### The dynamics of wave packets of highly-excited states of atoms and molecules

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A review is presented of a new field in optical spectroscopy, in which the dynamics of wave packets composed of stationary states of atoms and molecules is studied. The time limits of applicability of the correspondence principle and wave packet collapse caused by nonlinear effects are considered. A new phenomenon is discussed, in which the wave packets are revived during their long-term post-classical evolution. Recent experiments on wave packet generation and detection in Rydberg atoms and molecules are reviewed. The experiments provide new opportunities for investigation of the transition region between classical and quantum physics of atomic particles. The relations between the phenomena discussed and the problem of generation of non-classical states of light in nonlinear optical systems is considered.

### **1. INTRODUCTION**

Is it possible to observe the motion of an electron in a Kepler orbit around the nucleus? That is, is it possible to obtain experimentally a Rutherford atom? Is it possible to observe oscillations of the nuclei in a molecule that are similar to classical oscillations of point particles bound by a quasielastic force? An affirmative answer to these questions, as is well known, is given by the correspondence principle of quantum mechanics.<sup>1</sup> The mathematical expression of the statement that the predictions of quantum theory must coincide with the predictions of classical mechanics in the region of validity of the latter is that for sufficiently high energies, wave packets composed mainly of short-wavelength components must move according to the laws of geometrical optics.<sup>2,3</sup> This means, specifically, that the position of the "center of gravity" of a spatially localized wave packet (the average values of the particle coordinates) must vary in time according to Newton's law

$$M\ddot{\mathbf{x}} = -\frac{\partial U(\bar{\mathbf{x}})}{\partial \bar{\mathbf{x}}}; \qquad (1)$$

Here M is the mass of the particle, x is the coordinate of the center of gravity of the wave packet moving in a potential U(x). It is well known that for Eq. 1 to be applicable the following condition must hold<sup>4</sup>

$$\left| \frac{\partial U(\bar{x})}{\partial \bar{x}} \right| \gg \frac{1}{2} \left| \frac{\partial^3 U(\bar{x})}{\partial \bar{x}^3} \right| \bar{\Delta} \bar{x}^2, \qquad (2)$$
$$\bar{\Delta} \bar{x}^2 = \bar{x}^2 - (\bar{x})^2.$$

The overbar means the average value of the corresponding quantity. Inequality (2) places a limit on the duration of the classical dynamics of the packet. With the passage of time the dispersion  $\overline{\Delta x^2}$  of the coordinate increases because of the spreading of the wave packet, and condition (2) is violated. In all these discussions a natural and important question arises: What are the laws governing the long-term "postclassical" evolution of wave packets beyond the bounds of the dynamics according to the correspondence principle? Until recently, experiments in the field of atomic and molecular spectroscopy have bypassed these fundamental questions. Even though during the early development of quantum mechanics, data of atomic and molecular spectroscopy formed the foundation of the transition from classical con-

cepts to quantum mechanical concepts, a historical curiosity, or, perhaps more appropriately, a certain lack of logic in the development of physical understanding was the fact that it was not possible with experiments existing at that time to create conditions under which one could observe effects at the boundary region between quantum and classical physics. The methods of ordinary optical spectroscopy generally involve excitation of individual stationary (or quasistationary) states in atoms or molecules. Such states describe objects that are quantum mechanical by nature. For example, even for arbitrarily large quantum numbers a single stationary state of an electron in a Coulomb field does not describe (even approximately) the motion of a localized particle in a Kepler orbit, just as for any quantum number a stationary wave function of a harmonic oscillator does not describe the harmonic oscillations of a localized particle. Only recently, with the use of ultrashort optical pulses, has it become possible to create coherent superpositions of many highly-excited stationary states of atoms and molecules-localized wave packets that are particle-like objects whose dynamics obey quasiclassical laws.

Thus, in spite of the fact that at the time of the development of quantum mechanics classical ideas were used as a heuristic basis for the understanding of quantum phenomena, only recently has it been possible to create and study such objects as a Rutherford atom, a classical molecule, and the like. Immediate progress in this field was attained in certain directions associated with particular goals. First among these was the perfection of the technique of working with highly-excited Rydberg atoms and the commencement of experiments in the excitation and multi-photon ionization of the Rydberg atoms by short and ultrashort laser pulses. Second has been progress in the so-called laser femtochemistry, in which one of the major tasks is the use of femtosecond optical pulses to study short-lived molecular complexes that appear at intermediate stages of chemical reactions. Both these topics have moved forward rapidly in the last few years, both experimentally and theoretically, demonstrating results that are valuable from a general physics point of view, and making it meaningful to speak of a new field of atomic and molecular physics, which involves the study of phenomena at the boundary between quantum and classical physics.

Turning now to the history of the subject, we should note that the first attempts to observe wave packets of sta-

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tionary states of quantum systems undergoing bounded motion in the classical limit go back to the work of Schrödinger.<sup>5,6</sup> He introduced the so-called coherent state of a harmonic oscillator, which has minimum indeterminacy, and formulated the problem of the analogous wave packet for the electron moving in a Kepler orbit in a hydrogen atom.<sup>6</sup> Efforts to generalize the concept of a coherent state to the Kepler problem have continued up to the present time.<sup>7-15</sup> The main complicating factor in the introduction of these states in atomic physics is the nonlinearity of the atomic "oscillator." The problem of the evolution of wave packets in nonlinear systems arose independently in various fields of quantum physics. Active development has occurred almost simultaneously, as mentioned above, in the theoretical and experimental investigation of packets of highly-excited states of Rydberg atoms,<sup>16-48</sup> the dynamics of packets of the vibrational states of molecules excited by pulsed laser radiation,<sup>49-86</sup> packets of quantum states of the electromagnetic field, generated in nonlinear-optics systems,87-94 and the evolution of wave packets in an "atom + field" system.95-97

This review is an attempt to consider from a unified point of view a variety of questions related to this new field of optical spectroscopy. The temporal limits within which the correspondence is applicable to the description of wave packets are discussed, as is the breakup of the wave packets due to nonlinear effects. The recently observed universal mechanism of the revival of wave packets during long-term evolution of highly-excited quantum systems is discussed. We analyze a number of recent experiments on the generation and detection of wave packets in Rydberg atoms and molecules. It is shown that there is an intimate connection between this set of phenomena and the problem of the generation of nonclassical states of light in nonlinear systems.

### 2. LONG-TERM EVOLUTION OF PACKETS OF STATIONARY STATES IN VERY SIMPLE QUANTUM SYSTEMS

### 2.1. Coherent and squeezed states—packets of stationary states of the harmonic oscillator

First we shall discuss some general properties of the evolution of packets of quantum systems with discrete spectra, undergoing bounded motion. The simplest system of this type is the harmonic oscillator. Let us consider a linear superposition (packet) of stationary states of the harmonic oscillator.

$$\psi(x,t) = \sum_{n=1}^{\infty} c_n u_n(x) \exp\left(-\frac{i}{\hbar} E_n t\right); \qquad (3)$$

where  $u_n(x)$  is the stationary wave function corresponding to the quantum number *n* and energy  $E_n = \hbar\omega(n + 1/2)$ , where  $\omega$  is the oscillator frequency,  $c_n$  are arbitrary constants, whose dependence on the quantum number *n* determines the localization of the packet in energy space. If we take  $c_n$  in the form

$$c_n = \exp\left(-\frac{1}{2} |\alpha|^2\right) \frac{\alpha^n}{(n!)^{1/2}},$$
 (4)

then the packet (3) is the so-called coherent state of the harmonic oscillator.<sup>3</sup> With the use of (4) the series (3) can be summed explicitly. For t = 0 and real *a* this summation gives the following result

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$$\psi(x,0) = \left(\frac{1}{\pi\sigma^2}\right)^{1/4} \exp\left[-\frac{1}{2\sigma^2} \cdot (x - \sqrt{2}\alpha\sigma)^2\right]; \quad (5)$$

here  $\sigma = (\hbar/M\omega)^{1/2}$  and M is the mass of the oscillator. The wave function  $\psi(x,0)$  is the eigenfunction of the ground state of the oscillator, displaced from the equilibrium point, where it is centered, to the position  $x_{max} = \sqrt{2} \ a\sigma$  without change of shape (the wave packet is Gaussian with a dispersion of the order of the amplitude of the zero-point oscillations of the oscillator). The evolution of this packet, according to the rules of quantum mechanics, is given by the formula

$$\psi(x,t) = \int dx' G(x,t; x',0) \psi(x,0) , \qquad (6)$$

where G(x,t;x't') is the Green's function of the harmonic oscillator:<sup>98</sup>

$$G(x,t; x',0) = \left(\frac{M\omega}{2\pi i\hbar \sin \omega t}\right)^{1/2} \\ \times \exp\left\{\frac{iM\omega}{2\hbar \sin \omega t} \left[ (x^2 + x'^2) \cos \omega t - 2xx' \right] \right\}.$$
(7)

Using Eqs. (5)-(7) one can easily derive the following expression for the probability density of the distribution of the oscillator coordinate<sup>99,100</sup>

$$|\psi(x,t)|^2 = \left(\frac{1}{\pi\sigma^2}\right)^{1/2} \exp\left[-\frac{1}{\sigma^2}(x-\bar{x}(t))^2\right].$$
 (8)

The average value  $\bar{x}(t)$  of the oscillator coordinate depends periodically on the time, with the period of the classical motion,  $T = 2\pi/\omega$ :

$$\bar{x}(t) = \int dx \psi^*(x,t) \ x \psi(x,t) = x_{\max} \cos \omega t \ . \tag{9}$$

Thus, the coherent state given by the sum (3) with coefficients (4) is a spatially localized Gaussian wave packet with a characteristic localization dimension  $\sigma$ . The center of gravity of this packet, according to Eq. (9), undergoes harmonic oscillations in accordance with the classical equation of motion. As a rule, the oscillations of a particle in a potential well become quasiclassical for large values of the displacement amplitude. We note, however, that expression (9) is exact for any *a*. This fact is a property of the harmonic oscillator, for which condition (2), necessary for classical motion of the center of gravity, is satisfied automatically  $(\partial^3 U(\bar{x})/\partial \bar{x}^3 = 0)$ . The indeterminacy of the coordinate and the momentum of the oscillator  $\Delta x = [x^2 - (\bar{x})^2]^{1/2}$ and  $\Delta p = [\bar{p}^2 - (\bar{p})^2]^{1/2}$  in the coherent state do not depend on the time and are, respectively,

$$\Delta x = \frac{\sigma}{\sqrt{2}} , \ \Delta p = \frac{\hbar}{\sqrt{2}\sigma},$$

where the product  $\Delta p \Delta x = \hbar/2$  is a minimum. This means that the packet (3) and (4) evolves without any spreading whatsoever over an arbitrarily long time. The condition for the system to be quasiclassical,  $a \ge 1$ , means obviously, that the region of spatial localization of the packet ( $\sim \sigma$ ) is much smaller than the size of the classical orbit  $L = 2^{3/2}a\sigma$ , which corresponds to the energy

$$E = M\omega^2/2 \cdot (L/2)^2 = \alpha^2 \hbar \omega.$$

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The largest contribution to the sum (3) comes from the terms with  $n \sim \overline{n} = \alpha^2 \ge 1$ , since the distribution of  $c_n$  has a near-Gaussian shape with a dispersion  $\Delta n \propto \alpha \propto (\overline{n})^{1/2} \ge 1$ . Thus, to achieve strong spatial localization of a packet, the number  $\Delta n$  of states that form the packet must be quite large. In the classical limit  $\Delta n \propto (\overline{n})^{1/2} \rightarrow \infty$ . We note that packets composed of a small number of states also exhibit nonclassical behavior for large values of  $\overline{n}$ .<sup>101</sup>

