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# Chaotic Behavior in Quantum Systems

## Theory and Applications

Edited by

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Plenum Press
New York and London
Published in cooperation with NATO Scientific Affairs Division

Proceedings of a NATO Advanced Research Workshop on
Quantum Chaos: Chaotic Behavior in Quantum Systems, Theory and Applications,
held June 20–25, 1983,
in Como, Italy

Library of Congress Cataloging in Publication Data

NATO Advanced Research Workshop on Quantum Chaos: Chaotic Behavior in Quantum Systems, Theory and Applications (1983: Como, Italy)
Chaotic behavior in quantum systems.

(NATO ASI series. Series B, Physics; v. 120)
Proceedings of a NATO Advanced Research Workshop on Quantum Chaos: Chaotic Behavior in Quantum Systems, Theory and Applications, held 6/20–25/83 in Como, Italy.
"Published in cooperation with NATO Scientific Affairs Division."
Bibliography: p.
Includes index.
1. Chaotic behavior in systems—Congresses. 2. Quantum theory—Congresses. I. Casati, Giulio, 1942– . II. Title. III. Series.
QC174.84.N37 1983 530.1'2 84-26351
ISBN 0-306-41898-3


A Division of Plenum Publishing Corporation
233 Spring Street, New York, N.Y. 10013



Printed in the United States of America

# QUANTUM DIFFUSION LIMITATION AT EXCITATION OF RYDBERG ATOM IN VARIABLE FIELD

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Abstract

A computer simulation method is used to investigate excitation of a hydrogen-like atom in the field of an electromagnetic wave from the states with $n \sim 50$ and parabolic quantum numbers $n_1 >> n_2$ (or $n_1 << n_2$). It is shown that diffusion over levels is much slower than in the classical case. A significant part in atom excitation is played by multiphoton resonances with the number of photons $k \gtrsim 10$.

## 1. INTRODUCTION

For the last few years a number of interesting experiments was carried out on ionization and excitation of hydrogen atoms by the microwave field from the states with the main quantum number $n \sim 50$ [1-3]. The first of these experiments showed at the frequency of $\omega/2\pi = 9.9$ GHz, a strong ionization of atoms with $n \simeq 65$ at the electric field $\varepsilon \gtrsim 0.06\, n^{-4}$ and adiabatic tunneling parameter $\gamma = \omega/\varepsilon\, n \simeq 7$ [4] ($\omega \simeq 0.43\, n^{-3}$)*. To explain the results of the experiment a diffusion mechanism of atom ionization was offered in [5]. The cause of electron diffusion arising in an atom due to the purely monochromatic field effect can be understood in the following way. It is quite possible to assume [6] that at $n \simeq 50 >> 1$ the quasiclassical approximation is good enough and to pass on to investigation of the classical system. The latter is substantially nonlinear and at field $\varepsilon > \varepsilon_{cr}$ there arises a stochasticity leading to

* Here and below we use atomic units; $n, \ell, m$ are principal, orbital and magnetic quantum numbers.

a diffusion excitation of an electron [7]. In case of linear and circular polarizations and $\omega \approx \Omega = n^{-3}$ the estimation for $\varepsilon_{cr}$ was obtained in [7,8] on the grounds of the overlap criterium [9]. Dependences of critical field and diffusion rate on frequency are given in [10,11]. Despite all attractiveness of this classical approach, its only justification is the coincidence of the relative portion of ionized atoms at $\varepsilon \, n \approx 0.06$ [1] with the probability of ionization obtained through computer simulation of classical system's dynamics [6]. The accuracy of coincidence is about 30%.

At the same time a number of numerical experiments with simple quantum systems [12,13] have shown that statistical properties of quantum dynamics are much weaker than those of classical ones. Moreover, in the course of time the effect of quantum corrections increases [14] and leads to diffusion's slowing down and its final almost complete stopping. First this effect of quantum diffusion limitation was observed in the model of quantum rotator [12]. The same phenomenon can take place at diffusion ionization of atoms with $n \gg 1$. Preliminary estimates [11] show that relative magnitude of the quantum corrections can be compared to unity and so there may be a significant distinction between quantum dynamics of atom excitation and classical ones.

## 2. THE MODEL OF SURFACE-STATE ELECTRONS

When investigating quantum and classical dynamics we employed the computer simulation method. We chose an external field linear polarized along the axis z. The atom was initially excited into states with quantum numbers which satisfied the following conditions $n \sim 50$, $m = 0$, $n_1 \gg n_2$ (or $1 \lesssim \ell \ll n^{2/3}$). For these states the ratio of cross-size to longitudinal one is small and that is why it is possible to describe the dynamics of excitation in this case by the one-dimensional Hamiltonian

$$H = \frac{p^2}{2} - \frac{1}{|z|} + \varepsilon z \cos \omega t \qquad (1)$$

with the boundary condition $\Psi(0) = 0$, which corresponds to an infinite potential wall at $z = 0$. This Hamiltonian also describes the exitation of states of atom with $\ell = 0$ in the external field with the potential $V = \varepsilon |r| \cos\omega t$. Consequently for Equation (1) matrix elements $z_{nn'} = r_{nn'}$. To prove the fact that such a modification of the initial Hamiltonian will not lead to any significant change of the dynamics we can present the following arguments:

1. When $1 \leqslant \ell \ll n^{2/3}$ the matrix elements $z_{nn'}^{2\ell\pm1}$ do not depend on $\ell$ [15], as a result of which their sum $z_\varepsilon$ that determines the probability of transition from one shell into another in dipole approximation to within several per cent is equal to $r_{nn'}$ ($n, n' \sim 50$). So, for $n = 40$, $n' = 41$, $\ell = 2$, the ratio $z_\varepsilon / r_{nn'} = -0.996$ (here we use for $z_\varepsilon$ its quasiclassical value).

2. Due to Coulomb degeneration the transition frequency between adjoining sublevels of one and the same shell prove much smaller than Kepler frequency: $3\varepsilon n/2\Omega \lesssim 0.1 << 1$ for $\varepsilon_o = \varepsilon n^4 \lesssim 0.06$. So, despite the fact that the width of Stark multiplet exceeds the distance between the shells $\Delta\Omega \gtrsim \Omega$, the motion of sublevels proves synchronized and it is possible to substitute the whole shell for one level. A proof of such approximation is given by the experiments [2,3], where the observed resonant picture of excitation in the field with $\varepsilon_o \approx 0.06$ corresponded to the unperturbed spectrum of atom levels.

