an external potential (see also Ref. 15). We are considering one-dimensional oscillations in a direction perpendicular to the string. The Hamiltonian $\hat{H}$ takes in this case the form

$$\hat{H}=\int dx_1\left[\frac{\rho}{2}\dot{u}^2(x_1)-F(x_1,t)u(x_1)+U\left(\frac{\partial u}{\partial x_1}\right)+V(u)\right], \tag{28}$$

where x_1 is the coordinate along the string, $u(x_1)$ is the transverse deviation of the string, $\rho\dot{u}^2/2$ is the density of the momentum at the point x_1, $F(x_1,t)$ is the density of the external forces, $U(\partial u/\partial x_1)$ is the density of the potential energy of the interaction in the string, which is an even function of $\partial u/\partial x_1$, $V(u)$ is the external potential, an even function of u, in which the string is placed, and ρ is the density of the string. The wave function in the pseudo-coherent state is given by

$$\Psi_{sp}=\exp\left\{B-\frac{i}{\hbar}\int\dot{X}(x,t)y(x)\rho dx - \int A(x,x',t)y(x)y(x')dxdx'\right\},\quad u=X+y. \tag{29}$$

Then H_{eff} constitutes the Hamiltonian (28) averaged over (29), and is in essence a continual integral:

$$H_{\text{eff}}=\int\Psi^*\hat{H}\Psi Dy.$$

We carry out the continual integration in standard fashion (see, e.g., Ref. 16), using the fact that the integrals are Gaussian. As a result we obtain

$$H_{\text{eff}}=\int dx_1\left\{-F(x_1,t)X(x_1,t)+\frac{\hbar^2A_1(x_1,x_1)}{2\rho}+\frac{1}{2}\rho\dot{X}^2(x_1,t)+\frac{2\hbar^2}{\rho}\iint A_2(x_2,x_1)A_2(x_1,x_3)\times\lambda(x_3,x_2)dx_3dx_2+\sum_{n=0}^{\infty}\frac{1}{2n!!}\frac{\partial^{2n}U(z)}{\partial z^{2n}}\Big|_{z=\partial X/\partial x_1}\times\left(\frac{\partial^2\lambda(x_2,x_3)}{\partial x_2\partial x_3}\right)_{x_2=x_3=x_1}+\sum_{n=0}^{\infty}\frac{1}{2n!!}\frac{\partial^{2n}V}{\partial X^{2n}}\lambda^n(x_1,x_1)\right\}, \tag{30}$$

where

$$A(x,x')=A_1(x,x')+iA_2(x,x'),$$

$$\lambda(x,x')=\int y(x)y(x')|\Psi_{sp}|^2Dy,$$

$$\int A_1(x,x'')\lambda(x'',x')dx''=\tfrac{1}{4}\delta(x-x').$$

Choosing the canonical variables in analogy with (25) and taking the variational derivatives of the effective Hamiltonian (30) with respect to these variables, we obtain a closed system of integro-differential equations, which give the parameters of the pseudo-coherent state of the quantum string. These equations contain delta functions as well as their derivatives, and call for regularization.

Assume that at the initial instant of time at infinity ($x_1=\pm\infty$) the string is in the ground state. It is clear that sufficiently remote point of the string will be in this case in the ground state at any finite instant of time, since all the perturbations in the string propagate with finite velocities. In addition, the perturbation of the initial conditions above the ground state should be a smooth function of the coordinates. We carry out the regularization in the equations, by making the substitution

$$\delta(x-x')\to\frac{1}{(\pi\alpha)^{1/2}d}\exp\left\{-\frac{(x-x')^2}{\alpha d^2}\right\}, \tag{31}$$

where α is a dimensionless parameter, d is a small distance, and from physical considerations it can coincide in order of magnitude with the distance between the particles in a real system simulated by the string. We can then show that the quantities $\lambda(x,x')$, $A_1(x,x')$, and $A_2(x,x')$ can be represented in the form