In recent years there has been vigorous study of the socalled "squeezed" states of light,<sup>102</sup> which are packets of stationary states of the oscillator of the quantized electromagnetic field. In the squeezed states, as opposed to the coherent states, the dispersion of the coordinate (and of the momentum) depends periodically on the time and takes on values that are less than the amplitude of the zero-point oscillations. This property is of interest from the point of view of reducing the quantum noise in measuring systems and information transmission systems. A squeezed state can be obtained by, for instance, choosing the coefficients  $c_n$  in formula (3) in the form<sup>103-105</sup>

$$c_n = \left(\frac{\varkappa^2 - 1}{\varkappa^2}\right)^{1/4} \frac{\varkappa^{-n/2}}{\sqrt{2^n n!}} H_n\left(\frac{\varkappa + 1}{\sqrt{2\varkappa}}\right) \exp\left(-\frac{\varkappa + 1}{2\varkappa}\alpha^2\right); (10)$$

where the  $H_n(z)$  are the Hermite polynomials. The parameter  $\varkappa$  characterizes the degree of "squeezing." At t = 0 the direct sum in (3), with coefficients (10) gives<sup>104</sup>

$$\psi(x,0) = \left(\frac{1}{\pi\sigma^2}\right)^{1/4} \left(\frac{\varkappa+1}{\varkappa-1}\right)^{1/4} \exp\left[-\frac{(x-\sqrt{2}\alpha\sigma)^2}{2\sigma^2\cdot(\varkappa-1)/(\varkappa+1)}\right].$$
(11)

The function  $\psi(x,0)$  is a Gaussian, obtained by the displacement of the ground state of the oscillator by a distance  $\sqrt{2}a\sigma$ with a dispersion that is compressed by a factor of

$$\left(\frac{\varkappa+1}{\varkappa-1}\right)^{1/2}$$

Thus, the indeterminacy in the coordinate of the squeezed state (11), unlike in the case of the coherent state, is less than the amplitude of the zero-point oscillations of the oscillator. Clearly, in the state (11)

$$x = \sqrt{2}\alpha\sigma, \ \Delta x = \frac{\sigma}{\sqrt{2}} \left(\frac{\kappa-1}{\kappa+1}\right)^{1/2}, \ \Delta p = \frac{\hbar}{\sqrt{2}\sigma} \left(\frac{\kappa+1}{\kappa-1}\right)^{1/2},$$

i.e., to conserve the minimum indeterminacy  $\Delta p \Delta x = \hbar/2$ , the squeezing of  $\Delta x$  entails a corresponding increase in the indeterminacy of the momentum. For  $\varkappa \to \infty$  the squeezed state goes over into a coherent state. The squeezing is maximum for  $\varkappa \to 1$ . The evolution of the squeezed state for t > 0 is given by formulas (6), (7), and (11). It is easy to find the probability density distribution of the oscillator coordinate in the squeezed state with the use of these formulas<sup>99,105</sup>

$$|\psi(x,t)|^{2} = \left(\frac{1}{\pi\sigma^{2}}\right)^{1/4} (\operatorname{ch} 2r_{0} - \operatorname{sh} 2r_{0} \cdot \cos 2\omega t)^{-1/2} \\ \times \exp\left[-\frac{(x - \bar{x}(t))^{2}}{\sigma^{2}(\operatorname{ch} 2r_{0} - \operatorname{sh} 2r_{0} \cdot \cos 2\omega t)}\right], \quad (12)$$
$$r_{0} = \frac{1}{2} \ln \frac{\varkappa + 1}{\varkappa - 1}.$$

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The squeezed state thus evolves while maintaining the shape of the Gaussian wave packet, whose parameters depend periodically on the time.

The center of gravity of this packet moves along a classical trajectory (9) as in the case of the coherent state, but unlike the latter, the indeterminacy in the coordinate and momentum vary periodically<sup>105</sup>

$$\Delta x = \frac{\sigma}{\sqrt{2}} \cdot (\operatorname{ch} 2r_0 - \operatorname{sh} 2r_0 \cdot \cos 2\omega t)^{1/2},$$
  

$$\Delta p = \frac{h}{\sqrt{2}\sigma} (\operatorname{ch} 2r_0 + \operatorname{sh} 2r_0 \cdot \cos 2\omega t)^{1/2},$$
(13)

going through maxima and minima out of phase. During the course of each period  $\Delta x$  and  $\Delta p$  take on values that are smaller than the corresponding values in the coherent state. This property of the indeterminacy of the coordinate and momentum in the squeezed state is, in essence, a macroscopic quantum effect, since it is manifested for any amplitude, no matter how large, of the oscillations of the center of gravity of the packet.

The quantity  $|c_n|^2$  is the distribution of the oscillator quanta in the packet of stationary states (3). In the coherent state this is the Poisson distribution (see expression (4)). In the squeezed state (Expression (10)) the distribution is narrower than the Poisson distribution (sub-Poisson photon statistics), and for strong squeezing  $(x \rightarrow 1)$  it becomes an oscillating function of the number *n* of photons. The properties of the distribution of the number of photons for light in a squeezed state have been analyzed in detail in Ref. 104.

# 2.2. Classical stages in the evolution—the breakup and the revival of packets of stationary states of an anharmonic oscillator

Let us now consider the bounded one-dimensional motion of highly excited wave packets in an anharmonic potential. In the region of energy E corresponding to large quantum numbers, the energy spectrum is quasi-equidistant, with the frequency spacing  $\omega_{n+1,n} = (1/\hbar)(E_{n+1} - E_n)$  between adjacent energy levels determined by the reciprocal  $T_{\rm cl}$  of the period of classical motion

$$\omega_{n+1,n} \approx \omega_{\rm cl}(E) = \frac{2\pi}{T_{\rm cl}(E)} \,. \tag{14}$$

For times much shorter than the period of classical oscillations,  $T_{\rm cl}$ , the discreteness of the spectrum is not important and the initial spatially localized packet moves over a classical trajectory, in general spreading out in a manner similar to the spreading of a squeezed state (12). However, this broadening is not irreversible as it is in the case of free or infinite motion, and the packet almost completely recovers its shape after the period  $T_{\rm cl}$  because of quasi-equidistant nature of the energy spectrum of the states of which it is composed. This correspondence between the quantum and classical description is not preserved forever, however, as in the case of the harmonic oscillator. In the long-term evolution of the packet, because of the dependence of  $\omega_{\rm cl}$  on the energy

$$\rho_{n+1,n} - \omega_{n,n-1} \approx \hbar \omega_{cl} \frac{\partial \omega_{cl}}{\partial E}.$$
(15)

there will inevitably be some unequal spacings in the levels at high excitations. The dephasing contributions of the various stationary states in the superposition of the form (3) results

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in the decay of the packet after many classical periods, and thus a limit is imposed on the duration of the "classical" evolution of the packet:

$$t \ll T_{\rm cl} \left( \hbar \left| \frac{\partial \omega_{\rm cl}}{\partial E} \right| \right)^{-1}.$$
 (16)

A natural question arises regarding the laws of the evolution of a packet in the long-term post-classical stages. A very general answer to this question is that this dephasing is not totally irreversible, and in time scales much longer than  $T_{\rm cl}$ the packet almost exactly recovers its shape and again evolves for a time according to the classical laws. The necessity of this sort of behavior of wave packets in quantum systems with an arbitrary discrete spectrum can be proved in general by the following reasoning.<sup>106</sup> We shall demonstrate that for a quantum system with a discrete spectrum a wave packet of the form (3) is an almost periodic function of time. In other words, we shall show that for any arbitrarily small  $\varepsilon > 0$  there exists a sufficiently dense set of times  $\{\tau_{\varepsilon}\}$  such that for each  $\tau_{\varepsilon}$  of this set

$$\int dx |\psi(x,t+\tau_{\varepsilon}) - \psi(x,t)|^2 < \varepsilon$$
(17)

for all t. Actually, using the orthonormality of the stationary states  $u_n(x)$  and the fact that the wave packet as a whole is normalized,

$$\sum_{n=1}^{\infty} |c_n|^2 = 1 , \qquad (18)$$

it is easy to obtain

$$\int dx |\psi(x,t+\tau) - \psi(x,t)|^2$$
$$= 2\sum_{n=1}^{\infty} |c_n|^2 \left[1 - \cos\left(\frac{1}{\hbar}E_n\tau\right)\right].$$
(19)

It follows from (18) that for arbitrarily small  $\varepsilon > 0$  one can find a value  $n(\varepsilon) = n_0$  such that

$$\sum_{n=n_0+1}^{\infty} |c_n|^2 < \frac{\varepsilon}{8} \, .$$

Consequently,

$$\sum_{n=n_0+1}^{\infty} |c_n|^2 \left[1 - \cos\left(\frac{1}{\hbar} E_n \tau\right)\right] \le 2 \sum_{n=n_0+1}^{\infty} |c_n|^2 < \frac{\varepsilon}{4}.$$
(20)

For the remaining finite sum of periodic functions, it has been correctly pointed out<sup>107</sup> that for any  $\varepsilon > 0$  there exists a sufficiently dense set  $\{\tau_{\varepsilon}\}$  such that

$$\sum_{n=1}^{n_0} |c_n|^2 \left[ 1 - \cos\left(\frac{1}{h} E_n \tau_{\varepsilon}\right) \right] < \frac{\varepsilon}{4} .$$
(21)

Inequalities (20) and (21), together with Eq. (19) prove that inequality (17) is correct.

The absence of true irreversibility in systems with a discrete spectrum leads eventually to an arbitrarily exact restoration of the initial packet. This discussion does not, however, have anything to say about the structure of the wave function in the intermediate stages. The surprising fact is, as we have pointed out recently,<sup>31,32</sup> that for a broad class of physical systems the long-term evolution of quasiclassical packets proceeds (after a stage of dynamics in accordance

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with the correspondence principle) according to a universal scenario that does not depend on the shape of the packet or on the specific physical nature of the object. In this scenario, the wave function of the system, besides the almost complete "revival" of the initial packet, undergoes a deterministic sequence of changes corresponding to the formation of regularly bounded structures made up of condensations of probability density with a high degree of localization. The shape of each of these condensations is determined uniquely by the shape of the initial wave packet. The regular structures that are formed are purely quantum objects, having no classical analogs even at very high energies and macroscopic dimensions. In this sense one can speak of a new macroscopic quantum mechanical effect.

