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# Energy Approach to Atoms in a Laser Field and Quantum Dynamics with Laser Pulses of Different Shape

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## 1. Introduction

In most branches of physics, a controlled manipulation of the considered system has proven to be extremely useful to study fundamental system properties, and to facilitate a broad range of applications. A prominent example for this is quantum optics or laser physics in general, for instance related to light-matter interactions on the level of single quantum objects (Letokhov, 1977, 1984; Delone & Kraynov, 1984, 1995, 1999; Allen & Eberly, 1987; Kleppner, et al, 1991; Fedorov, 1995; Scully & Zubairy, 1997; Friedberg, et al 2003; Popov, 2004; Ficek & Swain, 2005; Shahbaz et al, 2006; Burvenich et al, 2006; Müller et al, 2008; Glushkov et al, 2003, 2004, 2005, 2008, 2009) . Similar control is also possible at lower driving field frequencies, e.g., with NMR techniques in the microwave frequency region. Towards higher frequencies, in particular the development and deployment of high-intensity lasers have opened the doors to new fascinating areas of physics of light-matter interactions. Laser fields reach and succeed the Coulomb field strength experienced by the electrons due to the nucleus and thus give rise to a plethora of exciting phenomena. The above examples have in common that they focus on the interaction of the driving fields with the outer electron shell of the atoms. Now it is clear that direct laser-atom and nucleus interactions may indeed become of relevance in future experiments employing x-ray lasers, opening the field of high-intensity atomic and nuclear quantum optics. In particular, the coherence of the laser light expected from new sources such as TESLA XFEL is the essential feature which may allow to access extended coherence or interference phenomena reminiscent of atomic quantum optics. Such laser facilities, especially in conjunction with moderate acceleration of the target atoms and nuclei to match photon and transition frequency, may thus enable to achieve nuclear Rabi oscillations, photon echoes or more advanced quantum optical schemes in atoms, nuclei, molecules, clusters, bose-condensate etc .

The interaction of the atomic systems with the external alternating fields, in particular, laser fields has been the subject of intensive experimental and theoretical investigation (Holt et

al, 1983; Delone & Kraynov, 1984, 1995, 1999; Ullrich et al, 1986; Allen & Eberly, 1987; Scully & Zubairy, 1997; Aumar-Winter, 1997; Becker & Faisal, 2002; Batani & Joachain, 2006; Glushkov, 2005, 2008; etc). The appearance of the powerful laser sources allowing to obtain the radiation field amplitude of the order of atomic field in the wide range of wavelengths results to the systematic investigations of the nonlinear interaction of radiation with atoms and molecules. Calculation of the deformation and shifts of the atomic emission and absorption lines in a strong laser field, definition of the k-photon emission and absorption probabilities and atomic levels shifts, study of laser emission quality effect on characteristics of atomic line, dynamical stabilization and field ionization etc are the most actual problems to be solved. Naturally, it is of the great interest for phenomenon of a multiphoton ionization. At present time, a progress is achieved in the description of the processes of interaction atoms with the harmonic emission field. But in the realistic laser field the according processes are in significant degree differ from ones in the harmonic field. The latest theoretical works claim a qualitative study of the phenomenon though in some simple cases it is possible a quite acceptable quantitative description. Among existed approaches it should be mentioned the Green function method (the imaginary part of the Green function pole for atomic quasienergetic state), the density - matrix formalism ( the stochastic equation of motion for density - matrix operator and its correlation functions), a time-dependent density functional formalism, direct numerical solution of the Schrödinger (Dirac) equation, multi-body multi-photon approach etc. Decay probabilities of the hydrogen atom states in the super-strong laser field are calculated by the Green function method under condition that electron- proton interaction is very small regarding the atom-field interaction. Note that this approach is not easily generalized for multielectron atoms. Alternative approach is using the double-time Gell-Mann and Low formalism for the investigation of line-shape of a multi-ionized atom in the strong field of electromagnetic wave. The effects of the different laser line shape on the intensity and spectrum of resonance fluorescence from a two-level atom are intensively studied (Bjorkholm & Liao, 1975; Grance, 1981; Georges & Dixit, 1981; Zoller, 1982; Kelleher et al, 1985; Sauter et al, 1986; Glushkov-Ivanov, 1992, 1993; Friedberg et al, 2003; Glushkov et al, 2005, 2008, 2009 et al).

The laser model considered is that of an ideal single-mode laser operating high above threshold, with constant field amplitude and undergoing phase-frequency fluctuations analogous to Brownian motion. As a correlation time of the frequency fluctuations increases from zero to infinity, the laser line shape changes from Lorentzian to Gaussian in a continuous way. For intermediate and strong fields, the average intensity of fluorescence in the case of a resonant broadband Lorentzian line shape is higher than that in the case of a Gaussian line shape with the same bandwidth and total power. This is in contrast to the weak- field case where the higher peak power of the Gaussian line shape makes it more effective than the Lorentzian line shape. In a case of a nonzero frequency correlation time (the non - Lorentzian line shape) an intensity of fluorescence undergoes the non-Markovian fluctuations . In relation to the spectrum of resonance fluorescence it is shown that as the line shape is varied from Lorentzian to Gaussian the following changes take place : in the case of off-resonance excitation, the asymmetry of the spectrum decreases; in a case of resonance excitation, the center peak to side-peak height ratio for the triplet structure increases. The predicted center - line dip, which develops in the spectrum in the case of broadband excitation when the Rabi frequency and the bandwidth are nearly equal, becomes increasingly deeper. In the modern experiment it has been found an anomalously

strong nonlinear coupling of radiation with atoms which can not fully explained by the modern theoretical models. In any case the problem requires a consistent quantum electrodynamic approach.

Another important topic is a problem of governing and control of non-linear processes in a stochastic, multi-mode laser field (Grance, 1981; Lompre et al, 1981; Zoller, 1982; Glushkov & Ivanov, 1992). The principal aim of quantum coherent control is to steer a quantum system towards a desired final state through interaction with light while simultaneously inhibiting paths leading to undesirable outcomes. This type of quantum interference is inherent in non-linear multiphoton processes. Controlling mechanisms have been proposed and demonstrated for atomic, molecular and solid-state systems (Goldansky-Letokhov, 1974, Letokhov, 1977; Delone-Kraynov, 1984). Theoretical studies of the laser-atom non-linear interactions are often based on solving the time-dependent Schrödinger equation or using the time-independent Floquet formalism or special perturbation theories (Brändas & Floelich, 1974; Hehenberger et al, 1977; Silverstone et al, 1979; Delone-Kraynov, 1984; Glushkov-Ivanov, 1992, 1993, 2004; Popov, 2004;). It has been extended the non-Hermitian multi-state Floquet dynamics approach to treat one-electron atomic system to the case of general multi-electron ones. The result is a generalization of the R-matrix Floquet theory, developed by Burke *et al*, that allows for pulse shape effects whilst retaining the *ab initio* treatment of detailed electron correlation. The approach based on the eigenchannel R-matrix method and multichannel quantum-defect theory, introduced by Robicheaux and Gao to calculate two-photon processes in light alkaline-earth atoms has been implemented by Luc-Koenig et al, 1997 in j-j coupling introducing explicitly spin-orbit effects and employing both the length and velocity forms of the electric dipole transition operator. For example, the two-photon processes including above-threshold ionization in magnesium have been in details studied (Luc-Koenig et al, 1997). Nevertheless in many calculations there is a serious problem of the gauge invariance, connected with using non-optimized one-electron representation (in fact provided by not entire account for the multi-body interelectron correlations). The known example is non-coincidence of values for the length and velocity forms of the electric dipole transition operator (Grant, 2007; Glushkov & Ivanov, 1992).

In whole one can note that a problem of correct description of the non-linear atomic dynamics in a stochastic, multi-mode laser field is quite far from the final solution. It requires developing the consistent, advanced approaches to description of multi-photon dynamics and new schemes for sensing the stochasticity and photon-correlation effects. In this paper we present a new consistent method for studying the interaction of atom with the realistic laser field, based on the quantum electrodynamics (QED) and S-matrix adiabatic formalism Gell-Mann and Low. In relativistic case the Gell-Mann and Low formula expressed an energy shift  $\delta E$  through the QED scattering matrix including the interaction with as the laser field as the photon vacuum field (Ivanova et al, 1985; Ivanov-Letokhov, 1986; Glushkov-Ivanov, 1992, 1993; Glushkov et al, 1986, 2004, 2008, 2009). It is more natural to describe the interaction of atom with the realistic laser field by means of the radiation emission and absorption lines (Glushkov-Ivanov, 1986, 1992). Their position and shape fully determine the spectroscopy of atom in a laser field. The radiation atomic lines can be described by moments of different orders  $\mu_n$ . The first moment are directly linked with the field shift and width of the corresponding resonances. The main contribution into  $\mu_n$  is given by the resonant range. The values  $\mu_n$  can be expanded into perturbation theory (PT) series, though in resonant range the PT can't be used for the transition probabilities. The powerful Ivanov-Ivanova method (Ivanov-Ivanova, 1981;

Ivanova et al, 1985, 1988; Ivanov et al, 1988) is used for calculating the corresponding QED PT second order sums. As example we use the presented method for numerical calculation of the three-photon resonant, four-photon ionization profile of atomic hydrogen (1s-2p transition; wavelength =365 nm) and multi-photon resonance shift and width for transition 6S-6F in the atom of Cs (wavelength 1059nm) in a laser pulse of the Gaussian form. We consider also a quite exact approach to calculation of the characteristics of multi-photon ionization in atomic systems, which is based on the QED PT and use it for numerical calculating the above threshold ionization (ATI) characteristics for atom of magnesium in a intense laser field.

## 2. Structure of the multi-mode laser pulse

As it is well known, for a laser with more than one longitudinal mode, mode beating gives rise to intensity fluctuations within the laser pulse (eg. Kelleher et al, 1985). The beat frequencies for n modes range up to  $nc/2L=B$ , where L is the optical length of the laser oscillator. A detailed analysis of the mode structure of the typical dye laser shows that it has about 15 modes, separated by 1 GHz with a Gaussian amplitude distribution. Classically, the field can be written as follows:

$$E(t)=\varepsilon(t)e^{-i\omega t} + c.c., \quad (1)$$

where

$$\varepsilon(t)=\sum_i 0.5a_i(t)e^{-i(\Delta\omega_i t+\phi_i)}.$$

Each mode has amplitude  $a_i$  containing a gaussian time envelope, a frequency detuning  $\Delta\omega_i$  from the central laser frequency and phase  $\phi_i$ . As experimental study (Lompre et al, 1981; Kelleher et al, 1985; ) of described laser pulse showed that there is no evidence of phase coherence in the temporal behavior of the laser pulse and thus it is usually assumed that the modes have random phases. Figure 1 shows the temporal variation of intensity for the multi-mode pulse of stochastic laser radiation with emission lines width  $b=0,1 \text{ cm}^{-1}$ , the coherence time  $-3 \cdot 10^{-10}\text{s}$ .

