

# Computational code in atomic and nuclear quantum optics: Advanced computing multiphoton resonance parameters for atoms in a strong laser field

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**Abstract.** The consistent relativistic energy approach to the finite Fermi-systems (atoms and nuclei) in a strong realistic laser field is presented and applied to computing the multiphoton resonances parameters in some atoms and nuclei. The approach is based on the Gell-Mann and Low S-matrix formalism, multiphoton resonance lines moments technique and advanced Ivanov-Ivanova algorithm of calculating the Green's function of the Dirac equation. The data for multiphoton resonance width and shift for the Cs atom and the  $^{57}\text{Fe}$  nucleus in dependence upon the laser intensity are listed.

## 1. Introduction

At the present time physics of multiphoton phenomena in atoms, molecules and nuclei has reached a great progress that is mostly stimulated by a development of new laser technologies and perspectives of new impressive applications (see Refs. [1,2]). 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 nuclear systems [1-16]. At the same time a direct laser-nucleus interactions traditionally have been dismissed because of the well known effect of small interaction matrix elements. 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.[2]) 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,  $^{153}\text{Sm}$ ,  $^{181}\text{Ta}$ ,  $^{223}\text{Ra}$ ,  $^{225}\text{Ac}$ ,  $^{227}\text{Th}$  etc) [9]. It means that the resonance condition ( $\omega \sim \Delta\varepsilon$ , where  $\Delta\varepsilon$  is a typical level spacing,  $\omega$  is a laser frequency) is fulfilled [9-12]. The coherence of the laser light expected from new sources (TESLA XFEL at DESY) may allow to access the extended complex coherence or interference phenomena. In particular, in conjunction with moderate acceleration of the target nuclei it allows principally to achieve realization of multi-photon phenomena, nuclear Rabi oscillations or more advanced quantum optical schemes in nuclei [9,10].

The interaction of atoms with the external alternating fields, in particular, laser field, has been the subject of intensive experimental and theoretical studies (see, for example, Refs. [1-27]). 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 current problems to be solved. At the present time, a progress is achieved in the description of the processes for atoms interacting with the harmonic emission field [9, 14-17]. But in a realistic laser field the corresponding processes differ significantly from the ones in a harmonic field. A substantial complexity is provided by the photon-correlation effects and an influence of the laser pulse multi-mode features. Notably, a number of different theoretical approaches have been developed in order to give adequate description of the atoms in a strong laser field. The corresponding methods are the standard perturbation theory (surely for low laser field intensities), the Green's function method, the density-matrix formalism, time-dependent density functional formalism, a direct numerical solution of the Schrödinger and/or Dirac equation, multi-body multi-photon approach, the time-independent Floquet formalism *etc.* (see [1,14-28] and references therein). The effects of the different laser line shape on an intensity and spectrum of resonance fluorescence from a two-level atom are studied in Refs. [1-5,15-17,19-23].

Earlier, a relativistic energy approach to studying the interaction of atoms with a realistic strong laser field, based on the Gell-Mann and Low S-matrix formalisms, has been developed. Originally, Ivanov has proposed an idea to describe quantitatively a behavior of an atom in a realistic laser field by means of studying the radiation emission and absorption lines and further the consistent theory of interaction of an atom with a Lorentzian laser pulse has been developed in Refs. [19-22]. The theory has been applied to computing the multi-photon resonance shifts and widths in hydrogen and caesium [20]. The consistent theory of interaction of an atom with the Gaussian and soliton-like laser pulses has been developed in Refs. [24-27], where the corresponding calculation results are listed.

Here we present an advanced generalized relativistic energy approach to computing multiphoton resonances parameters in the finite Fermi-systems (as example, atom of Cs and nucleus of Fe) in a strong laser field with using the Green's function technique for computing sums in expressions for a multiphoton resonance line moments.