Let us consider this topic in more detail. We shall reconsider a wave packet of the form (3), consisting of highly excited discrete states of a quantum system undergoing bounded motion in the energy region  $E \approx E_{\bar{n}} (\bar{n} \ge 1)$ , in which classical dynamics corresponds to regular periodic motion. We shall assume that at time t = 0 the packet is strongly localized in space (its spatial dimension  $\Delta x$  is much less than the characteristic dimension L of the classical orbit corresponding to  $E \approx E_{\bar{n}}$ ). It follows from the uncertainty principle that the energy width of the packet,  $\Delta E$  is of the order of

$$\Delta E \propto v \Delta p \propto \hbar \omega_{\rm cl} \frac{L}{\Delta x} ,$$

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where v and  $\Delta p$  are the characteristic values of the velocity and the uncertainty in the momentum. This means that the distribution of  $|c_n|^2$ , which has a sharp maximum for  $n \approx \bar{n}$ has the width

$$\Delta n \propto \frac{\Delta E}{\hbar \omega_{\rm cl}} \propto \frac{L}{\Delta x} \gg 1$$
.

For the coherent state of the harmonic oscillator,  $L \propto (E_{\bar{n}}/M\omega^2)^{1/2}$  and  $\Delta x \sim (\hbar/M\omega)^{1/2}$ , which gives us the estimate  $\Delta n \propto (\bar{n})^{1/2}$ , which coincides, of course, with the value obtained above with the use of the explicit expression for the wave function of the coherent state. In this way we have again verified that a strongly localized, particle-like packet must consist of a large number of stationary states: the larger the number, the greater the degree of localization of the packet.

If we take into account the fact that the energy levels of the quantum system are slightly nonequidistant at high values of  $\bar{n}$ , we can write the energy in the region  $E \propto E_{\bar{n}}$  in the form

$$E \approx E_n + 2\pi \hbar \frac{(n-\bar{n})}{T_{cl}} \pm 2\pi \hbar \frac{(n-\bar{n})^2}{T_{rev}} + \dots,$$
 (22)

where  $\pm$  refers to the sign of  $\partial \omega_{cl} / \partial E$  and

$$T_{\rm rev} = 2T_{\rm cl} \left( \hbar \left| \frac{\partial \omega_{\rm cl}}{\partial E} \right| \right)^{-1} \,. \tag{23}$$

Here, in addition to the classical period  $T_{\rm cl}$ , another, purely quantum mechanical time scale  $T_{\rm rev} \gg T_{\rm cl}$  appears, which plays a very important role in the long-term evolution of wave packets. For

$$t/T_{\rm cl} \ll \frac{1}{\hbar^2} \left| \frac{\partial}{\partial E} \left( \omega_{\rm cl} \frac{\partial \omega_{\rm cl}}{\partial E} \right) \right|^{-1}$$

we can restrict ourselves to the terms in the expansion given in (22) and write the wave function of the packet as

$$\psi(x,t) \approx \sum_{k} c_{k} u_{k}(x) \cdot \exp\left[-2\pi i (kt/T_{cl} + k^{2}t/T_{rev})\right], \quad (24)$$

$$k = n - \bar{n},$$

Here and below the energy is reckoned from  $E_n$ , and for the sake of definiteness it is assumed that  $\partial \omega_{\rm cl} / \partial E > 0$ . At times  $T_{\rm cl} \leq t \ll T_{\rm rev}$  the terms proportional to  $k^2$  in the exponent in expression (24) can be discarded and the time scale  $T_{\rm rev}$  will not be manifested. Thus,  $\psi(x,t+T_{\rm cl}) = \psi(x,t)$ . As a consequence, after a time equal to the classical period of oscillation,  $T_{\rm cl}$ , the packet, having undergone motion along a classical trajectory, returns to its earlier position in an unchanged form. Essentially, this means that the packet evolves according to the correspondence principle. At later times the dephasing of the individual terms in the superposition (24) begins to play a role because of the terms quadratic in k. It is easy to show that this dephasing for a packet composed of  $\Delta n$  levels takes place in a time

$$t \propto \frac{T_{\rm rev}}{(\Delta n)^2} \, .$$

An additional phase shift between the various components of the packet (24) (within the limits of energy width  $\Delta E \propto \hbar \omega_{\rm cl} \Delta n$ ) is of the order of unity. The dephasing leads to a decay of the packet after many periods of classical motion, that is, it limits the duration of the classical stage of evolution of the packet.

However, as shown above, in a system with a discrete energy spectrum the evolution of a wave packet cannot be completely irreversible. Actually, for  $t = T_{rev}$  the additional phases due to the terms quadratic in k are exactly integral multiples of  $2\pi$ , so that the wave packet can be completely restored. It is obvious that for  $t \gtrsim T_{rev}$  the classical evolution of the packet is again revived. This phenomenon has been called the "revival" of the packet.<sup>16</sup>

To answer the question of what sort of structure a wave packet has at intermediate times  $T_{\rm cl} \ll t \lesssim T_{\rm rev}$  we shall study the form of the packet (24) for  $t/T_{\rm rev} \approx m/n$ , where m and n are integers and m/n is an irreducible fraction. The additional phase shifts due to terms proportional to  $k^2$  are equal to  $2\pi\Theta_k$  where  $\Theta_k = \{mk^2/n\}$ . Here the curly brackets stand for the fractional part of the argument. It can be shown directly that the quantities  $\Theta_k$  form periodic sequences with a period  $l_0$  that depends only on the denominator n of the fraction

$$\Theta_{k+l_0} = \Theta_k;$$

where  $l_0 = n/2$  if n is divisible by 4, and  $l_0 = n$  otherwise.

The periodic sequences  $\exp(-2\pi i\Theta_k)$  can be decomposed into  $l_0$  fundamental sequences having the same period

$$\exp(-2\pi i\Theta_k) = \sum_{s=0}^{l_0-1} a_s \exp(-2\pi i sk/l_0), \qquad (25)$$

$$a_{s} = \frac{1}{l_{0}} \sum_{k=0}^{l_{0}-1} \exp\left(-2\pi i \Theta_{k} + 2\pi i \frac{sk}{l_{0}}\right) .$$
 (26)

Substituting (25) and (26) into (24) we have

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$$\psi(x,t) = \sum_{s,k}^{+} c_k a_s u_k(x) \cdot \exp{-2\pi i kt} / T_{cl} - 2\pi i \frac{sk}{l_0}, \quad (27)$$

or

$$\psi(x,t) = \sum_{s=0}^{l_0-1} a_s \psi_{cl}(x,t + \frac{s}{l_0} T_{cl}) , \qquad (28)$$

where  $\psi_{cl}(x,t)$  means

$$\psi_{\rm cl}(x,t) = \sum_{k} c_{k} u_{k}(x) \exp\left(-2\pi i k \, \frac{t}{T_{\rm cl}}\right) \,. \tag{29}$$

As follows from this analysis, expression (29) describes the evolution of the packet in a "classical" way. One can show that the number of nonzero coefficients  $a_s$  in formula (28) is  $q = (n/4) [3 - (-1)^n]$  with the moduli of all of them being identical.

Thus, for any rational number m/n the initial packet splits near  $t \approx T_{rev} (m/n)$  into q spatially separate packetfractions undergoing classical motion, shifted in time from each other by the qth part of the classical period. This structure was called in Refs. 31 and 32 fractional revival of order m/n. Of course, this structure will be clearly defined if the separate fractions do not overlap, that is, for  $q < L/\Delta x \sim \Delta n$ . The better the initial packet satisfies the conditions of classical behavior, the higher the order of splitting that can be observed.

Let us examine some specific structures. The simplest of them occurs for  $t \approx T_{rev}/2$ . Then  $a_0 = 0$ ,  $a_1 = 1$ , and

$$\psi(x,t) = \psi_{\rm cl}(x,t + \frac{1}{2}T_{\rm cl}), \qquad (30)$$

which is the original packet shifted in time by half the classical period.

Near 
$$t \approx T_{\rm rev}/4$$

$$\psi(x,t)$$

$$= \frac{1}{\sqrt{2}} \left[ \exp\left(-i\frac{\pi}{4}\right) \psi_{cl}(x,t) + \exp\left(+i\frac{\pi}{4}\right) \psi_{cl}(x,t + \frac{1}{2}T_{cl}) \right].$$
(31)

Expression (31) describes an essentially nonclassical object, consisting of a superposition of two correlated localized packets macroscopically separated by a length of the order of the dimension of the classical orbit. A similar structure arises for  $t \approx 3T_{rev}/4$ .

For  $T \approx T_{rev}/3$  the wave function is

$$\psi(x,t) = \frac{1}{3} \left[ 1 + 2\exp\left(-\frac{2\pi i}{3}\right) \right] \left[ \psi_{cl}(x,t) + \exp\left(\frac{2\pi i}{3}\right) \left( \psi_{cl}(x,t + \frac{1}{3}T_{cl}) + \psi_{cl}(x,t + \frac{2}{3}T_{cl}) \right) \right].$$
(32)

This kind of structure, consisting of three packets, occurs also for  $t/T_{\rm rev} \approx 1/6$ , 2/3, 5/6.

Let us illustrate these forms of behavior in the example of an anharmonic oscillator with a potential energy of the form

$$U(x) = \frac{1}{2} M\omega^2 x^2 + \Gamma x^4 .$$
 (33)

The eigenstates and the energy spectrum of this oscillator have been well studied. In the case of a weak anharmonicity



FIG. 1. Wigner distribution function of an anharmonic oscillator (from Refs. 32 and 47). Parameters:  $\eta = 0.001$ ,  $T_{rev} = 667 T_{cl}$ ;  $\alpha = 5$ ; a)  $t/T_{cl} = 0$ ; b)  $t/T_{cl} = 111$ ; c)  $t/T_{cl} = 166$ ; d)  $t/T_{cl} = 333$ .

 $\eta = \Gamma(\hbar/M^2 \omega^3 \ll 1)$  the following expression is valid<sup>108</sup>

$$E_n \approx \hbar\omega \left[ \left( n + \frac{1}{2} \right) + \frac{3}{2} \eta \left( n + \frac{1}{2} \right)^2 + \frac{3}{8} \eta \right].$$
(34)

A very clear picture of the evolution of the wave packets in this system under quasiclassical conditions can be obtained with the use of the Wigner distribution function<sup>109</sup>  $P_W(x,p,t)$ , defined in phase space

$$P_{W}(x,p,t) = \frac{1}{\pi\hbar} \int_{-\infty}^{+\infty} \mathrm{d}y \psi^{*}(x+y,t) \psi(x-y,t) \exp\left(\frac{2\pi i p y}{\hbar}\right) \,.$$
(35)

Figure 1 shows the results of numerical calculations<sup>32,47</sup> that depict the distribution  $P_W(x,p,t)$  at the initial instant of time (the initial wave packet is specified in the form of the coherent state of the harmonic oscillator, Eq. (3) with coefficients (4)) and also at succeeding instants of time that correspond to fractional revival of various orders. The smooth peaks in the Wigner distribution function correspond to the splitting of the initial packet, and the localized rapidly oscillating spikes in the phase plane appear as a result of the interference between the various packet-fractions and have no classical analog.

