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## SPECTROSCOPY OF ATOM AND NUCLEUS IN A STRONG LASER FIELD: STARK EFFECT AND MULTIPHOTON RESONANCES

The consistent relativistic energy approach to atom in a strong realistic laser field, based on the Gell-Mann and Low S-matrix formalism, is applied to studying the resonant multiphoton ionization of krypton by intense uv laser radiation and calculating the multiphoton resonances shift and width in krypton. An approach to treating the multiphoton resonances in nuclei is outlined on example of the <sup>57</sup>Fe nucleus.

### 1. Introduction

At the present time a physics of multiphoton phenomena in atoms, molecules ets has a great progress that is stimulated by development of new laser technologies (see Refs. [1-10]). 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 systematic investigations of the nonlinear interaction of radiation with atomic and molecular systems [1-14]. At the same time a direct laser-nucleus interactions traditionally have been dismissed because of the well known effect of small interaction matrix elements [9-11]. Some exceptions such as an interaction of x-ray laser fields with nuclei in relation to alpha, beta-decay and x-ray-driven gamma emission of nuclei have been earlier considered. With the advent of new coherent x-ray laser sources in the near future, however, these conclusions have to be reconsidered. From the design report (look table II in Ref. [10]) for SASE 1 at TESLA XFEL and parameters for current and future ion beam sources, the signal rate due to spontaneous emission after real excitations of the nuclei can be estimated. For nuclei accelerated with an energy resolution of 0.1% such that 12.4 keV photons produced by SASE 1 become resonant with the E1 transition

in a whole number of nuclei (for example, <sup>153</sup>Sm, <sup>181</sup>Ta, <sup>223</sup>Ra, <sup>225</sup>Ac, <sup>227</sup>Th etc). It means that the resonance condition (w~De, where De is a typical level spacing, w is a laser frequency) is fulfilled [10]. The coherence of the laser light expected from new sources (TESLA XFEL at DESY) may allow to access the extended coherence or interference phenomena. In particular, in conjunction with moderate acceleration of the target nuclei it allows principally to achieve realization of multiphoton phenomena, nuclear Rabi oscillations or more advanced quantum optical schemes in nuclei.

The interaction of atoms with the external alternating fields, in particular, laser fields, has been the subject of intensive experimental and theoretical studied (see, for example, Refs. [1-8, 12-24]). A definition of the k-photon emission and absorption probabilities and atomic levels shifts, study of dynamical stabilization and field ionization etc are the most actual problems to be solved. At present time, a progress is achieved in the description of the processes of interaction atoms with the harmonic emission field [1,12-14]. But in the realistic laser field the according processes are in significant degree differ from ones in the harmonic field. It has been proved a significant role of the photon-correlation effects and influence of the laser pulse multi-modity. Surely,

a number of different theoretical approached has been developed in order to give a adequate description of the atoms in a strong laser field. Here one could mention such approaches as the standard perturbation theory (surely for low laser filed intensities), Green function method, the densitymatrix formalism, time-dependent density functional formalism, direct numerical solution of the Schrödinger (Dirac) equation, multi-body multiphoton approach, the time-independent Floquet formalism etc (see [1-8,12-24] and Refs. therein). The effects of the different laser line shape on the intensity and spectrum of resonance fluorescence from a two-level atom are studied in Refs. [1-5,15-17,19-23]. Earlier the relativistic energy approach to studying the interaction of atom with a realistic strong laser field, based on the Gell-Mann and Low S-matrix formalism, has been developed. Originally, Ivanov has proposed an idea to describe quantitatively a behaviour of an atom in a realistic laser field by means studying the radiation emission and absorption lines and further the theory of interaction of an atom with the Lorenz laser pulse and calculating the corresponding lines moments has been in details developed in Ref. [19-25]. It has been checked in numerical simulation of the multiphoton resonances shifts and widths in the hydrogen and caesium. Theory of interaction of an atom with the Gauss and soliton-like laser pulses and calculating the corresponding lines moments has been in details presented in Refs. [23,26,27]. Here we apply this approach to studying the resonant multiphoton ionization of krypton by intense uv laser radiation and calculating the multiphoton resonances shift and width. Besides, at first we also outline the corresponding scheme to treating the multiphoton resonances in nuclei on example of <sup>57</sup>Fe nucleus.