## 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 are presented in Refs. [17-27] and here we are limited only by presenting the master moments. In the theory of the non-relativistic atom there is a convenient field procedure for computing the energy shifts  $\delta E$  of degenerate states, which is connected with the secular matrix  $M$  diagonalization. In constructing  $M$ , one could apply the known Gell-Mann and Low adiabatic formula for  $\delta E$  (look details in Refs. [19-22]). In relativistic theory, the Gell-Mann and Low formula  $\delta E$  is connected with electro-dynamical scattering matrices, which includes interaction with a laser field as a photon vacuum field. In relativistic theory the secular matrix elements are already complex in the second perturbation theory (PT) order and their imaginary parts are connected with radiation decay possibility. The total energy shift is usually presented in the form:  $\delta E = \text{Re}\delta E + i \text{Im}\delta E$ ,  $\text{Im} \delta E = -P/2$ , where  $P$  is the level width. Spectroscopy of an atom (nucleus) in a strong laser field is determined by position and shape of the radiation emission and absorption lines. The lines moments  $\mu_n$  are strongly dependent upon the laser pulse quality: intensity and mode constitution. We describe the interaction "nucleus-laser field" by the Ivanov potential [20]:

$$V(r,t) = V(r) \int d\omega f(\omega - \omega_0) \sum_{n=-\infty}^{\infty} \cos(\omega_0 t + \omega_0 n \tau). \quad (1)$$

Here  $\omega_0$  is the central laser radiation frequency,  $n$  is the whole number. The potential  $V$  represents the infinite duration of laser pulses with known frequency  $\tau$ . The function  $f(\omega)$  is a Fourier component of the laser pulse. For example, we consider a pulse with the Gaussian shape. A case of the Lorentzian shape has been earlier studied [20,21]. A case of the Gaussian and soliton-like shape is studied in Refs. [24-27]. The master program results in the calculating an imaginary part of an energy shift  $\text{Im}\delta E_\alpha(\omega_0)$

for any atomic level as the function of the central laser frequency  $\omega_0$ . An according function has the shape of the resonance, connected with the transition  $\alpha$ - $p$  with absorption (or emission) of the “ $k$ ” number of photons. For the resonance we calculate the following values [19-21]:

$$\delta\omega(p\alpha|k) = \int' d\omega \operatorname{Im} \delta E_\alpha(\omega) (\omega - \omega_{p\alpha}/k) / N, \quad (2a)$$

$$\mu_m = \int' d\omega \operatorname{Im} \delta E_\alpha(\omega) (\omega - \omega_{p\alpha}/k)^m / N, \quad (2b)$$

where  $\int' d\omega \operatorname{Im} E_\alpha$  is the normalizing multiplier;  $\omega_{p\alpha}$  is position of the non-shifted line for transition  $\alpha$ - $p$ ,  $\delta\omega(p\alpha|k)$  is the line shift under  $k$ -photon absorption;  $\omega_{p\alpha} = \omega_{p\alpha} + k \cdot \delta\omega(p\alpha|k)$ . The first moments  $\mu_1$ ,  $\mu_2$  and  $\mu_3$  determine an atomic line centre shift, its dispersion and asymmetry. In order to compute the values  $\mu_m$ , one should get an expansion of  $E_\alpha$  to the PT series [20]:  $E_\alpha = \sum E_\alpha^{(2k)}(\omega_0)$ . Here one may use the known Gell-Mann and Low adiabatic formula for  $\delta E_\alpha$  and the further representation of the  $S$ -matrix in the form of the PT [20]:

$$\delta E_\alpha(\omega_0) = \lim_{\gamma \rightarrow 0} \gamma \sum_{k_1 k_2 \dots k_n} a(k_1, k_2, \dots, k_n), \quad (3)$$

$$I_\gamma(k_1, k_2, \dots, k_n) = \prod_{j=1}^n (-1)^j \int_{-\infty}^0 dt_1 \dots \int_{-\infty}^{t_{m-1}} dt_j \langle \Phi_\alpha | V_1 V_2 \dots V_m | \Phi_\alpha \rangle,$$