## 2.3. Preparation and evolution of packets of stationary states in model multilevel quantum systems

Many features of the dynamics and the generation of wave packets with different physical systems (atoms, molecules) are reflected in a very simple model system that has been used many times in the theory of multi-photon ionization of atoms and molecules.<sup>110-114</sup> This same model has been used previously for the description of nonradiative transitions in complex molecules.<sup>115-118</sup> We shall examine a quantum system excited out of the ground state  $|0\rangle$  into a group of states  $|n\rangle$  with energies  $E_n$  by a laser pulse with an envelope f(t). The probability amplitudes  $a_n$  of finding the system in the states  $|n\rangle$  are given in the resonance approximation by the equation

$$i\hbar\dot{a}_{0} = \sum_{n} V_{0n}(t)a_{n},$$
  
 $i\hbar\dot{a}_{n} = \Delta_{n}a_{n} + V_{n0}(t)a_{0},$ 
(36)

where  $\Delta_n = (E_n - \overline{E})/\hbar$ ,  $\overline{E}$  is the energy of resonance with the ground state, the composite matrix element is  $V_{0n}(t) \sim [f(t)]^{k_0}$ , where  $k_0$  is the number of photons involved in the transition from the ground state to the group of states  $|n\rangle$ . The solution of the problem (36) for an arbitrary f(t) is unknown, but it is possible to identify three different regimes of excitation for which it is possible to predict the behavior of the system.

1. Let us assume that the pulse is sufficiently long so that the spectral width  $\hbar/\tau$  (where  $\tau$  is the pulse length) of the excitation  $V_{0n}(t)$  is much less than the energy spacing  $\delta$ between the excited levels in the neighborhood of the resonance. Let us also assume that  $|V_{0n}| \ll \delta$ . In this case a transition is induced to only one of the resonance levels. The system is described by a two-level model in which Rabi beats<sup>119</sup> occur between the ground and resonance excited states. If the excited level is ionized by the same pulse, then the pro-

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cess of multiphoton ionization through this intermediate state has a resonance nature.<sup>113,118</sup>

2. Let us assume that the pulse as before is weak  $(V_{0n} | \ll \delta)$  but quite short  $\hbar/\tau \gtrsim \delta$ . Then the probability amplitude  $\bar{a}_n$  for finding the system in the state  $|n\rangle$  after the pulse ends is

$$\left|\vec{a}_{n}\right| \approx \frac{1}{\hbar} \left| \int_{-\infty}^{+\infty} \mathrm{d}t \; V_{0n}(t) \exp\left(i\Delta_{n}t/\hbar\right) \right| , \qquad (37)$$

and the wave function of the system is a packet  $\sum_n \bar{a}_n \exp(-iE_n t/\hbar) |n\rangle$ . If the energy spectrum is quasiequidistant in the vicinity of  $\overline{E}$   $(E_n \approx \overline{E} + n\delta)$  and the matrix element  $V_{0n}$  depends only weakly on *n*, then in the case of a one-photon excitation  $\bar{a}_n$  is determined by the Fourier transform  $f_n$  of the envelope of the pulse f(t). The temporal dynamics of the packet that is formed can be followed from the time dependence of afterglow of the system resulting from the transition to the lower-lying level or from the time dependence of the ionization signal induced by a supplementary laser field. If the matrix element of the transition to the final state depends only weakly on the energy, then the instantaneous rate of the radiation (or ionization) process is

$$I(t) \propto \left| \sum_{n} f_{n} \exp\left(in\delta t/\hbar\right) \right|^{2}$$
(38)

and is determined by the quantum beats of a large number of transitions. If  $\hbar \tau \gg \delta$ , then, unlike the usual quantum beats associated with the coherent population of a small number of levels,<sup>120</sup> the intensity I(t) of the process at times  $t = 2\pi n \hbar/\delta$  exhibits sharp spikes (recurrences). The formation of these spikes is analogous to the appearance of the sharp diffraction peaks in the scattering of waves by a diffraction grating with a large number of lines.<sup>121</sup> Near the *n*-th recurrence the summation in (38) can be replaced by an integral, and as follows from expressions (37) and (38), the shape of each luminescence (or ionization) peak replicates the shape of the initial exciting pulse.<sup>16,121</sup>

3. If the exciting pulse is sufficiently strong  $(|V_{0n}| \ge \delta)$ and long, then the formation of the packet can no longer be described by perturbation theory. Here the behavior can be followed for the case of a step-function excitation that is switched on discontinuously at t = 0 and remains unchanged thereafter. For times  $\tau \ll \hbar/\delta$  the pulse does not distinguish the discrete nature of the energy spectrum but sees the levels  $E_n$  as a continuum with a density of states  $\rho = 1/\delta$ . The ground state component is destroyed exponentially<sup>115-118</sup>

$$|a_0(t)|^2 = \exp(-\gamma t)$$
, (39)

where (taking  $|V_{0n}| \equiv V$ ) the quantity

$$\gamma = 2\pi V^2 \rho/\hbar = \frac{2\pi V^2}{\hbar \delta}$$

is given by Fermi's Golden Rule. In this way there is a time scale  $\gamma^{-1}$  during which the excited states form a packet of "destruction." Because of the discrete nature of the spectrum, the system has a high probability of returning to the ground state near times  $t \propto n\hbar/\delta$ . If the excited levels are exactly equidistant, and the transition matrix elements are independent of the level index (the Bixon-Jortner model;

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Ref. 115) the exact solution for the time dependence of  $a_0(t)$  is known<sup>122,123</sup>

$$a_0(t) = \sum_{n=0}^{\infty} \exp\left(-in\Delta_0 t/\hbar\right)$$

$$\times \quad \exp\left[-\gamma(t-n\xi)/2\right] L_n(\gamma(t-n\xi))\Theta(t-n\xi) , \quad (40)$$

where  $\xi = 2\pi\hbar/\delta$ ,  $\Theta(z)$  is the unit step function,  $\Delta_0$  is the spacing of  $\overline{E}$  from the nearest level of the quasicontinuum, and the functions  $L_n(z)$  have the form

$$L_n(z) = L_0(z), \quad n = 0,$$
  
$$L_n(z) = L_n(z) - L_{n-1}(z), \quad n \ge 1;$$

where the  $L_n(z)$  are the Laguerre polynomials. For  $\hbar\gamma/\delta \ge 1$ , the coefficient  $a_n(t)$  falls off exponentially near t = 0 (compare with Eq. 39). Near  $t = t_1 = 2\pi\hbar/\delta$  the variation of  $a_0(t)$  is  $|a_0(t)|^2 = \gamma(t - t_1)\exp[-\gamma(t - t_1)]$ , and so forth. In Refs. 110 and 111 an approximate theory based on the existence of two different time scales ( $\gamma^{-1}$  and  $\hbar/\delta$ ) was developed for the description of the photoexcitation of a quasicontinuum by a long optical pulse. By this theory it is possible to describe the shape of each recurrence even if all the matrix elements of the transition are not exactly equal.

### 3. THE DYNAMICS OF PACKETS OF HIGHLY EXCITED STATES OF RYDBERG ATOMS

Because of the perfection of the technique of generating short light pulses it is possible in the excitation of atoms to employ an exciting pulse that is so short (or with a spectral width so broad) that a linear superposition (a packet) of a very large number of stationary states with different energies is created, rather than the excitation of only a single stationary state, as in conventional optical spectroscopy. The evolution of this packet, as shown above for simple models, must obey, at various stages, both classical and quantum mechanical laws. The most striking manifestation of this behavior will be observed in pulsed excitation of high-energy Rydberg states of atoms. Possible means of observing the dynamics of the wave packets that are created include detection of the spontaneous emission of the excited atoms or their ionization by a short optical pulse delayed in time relative to the exciting pulse. In the stage of classical motion in an elongated Kepler orbit the emission by the atom consists of regularly repeating peaks (with the period of the classical motion), corresponding to the passage of the packet at the minimum distance from the nucleus (or the atomic shell of a multielectron atom), where the acceleration of the electron is a maximum. Decay of the packet corresponds to the formation of an irregular pseudochaotic pattern that accompanies the falloff in intensity of the emission peaks. However, at later times the regular periodic pattern of emission peaks reappears, which corresponds to the revival of the localized particle-like packet in a Kepler orbit.

Parker and Stroud<sup>16</sup> made the first detailed numerical calculation of the excitation of high-energy states of the hydrogen atom by short optical pulses and the time dependence of the emission from the packet of stationary states formed in this way. They calculated the excitation of an atom by a laser pulse of duration  $\tau \propto 10$  ps into a group of states near the level with the principal quantum number  $\bar{n} = 85$ . The corre-



FIG. 2. Intensity of spontaneous emission by a Rydberg atom excited by a short laser pulse (from Ref. 16). The arrows indicate<sup>31,32</sup> the fractional revivals of various orders.

sponding period of classical motion is  $T_{\rm cl} = 2\pi \bar{n}^3 \hbar/\mathrm{Ry} \approx 94$ ps, which is an order of magnitude longer than the pulse length, so it is possible in the excitation to prepare a packet made up of many stationary states with various principal quantum numbers n near  $\overline{n}$ . Since the atom is excited from the ground state, the packet will contain states with small values of the angular momentum, which in classical language corresponds to particles moving in a highly elongated Kepler orbit. The results of the numerical calculation<sup>16</sup> are plotted in Fig. 2, which shows that sharp emission spikes are observed in the initial stage of evolution of the packet, and these spikes repeat with a time spacing equal to the period of classical motion in the Kepler orbit. After 35 periods of  $T_{\rm cl}$ the pattern of the emission in the first stage is repeated, which corresponds to a revival fraction of order 1/2  $(T_{rev} = 5.2 \text{ ns})$ . An analogous variation of the probability of an electronic Raman transition, induced by two consecutive laser pulses, as a function of the delay time between pulses was observed in the work reported in Refs. 19 and 22. The detailed picture of the time dependence of the emission reported in Ref. 16 was analyzed in detail in Refs. 31 and 32, where it was shown that for revival fractions of high order, shown in Fig. 2 by the arrows, the emission spikes correspond to the passage at the minimum distance from the nucleus by each of the fragments of the split packet. Therefore, near the instants of time  $T_{rev}/4$ ,  $T_{rev}/6$ ,  $T_{rev}/8$ , etc, the emission spikes appear twice, three times, four times, etc, more frequently than in the initial stage or at the stage  $T_{rev}/2$ , where the packet is reconstructed with a shift in time of  $T_{\rm cl}/2$ . Su<sup>80</sup> has carried out a numerical investigation of the time dependence of the intensity of the emission of a packet consisting of vibrational states of a two-atom ion, and observed analogous behavior of the breakup of the packet and its subsequent revival.