$$\begin{aligned}\lambda(x,x';t)&=\lambda^1(x-x';d)+\lambda^0(x,x';t),\\ A_1(x,x';t)&=A_1^1(x-x';d)+A_1^0(x,x';t),\\ A_2(x,x';t)&=A_2^0(x,x';t),\end{aligned} \tag{32}$$

where the functions λ_1 and A_1^1 can depend substantially on the cutoff parameter d and do not depend on the time, while λ^0, A_1^0, and A_2^0 are smooth functions of the variables, i.e., the characteristic scale of their variation is much larger than d, so that the dependence of these functions on the cutoff parameter can be neglected.

The "singular" parts λ^1 and A_1^1 are due entirely to the structure of the ground state of the string, and their contribution to the considered equations can be completely compensated by renormalization of the potentials U and V. Let the potentials U and V be represented in the form

$$U\left(\frac{\partial u}{\partial x},t\right)=\sum_{k=0}^{\infty}C_k\left(\frac{\partial u}{\partial x}\right)^{2k},\quad V(u,t)=\sum_{k=0}^{\infty}B_ku^{2k}.$$

Then the renormalization of the constant C_k and B_k in the form

$$\begin{aligned}C_k&\to\sum_{p=0}^{\infty}C_{k+p}\left(\frac{b}{2}\right)^p\frac{2(p+n)!}{2n!p!}=C_k^0,\\ B_k&\to\sum_{p=0}^{\infty}B_{k+p}\left(\frac{a}{2}\right)^p\frac{2(p+n)!}{2n!p!}=B_k^0,\end{aligned} \tag{33}$$

where

$$a=\lambda^1(0;d),\quad b=-\left[\frac{\partial^2\lambda^1(u;d)}{\partial u^2}\right]_{u=0}$$

(C_k^0 and B_k^0 are taken to be the observable quantities) eliminates completely the dependence of the equations on λ^1.

Changing over to the Fourier representation, we obtain

$$A_1^1(k)=\left[\frac{\rho}{2\hbar}(C_1^0k^2+B_1^0)\right]^{1/2}\exp\left\{-\frac{\alpha^2d^2k^2}{8}\right\}$$

$$\lambda^1(k)=\left[\frac{\rho}{2\hbar}(C_1^0k^2+B_1^0)\right]^{-1/2}\exp\left\{-\frac{\alpha^2d^2k^2}{8}\right\},$$

$$\lambda^1(x-x';0)=2\left(\frac{2\hbar}{\rho C_1^0}\right)^{1/2}K_0\left[\left(\frac{B_1^0}{C_1^0}\right)^{1/2}(x-x')\right].$$

Here $K_0(U)$ is a Bessel function of the second kind of imaginary argument. We finally obtain, putting $d=0$:

$$\int A_1^0(k,k'')\lambda^0(-k'',k')\,dk''+\left[\frac{\rho}{2\hbar}(C_1^0k^2+B_1^0)\right]^{1/2}\lambda^0(k,k')$$
$$+A_1^0(k,k')\left[\frac{\rho}{2\hbar}(C_1k^2+B_1^0)\right]^{-1/2}=0,$$
$$\dot\lambda^0(k,k')=-\frac{2\hbar}{\rho}\Big[\int A_2^0(k,k'')\lambda^0(-k'',k')\,dk''$$
$$+\int A_2^0(k',k'')\lambda^0(-k'',k')\,dk''\Big]+A_2^0(k,k')$$
$$\times\Big[\Big(\frac{\rho}{2\hbar}(C_1^0k^2+B_1^0)\Big)^{-1/2}+\Big(\frac{\rho}{2\hbar}(C_1^0k'^2+B_1^0)\Big)^{-1/2}\Big], \qquad (34)$$
$$A_2^0(k,k')=\frac{2\hbar}{\rho}\int A_2^0(k,k'')A_2^0(-k'',k')\,dk''$$
$$-\frac{2\hbar}{\rho}\int A_1^0(k,k'')A_1^0(-k'',k')\,dk''-\Big\{\Big[\frac{\rho}{2\hbar}(C_1^0k^2+B_1^0)\Big]^{1/2}$$
$$+\Big[\frac{\rho}{2\hbar}(C_1^0k'^2+B_1^0)\Big]^{1/2}\Big\}A_1^0(k,k')-\frac{1}{4}\zeta(k+k')(k+k')^2+\frac{1}{2}\eta(k+k');$$
$$\rho\ddot X=F(x,t)+\sum_{n=1}^{\infty}\frac{1}{[2n-2]!!}\frac{\partial}{\partial x}\Big[\Big(\frac{\partial^{2n-1}U_0(z)}{\partial z^{2n-1}}\Big)_{z=\partial X/\partial x}$$
$$\times\Big(\frac{\partial^2\lambda^0}{\partial x_1\,\partial x_2}\Big)^{n-1}_{x_1=x_2=x}\Big]-\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\frac{\partial^{2n-1}V_0}{\partial X^{2n-1}}(\lambda^0)^{n-1}(x,x), \qquad (35)$$

where

$$\zeta(k)=\int_{-\infty}^{+\infty}e^{ikx}\Big[\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\partial^{2n}U_0\Big(\frac{\partial X}{\partial x}\Big)$$
$$\times\Big(\frac{\partial^2\lambda^0}{\partial x_1\,\partial x_2}\Big)^{n-1}_{x_1=x_2=x}-C_1^0\Big]\,dx,$$
$$\eta(k)=\int_{-\infty}^{+\infty}e^{ikx}\Big[\sum_{n=1}^{\infty}\frac{1}{(2n-2)!!}\partial^{2n}V_0(X)(\lambda^0)^{n-1}(x,x)-B_1^0\Big]\,dx,$$

U_0 and V_0 are the renormalized potentials.

The system of integro-differential renormalized equations (34) and (35) provides us with complete information of the observable parameters of the pseudo-coherent state of a quantum nonlinear string in an external potential. We note that Eq. (35) goes over as $\hbar\to 0$ and correspondingly as $\lambda(x,x')\to 0$ into the usual classical equation for the vibrations of a nonlinear string. Equations (34) have no analog in the classical theory. To understand fully the physical meaning of the solutions described by these equations, we consider a situation wherein $F(x,t)\equiv 0$ and in addition $X(x,t)=\dot X(x,t)=0$. This condition means the absence of classical excitations. Equation (35) is then satisfied identically. However, as shown by the analysis of (34), perturbations $\lambda^0(x,x';t)$ can propagate in the string in this case. Physically this can be visualized in the following manner: if we alter at some point the width of the wave packet of the string particle, without displacing at the same time its center of gravity, and then remove the external action, then a wave will begin to propagate in the strong and the width of the wave packets will change in the wave without a change in the positions of their gravity centers. Since there are no classical displacements in such waves, we call them "null" waves.[17]

CONCLUSION

The article deals with the problem of exciting quantum nonlinear systems by an external, generally speaking, nonmonochromatic field. This problem is directly connected with the question of excitation of molecules by laser radiation. Both in the one-dimensional and in the multidimensional cases, closed systems of ordinary differential equations are obtained and permit the formulation of numerical experiments on molecule excitation. A method is indicated of searching for an optimal exciting signal, which has a time-dependent frequency and is capable, despite the anharmonicity, of "swaying" the system.

The approach proposed in the article for the investigation of multidimensional systems can be generalized also to include field problems. We have demonstrated the method of transition from multidimensional to field problems using as an example a quantum nonlinear string, for which a closed system of integro-differential renormalized equations was obtained, which go over as $\hbar\to 0$ into the ordinary equation for a nonlinear classical string.

