The Basis of "Atom in the External Field" Eigenfunctions to the Problem of High Harmonic and Terahertz Radiation Generation Study

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Abstract: The paper is devoted to the discussion of the main principles of the non-perturbative quantum-mechanical approach to the description of a single atom interaction with multicomponent laser fields. The main advantage of the theory is that the authors use a basis of "an atom in the field" eigenfunctions which are the exact solution of "an atom in the field" boundary value problem the Hamiltonain of which coincides with the one from the Schrodinger equation written in the velocity gauge. The theory is applied to analytical and numerical investigation of the high-order harmonic generation and the terahertz radiation generation phenomena.

1 INTRODUCTION

High order harmonic generation (HHG) is one of the most promising tools for generation of coherent ultraviolet and X-ray radiation (Popmintchev, 2012). The elementary act of harmonic generation lies in the scale of a single atom interaction with a laser field. There are a lot of theoretical approaches which are used to describe the HHG (see the introduction part of the (Andreev, 2012)). Intuitively the process can be understood in the frame of the "simple man model" (Krausz, 2009): an electron is ionized by an intense laser field, accelerated inside the oscillating laser field and gained kinetic energy, then it comes back to a bound state emitting a burst of photons with an attosecond pulse duration.

The terahertz (THz) radiation generation also has a lot of potential applications in molecular spectroscopy, imaging etc., that is why it is under an active study now. The process of atomic or molecular gas interaction with a multicolor laser field is one of the most effective tools for the generation of high intensity broadband pulsed THz radiation (Cook, 2000). The fundamental act of interaction with a laser field accompanied with the THz generation lies in the atomic (Karpowicz, 2009; Zhou, 2009; Zhang, 2012; Andreev, 2012; Andreev, 2013) or media (Kim, 2007; Couairon, 2007) scales. That is why different physical mechanisms have been used to describe this phenomenon: the four-wave mixing process (Cook, 2000), the photocurrent of free charges (Kim, 2007; Babushkin, 2011), the plasma current oscillation (Debayle, 2014) and intra-atomic nonlinearity mechanism (Andreev, 2013).

Here we discuss the quantum-mechanical non-perurbative theory of a single atom interaction with a multicomponent laser field which could simultaneously describe the HHG and the THz radiation generation phenomena. The main advantage of the theory is in the absence of the smallness parameter E/E_{at} (E_{at}= 5.1•10^9 V/cm being the intra-atomic field strength value). As a result, the theory can precisely describe the phenomena appearing in sub- and near-atomic laser fields. The main principles of the theory are discussed below (for more details, please, see (Andreev, 2011)).

2 BASIC PRINCIPLES OF THE THEORY

The process of a single atom interaction with a laser field can be described with the Schrodinger equation which has the form of:

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$$i\hbar\frac{\partial\psi(\vec{r},t)}{\partial t}=\left[\frac{1}{2m}\left(\vec{p}-\frac{q}{c}\vec{A}(t)\right)^2+U(r)\right]\psi(\vec{r},t), \quad (1)$$

where $\vec{A}(t)$ is the vector potential and $U(\vec{r})$ is the intra-atomic field potential.

To solve the eq. (1) we used a non-traditional basis of "an atom in the external field" wave-functions $\varphi_N(\vec{r},t)$ which is the exact solution of the boundary value problem of an atom in the external field:

$$\left[\frac{1}{2m}\left(\vec{p}-\frac{q}{c}\vec{A}(t)\right)^2+U(r)\right]\varphi_N(\vec{r},t)= =E_N\varphi_N(\vec{r},t). \quad (2)$$

The operator of the boundary value problem (2) coincides with the Hamiltonian of eq. (1), so these two equations have the same symmetry properties. The eigenfunctions $\varphi_N(\vec{r},t)$ can be analytically expressed in terms of eigenfunctions $u_n(\vec{r})$ for the free-atom boundary value problem:

$$\varphi_N(\vec{r},t)=u_n(\vec{r})\hat{V}^{-1}, \quad \hat{V}=\exp\left(-i\frac{q}{\hbar c}\vec{A}(t)\vec{r}\right).$$

Similar to a set of free-atom eigenfunctions $u_n(\vec{r})$ which form a complete basis of the orthonormal functions, the eigenfunctions of the boundary value problem (2) for "an atom in the external field" form also a complete basis of orthonormal functions $\varphi_N(\vec{r},t)$. There is a one-to-one correspondence between these two bases. Note that the eigenfunctions $\varphi_N(\vec{r},t)$ coincide exactly with the eigenfunctions $u_n(\vec{r})$ when the instant value of the external field amplitude is equal to zero. Hence, these two bases coincide at the time points when *I(t)=0*; and what is more important they coincide before the laser pulse arrival and after its termination.