The space-time structure of packets of Rydberg states moving in Kepler orbits has been studied in Ref. 46. The long-term evolution of packets of stationary hydrogen-atom states  $\psi_{n,l,m_l}$  with various principal quantum numbers *n*, the maximum orbital quantum number for a given *n*, l = n - 1and the maximum projection  $m_l = n - 1$  of the angular momentum was investigated in that paper. The distribution of

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the populations of the levels in the packet was taken to be Gaussian:

Here  $a_{\rm B}$  is the Bohr radius and  $N_n$  is a normalization constant. The packet of states (41) is localized both in the radial direction(r) and in the angular  $(\vartheta, \phi)$  variables. The evolution of this packet was followed for the parameter values  $\bar{n} = 320$  and  $\sigma_0 = 2.5$ . The expansion of the energy of the atom near  $n = \bar{n}$  has the form (compare with formula (22)):

$$-\frac{Ry}{2n^2} = -\frac{Ry}{2\bar{n}^2} \left[ 1 - 2\frac{\Delta n}{\bar{n}} + 3\left(\frac{\Delta n}{\bar{n}}\right)^2 - \dots \right], \qquad (42)$$
$$\Delta n = n - \bar{n}.$$

An investigation of the time dependence of the packet (41) demonstrates the sequential advent of the time scales that follow from the expansion (42) (Fig. 3). In times  $t \leq T_{cl} = 2\pi \bar{n}^3 \hbar/Ry$  the packet moves in a circular Kepler orbit lying in the plane perpendicular to the axis of quantization, undergoing dispersion because its Rydberg levels are not equidistant. For times  $t = (m_0/n_0) T_{rev}$  ( $m_0$  and  $n_0$  are relatively prime integers) it undergoes a succession of fractional revivals in complete accordance with the scenario<sup>31,32</sup> described in Section 2. Thus, the long-term evolution of wave packets of highly-excited Rydberg states of atoms inevitably exhibits both classical and quantum mechanical properties, associated with the discrete nature and the unequal spacing of the atomic spectrum (the nonlinearity of the atomic oscillator).

An interesting suggestion for compression (time squeezing) of optical pulses, using the properties of the time evolution of Rydberg wave packets, has been made by Noordam et al.<sup>37</sup> The phase-matching of the states in a linear superposition of the form (24), as noted above, is disrupted with time because the levels of the spectrum are not equally spaced. Spatially, this means that during the oscillations, the parts of the packet that are formed of states with the highest energies lag the lower-energy parts because the classical period increases with the energy. If the lower-energy parts of the packet are excited with a certain time delay relative to the excitation of the high-energy parts, then as time passes these parts will come into phase, forming a packet that is highly localized in space. This kind of excitation can be obtained by frequency chirping of the exciting pulse (decreasing the frequency (e.g., linearly) with time). It was shown by Noordam et al.<sup>37</sup> that as such a phased packet passes at the minimum distance from the nucleus of the atom, it can emit a pulse that is shorter than the excitation pulse. Thus, unlike the conventional methods of light-pulse compression,<sup>124</sup> this method is based on the dispersion properties of de Broglie waves in a single atom. Noordam et al.<sup>37</sup> also discussed the possibility of using chirp of the eigenfrequen-



FIG. 3. Evolution of the distribution of the probability distribution in the x-y plane for the wave packet of an electron moving in a circular Kepler orbit (from Ref. 46). The time t is measured in units of  $T_{rev}/2$ .

cies of an atom by means of the dynamic Stark effect instead of chirp of the frequency of the radiation exciting the atom.

It has also been proposed that the dynamics of the packet of Rydberg states formed as a result of the action of a short laser pulse on an atom might also be observed experimentally in the dependence of the probability of the electronic Raman transition induced by a second optical pulse as a function of the time delay of the second pulse relative to the first, <sup>19,22</sup> or from the analogous dependence of the delayed photoionization signal.<sup>26,42,43</sup> The semiclassical theory of two-photon excitation and ionization of Rydberg states of optical electrons in atoms has been developed in Refs. 18, 19, 22, 27, 28, and 41. The methods of quantum defect theory have been used in these papers to study the interaction of a Rydberg packet with the ion core, and interesting phenomena were observed as a result of the periodically repeating processes of elastic and inelastic scattering of the packet by the core.

The effect of the formation of linear superpositions of atomic states on the ionization of an atom in the strong field of laser radiation has been analyzed theoretically in a number of publications.<sup>26,42-44,125</sup> Parker and Stroud<sup>44</sup> have studied the situation where two closely-lying atomic levels are strongly shifted relative to each other because of resonance absorption and stimulated emission of photons through the continuum states. Because of these processes, in a strong field a linear superposition of states is formed for which the probability of a transition from these states to the continuum is small because of destructive interference. In a certain sense this result means that the atom is stabilized by the strong field. This effect is related to the well-known effect (since the classical work of Fano<sup>126</sup>) of the formation of nondecaying states in the model of two discrete levels lying on a background continuum and interacting with it. In essence, the same origin is associated with the imprisonment of the populations in three-level systems subjected to resonance excitation by two laser fields.<sup>127</sup> There is a great deal of interest in carrying over these results to the case of multi-level quantum systems, in particular, Rydberg atoms. Dealing with a real multilevel atomic spectrum and fields of arbitrary strength entails considerable mathematical difficulties, which certainly enhances the value of model calculations. In Refs. 26, 42, and 43 a model assuming a quasi-equidistant spectrum and a flat continuum was used to demonstrate the possibility of stabilization of a multilevel atom through the formation of packets of highly excited atomic states by a strong field. Analytic results have been obtained<sup>30</sup> in a study of the formation of Rydberg wave packets by strong optical pulses of a special shape. A detailed review of theoretical methods of describing laser excitation of packets of stationary states in Rydberg atoms can be found in Ref. 48.

One of the first experiments in which a wave packet was formed from Rydberg states of an atom is described in Ref. 25. By means of  $\approx$  3 ps laser pulses, (3 + 1)-photon ionization of xenon atoms was achieved (the energy of the three photons reached to the region of the Rydberg states near the ionization threshold for xenon). If the spectral width of the excitation pulse, associated with the finite pulse length, is less than the spacing between adjacent atomic levels, then the ionization can be enhanced by virtue of an intermediate resonance in the Rydberg region. If, on the other hand, the spectral width of the pulse overlaps several levels, (which is accompanied by the formation of a packet of stationary states), then the region of the Rydberg levels exhibits the properties of a continuum, and the resonance enhancement of the ionization probability does not happen. In an experiment Noordam et al.<sup>25</sup> observed a changeover from the resonance regime of ionization to a nonresonance regime as the frequency of the laser radiation was increased while the spectral width was maintained. The changeover of regimes was related to the increased density of Rydberg levels near the ionization threshold. Information on the preparation of a packet of Rydberg states is only implicit in the results of Ref. 25, since the packet does not move during the short time of the ionizing pulse, which is much shorter (in the nonresonance regime) than the classical period of the Keplerian motion, and consequently its most characteristic dynamic properties are not revealed. A theoretical investigation of the

near-threshold multi-photon ionization of atoms by short laser pulses, corresponding to the conditions of the experiment of Ref. 25, has been given in Ref. 38.

An electron wave packet localized in the angular variables was prepared for the first time in the work reported in Ref. 23 by means of the method described in Refs. 17 and 21. The packet was excited from the ground state of a sodium atom into a group of Rydberg states of principal quantum number n = 50, which were mixed with a circularly polarized rf field with an amplitude of 0.3 V/cm and a frequency of 65 MHz. The rf field was tuned near the 30-photon resonance between the 50d state and the state with n = 50 and orbital quantum number l = 32 (the level n = 50 in the sodium atom is easily split into a group of states with different values of l). During the pulsed optical excitation about ten of the "dressed" rf states were populated. After the rf field was turned off adiabatically (over a time of 5  $\mu$ s) a packet was formed, localized in the angular variables (but not in the radial direction) with an angular width of  $\sim 20^\circ$ . The packet at first was oriented along the electric vector of the rf field at the time of excitation, and then it began to precess slowly. Quantum mechanically, the reason for the precession was that a packet of states of different values of *l* is not a stationary state because of the lifting of the orbital degeneracy of the sodium levels. Yeazell and Stroud<sup>23</sup> interpreted this non-stationary behavior classically as the precession of the Kepler orbit due to the deviation of the potential acting on the optical electron from Coulombic. Figure 4 shows the distribution of the probability density for a packet localized in the angular variables.<sup>23</sup> The strong anisotropy in the angular distribution of the electron density was detected by means of ionization of the packet. The precession and dispersal of the packet were not observed under the conditions of the experiment<sup>23</sup> because these processes were too slow (with a characteristic time of the order of milliseconds).

The radial localization of a Rydberg wave packet was observed for the first time in the work of ten Wolde *et al.*<sup>24</sup> In this experiment, 6-ps laser pulses with two-photon excitation were used to prepare in the vicinity of the core of a rubidium atom Rydberg states of the outer electron with various principal quantum numbers in the region n = 42. The packet was formed of only s and d states (with more d states), which results in an angular delocalization of the probability density. The packet was like a spherical cloud with a radius that oscillated with the period of the Keplerian motion  $T_{\rm cl} = 2\pi \bar{n}^3 \hbar/Ry$  between the minimum and the maximum distances from the atom core, as defined by the classical turning points. Although indications of classical dynamics appear in this evolution with time, a packet of this sort is still far from being a particle moving in a Kepler orbit. The presence of the packet near the core was detected from the ionization signal induced by an additional probing pulse delayed in time relative to the pulse creating the packet. The probability of absorption of a photon is a maximum when the electron is near the core. Therefore, the maximum ionization signal occurs at a delay time that corresponds to the return of the electron to the core after it has receded from the core to the maximum distance. Ten Wolde et al. were able to observe two similar returns of the packet. The experimentally determined recurrence time was  $9.4 \pm 1.0$  ps, which is in good agreement with the theoretical value  $T_{cl} = 10.2$  ps (for  $\overline{n} = 42$ ). After two returns the packet decayed. The large value of the dispersion is due to the considerable variations in the energy spacings between the levels with different n in the region of  $\overline{n} = 42 \ (\omega_{n,n-1} - \omega_{n+1,n} \propto (\overline{n})^{-4} \text{Ry}/\hbar).$ 

In a similar experiment carried out with potassium atoms,<sup>33</sup> two-photon excitation was induced from the ground state to a group of levels (mainly *d*-levels) with *n* in the region of  $\bar{n} = 89$ , which corresponds to  $T_{cl} = 107$  ps. The spectral width of the laser pulse encompassed several levels ( $\Delta n = 5$ ), so that very sharp peaks with the period of the classical motion (Fig. 5) were observed in the delayed ionization signal. The decrease in the peak height, according to Yeazell et al.<sup>33</sup> is due to two causes: dispersal of the packet resulting from the slight nonequidistance of the levels, and fluctuations in the laser frequency from pulse to pulse.