## 2. Relativistic energy approach to atom in a strong laser field: Multiphoton resonances

The relativistic energy approach in the different realizations and the radiation lines moments technique is in details presented in Refs. [19-30]. So, here we are limited only by presenting the master elements. In the theory of the non-relativistic atom a convenient field procedure is known for calculating the energy shifts dE of degenerate states. This procedure is connected with the secular matrix M diagonalization. In constructing M, the Gell-Mann and Low adiabatic formula for dE is used [20-23,31]. In relativistic theory, the Gell-Mann and Low formula dE is connected with electrodynamical scattering matrice, which includes interaction with as a laser field as a photon vacuum field. A case of interaction with photon vacuum is corresponding to standard theory of radiative decay of excited atomic states. Surely, in relativistic theory the secular matrix elements are already complex in the second perturbation theory (PT) order. Their imaginary parts are connected with radiation decay possibility. The total energy shift is usually presented in the form [23]:

$$\delta E = \operatorname{Re}\delta E + \operatorname{i} \operatorname{Im}\delta E$$
,  $\operatorname{Im} \delta E = -P/2$  (1)

where P is the level width (decay possibility). Spectroscopy of an atom in a laser field is fully defined by position and shape of the radiation emission and absorption lines. The lines moments  $m_n$  are strongly dependent upon the laser pulse quality: intensity and mode constitution [15-23]. Let us describe the interaction "atom-laser field" by the Ivanov potential [21,23]:

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

Here  $w_0$  is the central laser radiation frequency, *n* is the whole number. The potential *V* represents the infinite duration of laser pulses with known frequency t. The function f(w) is a Fourier component of the laser pulse. The condition  $\partial dw f^2(w)=1$ normalizes potential V(rt) on the definite energy in the pulse. Let us consider the pulses with Lorentz shape (coherent 1-mode pulse):  $|(w) = b/(w^2+D^2)$ , Gaussian one (multi-mode chaotic pulse): |(w)| = $bexp[ln2(w^2/D^2)]$  and the soliton-like pulse: f(t) $= b ch^{-1} [t/D]$  (b -normalizing multiplier). A case of the Lorentz shape has been earlier studied [20-23]. A case of the Gauss and soliton-like shape is considered in Refs.[23,26,27]. The master program results in the calculating an imaginary part of energy shift  $\text{Imd}E_{a}(W_{0})$  for any atomic level as the function of the central laser frequency  $w_0$ . An according function has the shape of the resonance, which is connected with the transition a-p (a, pdiscrete levels) with absorption (or emission) of the "k" number of photons. For the resonance we calculate the following values [20-23]:

^

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

where  $\partial \notin dw \text{Im}E_a$  is the normalizing multiplier;  $w_{pa}$  is position of the non-shifted line for transition a-p, dw(pa|k) is the line shift under k-photon absorption;  $v_{pa} = w_{pa} + k \times dw(pa|k)$ . The first moments  $m_1$ ,  $m_2$  and  $m_3$  determine the atomic line centre shift, its dispersion and the asymmetry. To find  $m_m$ , we need to get an expansion of  $E_a$  to PT series:  $E_a = a E_a^{(2k)} (w_0)$ . One may use here the Gell-Mann and Low adiabatic formula for  $dE_a$ :

$$\delta E_{\alpha} = \lim_{\gamma \to 0} i\gamma g \ln \langle \Phi_{\alpha} | S_{\gamma}(0, -\infty | g) | \Phi_{\alpha} \rangle |_{g=1}$$
(4)

The representation of the *S*- matrix in the form of the PT series induces the expansion for  $dE_a$ :

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

$$I_{\gamma}(k_1, k_2, ..., k_n) = \prod_{j=1}^{n} S_{\gamma}^{(kj)},$$
(6)

$$S_{\gamma}^{(m)} = (-1)^{m} \int_{-\infty}^{0} dt_{1} \dots \int_{-\infty}^{t_{m}-1} dt_{m} \langle \Phi_{\alpha} | V_{1} V_{2} \dots V_{m} | \Phi_{\alpha} \rangle \quad (7)$$