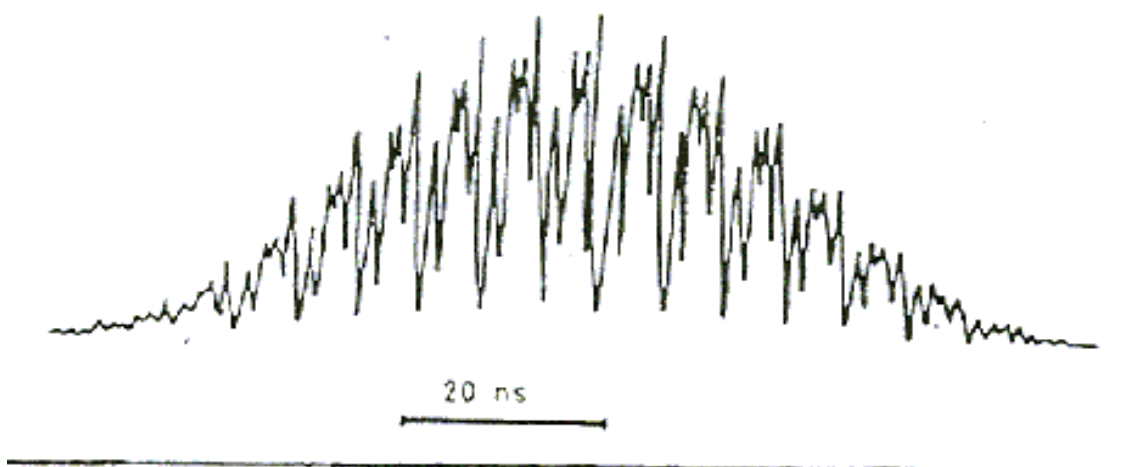


Fig. 1. The temporal variation of intensity for the multi-mode pulse of stochastic laser radiation with emission lines width  $b=0,1 \text{ cm}^{-1}$ , the coherence time  $-3 \cdot 10^{-10}\text{s}$ .



Further to make sensing a stochastic structure of the multi-mode laser pulse one can consider an interaction "atomic system – stochastic multi-mode laser pulse". Below it will be shown that this interaction is influenced by the specific chaotic, photon-correlation effects. New theoretical scheme for sensing stochasticity and photon-correlation features is based on the S-matrix energy approach (Glushkov & Ivanov, 1992, 1993) to calculating the multi-photon resonances spectra characteristics for atomic systems in a stochastic laser field.

### 3. S-matrix energy approach to atoms in a multi-mode laser field

Let us present the corresponding theoretical scheme. Following to (Glushkov & Ivanov, 1992, 1993; Glushkov et al, 2006, 2008, 2009), we describe the interaction of atom with the realistic laser field not by means the separated atomic levels shifts and by another set of characteristics, which are directly observed in the experiment. We are studying the radiation emission and absorption lines. Its position and shape fully determine the spectroscopy of atom in the field. It is natural to describe these lines by their moments of different orders  $\mu_n$ . The moments  $\mu_n$  are strongly dependent upon the laser pulse quality: intensity and the mode constitution. In particular, the k-photon absorption line center shift in the transition  $\alpha \rightarrow p$  can not be obtained from the corresponding expressions for the "one"-photon absorption by the change  $\omega_0 \rightarrow \omega_0/k$  and introduction of the multiplier  $1/k$  ( $\omega_0$  - the central laser emission frequency). The difference arises already in the first non-appearing perturbation theory (PT) order and connects with the unusual behaviour of the dynamic polarizability of atom in the resonant range (Glushkov-Ivanov, 1986, 1992). Let us describe the interaction of atom with laser radiation by means the potential:

$$V(r,t) = V(r) \int d\omega f(\omega - \omega_0) \sum_{n=-\infty}^{\infty} \cos [\omega_0 t + \omega_0 n \tau], \quad (2)$$

where  $n$  is the whole number. The potential  $V$  represents the infinite duration of laser pulses with known frequency  $\tau$ . Here we consider the effects of interaction of the atom with the single pulse. The representation  $V(rt)$  as the infinite sequence of pulses is a formal moment connected with the application of the stationary PT formalism. The function  $f(\omega)$  is a Fourier component of the laser pulse. The condition  $\int d\omega f^2(\omega) = 1$  normalizes potential  $V(rt)$  on the definite energy in a laser pulse. Let us consider the pulse with Lorentzian shape (coherent 1-mode pulse):  $f(\omega) = N/(\omega^2 + \Delta^2)$ , Gaussian shape (multi-mode chaotic laser pulse):  $f(\omega) = N \exp[-\ln 2(\omega^2/\Delta^2)]$ , and soliton-like pulse of the following shape:  $f(t) = N \operatorname{ch}^{-1}[t/D]$ . Further we will be interested by a case of the Gaussian and soliton-like pulses. A case of the Lorentzian shape has been considered by Glushkov & Ivanov (1992).

The further program resulted in the calculating an imaginary part of energy shift  $\operatorname{Im} E_\alpha(\omega_0)$  for any atomic level as the function of the laser pulse central frequency. An according function has the shape of the resonant curve. Each resonance is connected with the transition  $\alpha \rightarrow p$ , in which the definite number of photons are absorbed or radiated. Let us consider following situation:  $\alpha \rightarrow p$  transition with the absorption of  $k$  photons ( $\alpha, p$ -discrete levels). For the resonance which corresponds to this transition, we calculate the following values:

$$\delta\omega(p\alpha | k) = \int d\omega \operatorname{Im} E_\alpha(\omega) (\omega - \omega_{p\alpha} / k) / N, \quad (3)$$

$$\mu_m = \int d\omega \operatorname{Im} E_\alpha(\omega) (\omega - \omega_{p\alpha} / k)^m / N, \quad (4)$$

where  $\int' d\omega \operatorname{Im} E_\alpha$  is the normalizing multiplier;  $\omega_{p\alpha}$  is position of the non-shifted line for atomic transition  $\alpha$ - $p$ ,  $\delta\omega(p\alpha | k)$  is the line shift under  $k$ -photon absorption and  $\omega_{p\alpha} = \omega_{p\alpha} + k \cdot \delta\omega(p\alpha | k)$ . The first moments  $\mu_1$ ,  $\mu_2$  and  $\mu_3$  determine the atomic line center shift, its dispersion and coefficient of the asymmetry. To calculate  $\mu_m$ , we need to get an expansion of  $E_\alpha$  to PT series:  $E_\alpha = \sum E_\alpha^{(2k)}(\omega_0)$ . To get this expansion, we use method, based on the Gell-Mann and Low adiabatic formula for  $\delta E_\alpha$  (Ivanov et al, 1986, 1993; Ivanova et al, 1985, 1993). The representation of the  $S$ -matrix in the form of PT series induces the expansion for  $\delta E_\alpha$ :

$$\delta E_\alpha(\omega_0) = \lim_{\gamma \rightarrow 0} \gamma \sum_{k_1 k_2 \dots k_n} a(k_1, k_2, \dots, k_n), \quad (5)$$

$$I_\gamma(k_1, k_2, \dots, k_n) = \prod_{j=1} S_\gamma^{(k_j)}, \quad (6)$$

$$S_\gamma^{(m)} = (-1)^m \int_{-\infty}^0 dt_1 \dots \int_{-\infty}^{t_m-1} dt_m \langle \Phi_\alpha | V_1 V_2 \dots V_m | \Phi_\alpha \rangle, \quad (7)$$

$$V_j = \exp(1H_0 t_j) V(rt_j) \exp(-1H_0 t_j) \exp(\gamma t_j). \quad (8)$$

Here  $H$  is the atomic hamiltonian,  $a(k_1, k_2, \dots, k_n)$  are the numerical coefficients. The structure of the matrix elements  $S_\gamma^{(m)}$  is in details described (Glushkov & Ivanov, 1986, 1992, 1993). After sufficiently complicated one can get the expressions for the line moments. Let us present results for the Gaussian laser pulse:

$$\delta\omega(p\alpha | k) = \{\pi\Delta / (k+1)k\} [E(p, \omega_{p\alpha}/k) - E(\alpha, \omega_{p\alpha}/k)], \quad (9)$$

$$\mu_2 = \Delta^2/k$$

$$\mu_3 = \{4\pi\Delta^3 / [k(k+1)]\} [E(p, \omega_{p\alpha}/k) - E(\alpha, \omega_{p\alpha}/k)],$$

where

$$E(j, \omega_{p\alpha}/k) = 0,5 \sum_{p_i} V_{j p_i} V_{p_i j} \left[ \frac{1}{\omega_{j p_i} + \omega_{p\alpha}/k} + \frac{1}{\omega_{j p_i} - \omega_{p\alpha}/k} \right] \quad (10)$$

The summation in (10) is fulfilled on all states of atomic system. For the Lorentzian pulse the expressions were obtained by Glushkov & Ivanov (1986, 1992). In a case of the laser pulse with shape  $ch^{-1}[t/D]$  it is necessary to carry out a direct numerical calculation (we did it) or use different approximations to simplify the expressions. Indeed, the last procedure may result in a great mistake.

Each term in equations (9) for  $\delta\omega$  is formally similar to the known expression for off-resonant shift of atomic level ( $p$  or  $\alpha$ ) in the monochromatic emission field with frequency  $\omega_{p\alpha}/k$ . However, here these values have other physical essence. When  $k \rightarrow \infty$  (an infinite little laser pulse central frequency) the formula for  $\delta E$  gives the correct expression for energy level shift in the stationary field.

The expressions (9),(10) for  $\delta\omega$  and  $\mu_n$  describe the main characteristics of the absorption line near resonant frequency  $\omega_{p\alpha}/k$ . One can see that these characteristics are determined not only by the radiation frequency, but also by the quantiness of the process. For example, the line shift is proportional  $1/(k+1)$ , but not to value of  $1/k$ , as one can wait for. Under  $k=1$  there is an

additional non-standard term. It will be shown below that this approach allows getting the results in an excellent agreement with experiment. The details of the numerical procedure are given below and presented in refs. (Glushkov et al, 2004, 2005, 2006, 2008, 2009) too.