3. In the classical system when $\ell << n$ the dependence of $\varepsilon_{cr}$ and the rate of diffusion on $\ell$ proves insignificant [11]. The Hamiltonian (1) describes precisely the motion for the orbits with eccentricity $e = 1$ stretched along the field. In the quantum system the states with parabolic quantum numbers $n_1 = n - 1$, $n_2 = 0$ (or vice versa) and close to them ($n_1 \overset{<<>>}{} n_2$) correspond to such classical orbits. Let us see if this condition will be violated in the course of time. To do this we shall consider unperturbed ($\varepsilon = 0$) phase-action variables corresponding to parabolic quantum numbers. For these variables

$$Z = \frac{3}{2} n(n_1 - n_2) + \sum_{\substack{m_1,m_2 \\ m_1+m_2\neq 0}} B_{m_1m_2}\exp[i(m_1\lambda_1 + m_2\lambda_2)]$$

$$B_{m_1m_2} = \frac{1}{m_1+m_2}\left[\mu_2 \mathcal{J}_{m_1}(\mu_1(m_1+m_2))\mathcal{J}'_{m_2}(\mu_2(m_1+m_2)) - \right. \tag{2}$$

$$\left. - \mu_1 \mathcal{J}'_{m_1}(\mu_1(m_1+m_2))\mathcal{J}_{m_2}(\mu_2(m_1+m_2))\right],$$

$\mu_{1,2} = [n_{1,2}(n-n_{2,1})/n^2]^{1/2}$; $\lambda_1$, $\lambda_2$ - phases conjugated to $n_1, n_2$; where the derivative is taken over the argument of Bessel function. According to the correspondence principle, the values $B_{m_1m_2}$ determine matrix elements $z^{n_1'n_2'}_{n_1n_2}$ of the transition between the unperturbed levels with $m_{1,2} = n'_{1,2} - n_{1,2}$. When $n_1 >> n_2$ ($m = 0$) matrix elements for the transitions with the change of $n_2$ contain a supplementary small parameter $n_2/n$. The change magnitude of $n_2$ can be estimated in the course of time in the following way. For $\omega \eqsim \Omega$ and $\varepsilon > \varepsilon_{cr}$ there starts a diffusion electron excitation. According to the theoretical estimates [11] the rate of diffusion over $n$ and the value of $\varepsilon_{cr}$ are:

$$D_n = \frac{d(\Delta n)^2}{dt} \eqsim \frac{1}{3}\varepsilon^2 n^7; \quad \varepsilon_{cr} \eqsim \frac{1}{84n^4}. \tag{3}$$

When $\varepsilon > \varepsilon_{cr}$ the phase $\lambda_1$ changes randomly in the course of time, which makes it possible to estimate the rate of diffusion over $n_2$ : $D_{n_2} \sim (n_2/n)^2 D_n$. From the above we see that in the course of a numerical experiment $\tau = \omega t/2\pi \lesssim 50$ at $\varepsilon n^4 \eqsim 0.04$, $n_2 \sim 1$ the change

$\Delta n_2 \sim 1$ and so the one-dimensional approximation is not violated. It is interesting to note that when $\varepsilon n^4 n_2 \ll 1$ the dynamics over the second degree of freedom ($n_2$, $\lambda_2$) prove substantially quantum, since in this case the perturbation theory can be used [16]: $B_{m_1 m_2}/\Omega \ll 1$ ($m_2 \neq 0$). Let us point out the fact that when $m_1 + m_2 = \text{const} \sim 1$ the matrix elements $B_{m_1 m_2}$ with $m_1 - m_2 \gg 1$ are exponentially small. This proves still more rigorously the effect observed in the experiments where the shell shows itself as one level [2,3].

To determine in quantative terms how good is the selected one-dimension approximation, we made a comparison with the results of numerical experiments for a real atom in a linear polarized field carried out by F. M. Izrailev. The initial condition was given by parabolic quantum numbers $n_1 = n_o - 1$, $n_2 = 0$. The comparison was made for $n_o = 10$, 9 and showed a good agreement with the results obtained in a one-dimensional approximaton. So, for instance, the probabilities of excitation $W_{n>n_o}$ into states with $n > n_o$ coincide with the accuracy of 10 - 20%.

Hence, proceeding from the above arguments we can conclude that the system (1) will describe in qualitative and quantitative terms the dynamics of Rydberg atom excitation from states $n_1 \gg n_2$ or $n_1 \ll n_2$ (and qualitatively from $1 \lesssim \ell \ll n^{2/3}$). Besides, this model describes exactly the dynamics of surface-state electrons excitation [17] in a variable field, directed perpendicular to the surface of liquid helium and that is why its investigation is of interest in itself. It was offered to carry out an experiment with such a system in reference [18]*.

## 3. NUMERICAL EXPERIMENTS

Simulation of quantum dynamics of system (1) was carried out in the following way. At the initial moment of time one level of the unperturbed system with $n_o \sim 50$ was excited. The field frequency was close to the Kepler one: $\omega n_o^2 = \omega_o \sim 1$ and the field $\varepsilon n_o^4 = \varepsilon_o \sim 0.06$. Further on we carried out a numerical integration of equations for amplitudes $C_n$ of the unperturbed Hamiltonian's states,

$$i\dot{C}_n = -\frac{1}{2n^2} C_n + \sum_{n'=n_{min}}^{n_{max}} V_{nn'} C_{n'}. \qquad (4)$$

Thus, levels with $n_{min} \leqslant n \leqslant n_{max}$ were only involved in the dynamics. Since at the cited values of parameters $\varepsilon$ and $\omega$ diffusion in the classical system moves up along n [11], levels with $n < n_o$ are weakly

* We note that the functional dependence $\varepsilon_{cr}(\omega)$ in [18] was not found correctly (there is a mistake in f.(8)). See for a correct answer [11].