The long-term behavior of the wave packet in a Rydberg atom, the phenomenon of its decay and revival, was recently studied in Ref 45. In that work a group of levels of the potassium atom with  $\bar{n} = 65$  was populated by singlephoton excitation, and not by two-photon excitation as in Ref. 33. In this way it was possible during the excitation to avoid the shift of the atomic levels resulting from the highfrequency Stark effect. The frequency chirp due to the Stark effect has little effect on the behavior of the packet for short



FIG. 4. Distribution of the probability density in the x-y plane for the wave packet localized in the angular variables (from Ref. 23). The peak in the distribution is located 5000 Bohr radii from the nucleus. The packet is formed of states with large values of angular momentum  $(n = 50; 29 < l < 37; m_l = l)$ .

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FIG. 5. Experimental (points) and theoretical (solid curve) results for the ionization signal from potassium atoms (from Ref. 33). The delay time between the excitation and ionizing pulses is on the horizontal axis.





times, but it causes a substantial distortion of its long-term evolution. As in the previous works, these experiments<sup>45</sup> involved observation of a photoionization signal produced by a delayed probe pulse. Figure 6 shows clearly five oscillations corresponding to classical motion with  $\bar{n} = 65$ , followed by breakup of the packet because of dispersion. The experimental results are in good agreement with theoretical predictions (Fig. 6b). After about 15 periods after the excitation the delayed photoionization signal nearly completely recovers its initial shape, which is an indication of revival of order 1/2of the packet, as shown in Refs. 16, 31, and 32. The periods of classical Keplerian motion of the electron are marked off on the upper horizontal axis in Fig. 6. It follows from the experimental and calculated curves that the packet is regenerated with a shift of 1/2 a period relative to the position that a classical electron moving for this length of time would have. This result is clearly consistent with the scenario of fractional revival, discussed above. The calculated curve in Fig. 6 shows in its middle part oscillations with twice the Kepler frequency, which must correspond to the splitting of the packet into two components (fractional revival of order 1/4). In the experiment of Ref. 45 this region of delay was not sufficiently resolved, but in a recent and more careful experiment<sup>144</sup> showed with certainty the fractional revival of this type.

The dispersal of a wave packet can be substantially reduced if the levels are almost equidistantly spaced. Ten Wolde *et al.*<sup>34</sup> have reported the preparation of a long-lived wave packet formed of a coherent superposition of "parabolic" states of rubidium atoms in a dc electric field. Because of

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the field the rubidium levels of the hydrogenlike atom are split by the Stark effect. For weak fields the Stark shift is linear in the field and the level with the principal quantum number n is split into a group of sublevels with energies

$$E_{nk} = -\frac{1}{2n^2} \operatorname{Ry} + \frac{3}{2} e_0 a_{\mathrm{B}} Fnk;$$

where F is the electric field strength,  $k = n_1 - n_2$ , and  $n_1$ and  $n_2$  are the parabolic quantum numbers. The spacing between the sublevels is constant, which corresponds to beats in the linear superposition of the parabolic states with a single period  $T = 2\pi \hbar/3e_0 a_B Fn$ . During these beats the populations of the states with different l vary, and the angular part of the wave function oscillates between a more or less spherical distribution for small l and a distribution that is highly elongated in the direction of the field for large  $l \sim n - 1$ . In the experiment of Ref. 34, 7-ps laser pulses were used for two-photon excitation of a group of levels with n = 23 in a rubidium atom placed in a dc electric field of 247.5 V/cm. The evolution of the packet of parabolic states that was formed was observed by detecting the ionization signal with a time-shifted laser probe pulse. During the beats the ionization signal was a maximum when states with small values of l had the largest populations, and was minimum for high population of the level with l = n - 1. Figure 7 shows the dependence of the ionization signal on the delay time between pulses. Up to ten oscillations are observed with a period of 46 ps, which is in good agreement with the theoretical value of 45.8 ps.



FIG. 7. Ionization signal from rubidium atoms in a constant electric field, as a function of the delay time between the excitation and the ionizing pulses (from Ref. 34).

The experiments with the Rydberg atoms have thus made it possible to observe with good reliability the evolution of packets composed of highly excited states and observe indications of classical dynamics during this evolution. However at the present time the "Rutherford atom" discussed in the introduction has still not been realized. The packets that have been prepared are localized only in the radial or in the angular variables, whereas a packet corresponding to a classical particle must appear as a three-dimensional concentration of electron density, in which all the characteristic dimensions are less than those of the classical orbit.

### 4. LONG-TERM EVOLUTION OF PACKETS OF ELECTRON-VIBRATIONAL STATES OF MOLECULES. LASER FEMTOCHEMISTRY

Besides investigations of Rydberg wave packets in atoms, a new field of molecular spectroscopy, stimulated by the needs of chemistry, has been developed independently and with great success, and has been called femtosecond laser chemistry.<sup>49,50</sup> The investigation of the dynamics of chemical transformations in real time is a fundamental problem of chemistry. The characteristic time of molecular rearrangement is of the order of  $10^{-12}$  s or less. Therefore, only methods of femtosecond spectroscopy have sufficient time resolution to observe the individual states of the rearrangement of the chemical bonds. Since the period of oscillation of the nuclei of molecules is frequently hundreds of femtoseconds, the excitation of molecules with femtosecond optical pulses will create the conditions for formation of packets of stationary states corresponding to individual vibrational levels of the molecule. The evolution of these packets can, of course, be associated with the motion of the nuclei along classical trajectories during the chemical transformations of the molecules.

Figure 8 shows one of the possible schemes for observing the nuclear motion during chemical transformations.<sup>49</sup> The adiabatic potential  $V_0$  corresponds to the bound state of a molecule, and the two repulsion terms  $V_1$  and  $V_2$  correspond to dissociated states of the molecule with different degrees of excitation of one of the fragments. The initial packet of states of the  $V_1$  term is prepared by a short pump pulse at the wavelength  $\lambda_1$ . The repulsion that arises sets the



FIG. 8. Potential curves of a molecular system excited by two optical pulses separated by a delay time (from Ref. 49).

nucleus into motion according to the term  $V_1$  thereby increasing the internuclear distance. After some time delay the molecule is acted on by a short probe pulse of wavelength  $\lambda_2$ , the absorption of which is accompanied by a transition to term  $V_2$ . In accordance with the Franck-Condon principle, the absorption will be the greatest if after the delay time the packet is in the region where  $V_2(R) - V_1(R) = 2\pi \hbar c/\lambda_2$ (c is the speed of light). For  $\lambda_2 = \lambda_2^*$  (probing the transition state) this situation corresponds to a sharp peak in the dependence of the absorption probability on the delay. However, if the second pulse is tuned to a transition into the free reaction product  $(R \rightarrow \infty, \lambda_2 = \lambda_2^{\infty})$ , then, since any packet sooner or later must be found in the region of large internuclear distances, the absorption signal flattens out to a plateau with increased delay time. The degree of absorption of the probe pulse can be determined, for instance, from the integrated signal of subsequent luminescence from the spontaneous inverse transition  $V_2 \rightarrow V_1$ . The variation in the position of the absorption peak as the wavelength is scanned by the probe pulse affords the unique possibility of reconstructing the dependence of the adiabatic potentials on the internuclear distance. The idea behind this experiment was realized in the work of Dantus and his coworkers,<sup>51,52</sup> who, by means of the scheme described here observed the dissociation of iodine cyanide ( $\lambda_1 = 307$  nm,  $\lambda_2^{\infty} = 388.5$  nm). The wavelength of the probe pulse was scanned over 10 nm towards the red. The experimental dependence of the absorption signal on the delay time was found to be in good agreement with that expected (Fig. 9). The time of breakup of the molecule, evaluated from the experiment, was about  $205 \pm 30$  fs. A theoretical investigation of the dynamics of wave packets in the ICN molecule and the delayed absorption signals during its dissociation has also been reported in Refs. 58-60 and 62.

By means of this technique it is possible not only to monitor the transition stages of the formation and breaking of chemical bonds, but also to observe in real time the vibrational motion of a wave packet in the case where the term on which the system is pumped is not a repulsive term. A striking example of this comes from femtosecond experiments<sup>53,54</sup> on the dynamics of the predissociation of an excited NaI molecule into neutral fragments Na + I. The potential curves for this reaction are shown in Fig. 10. The

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FIG. 9. Absorption signal in experiments with ICN molecules (from Refs. 51 and 52). a)  $\lambda_2^{\infty} = 385.5$  nm, b)  $\lambda_2^{*} = 391.4$ , 390.4, 389.8, and 389.7 nm. The peak shifts to the right with decreasing wavelength.

ground state of the NaI molecule is ionic. Since the energy corresponding to Na  $^+$  + I  $^-$  is greater than that for Na + I when the atoms are widely separated, a quasicrossing of the ionic and covalent terms occurs at  $R_{\chi} \approx 6.93$  Å. The large interaction between these terms in the neighborhood of  $R_X$ results in the formation of the adiabatic terms shown in Fig. 10 by the dashed lines, with the upper one being a bonding term. A short laser pump pulse at a wavelength  $\lambda_1 = 310$  nm creates from the ground state of NaI a wave packet in the covalent state, which begins to oscillate in the upper adiabatic term. Each time it passes through the region of the quasicrossing the wave packet will make with a probability 0.1 a Landau-Zener nonadiabatic transition to the lower adiabatic term. As a result the wave function of the system in the asymptotic region  $(R \rightarrow \infty)$  in the covalent term will consist of a sequence of packets passing in succession. If the subsequent short laser probe pulse is tuned to resonance with the *D*-absorption line of the free sodium atom ( $\lambda_2^{\infty} = 549$  nm), the dependence of the integrated signal of the inverse luminescence on the delay time between the pump and the probe pulses has a staircase shape (the upper curve in Fig. 11). The steps correspond to the appearance of more new packets in the covalent term with the passage of time. If, however, the second pulse is tuned out of resonance with the D-transition, then the population of sodium atoms still locked in the transition state [Na...] is probed, and the time dependence of the signal is a periodic sequence of peaks (the lower curve of Fig. 11). The authors of Refs. 53 and 54 have shown that the repetition frequency of the peaks coincides with the frequency of classical oscillations in the upper adiabatic term at energies determined by the wavelength of the pump pulse. The falloff of the peak intensities is in accordance with the variation of the probability of the Landau–Zener transition as the system passes through the region of the quasicrossing. These experiments have stimulated a detailed theoretical investigation of nonstationary packet phenomena that occur during this reaction.  $^{53,54,61,64-69}$ 

Considerable success has been achieved in the observation of the long-term evolution of packets of vibrational states of molecules in the work of Refs. 55 and 56, which was focused on the pulsed optical excitation of iodine molecules



FIG. 10. Potential curves of the NaI molecule (from Refs. 53 and 54). The bell-shaped curve to the left of the vertical axis shows the spectral width of the excitation pulse. The motion of the packet, composed of vibrational states, is shown in the form of a distribution of the probability density moving along the horizontal axis.