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[1)] In view of the large number of papers, we refer only to those whose idea is close to that in this paper.

[2)] In Sec. 1 we show how to calculate a criterion that makes it possible to monitor the suitability of the proposed method in one concrete case or another. Actually, this criterion can be used to determine whether a pseudo-coherent state exists.

[3)] The possibility of "compensating" for the anharmonicity in excitation of molecules by changing the frequency of the exciting field was proposed in Ref. 1.

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Phasing of atomic velocities in the field of a traveling electromagnetic wave

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Resonant collisionless gas is considered in the field of a traveling monochromatic wave. The presence of a small parameter $\mu = \hbar k^2/m\gamma_\perp$ in the problem makes possible an asymptotic analysis of the equations that describe the action of the radiation pressure on the gas. It is shown that the selective character of the radiation force leads to a decrease of the random scatter of the atom velocities. The lower limit of the characteristic width $\delta v \sim (2\hbar\gamma/3m)^{1/2}$, of the nonequilibrium structure on the distribution function determined by the competition between the processes of phasing of the velocities and the diffusion spreading, is obtained. The possibility of eliminating the translational motion of the resonant ions in crossed optical and magnetic fields is noted.

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1. INTRODUCTION

Cooling of a gas in the field of a standing electromagnetic wave was predicted and analyzed in detail from various points of view in Refs. 1–9. In the present paper we show that in the field of a traveling electromagnetic wave it is also possible to eliminate in part the random motion of the atoms of a resonant gas and that a number of distinguishing features appear which are not encountered in the case of opposing waves. Observation of cooling in the field of a traveling electromagnetic wave is apparently simpler from the point of view of experimental realization.

When the action of resonant radiation pressure (RP) in the field of the plane traveling wave is considered, one must bear in mind the following aspects: the translational motion of the gas as a whole, the phasing of the atoms in velocity space, which leads to cooling, and diffusion in velocity space. The first of these factors is quite obvious. Induced absorption and emission of photons are characterized by a preferred direction, whereas scattered photons are on the average isotropically distributed.

Phasing in velocity space is due to the inhomogeneous character of the force of the spontaneous radiation pressure in the sense of its sharp dependence on the velocity, which manifests itself most strongly in weak fields ($|dE_0|/\hbar < \gamma$). The expression for the force in the case of exact tuning to resonance takes in this case the form

$$F \approx \hbar k \gamma \frac{1}{x^2+1} \frac{|dE_0|^2}{\hbar^2 \gamma_\perp^2}, \tag{1}$$

where $x = kv_x/\gamma_\perp$, v_x is the velocity of the atom in the direction of propagation of the radiation, k is the wave vector, $\gamma_\perp$ is the transverse relaxation rate, d is the dipole-moment matrix element, and E_0 is the amplitude of the field. It prevails for the resonant particles ($k|v_x| \lesssim \gamma_\perp$) and decreases rapidly with increasing Doppler shift.

We consider, at the instant of time t_0, two particles with mass m and somewhat different velocities $x_1, x_2 (x_1 - x_2 = \delta x(t_0))$ in the region $x > 0$. Then, obviously, at the instant of time t we have

$$\delta x(t) \approx \delta x(t_0) \exp\left(F'(x) m^{-1} (t - t_0)\right). \tag{2}$$

Since $F'(x) < 0 (x > 0)$, the distance between the chosen particles in velocity space decreases with time, and monochromatization in velocity (phasing of the velocities) takes place as a result. This can be easily understood with the aid of the following simple arguments. The atoms having a smaller velocity projection on the radiation-propagation direction and accordingly a smaller Doppler shift, move more rapidly in velocity space under the influence of the RP than the atoms having a larger velocity projection. Therefore the "slow" atoms ($x < 1$) gradually overtake the "fast" ones ($x > 1$), forming a narrow structure in the velocity distribution. This was first pointed out in Ref. 10, where the translational disequilibrium under the action of RP was in-

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