excited and that is why the accepted $n_o - n_{min} < 10$ proved sufficient. During all the time the probability $W_{n_{min}} = |C_{n_{min}}|^2$ was at the level $10^{-4}$, and a further decrease of $n_{min}$ did not influence the dynamics of excitation. The working value of $n_{max}$ equal to $\approx 2n_o$ was selected in most cases. At the same time a number of control experiments with $n_{max} \approx 2.7n_o$ was carried out. A good agreement of working and control points (see below) shows that in order to investigate electron excitation into states of the discrete spectrum within the time $\tau \approx 40$ periods of an external field, we can restrict ourselves to a finite number of levels with $n_{max} \approx 2n_o$. At values $n_{max} \approx 2n_o$, $n_o \approx 50$ and $\omega \approx \Omega$ to come out into a continuous spectrum it is necessary to absorb five photons more as compared to states $n \approx n_{max}$. That is why it is quite natural to expect that the probability of multiphoton ionization will be relatively small. Besides, the main aim of the experiments was to investigate the diffusion mechanism of excitation, occurring for discrete states only.

In the process of numerical integration probabilities $W_n = |C_n|^2$ were found, according to which the first $M_1 = \langle n - n_o \rangle / n_o$ and the second $M_2 = \langle (n - n_o)^2 \rangle / n_o^2$ moments of distribution were determined as well as $W_{n>n_o+4}$ - the probability of excitation in states with $n > n_o + 4$ and $W_{n \geqslant [1.5n_o]}$ - in states with $n \geqslant [1.5n_o]$ where brackets indicate an integer part. The step of integration $\Delta t$ was determined from the condition $\varepsilon z_{n_{max}n_{max}} \Delta t \lesssim 0.3$ and it was usually $\approx 200$ times less than an external field period. When it was reduced two times the relative change of cited values and probabilities $W_n > 10^{-2}$ during the time $\tau = 40$ was less than 1% (for $10^{-5} < W_n < 10^{-2}$ the change reached 10%). The accuracy of conservation of the complete probability $W = \Sigma W_n = 1$ was not worse than 0.1%. The calculation of matrix elements $z_{nn'} = r_{nn'}$ was done numerically according to the formules given in [19]. At $n \approx 40$ their agreement with classical values was 2 - 3%. The time of integration of one period of external field at $n_o = 30$, $n_{max} = 63$, $\varepsilon_o = 0.04$, $\omega_o = 1$ was $\approx 15$ seconds of computer ES-1060.

Together with the quantum dynamics simulation a numerical investigation of the classical system was carried out. To exclude the peculiarity at the point $z = 0$, it turned out to be suitable to pass on from unperturbed phase-action variables $(n,\lambda)$ to new variables $(n,\xi)$ and to a new "time" $\eta$, where the equations of motion are:

$$\frac{dn}{d\eta} = -\varepsilon n^2 \cos\omega t \sin\xi, \quad \frac{dt}{d\eta} = 1 - \cos\xi$$
$$\frac{d\xi}{d\eta} = -n^{-3} + 2\varepsilon n \cos\omega t (1 - \cos\xi), \quad \lambda = \xi - \sin\xi. \qquad (5)$$

The initial distribution of classical trajectories in the phase space was a line: $n = n_0$ and equipartition distribution along the phase variable $\lambda$, which corresponded exactly to the initial conditions in the quantum case (one level with $n = n$). The complete number of trajectories was $N = 1000$. In view of the fact that in the classical system there was a finite number of levels a reflection boundary condition at the corresponding $n_{max}$ was introduced in the classical model. However, this had a weak effect on the values $W_{n>n_0+4}$ and $W_{n\geqslant[1.5n_0]}$. So, for instance for $\varepsilon_0 = 0.04$, $\omega_0 = 1$, $\tau = 40$ when reflection was replaced by absorption, their change proved less than one standard deviation. The value of the latter was found by four groups, consisting of 250 classical trajectories equally distributed along $\lambda$. At the selected integration step the accuracy of conservation of energy in a constant field, i.e., at $\omega = 0$, $t \leqslant 80\ \pi n_0^3$, $\varepsilon_0 = 0.06$ was better than $10^{-5}$. It should be mentioned that the classical dynamics depend on scale variables only $\varepsilon_0 = \varepsilon n_0^4$, $\omega_0 = \omega n_0^3$ and at fixed values of the latter ones, they do not depend on $n_0$ [6,20].

## 4. THE RESULTS OF THE EXPERIMENTS

### a) One-Frequency Excitation

The main group of experiments was conducted for values $n_0 = 30$, 45, 66 in the range of parameters $\varepsilon_0$, $\omega_0$ where a strong stochasticity occurs, which leads to a diffusion excitation of a classical electron. However, despite the fact $n_0 >> 1$ the obtained results show that the exitation of an atom is of a fundamentally quantum nature. In fact, as one can see in Figure 1, quantum averages are close to classical ones only for a very short time $\tau < \tau^* \approx 5$ periods of an external field. When $\tau > \tau^*$ a quantum diffusion limitation is observed, because of which the excitation of a quantum atom proves much weaker than that of a classical one. We note that the given effect is not related to the finite number of selected levels. Indeed, for $n_0 = 45$ when $n_{max} - n_0$ is reduced one time and a half (to $n_{max} = 80$), the relative change $M_2$ increases from 4% when $\tau = 10$ to 24% when $\tau = 40$ and the ratio of the classical value $M_2$ to the quantum one changes from 2 to 6. Besides, the effective width of distribution $\Delta n \approx 5^* << n_{max} - n_0 = 52$ and the probability on levels with $n \approx n_{max} = 97$ is $\sim 10^{-6}$. The classical value $(\Delta n)^2$ agrees well with the theoretical estimate (3) $(\Delta n)^2 \approx 2\varepsilon_0^2 n_0^2 \tau$. But to check it in a more detailed way, it is necessary to calculate the local coefficient $D_n$ which goes beyond our paper.