#### 4. Ivanova-Ivanov approach to calculating the QED perturbation theory: second order sum

In this chapter we present the Ivanova-Ivanov approach to calculating sums of the second order of the QED perturbation theory (Ivanov & Ivanova, 1981; Ivanov et al, 1988, 1993; Ivanova et al, 1985, 1986; Glushkov et al, 2008, 2009). It will be used in calculation of the expressions (9), (10). In fact, speech is about determination of the matrix elements for operator of the interelectron interaction over an infinite set of virtual states, including the states of the negative continuum. A sum on the principal quantum number is defined in quadratures of the Dirac function and auxiliary functions  $x$ ,  $\tilde{x}$  (look below). All computational procedure results in solution of simple system of the ordinary differential equations with known boundary conditions under  $r=0$ . Exchange of the interelectron interaction operator  $1/r_{12}$  on one-electron operator  $V(r)$  decreases a brevity of summation on the virtual states. In a one-particle representation the cited sums are expressed through sums of the one-electron matrix elements:

$$\sum_{n_1} \langle n\chi m | V | n_1 \chi_1 m_1 \rangle \langle n_1 \chi_1 m_1 | V | n\chi m \rangle / (\varepsilon_{n_1 \chi_1 m_1} - \varepsilon), \quad (11)$$

where  $\varepsilon = \varepsilon_{n\chi m} + \omega_{p\alpha}/k$  is the energy parameter. One-electron energies  $\varepsilon_{n\chi m}$  include the rest energy  $(aZ)^{-2}$ . Let us note that here we use the Coulomb units (an energy in the Coulomb units [q.u.]: 1 q.u. =  $Z^2$  a.u.e. [Z - a charge of a nucleus; a.u.e. = 1 atomic unit of energy]). Consider a scheme of calculating the sum (11). Fundamental solutions of one-electron Dirac equations with potential  $V_C = U(r)$  have the same asymptotics as and the Dirac equation with Coulomb potential under  $r \rightarrow 0$  and  $r \rightarrow \infty$ . Let us consider a bi-spinor of the following form:

$$\Phi_{\chi_1 m_1} = \sum_{n_1} \varphi_{n_1 \chi_1 m_1} \langle n_1 \chi_1 m_1 | V | n\chi m \rangle / (\varepsilon_{n_1 \chi_1 m_1} - \varepsilon) \quad (12)$$

The radial parts  $F$ ,  $G$  of bi-spinor  $\Phi$  satisfy to system of differential equations:

$$\begin{aligned} -F' / \alpha Z + (1 + \chi_1)F / \alpha Z r + A_2 G &= \Lambda_2, \\ G' / \alpha Z + (1 - \chi_1)G / \alpha Z r + A_1 F &= \Lambda_1, \end{aligned} \quad (13)$$

$$\begin{aligned} A_1(r) &= U(r) + 1/(\alpha Z)^2 - \varepsilon \\ A_2(r) &= U(r) - 1/(\alpha Z)^2 - \varepsilon. \end{aligned} \quad (14)$$

The radial functions  $\Lambda_1$ ,  $\Lambda_2$  in a case of the dipole interaction are presented below. Solution of the system (13) can be represented as follows:

$$F(r) = \alpha Z [x(r)\tilde{f}(r) - \tilde{x}(r)f(r)] / 2\gamma,$$



$$G(r) = \alpha Z [x(r)\tilde{g}(r) - \tilde{x}(r)g(r)] / 2\gamma, \quad \gamma = [\chi^2 - \alpha^2 Z^2]^{1/2}, \quad (15)$$

A pair of functions  $f, g$  and  $\tilde{f}, \tilde{g}$  are two fundamental solutions of equations (13) without right parts; These functions satisfy to conditions:  $f \sim r^{-1}, \tilde{f}, g \sim r^{-1}, \tilde{g}$  under  $r \rightarrow 0$ . Here we introduce the following functions:

$$\begin{aligned} x &= \alpha Z \int_0^r dr' r'^2 [\Lambda_1(r')f(r') + \Lambda_2(r')g(r')] \\ \tilde{x} &= \alpha Z \int_0^r dr' r'^2 [\Lambda_1(r')\tilde{f}(r') + \Lambda_2(r')\tilde{g}(r')] + D. \end{aligned} \quad (16)$$

Further let us define a constant  $D$  in expressions (16). Let us suppose that  $\varepsilon < (\alpha Z)^{-2}$  (i.e. an energy lies below the boundary of ionization), but an energy does not coincide with any discrete eigen value of the Dirac equation. Then

$$D = -\alpha Z \int_0^\infty dr r^2 (\Lambda_1 \tilde{f} + \Lambda_2 \tilde{g}). \quad (17)$$

Let an energy  $\varepsilon$  coincides with energy of some discrete level  $n_0 \chi_1 m_1$ . It is supposed that this state excludes from (11) and (13). Then a constant  $D$  can be found from condition:

$$\int_0^\infty dr r^2 (F f_{n_0 \chi_1 m_1} + G g_{n_0 \chi_1 m_1}) = 0. \quad (18)$$

Now let  $\varepsilon > (\alpha Z)^{-2}$  (i.e. an energy lies above the boundary of ionization). Then a constant  $D$  can be found from the following condition:

$$\lim_{r \rightarrow \infty} r^2 \int_r^{T+r} dr' r'^2 (F f_{\varepsilon \chi_1 m_1} + G g_{\varepsilon \chi_1 m_1}) = 0, \quad (19)$$

Here  $\varepsilon \chi_1 m_1$  is one-electron state of scattering with energy  $\varepsilon$ ;  $T$  is a period of asymptotic oscillations of the functions  $f, g$ :

$$T = 2\pi [\varepsilon^2 - (\alpha Z)^{-2}]^{1/2}.$$

Let us give the corresponding expressions for functions  $\Lambda_1, \Lambda_2$  in the most typical case of the dipole interaction of an atom with the laser field. The corresponding potential is as follows:

$$V(\mathbf{r}) = (\mathbf{a}, \mathbf{a}), \quad (20)$$

Here  $\mathbf{a}$  is a vector of polarization of radiation;  $\mathbf{a}$  is a vector of the Dirac matrices.

Let us remember that usually the vectors  $\mathbf{a}_1 = (1, i, 0)$ ,  $\mathbf{a}_2 = (1, -i, 0)$  are corresponding to the circular polarization and the vector  $\mathbf{a}_3 = (1, 0, 0)$  is corresponding to linear one. Under definition

of the multi-photon resonance energies and widths there is a task of calculating the sums (11), where an index  $n_1$  runs the whole spectrum of states or some state  $n_0$  is excluded from the sum. In the first case the functions  $\Lambda_1$  and  $\Lambda_2$  are defined by the expressions:

$$\begin{aligned}\Lambda_1 &= B(a|j_1 l_1 m_1, \tilde{j} \tilde{l} m) g_{n\chi} / \alpha Z, \\ \Lambda_2 &= B(a|j_1 \tilde{l}_1 m_1, j l m) f_{n\chi} / \alpha Z\end{aligned}\quad (21)$$

In the second case one can substitute the following functions to the right parts of (13):

$$\bar{\Lambda}_1 = \Lambda_1 - f_{n_0 \chi_1 m_1} Y / \alpha Z,$$

$$\bar{\Lambda}_2 = \Lambda_2 - g_{n_0 \chi_1 m_1} Y / \alpha Z,$$

$$Y = \int dr r^2 \left[ f_{n_0 \chi_1 m_1} g_{n\chi m} B(a|j_1 l_1 m_1, \tilde{j} \tilde{l} m) - g_{n_0 \chi_1 m_1} f_{n\chi m} B(a|j_1 \tilde{l}_1 m_1, j l m) \right] \quad (22)$$

Here the functions  $\Lambda_1$  and  $\Lambda_2$  defined by the expressions (21).

The angle functions are dependent upon a polarization vector and defined by the following formula:

$$B(a_1|j l m, j' l' m') = (-1)^{j'+l'-1/2} \delta_{u'} \delta_{m', m-1} b(-m, m'),$$

$$B(a_2|j l m, j' l' m') = (-1)^{j+l-1/2} \delta_u \delta_{m', m+1} b(-m, m'),$$

$$B(a_3|j l m, j' l' m') = \delta_u \delta_{m m'} \left[ b(-m, -m) + (-1)^{j+j'} b(m, m') \right],$$

$$b(m, m') = \left[ 2 \frac{\chi + 1/2 + m}{2\chi + 1} - \frac{\chi' + 1/2 + m'}{2\chi' + 1} \right]^{1/2}. \quad (23)$$

The final expression for the sum (11) can be written as follows:

$$\int dr r^2 \left[ f_{n\chi_1} G B(a|j l m, j_1 \tilde{l}_1 m_1) + g_{n\chi} F \cdot B(a|\tilde{j} \tilde{l} m_1, j_1 l_1 m_1) \right]. \quad (24)$$

Finally the computational procedure results in a solution of sufficiently simple system of the ordinary differential equations for above described functions and integral (24). In concrete numerical calculations the block "Super-spinor" of the PC "Superatom" package (Ivanov-Ivanova, 1981; Ivanova et al, 1985, 1986, 2001; Glushkov et al, 2004, 2008, 2009) is used.

