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Spectroscopy of atom and nucleus in a strong laser field: Stark effect and multiphoton resonances

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Abstract. The consistent relativistic energy approach to atoms in a strong realistic laser field, based on the Gell-Mann and Low S-matrix formalism, is applied in the study of resonant multiphoton ionization of krypton by intense uv laser radiation and for the computation of the resonance shift and width in krypton. The approach to the treatment of the multiphoton resonances in nuclei is outlined for the ^{57}Fe nucleus.

1. Introduction

At the present time, the physics of multiphoton phenomena in atoms and molecules shows great progress that is stimulated by development of new laser technologies (see Refs. [1-10]). The availability of powerful laser sources allows one to obtain a radiation field amplitude on the order of atomic fields in the wide range of wavelengths. Consequently, systematic investigations of the nonlinear interaction of radiation with atomic and molecular systems can be possible [1-14]. At the same time, 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 considered earlier. 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 (see 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%, 12.4 keV photons produced by SASE 1 become resonant with the $E1$ transition in a whole number of nuclei (for example, ^{153}Sm , ^{181}Ta , ^{223}Ra , ^{225}Ac , ^{227}Th etc). In other words, the resonance condition ($\omega \sim \Delta\varepsilon$, where $\Delta\varepsilon$ is a typical level spacing, ω is a laser frequency) can be fulfilled [10]. The coherence of the laser light expected from new sources (TESLA XFEL at DESY) may allow one 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 multi-photon 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 studies (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 current problems to be solved. At present time, a progress is achieved in the description of the processes of atoms interacting with the harmonic emission field [1, 12-14]. But in a realistic laser field the according processes differ significantly from



the ones in the harmonic field. A substantial role is played by the photon-correlation effects and the influence of the multi-mode character of laser pulse. Notably, a number of different theoretical approaches have been developed in order to give adequate description of the atoms in a strong laser field. Methods of interest include standard perturbation theory (surely for low laser field intensities), Green function method, the density-matrix formalism, time-dependent density functional formalism, direct numerical solution of the Schrödinger and/or Dirac equation, multi-body multi-photon approach, the time-independent Floquet formalism *etc.* (see [1-8,12-24] and references 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 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 the behavior of an atom in a realistic laser field by means of studying the radiation emission and absorption lines and, further, the theory of interaction of an atom with a Lorentzian laser pulse and calculation of the corresponding line moments has been developed in detail in Ref. [19-25]. Multi-photon resonances shifts and widths in hydrogen and caesium have been confirmed in numerical simulations. Theory of interaction of an atom with the Gaussian and soliton-like laser pulses and calculations of the corresponding lines moments have been presented in detail in Refs. [23, 26, 27]. Here we apply this approach to the study of the resonant multiphoton ionization of krypton by intense uv laser radiation and compute the multiphoton resonances shift and width. We also outline the corresponding scheme to treating the multiphoton resonances in nuclei on example of ^{57}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 are presented in detail in Refs. [19-30]. Here we only present the essential elements. In the theory of the non-relativistic atom, a convenient field procedure is known for calculating the energy shifts δE of degenerate states. This procedure is connected with the secular matrix M diagonalization. In constructing M , the Gell-Mann and Low adiabatic formula for δE is used [20-23, 31]. In relativistic theory, the Gell-Mann and Low formula δE is connected with electrodynamic scattering matrices, which include interactions of a laser field with a photon vacuum field. The case of interaction with photon vacuum corresponds to standard theory of radiative decay of excited atomic states. 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]:

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

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 μ_n are strongly dependent upon the laser pulse quality, namely intensity and mode composition [15-23]. Let us describe the interaction “atom-laser field” by the Ivanov potential [21,23]:

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

Here, ω_0 is the central laser radiation frequency, n is an integer number. The potential V represents the duration of laser pulses, τ , with known frequency. The function $f(\omega)$ is a Fourier component of the laser pulse. The condition $\int d\omega f^2(\omega) = 1$ normalizes the potential $V(rt)$ on the definite energy in the pulse. Let us consider the pulses with Lorentzian shape (coherent 1-mode pulse): $f(\omega) = \beta/(\omega^2 + \Delta^2)$, Gaussian one (multi-mode chaotic pulse): $f(\omega) = \beta \exp[\ln 2(\omega^2/\Delta^2)]$ and the soliton-like pulse: $f(t) = \beta ch^{-1}[t/D]$ (β -normalizing multiplier). The case of the Lorentzian shape has been earlier studied [20-23]. The case of the Gauss and soliton-like shape is considered in Refs.[23,26,27]. The master program results in calculating an imaginary part of energy shift $\text{Im}\delta E_\alpha(\omega_0)$ for any atomic level as the function of the

central laser frequency ω_0 . An according function has the shape of the resonance, which is connected with the transition α - p (α , p -discrete levels) with absorption (or emission) of the “ k ” number of photons. For the resonance we calculate the following values [20-23]:

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

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

where $\int' d\omega \operatorname{Im} E_\alpha$ is the normalizing multiplier; $\omega_{p\alpha}$ is position of the non-shifted line for transition α - 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 μ_1 , μ_2 and μ_3 determine the atomic line centre shift, its dispersion and the asymmetry. To find μ_m , we need to obtain an expansion of E_α to PT series: $E_\alpha = \sum E_\alpha^{(2k)}(\omega_0)$. One may use here the Gell-Mann and Low adiabatic formula for δE_α :