It should be noted, however, that the proximity of the first and the second moments of quantum distribution to their classical values does not at all mean that the excitation occurs due to diffusion. An example of this is the excitation from $n_0 = 30$ at

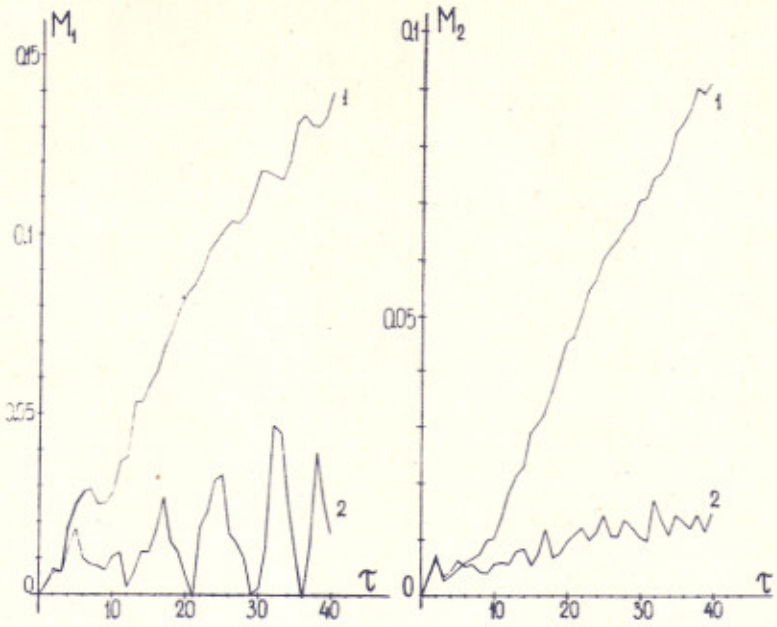


Fig. 1. Dependence of the moments of distribution on time for classical (1) and quantum (2) atoms with $\varepsilon_o = \varepsilon n_o^4 = 0.04$, $\omega_o = \omega n_o^3 = 1$, $n_o = 45$, $n_{max} = 97$.

$\varepsilon_o = 0.04$, $\omega_o = 1$. In this case for $\tau \leqslant 30$ the difference between quantum moments and classical ones is smaller than 20% (except the values $\tau = 4,5,6,7$ where it comes up to 100% for $M_1$). Nevertheless, in the quantum case the dependence of excitation probabilities $W_{n>n_o+4}$, $W_{n\geqslant[1.5n_o]}$ on frequency $\omega_o$ is of a pronounced resonance character and differs fundamentally from the classical one (Figure 2a,c). For $\omega_o = 0.9975$, $\tau = 30$, $n > 34$ maximus of probability distribution fall on levels with $n = 36,37,39,41,44,47,49,52,58$ and 66. These values ($n \neq 36$) approximately correspond to unperturbed resonant transitions from the initial level $n_o = 30$. For the last three values of n the necessary number of photons k is equal correspondingly to 10,11, and 12 (see Figure 2). For $\omega_o = 1.002$, $\tau = 30$ the main contribution (63%) into the probability $W_{n>n_o+4}$ comes from levels with $n = 35$ (33%), 36 (18%) and 37 (12%). Despite the fact that close to the resonance $W_{n\geqslant[1.5n_o]}$ makes up about 20% of the complete probability, control experiments have shown that when $n_{max} - n_o$ increased one time and a half, the change of probabilities of excitation proves insignificant (see Figure 2a,c).

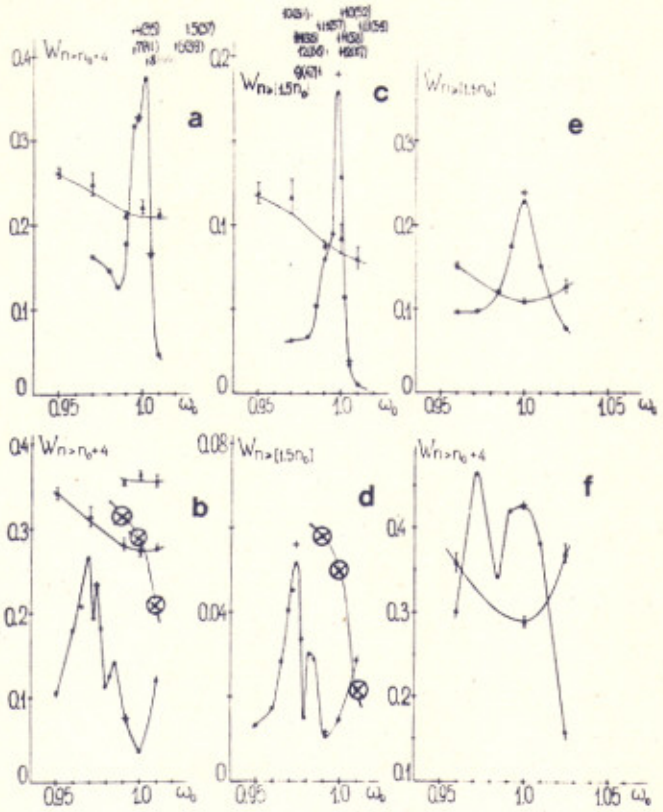


Fig. 2. Dependence of probabilities of atom excitation $W_{n>n_o+4}$ and $W_{n\geqslant[1.5n_o]}$ on frequency $\omega_o$ in the quantum (•) and classical (▲) case. For (a),(c): $\varepsilon_o = 0.04$, $n_o = 30$, $n_{max} = 63$ ((+)-control points for the quantum system with $n_{max} = 80$); (b),(d): $\varepsilon_o = 0.04$, $n_o = 45$, $n_{max} = 97$ ((+) - $n_{max} = 122$); (⊕) - quantum and (x) - classical values for $\varepsilon_o = 0.04$, $n_o = 66$, $n_{max} = 143$ (for $W_{n\geqslant[1.5n_o]}$ classical values (▲) are given in Figure 2c); the values of probabilities are taken at the moment of time $\tau = \omega t/2\pi = 30$. For (e),(f): $\varepsilon_o = 0.06$, $n_o = 45$, $\tau = 12$, $n_{max} = 97$ ((+) - the control point with $n_{max} = 122$). Arrows point to the position of resonances and figures alongside to the number of photons and the number of a resonant level. The curves are given to complete a graphic picture.

Thus, in the investigated case the mechanism of excitation proves not a diffusion but a multiphoton one. At that, up to k = 10 photons can be effectively absorbed. For $\omega_o = 0.9975$, $n_o = 30$,

$\tau = 30$, $0.03 \leqslant \varepsilon_o \leqslant 0.037$ the experimental value $k = 9.973 \pm 0.006$.