## 5. Energy QED approach to multiphoton resonances and above threshold ionization

In this section we consider a quite exact approach to calculation of the characteristics of multi-photon ionization in atomic systems, which is based on the QED perturbation theory

(Glushkov-Ivanov, 1992, 1993; Glushkov, 2006, 2008, 2009). Below we calculate numerically the above threshold ionization (ATI) cross-sections for atom of magnesium in a intense laser field. The two-photon excitation process will be described in the lowest QED PT order. This approach is valid away from any one-photon intermediate-state resonance. We start from the two-photon amplitude for the transition from an initial state  $\Psi_0$  with energy  $E_0$  to a final state  $\Psi_f$  with energy  $E_f=E_0+2\omega$  is:

$$T_{f0}^{(2)} = \lim_{\eta \rightarrow 0^+} \int d\varepsilon \langle \Psi_f | D \cdot e | \varepsilon \rangle (E_0 + \omega - \varepsilon + i\eta)^{-1} \langle \varepsilon | D \cdot e | \Psi_0 \rangle \quad (25)$$

Here  $D$  is the electric dipole transition operator (in the length  $r$  form),  $e$  is the electric field polarization and  $\omega$  is a laser frequency. It's self-understood that the integration in equation (25) is meant to include a discrete summation over bound states and integration over continuum states. Usually an explicit summation is avoided by using the Dalgarno-Lewis by means the setting (Luc-Koenig et al, 1997):

$$T_{f0}^{(2)} = C_f \langle \Psi_f | | D \cdot e | | A_p \rangle \quad (26)$$

where  $\langle || || \rangle$  is a reduced matrix element and  $C_f$  is an angular factor depending on the symmetry of the  $\Psi_f$ ,  $A_p$ ,  $\Psi_0$  states.  $A_p$  can be founded from solution of the following inhomogeneous equation (Luc-Koenig et al, 1997):

$$(E_0 + \omega - H) | A_p \rangle = (D \cdot e) | \Psi_0 \rangle \quad (27)$$

at energy  $E_0 + \omega$ , satisfying outgoing-wave boundary condition in the open channels and decreasing exponentially in the closed channels. The total cross section (in  $\text{cm}^4\text{W}^{-1}$ ) is defined as:

$$\sigma/I = \sum_J \sigma_J / I = 5,7466 \times 10^{-35} \cdot \omega_{au} \sum_J |T_{J,0}^{(2)}|^2 \quad (28)$$

where  $I$  (in  $\text{W}/\text{cm}^2$ ) is a laser intensity. To describe two-photon processes there can be used different quantities: the generalized cross section  $\sigma^{(2)}$ , given in units of  $\text{cm}^4\text{s}$ , by

$$\sigma_{\text{cm}^4\text{s}}^{(2)} = 4,3598 \times 10^{-18} \omega_{au} \sigma / I_{\text{cm}^4/\text{W}} \quad (29)$$

and the generalized ionization rate  $\Gamma^{(2)}/I^2$ , (and probability of to-photon detachment) given in atomic units, by the following expression:

$$\sigma / I_{\text{cm}^4/\text{w}} = 9,1462 \times 10^{-36} \omega_{au} \Gamma_{au}^{(2)} / I_{au}^2 \quad (30)$$

Described approach is realized as computer program block in the atomic numeric code "Super-atom" (Ivanov-Ivanova, 1981; Ivanova et al, 1985, 1986, 2001; Glushkov-Ivanov, 1992,1993; Glushkov et al, 2004, 2008, 2009), which includes a numeric solution of the Dirac equation and calculation of the matrix elements of the (17)-(18) type. The original moment is connected with using the consistent QED gauge invariant procedure for generating the atomic functions basis's (optimized basis's) (Glushkov & Ivanov, 1992). This approach allows getting results in an excellent agreement with experiment and they are more precise in comparison with similar data, obtained with using non-optimized basis's.

## 6. Some results and discussion

### 6.1 The multi-photon resonances spectra and above threshold ionization for atom of magnesium

Let us present the results of calculating the multi-photon resonances spectra characteristics for atom of magnesium in a laser field (tables 1,2). Note that in order to calculate spectral properties of atomic systems different methods are used: relativistic R-matrix method (R-метод; Robicheaux-Gao, 1993; Luc-Koenig E. et al, 1997), added by multi channel quantum defect method, K-matrix method (K-method; Mengali-Moccia,1996), different versions of the finite  $L^2$  method ( $L^2$  method) with account of polarization and screening effects (SE) (Moccia-Spizzo, 1989; Karapanagioti et al, 1996), Hartree-Fock configuration interaction method (CIHF), operator QED PT (Glushkov-Ivanov, 1992; Glushkov et al; 2004) etc.

Methods	$E$	$\Gamma$	$\sigma/I$
Luc-Koenig E. et al, 1997	Without	account	SE
Length form	68492	374	1,96 10 <sup>-27</sup>
Velocity form	68492	376	2,10 10 <sup>-27</sup>
Luc-Koenig E. et al, 1997	with	account	SE
Length form	68455	414	1,88 10 <sup>-27</sup>
Velocity form	68456	412	1,98 10 <sup>-27</sup>
Moccia and Spizzo (1989)	68320	377	2,8 10 <sup>-27</sup>
Robicheaux and Gao (1993)	68600	376	2,4 10 <sup>-27</sup>
Mengali and Moccia(1996)	68130	362	2,2 10 <sup>-27</sup>
Karapanagioti et al (1996)	68470	375	2,2 10 <sup>-27</sup>
Our calculation	68281	323	2,0 10 <sup>-27</sup>

Table 1. Characteristics for  $3p^2S_0$  resonance of atom of the magnesium:  $E$ - energy, counted from ground state ( $\text{cm}^{-1}$ ),  $\Gamma$ - autoionization width ( $\text{cm}^{-1}$ ),  $\sigma/I$ - maximum value of generalized cross-section ( $\text{cm}^4\text{W}^{-1}$ )

In table 1 we present results of calculating characteristics for  $3p^2S_0$  resonance of Mg;  $E$ - energy, counted from ground state ( $\text{cm}^{-1}$ ),  $\Gamma$ -autoionization width ( $\text{cm}^{-1}$ ),  $\sigma/I$ - maximum value of generalized cross-section ( $\text{cm}^4\text{W}^{-1}$ ). R-matrix calculation with using length and velocity formula led to results, which differ on 5-15%, that is evidence of non-optimality of atomic basis's. This problem is absent in our approach and agreement between theory and experiment is very good.

Further let us consider process of the multi-photon ATI from the ground state of Mg. The laser radiation photons energies  $\omega$  in the range of 0,28-0,30 a.u. are considered, so that the final autoionization state (AS) is lying in the interval between 123350  $\text{cm}^{-1}$  and 131477 $\text{cm}^{-1}$ . First photon provides the AS ionization, second photon can populate the Rydberg resonance's, owing to series  $4s_{nl}, 3d_{nl}, 4p_{np}$  c J=0 and J=2.

In table 2 we present energies ( $\text{cm}^{-1}$ ; counted from the ground level of Mg  $3s^2$ ) and widths ( $\text{cm}^{-1}$ ) of the AS (resonance's)  $4s_{nl}, 3d_{nl}, 4p^2 \ ^1D_2$ , calculated by the K-, R-matrix and our methods. In a case of  $^1S_0$  resonance's one can get an excellent identification of these resonance's. Let us note that calculated spectrum of to-photon ATI is in a good agreement with the R-matrix data and experiment. In a whole other resonances and ATI cross-sections demonstrate non-regular behaviour.

Studied system is corresponding to a status of quantum chaotic system with stochastization mechanism. It realizes through laser field induction of the overlapping (due to random

<sup>1</sup> D <sub>2</sub>	R-method		<sup>1</sup> D <sub>2</sub>	QED approach			K-method	
	E	Γ		E	Γ		E	Γ
4s3d	109900	2630	4s3d	109913	2645		110450	2600
3d <sup>2</sup>	115350	2660	3d <sup>2</sup>	115361	2672		115870	2100
4s4d	120494	251	4s4d	120503	259	(ds)	120700	170
3d5s	123150	1223	3d5s	123159	1235	(ds)	123400	2000
4p <sup>2</sup>	124290	446	4p <sup>2</sup>	124301	458		124430	500
3d4d	125232	400	3d4d	125245	430		125550	590
4s5d	126285	101	4s5d	126290	113	(ds)	126250	120
3d6s	127172	381	3d6s	127198	385	(ds)	127240	350
4s6d	127914	183	4s6d	127921	215		127870	1900
3d5d	128327	208	3d5d	128344	215			
4s7d	128862	18	4s7d	128874	24	(ds)	128800	30
3d5g	128768	4,5	3d5g	128773	5,2	3d5g	128900	2,2
3d7s	129248	222	3d7s	129257	235		129300	160
4s8d	129543	114	4s8d	129552	125	(ds)	129500	140
			3d6d	129844	115			
			4s9d	129975	64			
			4s10d	130244	5			
			3d8s	130407	114			
			4s11d	130488	118			
			4s12d	130655	28			
			3d7d	130763	52			
			4s13d	130778	36			
			4s14d	130894	14			
			4s15d	130965	7			

Table 2. Energies and widths (cm<sup>-1</sup>) of the AS (resonance's)  $4snl, 3dnl, 4p^2$  <sup>1</sup>D<sub>2</sub> for Mg (see text interference and fluctuations) resonances in spectrum, their non-linear interaction, which lead to a global stochasticity in the atomic system and quantum chaos phenomenon. The quantum chaos is well known in physics of the hierarchy, atomic and molecular physics in external electromagnetic field. Earlier it has been found in simple atomic systems H, He, and also Ca. Analysis indicates on its existence in the Mg spectrum. Spectrum of resonance's can be divided on three intervals: 1). An interval, where states and resonances are clearly identified and not strongly perturbed; 2) quantum-chaotic one, where there is a complex of the overlapping and strongly interacting resonances; 3). Shifted one on energy, where behaviour of energy levels and resonances is similar to the first interval. The quantitative estimate shows that the resonances distribution in the second quantum-chaotic interval is satisfied to Wigner distribution as follows:

$$W(x) = x \exp(-\pi x^2/4). \quad (31)$$

At the same time, in the first interval the Poisson distribution is valid.

## 6.2 The three-photon resonant, four-photon ionization profile of atomic hydrogen

Below we present the results of calculating the multi-photon resonances spectra characteristics for atomic systems in a stochastic laser field and show the possibilities for



sensing a structure of the stochastic, multi-mode laser pulse and photon-correlation effects for atomic (and nano-optical) systems in this field (figure 2). We start from results of the numerical calculation for the three-photon resonant, four-photon ionization profile of atomic hydrogen (1s-2p transition; wavelength =365 nm).

In figure 2 we present the shift  $S$  ( $=\delta\omega$ ) and width  $W$  of the resonance profile as the function of the mean laser intensity at the temporal and spatial center of the UV pulse: experimental data 3s, 3w (Kelleher et al, 1986; multi-mode Gauss laser pulse with bandwidth  $0.25 \text{ cm}^{-1}$ ; full width at half of one), theoretical calculation results on the basis of the stochastic differential equations method 1s and 1w by Zoller (1982) and results of our calculation: 2s, 2w.