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

The representation of the S -matrix in the form of the PT series induces the expansion for δE_α :

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

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

$$S_\gamma^{(m)} = (-1)^m \int_{-\infty}^0 d t_1 \dots \int_{-\infty}^{t_m-1} d t_m \langle \Phi_\alpha | V_1 V_2 \dots V_m | \Phi_\alpha \rangle, \quad (7)$$

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

Here, H is the atomic Hamiltonian, $a(k_1, k_2, \dots, k_n)$ are the numerical coefficients. The structure of matrix elements $S_\gamma^{(m)}$ is further described in [19-23]. Here we only note that one may simplify the considerations by accounting for the k -photon absorption contribution in the first two PT orders. Moreover, summation on laser pulse is exchanged with the integration. The corresponding $(l+2k+1)$ -times integral on $(l+2k)$ temporal variables and ρ ($l=0,2$) (integral I_γ) are calculated [19-23]. Finally, and only following some tedious transformations, one can obtain the expressions for the line moments. The corresponding expressions for the Gaussian laser pulse are as follows:

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

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

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

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 Equation (10) is over all atomic states. Note that these formulas for the Gaussian pulse differ from the expressions for Lorentzian-shape laser pulses [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]. To evaluate Eq. (10), we use an effective Ivanov-Ivanova technique [22,28] of calculating sums of QED PT second order, which has been applied earlier in calculations of atomic and meso-atomic parameters [26,27,30-32]. Finally, the computational procedure includes solving an ordinary differential equations system for above described functions and integrals. In specific

numerical calculations the PC “Superatom-ISAN” package is used. The construction of the operator wave functions basis within the QED PT, the technique of calculating the matrix elements in Eqs. (9, 10) and other details are presented in Refs. [19-30]. Special features of treating the multiphoton resonances in a nucleus within the outlined approach are obviously connected with estimating the corresponding matrix elements in the basis of the nuclear wave functions and some other details. In a modern theory of a nucleus, there are a sufficiently great number of the different models for generating the proton and neutron wave functions bases. At present time, it is acceptable that a quite adequate description of the nuclear density is provided by the relativistic mean-field (RMF) and other models of the nucleus [32-35]. 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

Here we present the results of the numerical simulation for the three-photon resonant, four-photon ionization profile of atomic krypton (the $4p \rightarrow 5d[1/2]_1$ and $4p \rightarrow 4d[3/2]_1$ three photon Kr resonances are considered). In Ref. [18], an experimental study has been presented of the resonant multi-photon ionization of krypton by intense uv (285-310 nm) laser radiation for the irradiance range of 3×10^{12} - 10^{14} W/cm². 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, transform-limited 0.1-nm-bandwidth beam, tunable between 285 and 310 nm. In these experiments, the corresponding parameters of the following transitions have been determined: (i) $4p \rightarrow 5d[1/2]_1$ and (ii) $4p \rightarrow 4d[3/2]_1$ 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 $\delta\omega_0(p\alpha|3)=aI$, $a_{\text{exp}}=3.9$ meV/(Tw·cm⁻²); width $b_{\text{exp}}=1.4$ meV/(Tw·cm⁻²); (ii) shift $\delta\omega_0(p\alpha|3)=aI$, $a_{\text{exp}}=8.0$ meV/(Tw·cm⁻²); width $b_{\text{exp}}=4$ meV/(Tw·cm⁻²). The authors have applied [18] a quite simple model for 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 increased. The important feature of the corresponding profiles is linked to 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 noted that while all resonances are “blue” shifted, ac Stark shift calculations are difficult to perform for excited states leading to both “blue” and “red” shifts. Our numerical simulation results for the (i) $4p \rightarrow 5d[1/2]_1$ and (ii) $4p \rightarrow 4d[3/2]_1$ three-photon Kr resonances are as follows: (i) the shift $\delta\omega_0(p\alpha|3)=aI$, $a_{\text{exp}}=3.95$ meV/(Tw·cm⁻²) and width $b_{\text{exp}}=1.5$ meV/(Tw·cm⁻²); (ii) shift $\delta\omega_0(p\alpha|3)=aI$, $a_{\text{exp}}=8.1$ meV/(Tw·cm⁻²) and width $b_{\text{exp}}=4.2$ 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 specific case of the “atom- single-mode pulse” (the Lorentzian pulse model) interaction. From the physical point of view, it is obviously provided by the action of the photon-correlation effects and influence of the multi-mode laser pulse. Of great interest is the possibility of quantitative construction of the corresponding resonance profiles with explanation of the asymmetric nature by means of calculating sufficiently “large” number of the multiphoton transition line moments. Such an approach may easily explain 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 ⁵⁷Fe nucleus subjected to radio-frequency field $\omega_0=30$ MHz. This scenario was experimentally observed in the Mössbauer spectra of ⁵⁷Fe nuclei in Permalloy by Tittonen *et al.* [35]. Really, the eight transitions are possible between the four hyperfine sub-states of the 14.4 keV excited level e and the two sub-states 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 $\omega_g > 0$ and $\omega_e > 0$, the transition frequencies corresponding to forbidden γ -ray transitions are $(E_e - E_g)/h \pm 3\omega_g/2 \pm \omega_e/2$, where E_e and E_g are respectively the energies of the 14.4-keV and ground states of the ^{57}Fe nucleus in an absence of any external field.

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