Quantum Geometry: Energy-amplitude approach to multiphoton resonances and above threshold ionization

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Abstract An advanced energy-amplitude approach to calculation of the characteristics of multi-photon ionization in atomic systems is presented and based on the many-body perturbation theory. It is used for numerical calculating the above threshold ionization characteristics for atom of magnesium in a intense laser field.

1 Introduction

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. 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, dinamical 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. t present time, a progress is achieved in the description of the processes of interaction atoms with the harmonic emission field [1]–[8]. 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. [1]–[8]. Decay probabilities of the hydrogen atom states in the super-strong laser field are calculated by the Green function method (see [2]) 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. In [2],[9] authors extended the non-Hermitian multi-state Floquet dynamics approach by Day 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 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 [9] in j-j coupling introducing explicitly spin-orbit effects and employing both the length and velocity forms of the electric dipole transition operator. 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 [1,2]. We consider an advanced amplitude approach to calculation of the characteristics of multi-photon ionization in atomic systems, which is based on the many-body perturbation theory (PT) and use it for numerical calculating the above threshold ionization (ATI) characteristics for atom of magnesium in a intense laser field.

2 Energy QED approach to multiphoton resonances and above threshold ionization

In this section we consider a quite exact approach based on the QED perturbation theory [2]–[7], which allow to calculate the characteristics of multi-photon ionization in atomic systems. 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 relativistic

PT order. This approach is valid away from any one-photon intermediate-sate resonance. We start from the two-photon amplitude for the transition from an initial state Ψ_0 with energy E_0 to a final state $|Psi_f$ with energy $E_f = E_0 + 2\omega$ is:

$$T_{f0}^{(2)} = \lim_{n \to 0_+} \int d\epsilon \langle \Psi_f | D \times e | \epsilon \rangle (E_0 + \omega - \epsilon + in)^{-1} \langle \epsilon | d \times e | \Psi_0 \rangle. \tag{1}$$

Here D is the electric dipole transition operator (in the length r form), e is the electric field polarization and ω is a laser frequency. It's self-understood that the integration in equation 1 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 [3]:

$$T_{f0}^{(2)} = C_f \langle \| D \times e \| \Lambda_p \rangle, \tag{2}$$

where $\langle \| ... \| \rangle$ is a reduced matrix element and C_f is an angular factor depending on the symmetry of the Ψ_f, Λ_p, Ψ_0 states. Λ_p can be founded from solution of the following inhomogeneous equation [3]

$$(E_0 + \omega \times H | \Lambda_p \rangle = (D \times e) | \Psi_0 \rangle \tag{3}$$

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 cm^4 W^{-1}) is defined as

$$\sigma/I = \sum_J \sigma_J/I = 5.7466 \times 10^{-35} \omega_{au} \sum_J |T_{J,0}^{(2)}|^2, \tag{4}$$

where I (in W/cm^2) is a laser intensity. To describe two-photon processes there can be used different quantities [9]: the generalized cross section $\sigma^{(2)}$, given in units of cm^4s, by

$$\sigma_{cm^4s}^{(2)} = 4.3598 \times 10^{-18} \omega_{au} \sigma / I_{cm^4W^{-1}} \tag{5}$$

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

$$\sigma/I_{cm^4W^{-1}} = 9.1462 \times 10^{-36} \omega_{au} \Gamma_{au}^{(2)} / I_{au}^2 \tag{6}$$

Described approach is realized as computer program block in atomic numeric code "Super-atom" (c.f. [2]–[7], which includes a numeric solution of the Dirac equation and calculation of the matrix elements of the Eqs. 1–5 type. The original

moment is connected with using the consistent QED gauge invariant procedure for generating the atomic functions basis's (optimized basis's) [8]. 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.

3 Some results and conclusion

Let us present the results of calculating the multi-photon resonances spectra characteristics for atom of magnesium in a laser field. Note that in order to calculate spectral properties of atomic systems different methods are used: relativistic R-matrix method (R-method; Robicheaux-Gao, 1993; Luc-Koenig E. etal, 1997), added by multi channel quantum defet 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.(c.f.[2,9]. In table 1 we present results of calculating characteristics for $3p^2$ $^1S_0$ resonance of Mg; E- energy, counted from ground state (cm^{-1}), Γ- autoionization width (cm^{-1}), σ/I- maximum value of generalized cross-section (cm^{4}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 ω 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 cm^{-1} and 131477cm^{-1}. First photon provides the AS ionization, second photon can populate the Rydberg resonance's, owning to series *4snl,3dnl,4pnp* where J=0 and J=2 [9]. In table 2 we present energies (cm^{-1} ; counted from the ground level of Mg 3s^2) and widths (cm^{-1}) of the AS (resonance's) $4snl, 3dnl, 4p^2$ $^1D_2$, 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 interference and fluctuations) resonances in spectrum,

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

Methods	E	Γ	σ/I
Luc-Koenig E. etal, 1997	without	account	SE
Length form	68492	374	$1,96\ 10^{-27}$
Velocity form	68492	376	$2,10\ 10^{-27}$
Luc-Koenig E. etal, 1997	With	Account	SE
Length form	68455	414	$1,88\ 10^{-27}$
Velocity form	68456	412	$1,98\ 10^{-27}$
Moccia and Spizzo (1989)	68320	377	$2,8\ 10^{-27}$
Robicheaux and Gao (1993)	68600	376	$2,4\ 10^{-27}$
Mengali and Moccia(1996)	68130	362	$2,2\ 10^{-27}$
Karapanagioti et al (1996)	68470	375	$2,2\ 10^{-27}$
Our calculation	68281	323	$2,0\ 10^{-27}$

Table 2 Energies and widths (cm^{-1}) of the AS (resonance's) $4snl, 3dnl, 4p^{2\,1}D_2$ for Mg (see text)

$^1D_2$	R−method $E\ \Gamma$	$^1D_2$	Our approach $E\ \Gamma$		K− method $E\ \Gamma$
4s3d	109900 2630	4s3d	109913 2645		110450 2600
$3d^2$	115350 2660	$3d^2$	115361 2672		115870 2100
4s4d	120494 251	4s4d	120503 259	(ds)	120700 170
3d5s	123150 1223	3d5s	123159 1235	(ds)	123400 2000
$4p^2$	124290 446	$4p^2$	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		

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 (c.f. refs. in [2,3,9]). Analysis indicates on its existence in the Mg spec-

trum. 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:

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

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

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