$$V_j = \exp(iH_0 t_j) V(rt_j)\exp(-iH_0 t_j)\exp(\gamma t_j). \quad (8)$$

Here *H* is the atomic hamiltonian,  $a(k_1, k_2, ..., k_n)$  are the numerical coefficients. The structure of matrix elements  $S_g^{(m)}$  is in details described in [19-23]. Here we only note that one may to simplify a consideration by account of the k-photon absorption contribution in the first two PT orders. Besides, summation on laser pulse is exchanged by integration. The corresponding (*l*+2k+1)-times integral on (*l*+2k) temporal variables and r (*l*=0,2) (integral  $I_g$ ) are calculated [19-23]. Finally, after some cumbersome transformations one can get the expressions for the line moments. The corresponding expressions for the Gaussian laser pulse are as follows:

$$\delta\omega(p\alpha \mid k) =$$

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

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

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

where

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

The summation in (10) is over all atomic states. Let us note that these formulas for the Gaussian pulse differ of the Lorenz shape laser pulse expressions [21-23]. For the soliton-like pulse it is necessary to carry out the numerical calculation or use some approximations to simplify the expressions [27]. In order to calculate (10), we use an effective Ivanov-Ivanova technique [28,29] of calculating sums of the QED PT second order, which has been earlier applied by us in calculations of some atomic and mesoatomic parameters [26,27,30-32]. Finally the computational procedure results in a solution of the ordinary differential equations system for above described functions and integrals. In concrete numerical calculations the PC "Superatom-ISAN" package is used. The construction of the operator wave functions basises within the QED PT, the technique of calculating the matrix elements in Egs. (9,10) and other details is are presented in Refs. [19-30]. The special features of treating the multiphoton resonances in a nucleus within the outlined approach are obviously connected with estimating the corresponding matrix elements on the basis of the nuclear wave functions and some other details. In a modern theory of a nucleus there is sufficiently great number of the different models for generating the proton and neutron wave functions basis's. At present time it is accepted that quite adequate description of the nuclear density is provided by the relativistic mean-field (RMF) and other models of the nucleus [32-36]. As alternative approach one could use the advanced RMF or shell models based on the effective Dirac-Wood-Saxon type Hamiltonian [32].

### 3. Results and conclusions

Further we present the results of the numerical simulation for the three-photon resonant, four-photon ionization profile of atomic krypton (the 4p  $\otimes$  5d[1/2], and 4p  $\otimes$  4d[3/2], three photon Kr resonances are considered). In Ref. [18] it has been performed the experimental studying the resonant multiphoton ionization of krypton by intense uv (285-310 nm) laser radiation for the intensity range 3'10<sup>12</sup>-10<sup>14</sup> W/cm<sup>2</sup>. The experiment consisted of the measurement of the number of singly charged Kr and Xe ions produced under collisionless conditions as a function of laser frequency and intensity. The output of a dye-laser system operating at 2.5 Hz is frequency doubled in a 1-cm potassium dihydrogen phosphate (KDP) crystal to give a 0.5-mJ, 1.3-ps, transformlimited 0.1-nm-bandwidth beam tunable between 285 and 310 nm. There have been determined the corresponding parameters of the 4p  $\otimes$  5d[1/2], (i) and  $4p \otimes 4d[3/2]$ , (ii) three photon Kr resonances. The resonance shift is proportional to intensity with a width dominated by lifetime broadening of the excited state. The corresponding shift and width have been found as follows: (i) the shift  $dw_0(pa|3)=aI$ ,  $a_{exp}=3.9$  meV/(Tw×cm<sup>-2</sup>); width  $b_{exp} = 1.4 \text{ meV}/(Tw \times cm^{-2});$  (ii) shift  $dw_0(pa|3) = aI$ ,  $a_{exp}^{rap}$ =8.0 meV/(Tw×cm<sup>-2</sup>); width  $b_{exp}$ =4 meV/(Tw× cm<sup>-2</sup>). The authors [18] have used quite simple model of an effective two-level atom with the assumption of a rate limiting three-photon excitation step followed by rapid one-photon ionization from the excited state. As expected, the three-photon resonances broaden and shift further as the laser pulse intensity is increases. The important feature of the corresponding profiles is linked with available asymmetry [18]. Naturally, it is easy to understand that the asymmetric profile is typical of realistic laser pulses with the spatially and temporally varying intensity. Besides, the authors of Ref. [18] have noticed that while all resonances are "blue" shifted, ac Stark shift calculations, which are difficult to perform for excited states lead to both "blue" and " red" shifts. Our numerical simulation results for the 4p  $\otimes$  5d[1/2], (i) and  $4p \otimes 4d[3/2]$ , (ii) three photon Kr resonances are as follows: (i) the shift  $dw_0(pa|3)=aI$ ,  $a_{exp}=3.95$ 