At first, one can see the excellent agreement between the theory and experiment. At second, a comparison of these results with analogous data for a Lorentzian laser pulse (Lompre et al, 1981; Glushkov & Ivanov, 1992) shows that the corresponding resonance shift obtained with the gaussian pulse is larger the shift, obtained with Lorentzian pulse at  $\sim 3$  times. This is an evidence of the photon-correlation effects and stochasticity of the laser pulse.

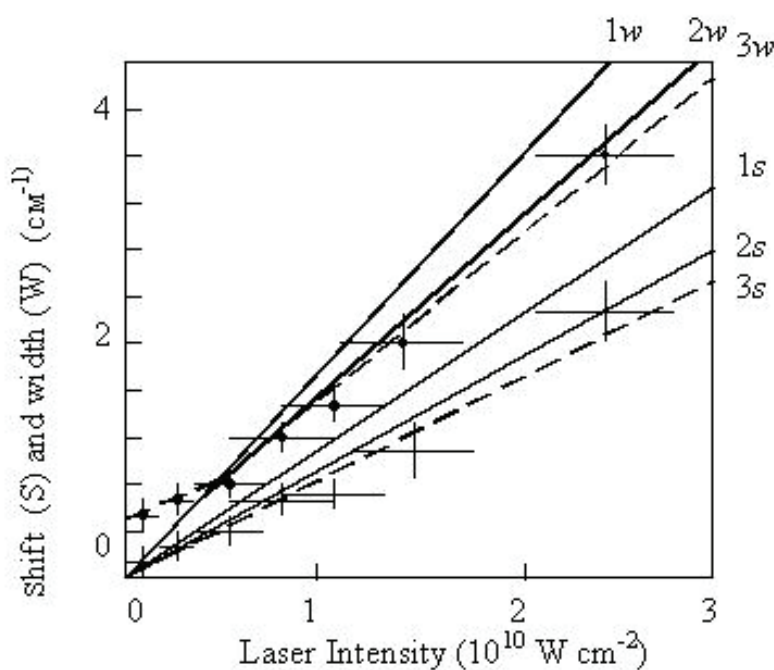


Fig. 2. Shift ( $S$ ) and width ( $W$ ) of resonant profile as laser intensity function: experiment -  $S_3$ ,  $W_3$  (Keller et al, 1981); theory of Zoller (1982)-  $S_1$ ,  $W_1$  and our results-  $S_2$ ,  $W_2$ .

### 6.3 Calculation results of the multi-photon resonance width and shift for transition 6S-6F in the atom of Cs

Further let us consider the numerical calculation results for three-photon transition 6S-6F in the Cs atom (wavelength  $1,059 \mu\text{m}$ ; see figure 3). The detailed experimental study of the multi-photon processes in Cs atom has been carried out by Lompre et al (1981). Lompre et al experimentally studied a statistics of the laser radiation and there are measured the characteristics of the multi-photon ionization.

The lines shift is linear to respect to the laser intensity (laser intensity is increased from 1,4 to 5,7 10(7) W/ cm<sup>2</sup>) and is equal (a case of the gaussian multi-mode laser pulse):  $\delta\omega(p\alpha | k) = bI$  with  $b=(5,6+-0,3)$  cm<sup>-1</sup>/GW·cm<sup>-2</sup> ( $b$  is expressed in terms of energy of the three-photon transition 6S-6F).

The corresponding shift obtained with coherent (one-mode) laser pulse is defined as follows:  $\delta\omega(p\alpha | k) = aI$ ,  $a=2$  cm<sup>-1</sup>/GW·cm<sup>-2</sup>. Theoretical values, obtained with using no-optimized atomic basises, are as follows: i). for soliton-like laser pulse:  $\delta\omega(p\alpha | k) = bI$ ,  $b=6,7$  cm<sup>-1</sup>/GW·cm<sup>-2</sup>; ii). for the gaussian multi-mode pulse (chaotic light):  $\delta\omega(p\alpha | k) = bI$  with  $b=5,8$  cm<sup>-1</sup>/GW·cm<sup>-2</sup>; iii). for the coherent one-mode pulse:  $\delta\omega(p\alpha | k) = aI$ ,  $a=2,1$  cm<sup>-1</sup>/GW·cm<sup>-2</sup>.

The analogous theoretical values, obtained in our calculation within described above S-matrix formalism, are the following:

- i. the gaussian multi-mode pulse (chaotic light)

$$\delta\omega(p\alpha | k) = bI, \quad b=5,63 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2};$$

- ii. the coherent one-mode pulse:

$$\delta\omega(p\alpha | k) = aI, \quad a=2,02 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2};$$

- iii. the soliton-like laser pulse:

$$\delta\omega(p\alpha | k) = bI, \quad b=6,5 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2}.$$

One can see that for the with multi-mode pulse, the radiation line shift is significantly larger (in ~ 3 times), then the corresponding shift, which is obtained for single-mode pulse. In fact the radiation line shift is enhanced by the photon-correlation effects. In figure 3 we present the results of calculation for the multi-photon resonance width for transition 6S-6F in the atom of Cs (wavelength 1059nm) in dependence upon the laser intensity.

We use the following denotations: S- for single-mode Lorentz laser pulse; M<sub>1</sub>, M<sub>3</sub>, M<sub>4</sub>- for multi-mode Gauss laser pulse respectively with line band 0.03cm<sup>-1</sup>, 0.08cm<sup>-1</sup> and 0.15cm<sup>-1</sup>; M<sub>2</sub>, M<sub>5</sub>- for multi-mode soliton-type with line band 0.03 cm<sup>-1</sup> and 0.15cm<sup>-1</sup>; †-experimental data (Lompre et al, 1981). Lompre et al presented the experimental data for laser pulse of the Gaussian form with line band respectively 0.03cm<sup>-1</sup>, 0.08cm<sup>-1</sup>, 0.15cm<sup>-1</sup>. In general there is a physically reasonable agreement between theory and high-qualitative experiment.

The detailed analysis shows that the shift and width of the multi-photon resonance line for interaction of atomic system with multimode laser pulse is greater than the corresponding resonance shift and width for a case of interaction between atom and single-mode laser pulse. This is entirely corresponding to the experimental data by Lompre et al. From physical point of view it is provided by action of the photon-correlation effects and influence of the multi-modity of the laser pulse (Lompre et al, 1981; Zoller, 1982; Kleppner, et al, 1991; Glushkov-Ivanov, 1992; Glushkov, 2004, 2005, 2008).

## 7. Modeling a population differences dynamics of the resonant levels in a rectangular form laser pulse: Optical bistability effect

Here we consider the following tasks (i) to simulate numerically a temporal dynamics of populations' differences at the resonant levels of atoms in a large-density medium in a nonrectangular form laser pulse and (ii) to determine possibilities that features of the effect of internal optical bistability at the adiabatically slow modification of effective filed intensity appear in the sought dynamics.

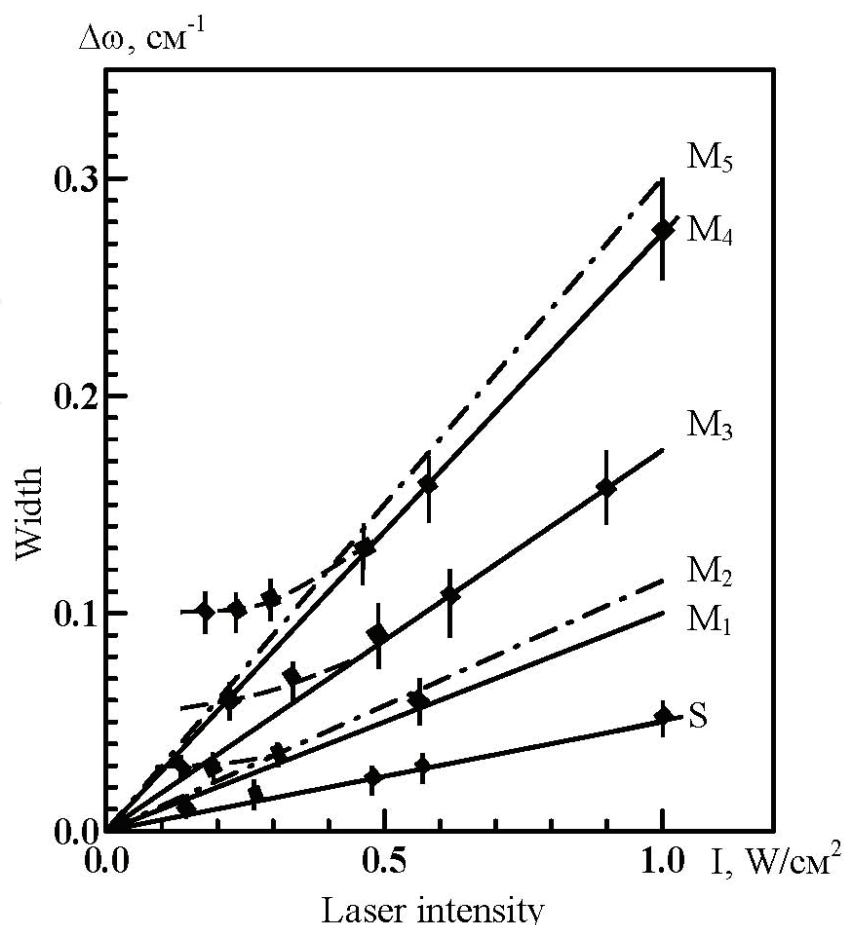


Fig. 3. The multi-photon resonance width for transition 6S-6F in the atom of Cs (wavelength 1059nm) in dependence upon the laser intensity  $I$ : theoretical data by Glushkov-Ivanov, 1992; Glushkov et al, 2008, 2009) S- for single-mode Lorentz laser pulse;  $M_1, M_3, M_4$ - for multi-mode Gauss laser pulse respectively with line band  $0.03\text{cm}^{-1}, 0.08\text{cm}^{-1}$  and  $0.15\text{cm}^{-1}$ ;  $M_2, M_5$ - for multi-mode soliton-type with line band  $0.03\text{cm}^{-1}$  and  $0.15\text{cm}^{-1}$ ;  $\blacklozenge$ -experiment (Grance, 1981; Lompre et al, 1981).

It is known that the dipole-dipole interaction of atoms in dense resonant mediums causes the internal optical bistability at the adiabatically slow modification of radiation intensity (Allen & Eberly, 1987; Scully & Zubairy, 1997; Afanas'ev & Voitikova, 2001; Ficek & Swain, 2005; Glushkov et al, 2008). The experimental discovery of bistable cooperative luminescence in some matters, in crystal of  $\text{Cs}_3\text{Y}_2\text{Br}_9\text{Yb}^{3+}$  particularly, showed that an ensemble of resonant atoms with high density can manifest the effect of optical bistability in the field of strong laser emission.