For $n_o = 45$, $\varepsilon_o = 0.04$ excitation also is of a pronounced resonant character (Figure 2b,d). However, we cannot connect the observed resonances with multiphoton transitions between unperturbed levels. For $\omega_o = 0.974$, $n > 49$, $\tau = 30$ levels with n = 50-55,57,59, 63,67,72,86 and 93 are the most excited. Even in resonance, absolute values of probabilities of excitation happen to be much smaller than their classical values (Figure 2). However, so far as the field $\varepsilon_o$ grows, this difference decreases and at $\varepsilon_o \approx 0.06$ the probabilities become comparable (Figure 3). Nevertheless, as is seen in Figure 2e,f, even if $\varepsilon_o = 0.06$, the mechanism of excitation is of a resonant nature and results in a more effective excitation than classical diffusion does.

The dependence of the probability of excitation on the frequency of an external field proves rather strong. So, for $n_o = 45$, $\varepsilon_o = 0.03$, $\omega_o = 1$, 1.3 for $\tau = 40$, the value $W_{n \geqslant [1.5n_o]} < 2 \cdot 10^{-3}$ and the probability of excitation is two orders of magnitude higher for

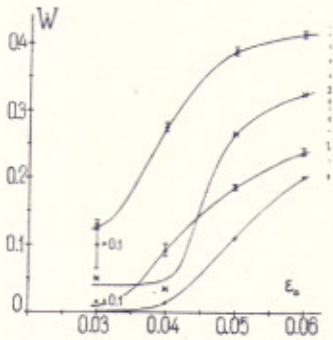


Fig. 3. Dependence of the probability of atom excitation on field $\varepsilon_o$ for $\omega_o = 1$, $n_o = 45$, $\tau = 30$, $n_{max} = 97$. Symbols (●), (*) correspond to quantum probabilities $W_{n \geqslant [1.5n_o]}$, $W_{n > n_o+4}$ and (▲), (♦) to classical ones.

$\omega_o = 1\sqrt{2} \approx 0.707$, $\tau = 30$. In the classical system $W_{n \geqslant [1.5n_o]}$ changes in order of magnitude from 0.016 at $\omega_o = 1$, $\tau = 40$ to 0.12 at $\omega_o \approx 0.707$, $\tau = 30$. However, despite the fact that at $\omega_o \approx 0.707$ the probabilities of excitation and the moments of distribution $M_1$ and $M_2$ are close to their classical values (for $M_1$, $M_2$ at $\tau \leqslant 30$ the difference $\leqslant 30\%$), the dependence $W_{n>n_o+4}$, $W_{n \geqslant [1.5n_o]}$ on the frequency $\omega_o$ differs greatly from the classical one. As it is evident in Figure 4 the excitation of levels with $n \geqslant 67$ occurs in a resonant way, levels with n = 69,76,78,79,80,81 and 82 for $\omega_o \approx \approx 0.707$ being the most excited. All of them except n = 76 ($W_{76} \approx \approx 0.03$) have about the same probability $W_n \approx 0.014$. For the frequency $\omega_o = 0.697$ the probabilities of excitation of these levels are one order less and do not correspond to the positions of maximums $W_n$. An emergence of such a group of strongly excited close states can be qualitatively explained in the following way. After a small number of periods ($\tau \approx 5$) the initial distribution spreads quickly. The width of spreading is equal $\Delta n \approx 7$, and $\langle n \rangle \approx 49$. As when neighboring levels are nearly equidistant for all of them a simultaneous

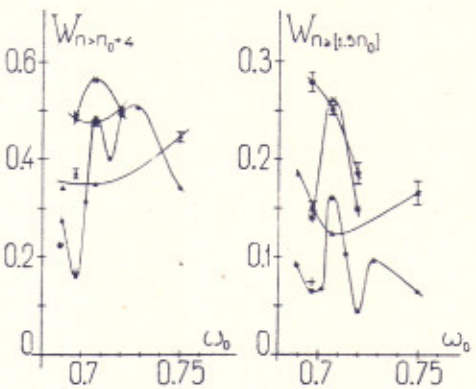


Fig. 4. Dependence of probabilities of atom excitation on the frequency $\omega_o$ when $n_o = 45$, $n_{max} = 97$. Points (●) are quantum values for $\varepsilon_o = 0.03$, $\tau = 30$, (▲) – classical values. The control point (+) corresponds to $n_{max} = 122$. Quantum (*) and classical (♦) values correspond to $\varepsilon_o = 0.04$, $\tau = 15$.

multiphoton excitation can take place. However, it should be noted that the number of photons necessary for that is rather great: $k = (E_{80} - E_{45})/\omega \approx 22$.

As the field increases up to $\varepsilon_0 = 0.04$ the probability of excitation $W_{n \geq [1.5n_0]}$ grows sharply from 0.062 to 0.26 ($\omega_0 = 0.707$, $\tau = 15$). When the frequency changes the values $W_{n>n_0+4}$ are close to classical ones, but for $W_{n \geq [1.5n_0]}$ a resonance dependence on $\omega_0$ is still observed, and the difference from the classical value of probability is 100% (Figure 4).

As the frequency grows, the probability of quantum system excitation becomes smaller than in the classical case. So, when $n_0 = 45$, $\varepsilon_0 = 0.04$, $\omega_0 = 1.3$, $\tau = 50$ in the classical system $W_{n>n_0+4} = 0.359 \pm 0.013$, $W_{n \geq [1.5n_0]} = 0.136 \pm 0.003$ and in the quantum one ($n_{max} = 97$) - 0.181 and 0.051 correspondingly. Quantum moments of distribution $M_1$ and $M_2$ are 2.5 times smaller than classical ones. The same is valid for $\omega_0 = 1.25$.