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

Here  $H$  is the atomic hamiltonian,  $a(k_1, k_2, \dots, k_n)$  are the numerical coefficients. The structure of matrix elements  $S_\gamma^{(m)}$  and integral technique are in details described in [17-23]. The corresponding expressions for the Gaussian laser pulse are as follows [24,25]:

$$\delta\omega(p\alpha|k) = \{\pi\Delta / (k+1)k\} [E(p, \omega_{p\alpha}/k) - E(\alpha, \omega_{p\alpha}/k)], \quad (4)$$

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

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

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

where the summation in (5) is over all atomic states. Note that these formulas for the Gaussian pulse [24,25] differ from the expressions for Lorentzian shape laser pulse [19-22]. For the soliton-like pulse, it is necessary to fulfil the numerical calculation or use some approximations to simplify the expressions [27]. Earlier to determine the the PT second order sum in (5) in different calculations of atomic and meso-atomic characteristics [28-36], it has been applied the effective Ivanov-Ivanova technique [18]. The alternative approach can be provided by method of the Green's function of the Dirac equation (look below). The corresponding computational procedure includes solving an ordinary differential equations system for above mentioned functions and integrals. The scheme of computing electron c wave functions within the relativistic many-body PT with the Dirac-Kohn-Sham zeroth approximation, the corresponding technique of computing all matrix elements in the expressions (2,5) and other details are presented in Refs. [20-30]. In all numerical calculations the PC “Superatom-ISAN” package is used.

Further it is important to underline that above presented expressions remain the same for treating the multiphoton resonances in a nucleus in a strong laser field, however there is some difference in comparison with an atomic case. It is obviously connected with calculating the corresponding matrix

elements in formulae for resonance parameters with substituting 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 (c.f. [37,38]). At present time, it is acceptable that a quite adequate description of the nuclear systems is provided by the relativistic mean-field (RMF) and other similar models [37,38]. As an alternative approach one could use the advanced shell models based on the effective Dirac-Wood-Saxon type Hamiltonian [26,39]. This model [39] has been used in our work.

### 3. Method of the Green's function of the Dirac equation with complex energy: Advanced Ivanov-Ivanova computational algorithm

The details of the method of computing the sum in (5) are presented in Refs. [18,20,23,27]. The fundamentals of the method have been developed by Ivanov-Ivanova et al (c.f. [18-20]). The advanced version of this approach has been presented in Ref. [23]. Here we are limited only by the principal points of an advanced computational algorithm [23]. As usually, the radial Dirac equations can be written as follows (in the Coulomb units):

$$\begin{aligned} f' &= -(\chi + 1)f / r - V^- g \tilde{\alpha}; \\ g' &= (\chi - 1)g / r + V^+ f \tilde{\alpha}; \\ V^\mp &= V(r) - i\xi \mp \tilde{\alpha}^{-2}; \end{aligned} \quad (6)$$

where  $V(r)$  is the potential of a nucleus,  $\tilde{\alpha} = \alpha Z$  ( $\alpha$  is the fine structure constant),  $f$  and  $g$  are the Dirac equations solutions. We are interested by the case, when the potential is regular for  $r \rightarrow 0$ . It is easy to show (c.f. [18]) that for such a potential the solutions of two types (regular and non-regular at  $r \rightarrow 0$ ) exist for each value of  $\xi$  and  $\chi$ :

for  $\chi < 0$

$$f \sim r^{|\chi|-1}, \quad g \sim r^{|\chi|}, \quad \tilde{f} \sim r^{-|\chi|}, \quad \tilde{g} \sim r^{-|\chi|-1},$$

for  $\chi > 0$

$$f \sim r^{|\chi|}, \quad g \sim r^{|\chi|-1}, \quad \tilde{f} \sim r^{-|\chi|-1}, \quad \tilde{g} \sim r^{-|\chi|}. \quad (7)$$

The regular solution  $(f, g)$  at  $r \rightarrow 0$  is simply defined by the condition (6) with the accuracy to a normalization. At the same time the singular solutions are not defined by these conditions.