The Z-shaped effect is actually caused by the first-type phase transfer. Most attractive potentialities of sought effect are associated with the development of new system for optical information processing as well as with the creation of optical digital and analog processors. The creation of optical computer with an optical radiation as the data carrier excludes the necessity in the multiple transformation of electric energy into optical one and vice-versa. This consequently leads to the energy saving and abrupt increase of computer speed. The progress in the stated areas is especially defined by the creation of optical elements for the computer facilities on basis of optical bistability phenomenon.

On basis of the modified Bloch equations, we simulate numerically a temporal dynamics of populations' differences at the resonant levels of atoms in the field of pulse with the nonrectangular  $ch^{-1}t$  form. Furthermore, we compare our outcomes with the results (Afanas'ev & Voitikova, 2001; Glushkov et al, 2008), where there are considered the interaction between the ensemble of high-density atoms and the rectangularly- and sinusoidally-shaped pulses. The modified Bloch equations describe the interaction of resonance radiation with the ensemble of two-layer atoms taking into account the dipole-dipole interaction of atoms.

A fundamental aspect lies in the advanced possibility that features of the effect of internal optical bistability at the adiabatically slow modification of effective field intensity for pulse of  $ch^{-1}t$  form, in contrast to the pulses of rectangular form, appear in the temporal dynamics of populations' differences at the resonant levels of atoms.

The modified Bloch equations, which describes the interaction of resonance radiation with the ensemble of two-layer atoms subject to dipole-dipole interaction of atoms, are as follows:

$$\begin{aligned}\frac{dn}{d\tau} &= \frac{i2\mu T_1}{\hbar}(E^*P - P^*E) + (1-n) \\ \frac{dP}{d\tau} &= \frac{i2\mu T_1 n}{\hbar} - PT_1 \frac{1-i(\delta+bn)}{T_2},\end{aligned}\quad (32)$$

where  $n = N_1 - N_2$  are the populations' differences at the resonant levels,  $P$  is the amplitude of atom's resonance polarization,  $E$  is the amplitude of effective field,  $b = 4\pi\mu^2 N_0 T_2 / 2\hbar$  is the constant of dipole-dipole interaction,  $T_1$  is the longitudinal relaxation time,  $\delta = T_2(\omega - \omega_{21})$  is the offset of the frequency  $\omega$  of effective field from the frequency of resonance transition  $\omega_{21}$ ,  $N_0$  is the density of resonance atoms,  $\mu$  is the dipole moment of transition,  $\tau = t/T_1$ . Analytical solution of the set (32) cannot be found in general case.

Therefore we carried out the numerical modeling using the program complex "Superatom" (Ivanov-Ivanova, 1981; Ivanova et al, 1985, 1986, 2001; Glushkov-Ivanov, 1992,1993; Glushkov et al, 2004, 2008, 2009). The temporal dynamics for the populations' differences at the resonant levels of atoms in a nonrectangular form pulse field:

$$E(\tau) = |E_0|^2 ch^{-1} \frac{\pi\tau T_1}{T_2}. \quad (33)$$

was calculated.

In the numerical experiment  $\tau$  varies within  $0 \leq \tau \leq T_p/T_1$  and  $T_p$  is equal to  $10T_1$ . It is known (c.f. Afanas'ev & Voitikova, 2001) from general examination of set (32) that on the assumption of  $b > 4$  and  $b > |\delta|$  with  $\delta < 0$  (the long-wavelength offset of incident light frequency is less than Lorenz frequency  $\omega_L = b/T_2$ ) and if the intensity of light field has certain value ( $I_0 = 4|E_0|^2\mu^2 T_1 T_2 / \hbar^2$ ) then there are three stationary states  $n_i$  (two from them with maximal and minimal value of  $n$  are at that stable). This can be considered as evidence and manifestation condition of the internal optical bistability effect in the system.

Figure 4 shows the results of our numerical modeling the temporal dynamics of populations' differences at the resonant levels of atoms for the nonrectangular form pulse (2).

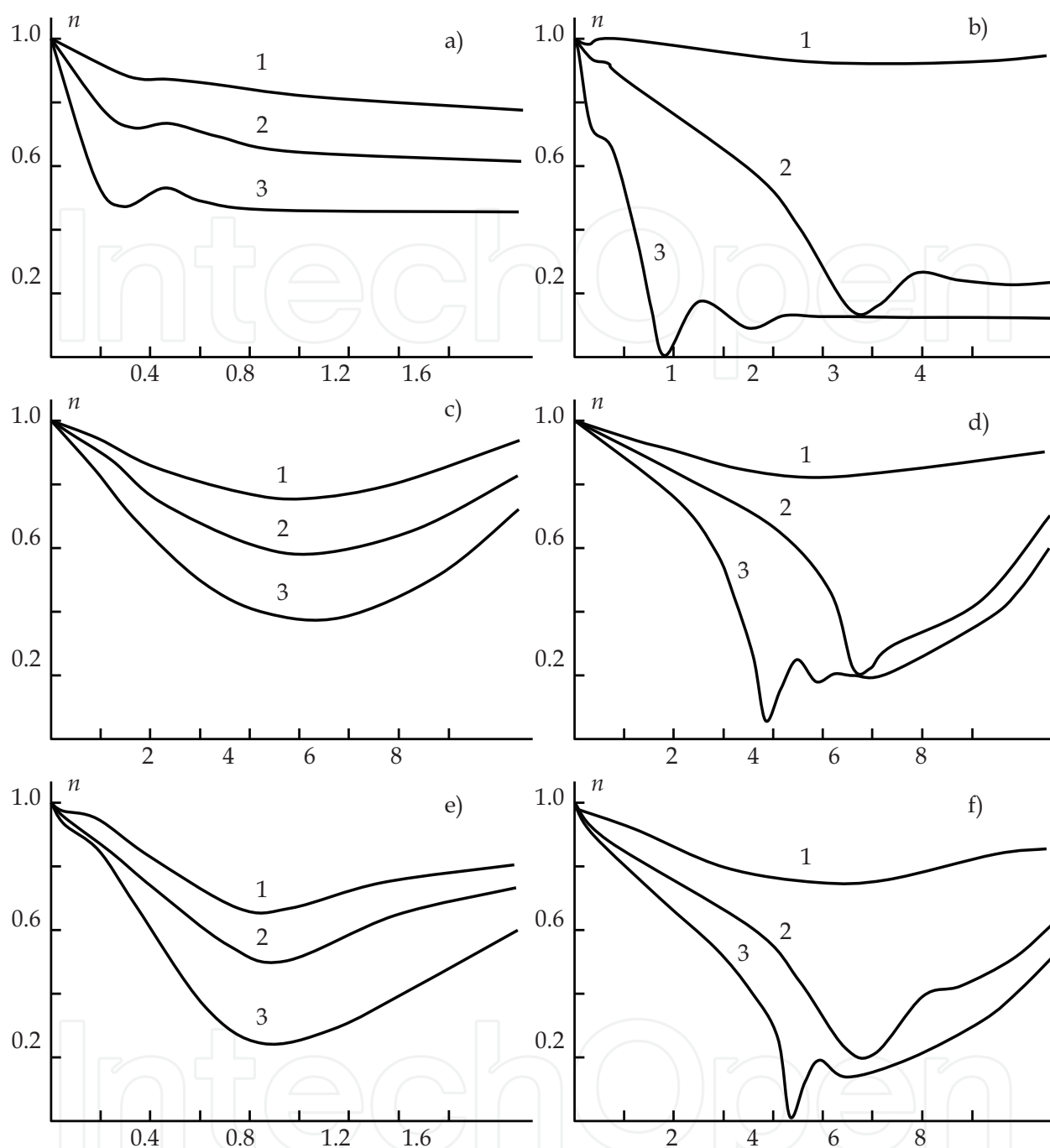


Fig. 4. Results of modeling temporal dynamics of populations' differences  $n(\tau)$  at resonant levels of atoms for the pulses of rectangular (a, b) and sinusoidal (c, d) forms by method of Allen & Eberly (1987) and Afanas'ev & Voitikova (2001), and for the pulse calculated by Eq. (32) with  $\delta = 2$ ,  $T_1 = 5T_2$ ;  $b = 0$  (a, c, e);  $b = 6.28$  (b, d, f);  $I_0 = 2$  (1), 5 (2), and 10 (3)

For collation, Figure 4 also shows similar results but for rectangularly- and sinusoidally-shaped pulses. The increase of field intensity above certain value  $I_0 = 2.5$  for selected parameters (shown in Fig. 4) leads to the abrupt increase of populations' differences. This fact represents the Z-shaped pattern of dependence  $n(I)$  observed in the stationary mode. It is important to note that there is the significant difference between the model results for the



pulses of various forms. For given values of rectangularly-shaped pulse intensity, which is equal to several values of  $T_1$ , the dependence  $n(\tau)$  tends to stationary state with magnitude defined by zero values of right-hand terms in the set (32). For the sinusoidally-shaped pulse, the slow rise of intensity is typical, and the explicit hysteresis pattern for the dependence of populations' differences from the field intensity is obtained.

For the pulse calculated by Eq. (33), the sought effect looks more explicitly. This is especially important from the standpoint of using the sought effect to simulate efficient neural networks and their components. Substantial fact also is the implementation of hysteresis in the dependence of populations' differences from the field intensity if a threshold values for  $b$  and  $\delta < 0$  have a place. This corresponds to the situation when the frequency of radiation  $\omega$  is within the range, which is formed by the proper frequency  $\omega_{21}$  and a frequency with the local-field correction:

$$\omega_{21}^L = \omega_{21} - 4\pi\mu^2 N_0/3h \quad (34)$$

Note that if above mentioned frequencies are almost equal or, e.g., a multimode electromagnetic field (chaotic light) is used, a stochastic resonance can be observed in the analyzed system.