FIG. 11. Absorption signal in experiments with NaI molecules, as a function of the delay time between the excitation and the probe pulses (from Refs. 53 and 54). The explanation is in the text.

 $(I_2)$ . Figure 12 shows the ground term and two excited terms of this molecule. The molecule was excited from the ground state X to the term B at the wavelength  $\lambda_1 = 620$  nm by a short 50 fs pulse. A packet was created out of several vibrational levels in the region of the vibronic quantum number v = 3 of this term. The presence of the packet was established by a delayed pulse with a wavelength  $\lambda_2^*$  with the observation of, for example, the dependence of the luminescence intensity at the wavelength  $\lambda_{det}$  on the delay time. The results of the experiment are shown in Fig. 13, where one sees a large number of oscillations of the detected signal, corresponding to long-term oscillations of a particle-like packet. Besides the oscillations with the classical period of  $\sim$  300 fs, well-defined pulses in the envelope of the signal are observed. The interpretation of these results is given in Ref. 56, and is based on the fact that the vibrational spectrum of term B in the group of levels populated by the pump pulse at  $\lambda_1$  are not equally spaced. In this case the detected signal I(t) is represented in the form of a Fourier expansion

$$I(t) = \sum_{i,i} A_{ij} \cos \omega_{ij} t, \qquad (43)$$

where the values of  $A_{ii}$  depend on the parameters of the excitation and the delayed laser pulses and of the three terms that are involved in the experimental scheme. The frequency  $\omega_{ii}$  depends only on the mutual arrangement of the vibrational levels in the term B. It is obvious that in the case of a spectrum with equal spacing we have  $\omega_{ij} = \omega$ ,  $2\omega$ ,  $3\omega$ ,..., where  $\omega$  is the frequency of the harmonics of the nuclear vibrations. The inverse Fourier transformation of the measured signal in fact showed that there were several different vibrational frequencies in the expansion (43) (Fig. 14). The beats on the envelope of the signal are a consequence of this multi-frequency character (the non-equidistance of the vibrational spectrum). We note that the time resolution in the experiment of Ref. 56 was not sufficient to observe frequencies at multiples of the frequency spacing between adjacent energy levels, since the length of the readout pulse was not short enough. The interesting possibility of solving the inverse problem of spectroscopy-reconstructing the adiabatic potentials of molecules from the data of the time variation I(t)—has been discussed in Refs. 56 and 59. For instance, by taking the inverse Fourier transform of the signal I(t) it is

possible to find the mutual positions of the vibrational levels over an extremely broad energy region<sup>56</sup> (of the order of the energy width of the excitation pulse), which permits the solution of the inverse problem with the use of well-known methods.<sup>128</sup> A significant advantage of this "time-domain Fourier spectroscopy" over direct spectroscopic methods of determining the vibrational spectrum in the term B is, for example, that I(t) contains information on the interlevel beats in the term B, independently of the positions of the analogous levels in the other terms that are involved in the experimental scheme. It is difficult to obtain independent information by ordinary spectroscopic methods that investigate the resonance transitions between pairs of levels that belong to different terms. The advantages and disadvantages of the spectroscopy described in this section, based on pulsed excitation, and the study of the time dependence of the responses of the molecule have been discussed thoroughly in Ref. 70.



FIG. 12. Potential curves and the excitation scheme for the  $I_2$  molecule (from Ref. 55).

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FIG. 13. Dependence of the absorption signal on the delay time between the excitation and probe pulses in experiments with  $I_2$  molecules (from Ref. 55).

The experiments with molecules, described above, thus give an affirmative answer to the question posed in the introduction as to whether it is possible to observe classical vibrations of nuclei, and they also demonstrate the behavior of the post-classical stages of the nuclear motion. The situation with regard to the observation of these effects in molecules is, for a variety of reasons, simpler than in the case of the Rydberg atoms, discussed in Section 4. First, the presence of a single vibrational degree of freedom in a diatomic molecule simplifies considerably the conditions of the excitation of particle-like spatially localized packets. The only requirement is essentially that a large enough number of levels be involved in the formation of the packet. This is achievable by virtue of the spectral width of a short excitation pulse. This discussion also pertains to multiatom molecules if one-dimensional bonds are optically excited in them. We recall that the main obstacle to the solution of the analogous problem in hydrogen-like atoms is its three-dimensional nature.

Second, a feature of molecular vibrational spectra is the presence of extremely broad energy regions containing a large number of practically equidistant levels. This circumstance makes it possible to create packets that undergo periodic oscillations for a long time without dispersing. These groups of levels can be found far from the dissociation threshold, which distinguishes molecules favorably in this respect from Rydberg atoms, where the necessary levels lie at the edge of the continuum.

We note also that the general arguments in Section 2, based on very simple assumptions about the energy spectrum, can be transferred unconditionally to one-dimensional molecular wave packets. The experiments to which we have referred also serve to illustrate these arguments, but of course they do not exhaust all the aspects of laser femtochemistry. The majority of theoretical works in this field contain calculations that take into account the features of the spectra of the specific molecules for various methods of excitation. Calculations of this sort are necessary for a quantitative analysis of experimental results and are dictated by the large variety of molecular spectra. This also distinguishes the situation with molecules from that of hydrogen-like atoms, where the Rydberg states have a universal character. Readers that are interested in specific calculations will find them in the works cited above. A detailed discussion of these calculations is beyond the scope of this review, which is devoted to the general aspects of the dynamics of wave packets.

### 5. PACKETS OF QUANTUM STATES OF THE ELECTROMAGNETIC FIELD IN NONLINEAR-OPTICS SYSTEMS. THE EVOLUTION OF PACKETS OF STATIONARY STATES IN A STRONGLY COUPLED "ATOM + FIELD" SYSTEM

As we have noted above, the features of the long-term evolution of packets of stationary states of nonlinear quantum systems (with nonequidistant spectra) have a quite uni-



FIG. 14. Fourier transform of the absorption signal in experiments with  $I_2$  molecules (from Ref. 56). The arrows indicate the vibrational frequencies that correspond to adjacent energy levels with vibrational quantum numbers v. versal character, and show up in many details even in the very simple case of a one-dimensional oscillator with a weak anharmonicity. It therefore appears natural and reasonable that these phenomena were first encountered in investigations of the states of the electromagnetic field generated by nonlinear-optics media, where these states can be described by a model of a weakly anharmonic oscillator. In connection with this topic a number of publications<sup>87–91,93,94</sup> have dealt with the long-term behavior of packets of stationary states of a field oscillator with the Hamiltonian

$$H = \hbar \{\omega(a^{+}a) + \lambda(a^{+}a)^{2}\},$$
(44)

where  $\omega$  is the frequency of the mode of the electromagnetic field and  $a^+$  and a are the creation and annihilation operators. The Hamiltonian (44) is used to describe propagation of light through a nonlinear light guide, where the nonlinear term, proportional to  $\lambda$ , is due to the Kerr nonlinearity of the medium.<sup>129</sup> In Refs. 87 and 88 Milburn and Holmes studied the time dependence of the so-called *Q*-function

$$Q(\alpha, \alpha', t) = \frac{1}{r(\beta(t)|\alpha|, \alpha|)}.$$

where  $\hat{\rho}(t)$  is the density matrix of the system with the Hamiltonian (44) and  $|a\rangle$  is a coherent state of the harmonic oscillator. It was assumed that at the initial instant of time the system was in the coherent state  $|a_0\rangle$ . The quantity  $Q(a,a^*,t)$  represents the localization of the wave function of the system in phase space. A direct numerical calculation carried out in Ref. 87 showed that the initial localization Q is a function that disperses with time, undergoing interference beats, but then recovers its localization, forming regular spatial structures (we note also the work of Ref. 92, in which analogous behavior was observed for a nonlinear oscillator subjected to an external periodic force). The physical reasons for this behavior were discovered in Ref. 89, where it was shown that in the evolution of the initial coherent state in a system with the Hamiltonian (44) a linear superposition is formed of two coherent states shifted from each other by  $\pi$ (one of them is retarded from the other by half an oscillation period  $T = 2\pi/\omega$ ). In accordance with the analysis performed in Refs. 31 and 32 (see also Section 2 of this review), this corresponds to fractional revival of order 1/4. States of this kind have been examined previously from a formal point of view in Ref. 130, where they were called generalized coherent states. Yurke and Stoler<sup>89,91</sup> suggested the use of an effect that they observed for the generation of nonclassical states of light that maintain their quantum mechanical properties for a macroscopically large average number of photons. A similar effect was also observed in the work of Mecozzi and Tombezi<sup>93,94</sup> in a study of quantum systems simulating optical processes in media with a nonlinear birefringence, and by Yurke et al.<sup>90</sup> in their analysis of a quantum mechanical model of a four-wave mixer. In Ref. 94 Tombezi et al. suggested the possibility of multipacket structures that form during the evolution of the initial state at times  $t \approx T_{rev}/2^k$ , where k = 2,3,... although they were unable to give them an analytical description. The solution of this problem is also a part of the universal scenario of fractional revival.31,32

To detect such macroscopic states of light that preserve their essentially quantum mechanical properties (for this reason they are sometimes called "optical Schrödinger

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cats<sup>\*\*89-91</sup>), Yurke and Stoler<sup>89-91</sup> proposed to use the phasesensitive technique of homodyne detection.<sup>131-133</sup> In this method of measurement the electromagnetic field that is studied is mixed at the surface of the photodetector with a powerful reference beam at the same frequency with a phase  $\vartheta$  that is monitored by the experimenter and can be varied. The statistical distribution of the total number of photoelectrons counted by the detector in a specified time reflects the multipacket character of the state of the initial field in Fock space. The quantum mechanical coherence between the packet fractions shows up as interference beats in the statistical distribution of the total number of photon counts for specific values of  $\vartheta$ .<sup>89,91,94</sup>

The problem discussed in this review should also include investigations of the long-term evolution of packets comprised of stationary states of a strongly coupled "atom + field" system (see. e.g., the review in Ref. 95). With the use of a two-level (or a several-level) model of the atom it is not possible, of course, to follow the transition to a completely classical description of the resonance interaction of light with matter. However, the features of the long-term evolution of packets, their destruction and revivals are in many ways similar to those that have been discussed above, and they have similar causes and manifestations.