meV/(Tw×cm<sup>-2</sup>) and width  $b_{exp}$  = 1.5 meV/ (Tw×cm<sup>-2</sup>); (ii) shift  $dw_0(pa|3)=aI$ ,  $a_{exp}=8.1 \text{ meV}/$ (Tw×cm<sup>-2</sup>) and width  $b_{exp} = 4.2 \text{ meV/}(Tw×cm^{-2})$ . One could conclude that there is a physically reasonable agreement of the theoretical and experimental data.. Analysis shows that the shift and width of the multi-photon resonance line for the interaction "atom- multimode laser pulse" is greater than the corresponding shift and width for a case of the "atom- single-mode pulse" (the Lorenz pulse model) interaction. From the physical point of view it is obviously provided by action of the photon-correlation effects and influence of the laser pulse multi-modity. A great interest represents the possibility of the quantitative construction of the corresponding resonances profiles with explanation of the asymmetric nature by means calculating sufficiently "large" number of the multiphoton transition line moments. It is interesting to note that such an approach easily explains the qualitative features of the multiphoton resonances lines in the 57Fe nucleus. According to Ref. [34], the nuclear multiphoton transitions are taking a place in <sup>57</sup>Fe nucleus subjected to radio-frequency electromagnetic field  $w_0$ =30MHz. This picture was experimentally observed in the Mössbauer spectra of <sup>57</sup>Fe nuclei in Permalloy by Tittonen et al [35]. Really, the eight transitions are possible between the four hyperfine substates of the 14.4 keV excited level e and the two substates of the ground state g in the radio-frequency magnetic field [34]. If the static magnetic hyperfine splitting of the ground and excited states are respectively  $w_{o} > 0$  and  $w_{o} > 0$ , the transition frequencies corresponding to forbidden g-ray transitions are  $(E_e - E_e)/\hbar \pm 3w_e/2 \pm w_e/2$ , where  $E_e$ ,  $E_e$ are respectively the energies of the 14.4-keV and ground states of the 57Fe nucleus in an absence of any external field.

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### Abstract

The consistent relativistic energy approach to atom in a strong realistic laser field, based on the Gell-Mann and Low S-matrix formalism, is applied to studying the resonant multiphoton ionization of krypton by intense uv laser radiation and calculating the multiphoton resonances shift and width in krypton. An approach to treating the multiphoton resonances in nuclei is outlined on example of the <sup>57</sup>Fe nucleus.

Key words: electromagnetic and strong interactions, laser field, multiphoton resonances

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### СПЕКТРОСКОПИЯ АТОМА И АТОМНОГО ЯДРА В СИЛЬНОМ ЛАЗЕРНОМ ПОЛЕ: ЭФФЕКТ ШТАРКА И МНОГОФОТОННЫЕ РЕЗОНАНСЫ

### Резюме

Последовательный релятивистский энергетический подход к атому в сильном реалистичном лазерном поле, основанный на S-матричном формализме Гелл-Манна и Лоу, применяется для изучения резонансной многофотонной ионизации криптона интенсивным ультрафиолетовым лазерным излучением и вычисление многофотонных резонансных смещений и ширины в криптоне. Подход к рассмотрению многофотонных резонансов в ядрах изложен на примере ядра 57Fe.

Ключевые слова: электромагнитное и сильное взаимодействия, лазерное поле, многофотонные резонансы.

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В. В. Буяджи, О. В. Глушков, Л. Ловетт

### СПЕКТРОСКОПІЯ АТОМА І АТОМНОГО ЯДРА В СИЛЬНОМУ ЛАЗЕРНОМУ ПОЛІ: ЕФЕКТ ШТАРКА І БАГАТОФОТОННІ РЕЗОНАНСИ

#### Резюме

Послідовний релятивістський енергетичний підхід до атома в сильному реалістичному лазерному полі, оснований на S-матричному формалізмі Гелл-Манна і Лоу, застосовується для вивчення резонансної багатофотонної іонізації криптона інтенсивним ультрафіолетовим лазерним випромінюванням і обчислення багатофотонних резонансних зміщень і ширини в криптоні. Підхід до розгляду багатофотонних резонансів у ядрах викладений на прикладі ядра 57Fe.

Ключові слова: електромагнітна і сильна взаємодія, лазерне поле, багатофотонні резонанси.