## 8. Modeling Laser photoionization isotope separation technology and new principal scheme for $\gamma$ -laser on quickly decayed nuclear isomers with autoionization sorting of highly excited atoms

To number of the very actual problem of modern nuclear technology, quantum and photoelectronics is related a search of the effective methods for isotopes and nuclear isomers separation and obtaining especially pure substances at atomic level (Letokhov, 1969, 1977; Letokhov et al, 1975-1977, 1979, 1980, 1985, 1986, 1992, 1990; Basov et al. 1969, 1977; Prokhorov, 1979, 2001; Janes et al, 1975; Solarz et al, 1976; Lisitsa, 1977; Singh et al, 1994; Duarte et al, 1990, 2002, 2003, 2010; Bokhan et al, 2006; etc). The basis for its successful realization is, at first, carrying out the optimal multi stepped photo-ionization schemes for different elements and, at second, availability of enough effective UV and visible range lasers with high average power (Letokhov, 1977, 1979, 1983; etc). The standard laser photo-ionization scheme may be realized with using processes of the two-step excitation and ionization of atoms by laser pulse. The scheme of selective ionization of atoms, based on the selective resonance excitation of atoms by laser radiation into states near ionization boundary and further photo-ionization of the excited states by additional laser radiation, has been at first proposed and realized by Letokhov et al (Letokhov, 1969, 1977). It represents a great interest for laser separation of isotopes and nuclear isomers. The known disadvantage of two-step laser photoionization scheme a great difference between cross-sections of resonant excitation  $\sigma_{exc}$  and photo-ionization  $\sigma_{ion}$  ( $[\sigma_{exc}/\sigma_{ion}] > 10^4 \div 10^8$ ). It requires using very intensive laser radiation for excited atom ionization. The same is arisen in a task of sorting the excited atoms and atoms with excited nuclei in problem of creation of  $\gamma$ -laser on quickly decayed nuclear isomers.

Originally, Goldansky and Letokhov (1974) have considered a possibility of creating a  $\gamma$ -laser, based on a recoilless transition between lower nuclear levels and shown that a  $\gamma$ -laser of this type in the 20-60 keV region is feasible. A feature of design is operation based on relatively short-lived isomer nuclear states with lifetime of 0,1 to 10 sec. These authors has

estimated the minimal number of excited nuclei required for obtaining appreciable amplification and possibility of producing sufficient amounts of excited nuclei by irradiation of the target with a thermal neutron beam or by resonant  $\gamma$ -radiation. It is important that low-inertia laser selection of a relatively small fraction of excited nuclei of a given composition from the target by the two-step method of selective laser photoionization of atoms with excited nuclei by the radiation from two lasers is principally possible. But, it is obvious that here there is a problem of significant disadvantage of the two-step selective ionization of atoms by laser radiation method. The situation is more simplified for autoionization resonance's in the atomic spectra, but detailed data about characteristics of these levels are often absent (Letokhov, 1977, 1983; Glushkov & Ivanov, 1986, 1992).

The key problems here are connected with difficulties of theoretical studying and calculating the autoionization resonance characteristics. Several new optimal schemes for the laser photo-ionization sensors of separating heavy isotopes and nuclear isomers are proposed (Letokhov, 1983; Glushkov et al, 2004, 2008). It is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further autoionization and DC electric field ionization mechanisms. To carry out modelling the optimal scheme of the  $U$  and  $Tm$  isotopes (nuclei) sensing, the optimal laser action model and density matrices formalism were used. The similar schemes of laser photo ionization method are developed for control and cleaning the semiconductor substances (Glushkov et al, 2008). The optimal laser photo-ionization schemes for preparing the films of pure composition on example of creation of the 3-D hetero structural super lattices (layers of  $Ga_{1-x}Al_xAs$  with width  $10\text{\AA}$  and  $GaAs$  of  $60\text{\AA}$ ) have been proposed and new models of optimal realization of the first step excitation and further ionization of the  $Ga^+$  ions in Rydberg states by electric field are calibrated. In this paper we give the further development of approach to construction for the optimal schemes of the laser photo-ionization isotope separation technology and to creation of new possible principal scheme of  $\gamma$ -laser on quickly decayed nuclear isomers with laser autoionization or electromagnetic field ionization sorting the excited atoms.

Let us remind that in a classic scheme the laser excitation of the isotopes and nuclear isomers separation is usually realized at several steps: atoms are resonantly excited by laser radiation and then it is realized photo ionization of excited atoms. In this case photo ionization process is characterized by relatively low cross section  $\sigma_{ion}=10^{-17}-10^{-18}\text{cm}^2$  and one could use the powerful laser radiation on the ionization step. This is not acceptable from the energetics point of view (Letokhov, 1983; Buchanov, 2001; Stoll, 2001; Glushkov, 2005).

The alternative mechanism is a transition of atoms into Rydberg states and further ionization by electric field or electromagnetic pulse. As result, requirements to energetic of the ionized pulse are decreased at several orders. The main feature and innovation of the presented scheme is connected with using the DC electric field (laser pulse) autoionization on the last ionization step of the laser photoionization technology. There is a principal difference of the simple ionization by DC electric field. The laser pulse ionization through the auto ionized states decay channel has the advantages (more high accuracy, the better energetics, universality) especially for heavy elements and isotopes, where the DC electric field ionization from the low excited states has not to be high effective. This idea is a key one in the realization of sorting the definite excited atoms with necessary excited nuclei of the  $A^+$  kind, obtained by optimal method of selective photo-ionization of the  $A$  kind atoms at the first steps. The suitable objects for modelling laser photoionization separation technology are the isotopes of alkali element Cs, lanthanides and actinides.

We considered the isotopes of  ${}_{55}\text{Cs}^{133}$  and  ${}_{70}\text{Yb}^{171}$ . For example, the resonant excitation of the Cs can be realized by means dye lasers with lamp pumping (two transitions wavelengths are:  $6^2S_{1/2} \rightarrow 7^2P_{3/2}$  4555Å and  $6^2S_{1/2} \rightarrow 7^2P_{1/2}$  4593Å). In table 3 there are listed the energy parameters for different states of the caesium, obtained in the different approximations (Derevianko & Porsev, 2003; Glushkov et al, 2008; Khetselius, 2009). It is useful to remind the corresponding hyperfine splitting energy ( $6^2S_{1/2}$ , transition 4-3) of Cs: experimental data-  $\Delta\nu(F,F')= 9192,64\text{MHz}$  ;  $\Delta E(F,F')= 306,630 \cdot 10^{-3} \text{ cm}^{-1}$  ; theoretical data -  $\Delta\nu(F,F')= 9177,80\text{MHz}$  ;  $\Delta E(F,F')= 306,135 \cdot 10^{-3} \text{ cm}^{-1}$  (Khetselius, 2009).

The next step is in the further excitation to the Rydberg S,P,D states with main quantum number  $n=31-37$  (the optimal value  $n=35$ ). Final step is the autoionization of the Rydberg excited atoms by a electromagnetic field pulse and output of the created ions. The scheme will be optimal if an atom is excited by laser radiation to state, which has the decay probability due to the autoionization bigger than the radiation decay probability. In figure 5 we present the numeric modeling results of the optimal form of laser pulse in the photoionization scheme with auto-or electric field ionization by solving the corresponding differential equations system (Glushkov et al, 2008).

State	$\varepsilon^{RHF}$	$\varepsilon^{RHF} + \delta\varepsilon^{RHF}$	$\varepsilon^{QED}$	$\varepsilon^{Exp}$
$6s_{1/2}$	0,12737	0,14257	0,14295	0,14310
$6p_{1/2}$	0,08562	0,09198	0,09213	0,09217
$6p_{3/2}$	0,08379	0,08951	0,08960	0,08964
$7s_{1/2}$	0,05519	0,05845	0,05862	0,05865
$7p_{1/2}$	0,04202	0,04385	0,04391	0,04393
$7p_{3/2}$	0,04137	0,04303	0,04309	0,04310

Table 3. Valent electron ionization energies (in atom. units) of the  ${}^{133}\text{Cs}$ :  $\varepsilon^{RHF}$  -one-configuration Hartree-Fock data, relativistic Hartree-Fock (RHF);  $\varepsilon^{RHF} + \delta\varepsilon^{RHF}$  - the same data, but with account for the correlation corrections (Derevianko & Porsev, 2005;  $\varepsilon^{QED}$  - QED perturbation theory data (Khetselius, 2009);  $\varepsilon^{Exp}$ - experimental data (see text)

The following definitions are used:  $\delta$ -dashed line is corresponding to optimal form of laser pulse, curves 1 and 2 are corresponding to populations of the ground and excited states of Cs. The  $\delta$  -pulse provides maximum possible level of excitation (the excitation degree is about  $\sim 0,25$ ; in experiment (Letokhov, 1983) with rectangular pulse this degree was  $\sim 0,1$ ). It is in great degree similar to analogous scheme with the DC electric field and stochastic collisional ionization mechanisms.

In fig.5 there is also presented the typical behaviour of the ground (curve 1) and highly excited (curve 2) states population. Let us remember data regarding the excitation and the ionization cross sections for studied system: the excitation cross section at the first step of the scheme is  $\sim 10^{-11}\text{cm}^2$ ; the ionization cross-section from excited  $7^2P_2$  state:  $\sigma_2=10^{-16}\text{cm}^2$ , from ground state  $\sigma_1=10^{-18}\text{cm}^2$  (Letokhov, 1983). One can see that the relation of these cross sections is  $10^5$  and  $10^7$  correspondingly. This fact provides the obvious non-efficiency of standard photoionization scheme.