For large values of  $|\chi|$  the functions (7) have a strong degree dependence at  $r \rightarrow 0$  that is a reason of the known computational difficulties during the numerical integration of the Dirac equations. At large  $\chi$  the radial functions  $F$  and  $G$  vary rapidly at the origin of co-ordinates (c.f.[19,28]):

$$\begin{aligned} F(r), G(r) &\approx r^{\gamma-1} \\ \gamma &= \sqrt{\chi^2 - \alpha^2 z^2} \end{aligned} \quad (8)$$

and further it is convenient to introduce the new functions isolating the main power dependence:

$$(F, G) = (f, g) \cdot r^{-|\chi|};$$

$$(\tilde{F}, \tilde{G}) = (\tilde{f}, \tilde{g}) \cdot r^{|\chi|+1}. \quad (9)$$

The Green's function is a combination of the Dirac equation modified (the power dependence is separated) fundamental solutions:

$$F' = -(\chi + |\chi|)F/r + V^- \tilde{\alpha}G; \quad G' = (\chi - |\chi|)G/r - V^+ \tilde{\alpha}F; \quad (10a)$$

$$\tilde{F}' = -(\chi + |\chi|)\tilde{F}/r + V^- \tilde{\alpha}\tilde{G}; \quad \tilde{G}' = (|\chi| + \chi)\tilde{G}/r - V^+ \tilde{\alpha}\tilde{F}. \quad (10)$$

The functions  $(F, G)$  represent the first fundamental solution, which is regular for  $r \rightarrow 0$  and singular for  $r \rightarrow \infty$ . Any combination  $(\tilde{F}, \tilde{G}) + Cr^{2|\chi|}(F, G)$  satisfies to above written equations for  $(\tilde{F}, \tilde{G})$  and represents singular solution at zero [18,19,23]. The right chosen combination  $(\hat{F}, \hat{G})$  for the single value of the mixing coefficient  $C$  (regular for  $r \rightarrow \infty$ ) is second fundamental solution  $(\hat{f}, \hat{g})$ . Finally, the Green's function electron function is a four-component matrix with the functions  $(F, G)$  and  $(\hat{F}, \hat{G})$ , which are the Dirac equations solutions with account of the corresponding asymptotic conditions. Further one can obtain from Eqs. (10) that for  $r \rightarrow \infty$  :

$$(F, G) \sim \exp Ar; \quad (\hat{F}, \hat{G}) \sim \exp(-Ar); \quad A = (\tilde{\alpha}^{-2} + \xi^2 \tilde{\alpha}^2)^{1/2}. \quad (11)$$

As the bi-linear combinations of the Dirac function components are presented in the Green's function, only their relative normalization is important. It is determined by the known "Wronscian" condition ( $W = F\hat{G} - \hat{F}G \equiv 1$ ). The electron radial Green's function is the four component matrix as follows:

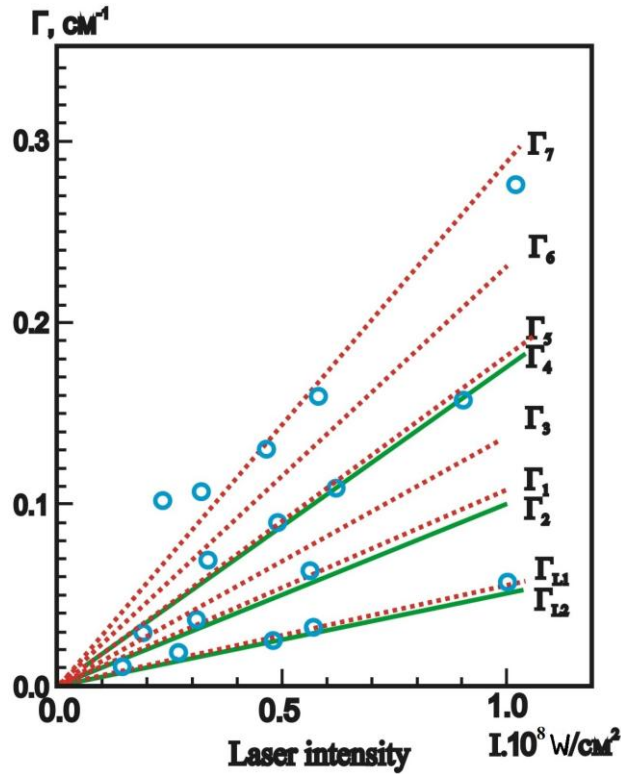
$$G(r_1 r_2 | E, \chi) = \begin{pmatrix} \hat{F}(r_>)F(r_<) & \hat{F}(r_>)G(r_<) \\ \hat{G}(r_>)F(r_<) & \hat{G}(r_>)G(r_<) \end{pmatrix}, \quad (12)$$

where  $r_>$  ( $r_<$ ) is more (or less) value of  $r_1, r_2$ ; the functions  $(F, G)$  and  $(\hat{F}, \hat{G})$  satisfy the Dirac equations (10), their asymptotical conditions (11) and the "Wronscian" normalization condition. The corresponding computational procedure, which includes solving an ordinary differential equations system for relativistic wave functions, computing all matrix elements and other details can be found in Refs. [23-28,39].

#### 4. Some results and conclusions

Below we present the calculation results for the three-photon transition 6S-6F ( $p-\alpha$  transition) in the  $^{133}\text{Cs}$  atom (wavelength 1,059  $\mu\text{m}$ ). The detailed experimental studying the multi-photon processes in Cs atom has been fulfilled in Ref. [15]. In this paper it has been experimentally studied a statistics of the laser radiation and there have been measured the characteristics of the multi-photon ionization. According to Ref.[15], the multiphoton line shift is linear to respect to a laser intensity (the laser intensity is increased from 1.4 to  $5.7 \cdot 10^7 \text{ W/cm}^2$ ) and is equal (a case of the Gaussian multi-mode laser pulse):  $\delta\omega(p\alpha|k) = bI$  with  $b = (5.6 \pm 0.3) \text{ cm}^{-1}/\text{GW} \cdot \text{cm}^{-2}$  ( $b$  is expressed in terms of energy of the three-photon transition 6S-6F). The corresponding shift for the coherent (one-mode) laser pulse is equal as follows [15]:  $\delta\omega_0(p\alpha|k) = aI$ ,  $a = 2 \text{ cm}^{-1}/\text{GW} \cdot \text{cm}^{-2}$ . The theoretical values, obtained within the

relativistic energy approach [22,24,25] are as follows: i).  $\delta\omega(p\alpha|k)=bI$  with  $b=5.8 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2}$  for the Gaussian multi-mode pulse (chaotic light); ii).  $\delta\omega_0(p\alpha|k)=aI$ ,  $a=2.1 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2}$  for the coherent one-mode pulse. The corresponding theoretical values, obtained in this work, are as follows: i).  $\delta\omega(p\alpha|k)=bI$ ,  $b=5.62 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2}$  for the Gaussian multi-mode pulse; ii).  $\delta\omega_0(p\alpha|k)=aI$ ,  $a=2.03 \text{ cm}^{-1}/\text{GW}\cdot\text{cm}^{-2}$  for the coherent one-mode pulse. One should note that the radiation line shift for the multi-mode pulse is significantly larger (in  $\sim 3$  times) than the corresponding shift for the single-mode pulse. In fact the resonance line shift is enhanced by the photon-correlation effects. In figure 1 we present the results of computing the multi-photon resonance width for transition 6S-6F in the atom of Cs (wavelength 1059nm) in dependence upon a laser intensity. We use the following denotations: experimental data- o (Grance ; Lompre etal, 1981); theory – by Glushkov-Ivanov (the energy approach plus method of the differential equations; continuous lines)  $\Gamma_{L2}$  (1-mode Lorentzian pulse),  $\Gamma_2, \Gamma_4$  - multimode Gaussian laser pulse (the linewidth 0.03, 0.08  $\text{cm}^{-1}$ ); this work: the dotted lines  $\Gamma_{L1}$  (the Lorentzian pulse),  $\Gamma_1, \Gamma_3, \Gamma_5, \Gamma_6, \Gamma_7$  - multimode Gaussian laser pulse (the linewidth 0.03, 0.055, 0.08, 0.115, 0.15  $\text{cm}^{-1}$  correspondingly). In Ref. [15] there are presented experimental data for laser pulse of the Gaussian shape with line band respectively 0.03 $\text{cm}^{-1}$ , 0.08 $\text{cm}^{-1}$ , 0.15 $\text{cm}^{-1}$ .