The growth of the number of initially excited levels up to $n_0 = 66$ does not lead to a qualitative change of dynamics. So, for $\varepsilon_0 = 0.04$, $\omega_0 = 1.01$, $\tau = 35$ the moments of quantum distribution are rather ($\approx 4$ times) smaller than the classical ones. Besides, the quantum distribution breaks sharply at $n > 76$. This is its fundamental distinction from the classical one, which diminishes very slowly at great n. As a result the classical value of $W_{n \geq [1.5n_0]}$ is about 5 times greater than the quantum one (Figure 2d) and $W_{n>76} = 0.23$ is 5.4 times greater. The dependence of the probability of excitation on frequency also differs from the classical one (Figure 2b,d). A significant role belongs here to the multiphoton mechanism of excitation. So, for $\omega_0 = 1$, $\tau = 20$ maximums of distribution $W_n$ at n = 60,76 and 89 approximately correspond to k = 7,8 and 15-photon transitions from initial level (Figure 5).

b) Two-Frequency Excitation

A number of experiments was carried out for the case when an external field has two harmonics: $\varepsilon(t) = \varepsilon(\cos\omega t + \cos\nu t)$. One frequency has been fixed $\omega n_0^3 = \omega_0 = 1$ and the second was being changed in a small range near the values $\omega_1 = \nu n_0^3 = 0.7, 1.3$. For the former one a visible excitation of the classical system occurs at $\varepsilon_0 = \varepsilon n_0^4 \geq 0.015$. For smaller values spreading of the classical packet during the time $\tau = \omega t/2\pi \leq 40$ proves insignificant (Figure 6a). But at $\varepsilon_0 = 0.02$, $\tau = 20$ there appears already a developed stochastisity, leading to a substantial excitation of the system (Figure 6b,c). In the quantum case when $\varepsilon$ grows the width of distribution grows as

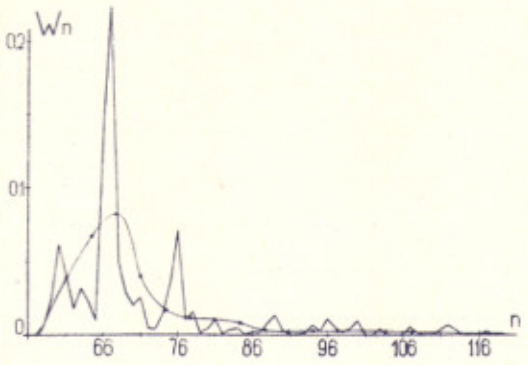


Fig. 5. Distribution over levels in the classical and quantum cases when $\varepsilon_o = 0.04$, $\omega_o = 1$, $n_o = 66$, $\tau = 20$, $n_{max} = 143$. The broken line is the quantum distribution; points correspond to cells of histogram of classical distribution with $\Delta n/n_o = 0.05$, the smooth curve is given to complete a graphic picture.

well (Figure 6) which agrees qualitatively with the idea of the diffusion mechanism of excitation. The probability of excitation of levels close to $n_o$ ($W_{n>n_o+4}$) is close to the classical one (see Figure 7a), and the probability of excitation of high levels of $W_{n\geqslant[1.5n_o]}$ is much greater than that and is of a marked resonance nature (Figure 7c). The latter points to the fact that multiphoton excitation may also be rather substantial in the case of a wide distribution over levels (Figure 6b). In all the investigated cases the quantum distribution turned out to be more cutting than the classical one, peaks $W_n$ in a weak field correspond to resonance transitions (e.g., in Figure 6a the peak at $n = 51$ corresponds to 5-photon resonance with $n_o = 45$).

For $\omega_1 \approx 1.3$, $n_o = 45$, $\varepsilon_o = 0.02$ the quantum system is less excited than the classical one (Figure 7b,d). So, for instance, for $\omega_1 = 1.27$, $n_o = 45$, $\varepsilon_o = 0.02$ the classical value $M_2$ is 3 times less than the classical one, and $M_1$ is 300 times less. When $n_o$ comes up to 66 probabilities of excitation are close to classical ones (see Figure 7b,d for $\omega_1 = 1.31$, $\tau = 40$), but the first moment of quantum

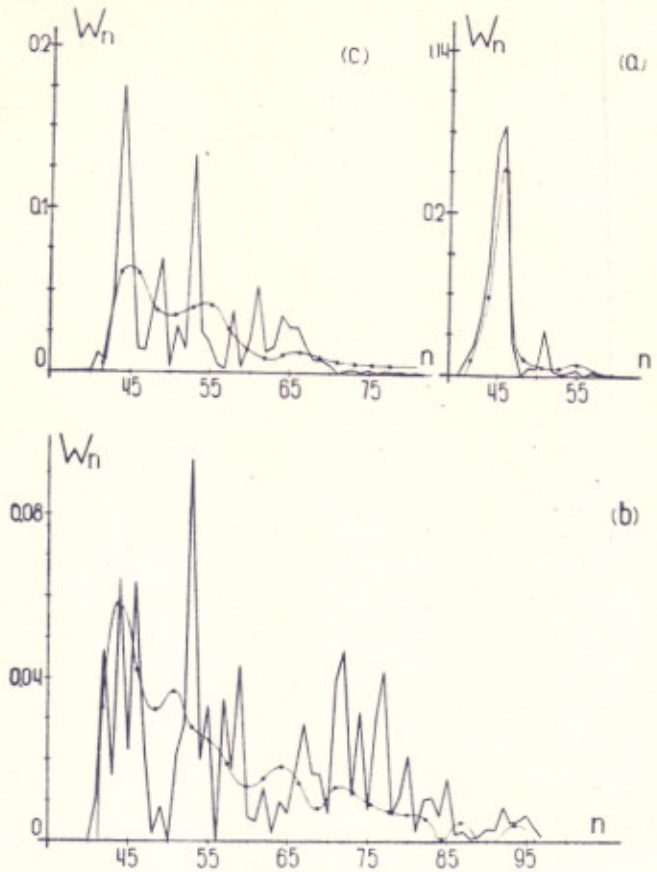


Fig. 6. The same as In Figure 5 but in case of the two-frequency excitation for $n_o = 45$, $\omega_o = 1$, $\tau = \omega t/2\pi = 20$, $n_{max} = 97$. (a) $\varepsilon_o = 0.01$, $\omega_1 = 0.70711$; (b) $\varepsilon_o = 0.02$, $\omega_1 = 0.735$; (c) $\varepsilon_o = 0.02$, $\omega_1 = 0.70711$.

distribution at $\tau > 10$ differs 2 or 4 times from the classical ones and at $10 < \tau < 15$ has another sign. Sharp peaks ($0.08 < W_n < 0.12$ for n = 61,67,70 and 71) about 1.5 - 3 times exceeding the classical value $W_n$ are observed in distribution for $\tau = 40$. So, despite the fact that in the quantum case a substantial spreading of the packet occurs (the excited number of levels $\Delta n \simeq 20$, $M_2^2/M_2^{cl} \simeq 0.8$) quantative characteristics of distribution somehow differ from classical ones.