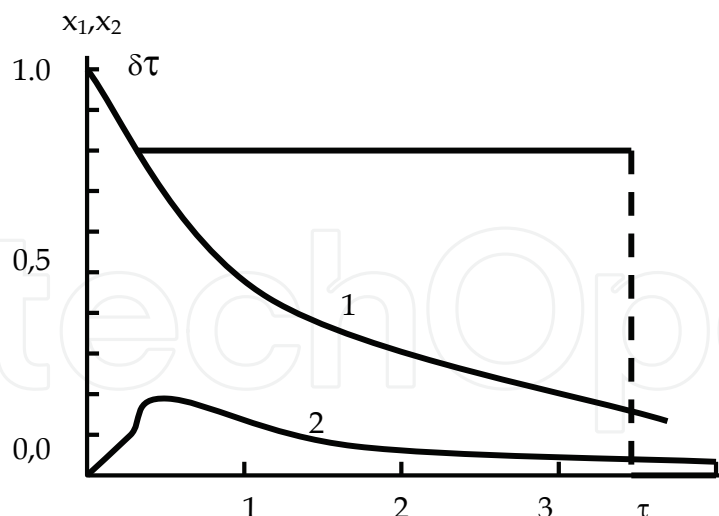


Fig. 5. Results of modelling Cs isotopes separation process by the laser photo-ionization method ( $\delta$ +dashed - laser pulse optimal form; see text)

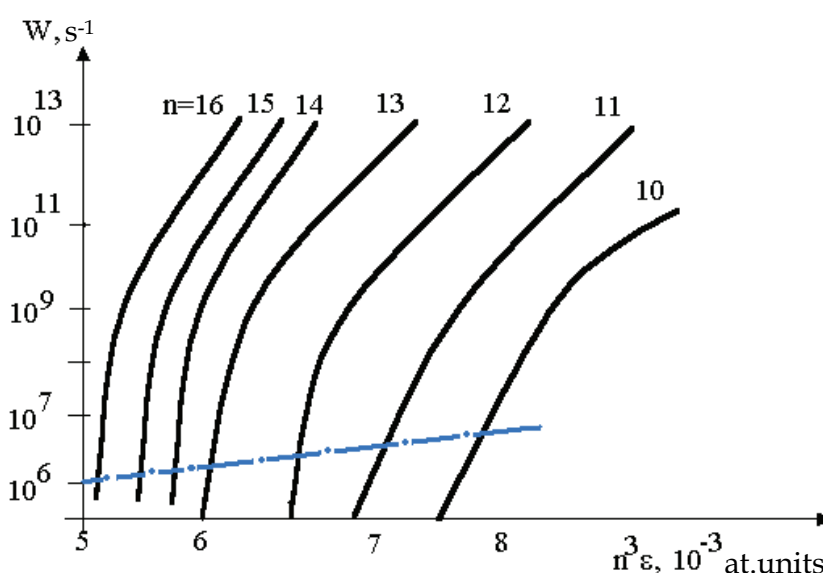


Fig. 6. Dependence of the velocity of ionization for high excited atoms of Cs upon the electric field strength for states with quantum numbers  $n=10-16$ ,  $m=0, n_2=n-1$ .

In figure 6 we present the results of our calculating dependence of the ionization velocity for high excited atoms of Cs upon the electric field strength for states with quantum numbers  $n=10-16, m=0, n_2=n-1$ . The dashed line is corresponding to velocity of the radiative decay. The decay of Cs atoms and ions in the high-excited state demonstrates the properties of the H-like systems at the qualitative level. But, there is quite significant quantitative difference. We have found that the ionization velocity for states with  $n > 14$  is more than the radiative decay velocity in electric field with strength  $E$  less than  $15 \text{ kV/cm}$ . Our estimate for the Ga atom ionization cross section is  $1,5 \cdot 10^{-13} \text{ cm}^2$  that is higher than the corresponding cross section of ionization process by laser pulse in the two-stepped photo ionization (Letokhov, 1977) scheme ( $\sim 10^{-17} \text{ cm}^2$ ).

Using  $\delta$ -pulse provides a quick ionization, but the ionization yield will be less than 100% because of the sticking on intermediate levels. Experimentally obtained dependence of the critical ionization field strength  $E$  upon the effective quantum number  $n^*$  is usually approximated by simple theoretical dependence  $E_{cr}=(2n^*)^{-4}$ .

Using the autoionization mechanisms at the final step for ionization of the Rydberg excited atoms provides more optimal scheme from energetic point of view. For example, for the  $35^2S_{1/2}$  transition the corresponding cross section can reach the value  $\sim 10^{-13}\text{cm}^2$ . So, from energetic point of view, this type of ionization can be very perspective alternative to earlier proposed classical two-step and more complicated photoionization schemes (Letokhov, 1983). More suitable situation takes a place for the for Yb isotope separation.

It is very important that the proposed scheme can be easily implemented to the possible advanced scheme of the  $\gamma$  - laser on quickly decayed nuclear isomers with using laser photoionization sorting excited nuclei  $M_{k+1}^*$  with autoionization mechanism through the Rydberg states.

Figure 7 easily explains the principal moments of this scheme. It generalizes the known Goldansky-Letokhov (Goldansky & Letokhov, 1974) and other (Baldwin et al, 1981; Glushkov, 2005) schemes and has to be more efficient especially from energetics point of view. In this context it is worth to remind very impressive results of the last years, connected with

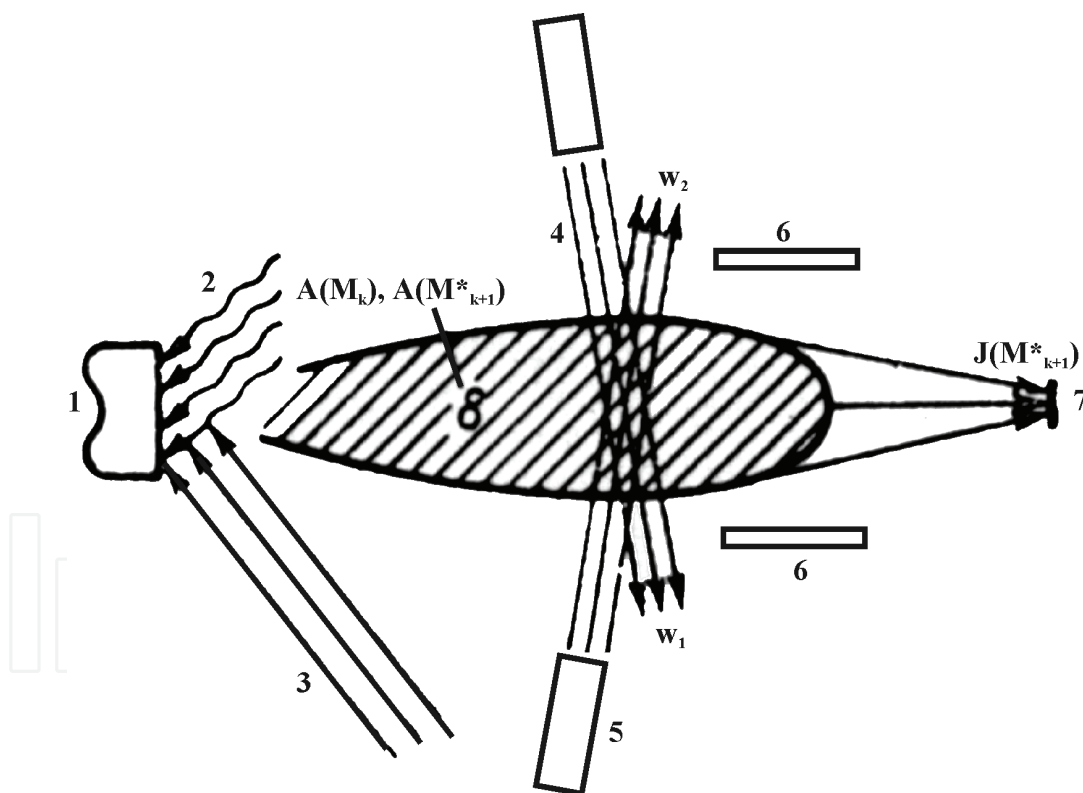


Fig. 7. Possible scheme  $\gamma$  - laser on quickly decayed nuclear isomers with using laser photoionization sorting excited nuclei  $M_{k+1}^*$  with electric field and auto- and electric field ionization mechanisms: 1 - target of atoms  $M_k$ ; 2- flux of slow neutrons; 3 - laser ray for evaporation of target; 4 - laser ray for the first step excitation of atoms with excited nucleus  $A(M_{k+1}^*)$ ; 5 - laser ray for second-step excitation to highly excited atomic states and Rydberg autoionization by electromagnetic field; 6 - collector system; 7 - atoms with excited nucleus  $A(M_{k+1}^*)$ ; 8 - flux of evaporated atoms;



engineering atomic highly excited Rydberg states and correspondingly cooperative laser-gamma-muon-electron- nuclear states (transitions) with the laser (and raser) pulses. It is quite possible that cited new effects can be realized in the tasks considered here.

The laser photo ionization scheme with autoionization of the highly excited atoms (with optimal set of energetic and radiative parameters: pulse form, duration, energetic for laser and electric field pulses etc.) could provide significantly more high yield and effectiveness of the whole process of the isotope separation. It is especially worth for implementation to the possible principal scheme of  $\gamma$ -laser on quickly decayed nuclear isomers with autoionization sorting the excited atoms.

## 9. Conclusions

We presented a new consistent method for studying the interaction of atom with a realistic laser field, based on the quantum electrodynamics (QED) and S-matrix adiabatic formalism Gell-Mann and Low. In relativistic case the Gell-Mann and Low formula expressed an energy shift  $\delta E$  through QED scattering matrix including the interaction with as the laser field as the photon vacuum field. It is natural to describe the laser field-atom interaction by means of the radiation emission and absorption lines. Their position and shape fully determine the spectroscopy of atom in the field. The radiation atomic lines can be described by moments of different orders  $\mu_n$ . The main contribution into  $\mu_n$  is given by the resonant range. The values  $\mu_n$  can be expanded into perturbation theory (PT) series. As example, the method has been used for numerical calculation of the three-photon resonant, four-photon ionization profile of atomic hydrogen (1s-2p transition; wavelength =365 nm) and multi-photon resonance width and shift for transition 6S-6F in the atom of Cs (wavelength 1059nm) in a laser pulse of the Gaussian and soliton-like forms.

The results of numeric calculation of population kinetics of resonant levels for atoms in the non-rectangular form laser pulse on the basis of the modified Bloch equations are presented. Cited equations describe an interaction between two-level atoms ensemble and resonant radiation with an account of the atomic dipole-dipole interaction. It has been found for a case of  $ch^{-1}t$  laser pulse a strengthen possibility of manifestation of the internal optical bistability effect special features in the temporary dynamics of populations for the atomic resonant levels under adiabatic slow changing the acting field intensity in comparison with a case of the rectangular form pulses.

At last, we shortly presented an optimal scheme of the laser photo-ionization heavy isotopes (isomers) separation technology and the new possible principal scheme of  $\gamma$ -laser on quickly decayed nuclear isomers with autoionization sorting the highly excited heavy atoms.

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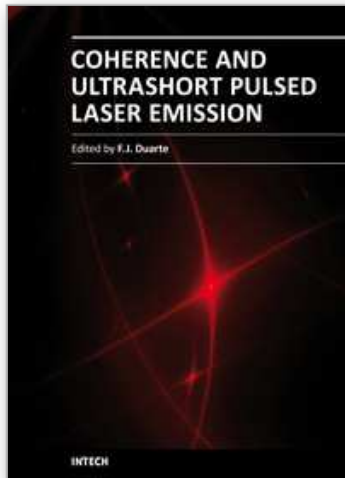
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