**Figure 1.** The results of computing the multi-photon resonance width for transition 6S-6F in the atom of Cs (wavelength 1059nm) in dependence upon laser intensity: experimental data- o (Grance, Lompre et al [15]); theory – Glushkov-Ivanov (the energy approach plus method of the differential equations; continuous lines)  $\Gamma_{L2}$  (1-mode Lorentzian pulse),  $\Gamma_2, \Gamma_4$  - multimode Gaussian laser pulse (the linewidth 0.03, 0.08  $\text{cm}^{-1}$ ) [20,24]; This work: the dotted lines  $\Gamma_{L1}$  (the Lorentzian pulse),  $\Gamma_1, \Gamma_3, \Gamma_5, \Gamma_6, \Gamma_7$  - multimode Gaussian laser pulse (the linewidth 0.03, 0.055, 0.08, 0.115, 0.15  $\text{cm}^{-1}$  correspondingly).

In general there is a physically reasonable agreement between theoretical and experimental data. The analysis shows that the shift and width of the multi-photon resonance line in atomic system for the multimode laser pulse are greater than the corresponding resonance shift and width for the one-mode

laser pulse. This fact is entirely corresponding to the experimental result [15]. From physical point of view it is provided by action of the photon-correlation effects and influence of the laser pulse multimodality (c.f.[14,15,20-27]).

Further let us present the first results of estimating the multiphoton resonance parameters in the  $^{57}\text{Fe}$  nucleus in a super strong laser field. In order to estimate the corresponding matrix elements on the nuclear wave functions it was used the shell model based on the effective Dirac-Wood-Saxon type Hamiltonian, which contains the Bloumkvist-Wahlborn nuclear potential [39]. According to Ref. [40], the nuclear multiphoton transitions take a place in  $^{57}\text{Fe}$  nucleus subjected to radio-frequency electromagnetic field  $\omega_0=30\text{MHz}$ . This picture was experimentally observed in the Mössbauer spectra of  $^{57}\text{Fe}$  nuclei in Permalloy by Tittonen et al [41]. 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 [16]. If the static magnetic hyperfine splitting of the ground and excited states are respectively  $\omega_g>0$  and  $\omega_e>0$ , the transition frequencies corresponding to forbidden  $\gamma$ -ray transitions are  $(E_e-E_g)/h\pm 3\omega_g/2\pm\omega_e/2$ , where  $E_e$ ,  $E_g$  are respectively the energies of the 14.4-keV and ground states of the  $^{57}\text{Fe}$  nucleus in an absence of any external field. The estimates of the  $^{57}\text{Fe}$  resonance shift in dependence upon the mean laser intensity are as follows:  $I=10^{25}\text{W/cm}^2$ ,  $\delta\omega=5\cdot 10^{-12}\text{keV}$ ,  $I=10^{28}\text{W/cm}^2$ ,  $\delta\omega=10^{-9}\text{keV}$ . The corresponding asymmetric nature of the multiphoton resonance line in  $^{57}\text{Fe}$  nucleus is naturally explained within the presented theoretical approach.

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