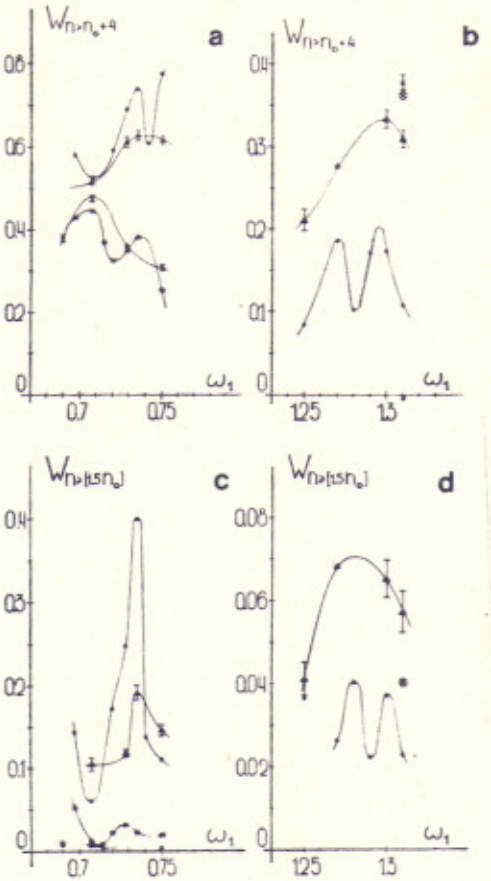


Fig. 7. Dependence of the probability of atom excitation on frequency in case of two-frequency excitation when $n_0 = 45$, $\omega_0 = 1$, $n_{max} = 97$. (a),(c): for $\varepsilon_0 = 0.015$ with quantum (*) and classical (♦) values; for $\varepsilon_0 = 0.02$ correspondingly (●) and (▲), $\tau = 20$; (b),(d): the same as (a),(c) but for $\tau = 40$. Symbols (⊕), (x) correspond to the quantum and classical systems when $n_0 = 66$, $\varepsilon_0 = 0.02$, $\tau = 40$, $n_{max} = 143$. For $W_{n \geqslant [1.5n_0]}$ the classical value is (▲).

## 5. DISCUSSION

Thus the carried out experiments have shown that the diffusion excitation of atoms in a field of the monochromatic wave from states with $n \sim 50$ is rather suppressed. A simple estimate of values n, starting with that which on the diffusion excitation will be close to the classical one, can be obtained in the following way [13]. According to Equation (3) the number of diffusively excited levels

$\Delta n$ grows with the number of periods $\tau$ ($\omega \approx \Omega$) according to the law $(\Delta n)^2 \approx D_o \tau \approx 2(\varepsilon_o n_o)^2 \tau$. At the same time $\Delta n$ determines an effective number of lines in the spectrum of the wave function $N_L \sim \Delta n$. The spectrum can be considered continuous as far as $N_L \gtrsim \tau$. As diffusion needs a continuous spectrum the time of quantum diffusion limitation is determined by the condition $N_L(\tau^*) \sim \tau^*$ which gives

$$\tau^* \sim 2(\varepsilon_o n_o)^2 \sim D_o. \qquad (6)$$

At $\varepsilon_o \sim \varepsilon_{cr}$ this time $\tau^* \propto n_o^2$, which agrees with the estimate given in [11]. Since $\tau \gtrsim 1$, $\varepsilon_o \gtrsim \varepsilon_{cr}$ as well as $\varepsilon_o > 1/n_o$ (or $\varepsilon > n^{-5}$) are the necessary conditions for diffusion, which corresponds to going beyond the perturbation theory and exceeding the quantum border of stability [16]. Thus, for the diffusion to go over a large time interval $\tau^* >> 1$ in the field $\varepsilon_o$ (at $\omega_o \approx 1$) the following values are needed

$$n_o >> 1/\varepsilon_o. \qquad (7)$$

At $\varepsilon_o \approx 0.03$ this condition is satisfied by $n_o >> 30$. Let us note that the number of levels in one non-linear resonance is $(\Delta n)_r \gtrsim 1$ for $\varepsilon \gtrsim n^{-6}$ [11]. For $\varepsilon_o = 0.04$, $n_o = 45$ the value $\tau^* \sim 6$ agrees well with the results of a numerical experiment (Figure 1). But for $\tau^*$ to become comparable with the characteristic diffusion time $\tau_D \sim \sim n_o^2/D_o \sim 1/2\varepsilon_o^2$, we need to satisfy this condition and it is necessary to increase either the number of level $n_o$ or $\varepsilon_o$. At $n_o = 45$, the necessary value $\varepsilon_o \sim (2n_o)^{-1/2} \sim 0.1$. Computer experiments really point to the fact that with the growth of the field, the probabilities of excitation in the classical and quantum cases become comparable. But with the growth of $\varepsilon$ the probability of various multiphoton transitions grows also and can lead to a quicker atom excitation (Figure 2e,f). A marked difference of the moments $M_1$, $M_2$ from the classical values also points to the quantum nature of excitation in the field with $\varepsilon_o = 0.06$. Thus for $n_o = 45$, $\varepsilon_o = 0.06$, $\tau = 12$, $\omega_o = 1$ the quantum moments are greater than the classical ones ($M^2_{1,2}/M^{c\ell}_{1,2} = 1.8, 1.7$) and for $\omega_o = 1.025$ smaller than the classical ones ($M^2_{1,2}/M^{c\ell}_{1,2} = 0.41, 0.56$). The given estimates belonged to the case when a one-dimensional approximation can be used, i.e., $n >> n_2 \sim 1$. If $n_{1,2} \sim n_2$ the number of lines in the spectrum $N_L \sim \Delta n_1 \Delta n_2 \sim (\varepsilon_o n_o)^2 \tau$. From which it is clear that the necessary condition of diffusion excitation is $\varepsilon_o \gtrsim 1/n_o$. However when $\varepsilon > \varepsilon_{cr}$ the fulfilment of this condition does not at all denote that there will be no quantum diffusion limitation in the system (although the condition $N_L >> \tau$ is fulfilled). This involves the fact that due to Kepler degeneration $N_L$ contains groups of very close lines, corresponding to one shell, which decreases its efficient value. That is why to find the conditions of diffusion in the range $n_{1,2} \sim n$, $\varepsilon > n^{-5}$, $\varepsilon > \varepsilon_{cr}$ ( i.e., for $n \gtrsim 50$) we will need further investigation.