As we have mentioned above, the eigenfunctions $\varphi_N(\vec{r},t)$ have the same symmetry properties as the wavefunction of the Schrodinger equation (1). Therefore, it looks quite natural to use the basis of these functions for solving the eq. (1). However, due to the time derivative in the left-hand-side of equation (1) the equations for the probability amplitudes of such expansion will inevitably include the integrals over the products of these eigenfunctions and their time derivatives. But the operator of the boundary value problem (2) is time dependent; hence, the eigenfunctions of this problem and their time derivatives are not orthogonal. To overcome this problem we can initially expand the wavefunction $\psi(\vec{r},t)$ into a series of eigenfunctions $u_n(\vec{r})$:

$$\psi(\vec{r},t)=\sum_{n,l}a_{n,l}(t)u_{n,l}(\vec{r})+\int a(k,l,t)u(k,l,\vec{r})dk$$

and then make use of the one-to-one correspondence of these two bases. Moving from eq. (1) to a set of equations for the probability amplitudes we should calculate the following integral:

$$\int u_n^*\left(\frac{1}{2m}(\vec{p}-\frac{e}{c}\vec{A}(t))^2+U(r)\right)u_m dV.$$

Decomposing $u_n(\vec{r})$ through the set of $\varphi_N(\vec{r},t)$ we can find this integral analytically:

$$\int u_n^*\left(\frac{1}{2m}(\vec{p}-\frac{e}{c}\vec{A}(t))^2+U(r)\right)u_m dV= =\sum_p V_{np}^{-1}(t)E_pV_{pm}(t)$$

and then write a set of differential equations for the population amplitudes of discrete states and continuum quasistates:

$$i\hbar\frac{da_n(t)}{dt}=\sum_{m,k}V_{nk}^{-1}(t)E_kV_{km}(t)a_m(t), \quad (3)$$

where E_k are the energy eigenvalues.

To calculate the spectrum of atomic response we should calculate previously the atomic current density:

$$\vec{j}(\vec{r},t)= =\frac{q}{2m}\left[\psi^*\cdot\left(\vec{p}-\frac{q}{c}\vec{A}\right)\psi+\left(\left(\vec{p}-\frac{q}{c}\vec{A}\right)\psi\right)^*\cdot\psi\right]. \quad (4)$$

In the far-field zone the spectrum of atomic response coincides with the spectrum of atomic current (Landau, 1981), which is:

$$\vec{J}(t)=i\sum_{n,m,p,q}a_n^*(t)a_m(t)\omega_{pq}V_{np}^{-1}(t)\vec{d}_{pq}V_{qm}(t) \quad (5)$$

where $a_n(t)$ are the probability amplitudes of atomic states, $\vec{d}_{pq}$ are the matrix elements of the dipole operator and $\omega_{pq}=\left(E_p-E_q\right)/\hbar$.

Notice that in all the equations above the atomic states were designated by the one-letter symbol (*n*). However, the atomic states of the three-dimensional

spherically symmetric boundary value problem depend on three quantum numbers: a principle quantum number n, an orbital quantum number l, and its projection m. By writing only a one-letter symbol we mean all the three quantum numbers from the previous formulas.

The equations (3, 5) enable to calculate the photoemission spectrum at given parameters of the laser field interacting with an atom and describe the features of the HHG spectrum (the short wavelength part of the photoemission spectrum) as well as the THz spectrum (the long wavelength part of the photoemission spectrum). However the set (3) include the infinite number of equations. The infinite set of equations (3, 5) cannot be solved neither analytically nor numerically. On the other hand, at any finite amplitude of the laser field only some finite number of atomic levels makes an appreciable input in the atomic response. The main advantage of the "an atom in the external field" basis is the following: the input of each state can be numerically calculated before we solve the set of equations for probability amplitudes. We can exactly estimate the accuracy of calculations with the help of truncated basis at any amplitude of the laser field. It should be also noted that the number of states in "an atom in the external field" basis is truncated, but each eigenfunction of this basis is the infinite series over the eigenfunctions of the "free atom" basis and the coefficients of this decomposition depend on the laser field amplitude.