The simplest completely quantum mechanical model of the interaction of light with matter, the Jaynes–Cummings model, describes the behavior of a single two-level atom that is coupled to a single mode of the quantized electromagnetic field.<sup>134,135</sup> In the resonance approximation the Hamiltonian of the model has the form

$$H = \frac{1}{2} \hbar \omega_0 \sigma_3 + \hbar \lambda (\sigma_+ a + \sigma_- a^+) + \hbar \omega (a^+ a + \frac{1}{2}); \qquad (45)$$

where  $\sigma_3$  and  $\sigma_{\pm}$  are the Pauli matrices,  $a^+$  and a are the Bose creation and annihilation operators for quanta of the field mode, and the frequency  $\omega$  of the field is close to the frequency  $\omega_0$  of the atomic transition  $|\Delta| \ll \omega_0$ ,  $\omega$ ,  $\Delta = \omega_0 - \omega$ , and  $\lambda$  is the coupling constant. For  $\lambda = 0$  the eigenstates of the Hamiltonian (45) are  $|1,n\rangle = |1\rangle|n\rangle$  and  $|2,n\rangle = |2\rangle|n\rangle$ , where the vectors  $|1\rangle$  and  $|2\rangle$  describe an atom in the ground and excited states, respectively, and  $|n\rangle$ is the *n*-photon state of the field oscillator. Since the Hamilton (45) couples states of type  $|2, n\rangle$  only to states  $|1, n + 1\rangle$ , the eigenvectors, the energy spectrum, and the time dependence of the wave function of the system can be found exactly. If, for example, at time t = 0 the state vector is

$$|\psi(0)\rangle = |1\rangle \sum_{n=0}^{\infty} a_n |n\rangle, \tag{46}$$

then in the simplest case of exact resonance ( $\Delta = 0$ )

$$|\psi(t)\rangle = \sum_{n=0}^{\infty} \{a_n \cos(\Omega_n t) | 1, n\} \exp(-in\omega t)$$
$$-ia_{n+1} \sin(\Omega_n t) | 2, n\} \exp[-i(n+1)\omega t]\}, \qquad (47)$$

 $\Omega_n = \lambda n^{1/2}.$ 

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It follows from (47) that the population of the ground state of the atom varies with time as

$$n_1(t) = \sum_{n=0}^{\infty} |a_n|^2 \cos^2 \Omega_n t.$$
 (48)

We shall assume that the coefficients  $a_n$  of the initial field packet have maximum moduli for  $n = \bar{n} \ge 1$  and fall off rapidly with distance from this central value by an amount  $\Delta n$ ;  $(1 \le \Delta n \le n)$ . As long as the cosines in expansion (48), corresponding to transitions between the various components of the packet, oscillate in phase, we can take this oscillating function out from under the summation sign with the average value  $n = \bar{n}$ , and using the normalization condition  $\sum_n |a_n|^2 = 1$  obtain

$$n_1(t) \approx \cos^2(\Omega_{\bar{n}}t). \tag{49}$$

Expression (49) describes the well-known Rabi beats<sup>113,119</sup> at the frequency  $\Omega_{\bar{n}} = \lambda \bar{n}^{1/2}$  in the population of a two-level system subjected to the action of a classical electromagnetic field. The condition for the validity of this formula here is

$$\left. \frac{\mathrm{d}\Omega_n}{\mathrm{d}n} \right|_{\bar{n}} (\Delta n) t \ll 1,\tag{50}$$

that is,

$$t \ll t_0 \propto \left( \frac{\mathrm{d}\Omega_n}{\mathrm{d}n} \bigg|_{\bar{n}} \Delta n \right)^{-1}.$$
 (51)

For an initial packet that has the form of a coherent state of the field mode,  $t_0 \sim \lambda^{-1}$  and does not depend on  $\bar{n}$  (this is correct only in the case of exact resonance<sup>96,97</sup>).

For  $t \gtrsim t_0$  large phase shifts accumulate between the various terms in (48) because  $\Omega_n$  is not constant, and as a result the contributions of all these terms become averaged over a background of a smooth distribution of  $a_n$ . The Rabi beats cease (the so-called collapse<sup>136</sup>), and the populations of the states of the atom no longer depend on the time  $(n_2(t) = n_1(t) = 1/2)$ . The form of the envelope of the collapsing Rabi beats in the case of exact resonance was studied in Ref. 136, and for the case of detuning in Refs. 96 and 97. The phenomenon of collapse, like the destruction of the wave packet discussed above, is due to the dependence of  $\Omega_n$  on n

$$\Omega_n = \Omega_{\bar{n}} + \frac{\mathrm{d}\Omega_n}{\mathrm{d}n} \Big|_{\bar{n}} \Delta n + \dots$$
 (52)

It was first pointed out in Refs. 96 and 97 that for  $t \sim T_{\rm rev} = \pi (d\Omega_n/dn|_{\bar{n}})^{-1} \gg t_0$  additional phase shifts between the various contributions to the sum (48), due to  $\Omega_n$ not being a constant, become multiples of  $\pi$ . The initial time variation of the beats in the population is approximately recovered (they are revived) and then they again collapse. Approximate revivals also occur for  $t = mT_{rev}$  (m = 2, 3, 4,...;Fig. 15). The form of the envelopes of the beats near each revival, and also the pulsations that occur where these envelopes begin to overlap, have been studied in detail in Refs. 97, 137, and 138. Similar behavior appears also in the time-dependent dynamics of the average dipole moment of an atom. In the case of two-photon resonance of an atom with a field mode, the approximate periodicity of the revival becomes exact.<sup>139</sup> The revival of the Rabi pulsations in the Jaynes-Cummings model is due to the discrete nature of the energy of the quantized electromagnetic field and is a purely quantum mechanical phenomenon. The collapse and revival of the Rabi nutations were first observed in the experiments of Rempe et al.<sup>140</sup> (see also the review of Ref. 141), in which a beam of rubidium atoms in the  $63p_{3/2}$  state was transmitted





FIG. 15. Long-term evolution of the atomic inversion  $W(t) = n_2(t) - n_1(t)$  (from Ref. 97) a) collapse; b) revival of the Rabi beats.

through a high-Q superconducting microwave cavity. The transition between the Rydberg states  $63p_{3/2} - 61d_{5/2}$ , to which the cavity was tuned, corresponded to the two-level system of the Jaynes-Cummings model. The number of nonequilibrium photons in the mode arising in the transition of the atoms to the lower level is determined by the flux of atoms through the cavity. The measurements were made for various values of the flux while no more than one atom was present in the cavity at one time. The time of interaction of the atoms with the field (the transit time of an atom through the cavity) was controlled by varying the velocity of the atom beam. The population of the atoms in the upper state after passing through the cavity was monitored by the ionization signal. For low fluxes Rabi beats were observed in the time dependence of the atomic inversion. As the flux was increased the beats smoothed out, and then they appeared again, which was interpreted as collapse and revival.

The nontrivial time dynamics of the averaged quantities (level population, average dipole moment of the atom) is directly connected to the restructuring of the packet of states of the field oscillator. Let us demonstrate this point by arguments similar to those used in the analysis of fractional revival in Section 2. We shall consider as an example the shape of that part of the wave function of the field that corresponds to an atom in state 1 after a time t

$$|\psi_{1}(t)\rangle = \frac{1}{2}|1\rangle \sum_{n=0}^{\infty} a_{n} \exp(-i\Omega_{n}t - in\omega t)|n\rangle + \frac{1}{2}|1\rangle \sum_{n=0}^{\infty} a_{n} \exp(i\Omega_{n}t - in\omega t)|n\rangle$$
(53)

(one can determine the intracavity field in this state by detecting the atoms exiting from the cavity after an interaction time t, and selecting only those events for which the atom is in state 1). The probability of detecting an atom in the lower energy state is, of course equal to  $|\langle \psi_1(t) | \psi_1(t) \rangle|^2$ . If we take into account that at the initial instant of time the field is in a coherent state and if the deviation of  $\Omega_n$  from  $\Omega_{\bar{n}}$  is ignored, then we see that  $|\psi_1(t)\rangle$  is a superposition of two coincident packets that are Gaussian in the "space" of the field oscillator and are oscillating in phase

$$\begin{aligned} \langle 1 | \psi_1(t) \rangle &\approx \frac{1}{2} \exp(-i\Omega_{\tilde{n}}t) | \alpha \exp(-i\omega t) \rangle \\ &+ \frac{1}{2} \exp(i\Omega_{\tilde{n}}t) | \alpha \exp(-i\omega t) \rangle. \end{aligned}$$

If we keep the terms in (52) that are of first order in  $(n - \overline{n})$ , the resulting expression gives an effective renormalization of the oscillation frequency of the packet  $\omega \rightarrow \omega \pm d\Omega_n/dn|_{\overline{n}}$ . As a result, the initial in-phase packets in (53) move apart and no longer overlap, so their interference is eliminated and the population beats collapse. After a time  $T_{rev}$  the packets again intersect, and the beats are regenerated. A similar relation between the phenomena of collapse and revival in the Jaynes-Cummings model with the splitting of the field packet was recently examined theoretically in Refs. 142 and 143 in studies of the long-term behavior of the Q-function of an electromagnetic mode.

#### 6. CONCLUSIONS

The study of the dynamics of packets composed of highly excited stationary states of various quantum systems has produced a number of extremely important results in recent years. In experiments with pulsed laser excitation of atoms and molecules it became possible to observe effects that belong to the boundary between quantum- and classical physics. It became practical to prepare localized wave packets moving according to quasiclassical laws and describing almost classical oscillations of nuclei in molecules and Keplerian motions of an electron in an atom. Essentially for the first time since the development of the quantum mechanics of atomic particles the correspondence principle and the bounds of its applicability became a subject for direct experimental investigation. It has been shown theoretically and experimentally that the long-term "post-classical" evolution of wave packets follows a very universal course, with the formation of macroscopic quantum structures that replace one another with the passage of time. The mechanisms for the formation of coherent packet structures in wave functions of atomic particles prove to be closely tied to the mechanisms, related to the essentially quantum mechanical nature of the electromagnetic field, for the generation of nonclassical states of light in nonlinear-optics systems. These advances as a whole open up new prospects for testing the fundamental propositions of quantum mechanics by optical methods. In addition, potential practical applications of this set of phenomena have been found.

The investigations of packet dynamics in the aspect described above are far from complete. It would, therefore, be rash to make extensive predictions about their further development. We note only certain problems. It would be of importance to prepare a packet of Rydberg states of an atom, localized in both the radial and the angular variables, and thus comprising a particle-like entity moving in a Kepler orbit. This, of course, would at the same time be the first realization of the Rutherford atom, a concept that played such an important role in the formulation of quantum mechanics. Also of great importance are investigations of the long-term evolution of packets from the point of view of the relation between the phenomena of dynamic chaos in classical and quantum mechanical systems. Considering the universality of the laws of evolution of wave packets in quantum mechanical systems with a discrete spectrum, it would be of considerable interest to include in the list of investigated topics semiconducting quantum-size-effect structures such as quantum wells. The use of these microelectronics structures with quantum mechanical properties presents still another route for passing into this boundary region by way of the physics of the so-called mesoscopic phenomena. This route is in essence an alternative to that described in this review, since the transition to the mesoscopic region is from the direction of the macroscopic region.

We are grateful to N. B. Delone, who suggested this review, and also to J. Eberly, H. Metiu, K. Stuart, A. Mecozzi, P. Tombezi, B. Yurke, and P. Zoller for kindly providing reprints of their latest publications, including those in press.

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Translated by J. R. Anderson

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