When the frequency of excitation goes up the rate of diffusion slows down [11], which leads to a decrease of $\tau^* \sim D_0(\omega)/\omega$ ($n_2 \ll n$). The marked growth of the probability of excitation at the decrease of frequency can be qualitatively explained in the following way. At $\varepsilon_0 \simeq 0.04$, $\omega_0 \simeq 0.7$ the value $n = n_0$ is in resonance, the center of which is at $n_r = n_0\omega_0^{-1/3}$. This leads to an efficient increase of the field $\varepsilon_0 \to \varepsilon_0(n_r/n_0)^4 = \varepsilon_0\omega_0^{-4/3} \simeq 1.6\varepsilon_0$ and a more fast classical diffusion. This effect was observed for the classical atom in [20]. This sort of explanation is suitable for the diffusion mechanism of excitation only, since in case of the multiphoton mechanism the probability of excitation should decrease with the lowering of frequency. So, the resonance dependence on $\omega_0$, observed in the experiment (Figure 4) is still being obscure. A further theoretical explanation is also needed for the results on excitation from the states $n_0 = 30,45$ and 66 at $\omega_0 \simeq 1$. So, for instance, the theoretical formula, obtained in [21,11], gives value $W_{n \geq [1.5n_0]}$ 10 orders less than the experimental one ($n_0 = 30$, Figure 2c).

In case of the two-frequency excitation a substantial spreading of the initial distribution occurs (Figure 6), which corresponds qualitatively to the picture of diffusion excitation. However, in this case as well, a qualitative agreement with classics is observed for the probabilities $W_{n>n_0+4}$ only, while the dependence on frequency for $W_{n \geq [1.5n_0]}$ is of a marked resonance nature (Figure 7). Since the quantum diffusion limitation for quasiperiodical excitation shows much weaker than for the periodical one [22], the most optimum case for the diffusion mechanism is the one of two-frequency excitation. But in this case as well, at $\varepsilon_0 \simeq 0.02$, $\omega_0, \omega_1 \sim 1$ the values $n_0 \simeq 66$ are still in the quantum range ($M_1$ differs greatly from the classical one). The reason for this difference is evidently connected with the fact that even for $n_0 = 66$ the exceeding of the quantum border of stability $\varepsilon_0 >> \varepsilon_q = 1/n_0$ at $\varepsilon_0 = 0.02$ proves rather small. According to the results obtained for simple models [22], it should be expected that when $n_0$ increases the time of quantum diffusion limitation will grow exponentially. This question, however, needs further investigation.

For one-frequency excitation quantum corrections lead to a substantial difference of distribution characteristics from the classical ones. This result testifies to the fact that the coincidence of the ionization probability of the classical system [6] with the one obtained in the experiment [1] is evidently accidental. In fact computer experiments have shown that for $n_0 = 66$, $\omega_0 = 0.4327$, $\varepsilon_0 = 0.061$ already at $\tau = 3$ quantum moments are $1.7(M_1)$, $1.8(M_2)$ times smaller than the classical ones, and therefore the classical description of ionization is not applicable in this case. The probabilities of excitation are also smaller than the classical ones

(for $\tau = 3$ the probabilities $W_{n>n_0+4} = 0.45$, $W_{n\geqslant[1.5n_0]} = 0.23$ and corresponding quantum values are equal to 0.29, 0.1).

## 6. SUMMARY

The investigations carried out have shown that the excitation of atom from states with $n \lesssim 66$ by a monochromatic field with the frequency close to the Kepler one ($\omega \approx n^{-3}$) and amplitude $\varepsilon \sim 0.04n^{-4}$ is of a fundamentally quantum character. The reason for this lies in the fact that on the one hand quantum corrections lead to a quantum diffusion limitation [12,13], and on the other hand there may be strong multiphoton transitions in the atom, that result in many cases in a stronger excitation than the classical diffusion. When $\varepsilon_0 \lesssim 0.04$, $\omega_0 \approx 1$ the probability of excitation out of the resonance may be one order of magnitude smaller than the classical one. So far as the field grows the probabilities become comparable, but differ 2 - 3 times from each other depending on the frequency $\omega_0$. A more efficient atom excitation is observed in the case when the frequency $\omega_0 < 1$. So, when $\varepsilon_0 = 0.03$, $n_0 = 45$ the decrease of the frequency from $\omega_0 \approx 1$ to $\omega_0 \approx 0.7$ involves an increase of the probability $W_{n\geqslant[1.5n_0]}$ by 2 orders of magnitude. At an increase of the there is no such effect.

In the case of the two-frequency excitation in the range of stochasticity there occurs a significant spreading of the packet over unperturbed levels. But, in these conditions as well, when up to 20 levels are excited efficiently, the probability of excitation may have a resonant dependence from the frequency. Quantitative characteristics of quantum distribution differ from the classical ones.

Thus, quantum diffusion limitation leads to the localization of the distribution over a relatively small group of unperturbed levels, from which strong multiphoton transitions start later on. The probability of the latter is significant and cannot be explained by available theoretical estimates. The investigation of the dynamics of excitation in the region where the quantum diffusion limitation takes place can be conducted in experiments similar to [1-3]. Conditions of the one-dimensional approximation will be satisfied if the states with strongly different parabolic quantum numbers ($n_1 \gg n_2$) are excited. Modern experiment methods make it possible to perform this with a high accuracy [23].

The author expresses his sincere gratitude to B. V. Chirikov for his attention to this work and valuable comments, and to N. B. Delone, F. M. Izrailev and V. P. Krainov for helpful discussions.

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