2.1 Matrix Elements of the V Operator

Let us have a look at the matrix elements of the V operator. To calculate it analytically we will write the free-atom boundary value as a multiplication of its radial part $R_{n,l}(r)$ and its angular part $Y_{l,m}(\theta,\varphi)$: $u_{n,l,m}(\vec{r}) = R_{n,l}(r)Y_{l,m}(\theta,\varphi)$. The matrix element of the transition between two states described by two sets of quantum numbers $|n_1 l_1 m_1\rangle$ and $|n_2 l_2 m_2\rangle$ has the form of

$$\begin{aligned}\langle n_2 l_2 m_2 | V^{-1} | n_1 l_1 m_1 \rangle = \sum_{l=|l_1-l_2|}^{l_1+l_2} Y_{lm}^{*}(\vec{e}(t))\cdot \\ \cdot(-1)^{l+m_2}\begin{pmatrix} l_2 & l & l_1 \\ -m_2 & m & m_1 \end{pmatrix}\begin{pmatrix} l_2 & l & l_1 \\ 0 & 0 & 0 \end{pmatrix}\cdot \\ \cdot\sqrt{4\pi(2l+1)(2l_1+1)(2l_2+1)}\cdot \\ \cdot\int_0^{\infty} R_{n_2 l_2}(r)\, j_l(\mu(t))\, R_{n_1 l_1}(r) r^2 dr,\end{aligned} \quad (6)$$

where $\mu(t) = \frac{q}{\hbar c}A(t)r$, $j_l(\mu(t))$ are generalised Bessel functions, $\vec{e}(t)$ is the unit polarization vector of the laser field. Assuming that the vector-potential can be presented by the following

$$A(t) = \sum_i A_{0i} e^{-\left(\frac{t-t_{0i}}{\tau_i}\right)^2} \sin\left((\omega_i + \alpha_i t)(t-t_{0i}) + \sigma_i\right),$$

where A_{0i} is the amplitude of the vector potential of the components of the laser fields, τ_i are the temporal widths of the pulses, ω_i, α_i, σ_i, t_{0i} are the frequencies of the components of the laser fields, their chirps, phases and delays, respectively; we can set the control parameter of the theory as

$$\mu_{0i} = \frac{q}{\hbar c}A_{0i}r.$$

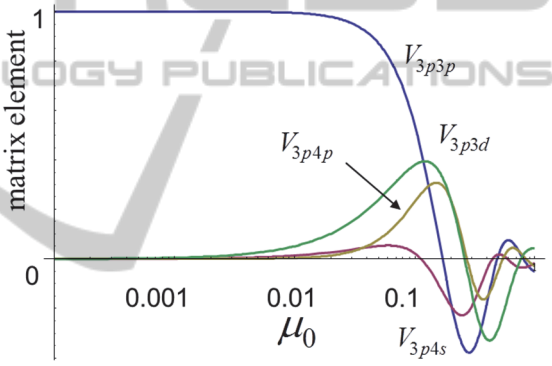

Figure 1: Matrix elements for discrete-discrete transitions as a function of the field strength μ_0

Using the hydrogen-like wave-functions $u_{n,l,m}(\vec{r})$ we can analytically integrate (6) and investigate the properties of the matrix elements. Figure 1 represents the behaviour of some matrix elements calculated between the discrete states as a function of the control parameter value. It is clearly seen that the matrix elements demonstrate non-linear and non-monotonical behaviour. As a result, the atomic response has also the non-linear dependency on the laser field amplitude which is qualitatively different in subatomic and near-atomic regions. The expansion of the matrix elements into the series of the laser field amplitudes includes all the powers of the ratio E/E_{at}. So, any multiquantum process is accounted in a consecutive manner.

Some matrix elements calculated between discrete and continuum states as a function of electron energy calculated at two values of the control parameter are presented in figure 2. It is

clearly seen that the non-monotonical behaviour of the matrix elements strongly depends on the value of the laser field strength (the value of the control parameter). Moreover, in the region of high electron energy the value of the matrix elements decreases and we can estimate the upper boundary of the of photoelectron energy region which must be taken into account for the calculation of the system of equations (3) with a given accuracy.

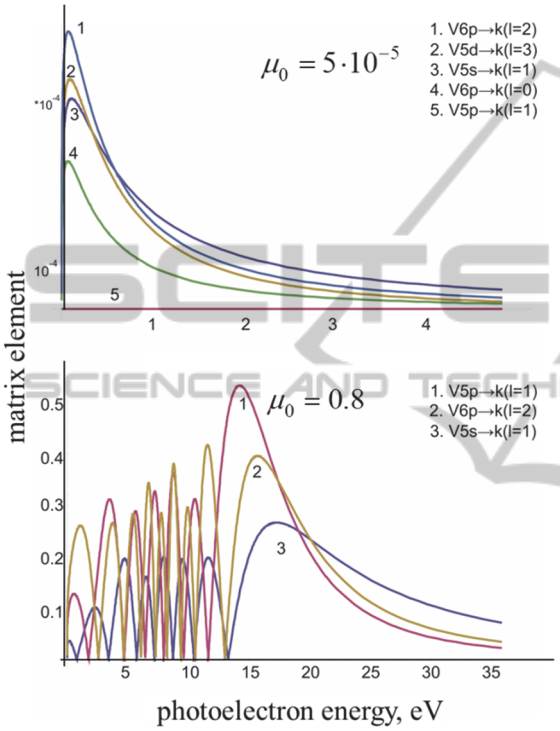

Figure 2: Matrix elements for discrete-continuum transitions as a function of the photoelectron energy calculated at a given value of the control parameter $\mu_0 = 5\cdot10^{-5}$ (a), $\mu_0 = 0.8$ (b).

2.2 Matrix Elements of the J Operator

The mathematical formalism provides us with a possibility to calculate the angular-frequency spectrum (AFS) of the atomic response for the case of an arbitrary mutual orientation of the atomic angular momentum and the laser field polarization vector. The polarization of the AFS components depends on both the angular momentum direction and the polarization of the incident field. In the non-polarized ensemble of atoms the response field polarization depends only on the polarization state of the laser field.

In order to investigate the convergence of the usage of the truncated basis of wave-function let us have a look at the atomic current calculated for only one level (ground state-ground state $\langle 0|\vec{j}|0\rangle$) transition.

$$P_{00} = \langle 0|\vec{j}|0\rangle = \\ = i\sum_{n_1 l_1 m_1}\sum_{n_2 l_2 m_2}\omega_{n_1 l_1 m_1, n_2 l_2 m_2}\langle 0|V^{-1}|n_1 l_1 m_1\rangle\cdot \quad (7) \\ \cdot\langle n_1 l_1 m_1|\vec{d}|n_2 l_2 m_2\rangle\langle n_2 l_2 m_2|V|0\rangle.$$

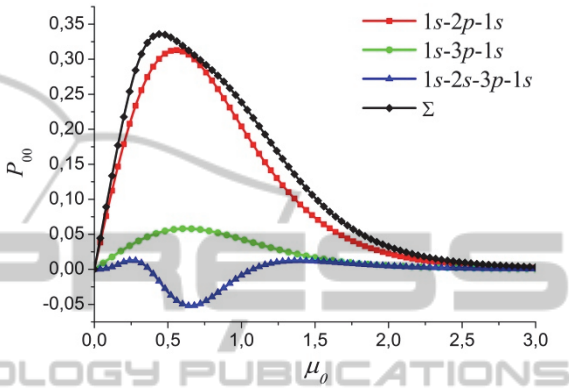

Figure 3: Matrix elements for J operator as function of field strength μ_0.

Despite the fact that the initial and the final states of this transition are fixed, the value of this matrix element depend on the impact of the exited states. Figure 3 shows the dependence of this matrix element calculated for the case of the hydrogen atom (1s ground state), with taking into account only one exited state (2p – a curve with squares, 3p – a curve with circles), two exited states (2s and 3p – a curve with triangles), three exited states (2s, 2p, 3p – a curve with rhombuses). Figure 3 demonstrates fast convergence of the sum (7) since the curve calculated with taking into account the first exited state of the atom (the curve with squares) almost perfectly represents the behavior of the matrix element calculated with taking into account the impact of three states (the curve with rhombuses).

3 APPLICATION OF THE THEORY

Figure 4 represents the typical photoemission spectrum calculated for the case of an Ar atom interaction with a two-colour laser field formed by the fundamental and the second harmonics of the Ti:Sapphire laser, the parameters of which have the form of $\mu_{01} = \mu_{02} = 0.1,\ \tau_1 = \tau_2 = 26.6 fs,$

$t_{02}-t_{01}=0,\ \alpha_i=0,\ \sigma_i=0$, the angle between the polarization of the components of the field being equal to $\theta=\frac{21\pi}{48}$ (Andreev, 2013). We assume here that the fundamental harmonic is polarized along the z-axis, and the second harmonic is polarized in zy-plane. It is clearly seen that the spectrum consists of both odd and even harmonics which have non-zero projections on the two perpendicular axes. The information about the polarization properties of the generated harmonics can be directly extracted from the photoemission spectrum with the help of the Stokes parameters.

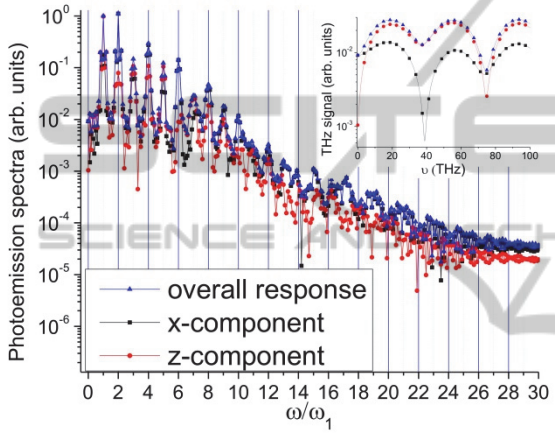

Figure 4: The photoemission spectrum of an Ar atom interacting with the two-colour laser field formed by the fundamental and the second harmonics of the Ti:Sapphire laser: the integral intensity of response (triangles) and the intensities of the two orthogonally polarized components (squares and circles). The parameters of the two-colour laser field are the following: $\mu_{01}=\mu_{02}=0.1$, $\tau_1=\tau_2=26.6fs,\ t_{02}-t_{01}=0,\ \alpha_i=\sigma_i=0,\ \theta=21\pi/48$. (Inset) The THz part of the photoemission spectrum (Andreev, 2013).

The inset in the figure 4 demonstrates the THz (long wavelength) part of the photoemission spectrum. The signal has also non-zero projections on the two perpendicular axes.

The theory described above was applied for the investigation of some features of the HHG and the THz radiation phenomenon. We theoretically explained the saturation of the cut-off frequency in near-atomic laser field (Andreev, 2011; Andreev, 2013). The value of the cut-off frequency coincides with the experimentally measured one (Andreev, 2013). We also theoretically investigated the HHG (Andreev, 2013) and the THz radiation generation (Andreev, 2013) in the ionization-free regime in the case of a two-colour laser field interaction with an atom. What is more interesting in this investigation is that the HHG spectra are not limited to below-threshold and near-threshold harmonics which are effectively generated in the same region of the laser field intensities (Sofier, 2010; Yost, 2009). The specific features of the THz radiation emitted by the extended gas interacting with a two-color laser field is been investigated in (Stremoukhov, 2015). It is shown that spatial oscillations of the THz radiation efficiency appearing during the dispersion effects in the gas change the conical structure of the THz radiation. The theory was also applied for the interpretation of the resent experiment of the effective generation of high intensity high ellipticity harmonics in two-colour orthogonally polarized laser fields (Lambert, 2015)

4 CONCLUSIONS

The basic principles of the quantum-mechanical non-perturbative theory based on the usage of the bases of "an atom in the external field" eigenfunctions are described and discussed in the application to the description of the HHG and the THz radiation generation phenomena. It is shown that the usage of these bases of functions enables taking into account the symmetry properties of the problem and, thus, brings the numerical investigation of the light-atom interaction to a new level. What is more important, with the help of the theory ones can calculate the atomic response for the case of an arbitrary mutual orientation of the atomic angular momentum and the laser field polarization vector. The recent applications of the theory are named and discussed shortly.

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