High harmonic generation under dynamical confinement: An atom-in-box model

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We consider optical high harmonic generation in a hydrogen atom confined in a breathing spherical box by considering atomic nucleus as fixed at the center of sphere. In such spherically symmetric, dynamical trap, the high harmonic generation spectrum is calculated at different values of the oscillation amplitude, confinement size and atomic nucleus charge.

I. INTRODUCTION

Study of the nonlinear optical phenomena in the interaction of atoms and molecules with external optical fields is of practical and fundamental importance for the problems, e.g., attosecond physics, high-power laser generation, optical materials design and optoelectronic device fabrication, etc. An interesting aspect of this topic is optical harmonic generation in quantum regime, which attracted much attention recently \textsuperscript{1-10}. One of the main tasks in this field is achieving non-, or slowly-decaying intensity of the generated high harmonics. Usually, in free atoms and molecules interacting with external optical fields intensity of generated harmonic decreases, as the order of harmonic increases. Such effect makes difficult generating of very high order harmonics and ultrashort pulses, as their intensity becomes very small. Therefore revealing the regimes for high harmonic generation, providing non-decaying intensity (as a function of harmonic order) is of importance for different practical tasks. One of such ways could be confining of atoms and molecules in finite spatial domain, where changing confinement size and geometry one can control the harmonic generation.

In this paper we discuss a model for high harmonic generation in an atom confined in a dynamical (time-dependent) trap. The latter presents a spherical box with harmonically breathing radius. The nucleus of atom is considered as fixed at the center of sphere, so that spherical symmetry of whole system is not broken. It is well known that confined systems have completely different physical properties compared to the free ones. Such difference is caused by the boundary conditions imposed for the Schrödinger equation, which cause modification of the energy and wave function spectrum.

Atoms and molecules confined in nanoscale domains have the The earlier studies of atom-in-box system date back to the Refs. \textsuperscript{16-17}, where effect of the pressure on an atom was explored in quantum approach. Later, Wigner studied the problem within the Rayleigh-Schrödinger perturbation theory and showed that in the limit of infinite box size, the result does not converge into that for the free atom. Considerable number of papers on the atom-in-box problem (see, e.g., \textsuperscript{14-33} and review paper \textsuperscript{34} for more references) has been published, since from these pioneering works. In \textsuperscript{19} the problem of hyperfine splitting in such system is treated, Ref.\textsuperscript{20} presents first numerical solution of the problem. More comprehensive treatment of atom-in-box system can be found in a series of papers by Burrows et.al \textsuperscript{22-25,32,33}, where the authors used different analytical and numerical methods for finding eigenvalues of the system. In \textsuperscript{28,30,31} the quantum dynamics of hydrogen atom confined in a spherical box and driven by external electric field is studied.

Experimentally, atom-in-box system can be realized, e.g., in so-called atom optic billiards which represent a rapidly scanning and tightly focused laser beam creating a time-averaged quasi-static potential for atoms \textsuperscript{35-41}. By controlling the deflection angles of the laser beam, one can create various box (billiard) shapes. Another method is putting the hydrogen atom inside the fullerene \textsuperscript{13,14}. Different ways for confining of atom inside a cage in experiment are discussed in \textsuperscript{34}. We note that all the studies of atom-in-box system are mainly focused by considering the case of box static boundaries, while modern technologies in quantum and atom optics provide different tools for creating dynamical confinement. Some versions for experimental realization of such dynamical traps have already been discussed in the literature (see, e.g., the Refs.\textsuperscript{42,43}). In such cases the dynamics of atomic electron is completely different than that for fixed boundaries.

In this paper we consider high harmonic generation in quantum regime by an atom confined in a "breathing" spherical box. To solve the time-dependent Schrödinger equation for Coulomb potential with dynamical boundary conditions we use the same approach as that in our recent paper \textsuperscript{44}.

We note that the problem of moving boundaries in quantum mechanics is treated in terms of the Schrödinger equation with time-dependent boundary conditions. Earlier, the quantum dynamics of a particle confined in a time-dependent box was studied in different contexts (see Ref. \textsuperscript{47-62}). Here we consider similar problem for an electron moving in a Coulomb field of the atomic nucleus, confined in a harmonically breathing spherical box.

This paper is organized as follows. In the next section...
we give brief description of a quantum system, consisting of one-electron atom confined in a breathing spherical box. Section 3 presents detailed treatment of high harmonic generation in such system. Finally, section 4 provides some concluding remarks.

II. QUANTUM DYNAMICS OF BY HYDROGEN-LIKE ATOM CONFINED IN A TIME-DEPENDENT SPHERICAL BOX

Consider atom confined in a spherical box with time-varying radius given by \( r_0 = r_0(t) \). In this case the sphere retains its shape during the expansion(contraction), so that the central symmetry is not broken. Therefore, if atomic nucleus is fixed at the center of sphere, the electron dynamics is described by the time-dependent radial Schrödinger equation which is given as

\[
i \frac{\partial R(r, t)}{\partial t} = \hat{H} R(r, t),
\]

where

\[
\hat{H} = \frac{1}{2} \frac{\partial^2}{\partial r^2} - \frac{1}{r} \frac{\partial}{\partial r} + \frac{l(l+1)}{2r^2} - \frac{Z}{r}.
\]

For such regime, the boundary conditions for Eq. (1) are imposed as

\[
R(r, t)|_{r=r_0(t)} = 0.
\]

To solve Eq. (1) one should reduce the boundary conditions into time-independent form. This is can done by using the following coordinate transformation [51, 52]:

\[
y = \frac{r}{r_0(t)}.
\]

In terms of new coordinate, \( y \) Eq. (1) can be rewritten as

\[
i \frac{\partial R(y, t)}{\partial t} = \left[ -\frac{1}{2r_0^2} \frac{\partial^2}{\partial y^2} - \left( \frac{1}{2r_0^2} - \frac{i}{r_0} y \right) \frac{\partial}{\partial y} + \frac{1}{2r_0^2} \frac{\partial^2}{\partial y^2} \right] R(y, t) \equiv \hat{\tilde{H}} R(y, t).
\]

In Eq. (4) the self-adjointness is broken, i.e., operator \( \hat{\tilde{H}} \) is not Hermitian. In addition, due to the first-order derivative in this equation makes complicated its solution. Therefore, to restore Hermitticity and remove the first order derivative, one can use the transformation of the wave function which is given by

\[
R(y, t) = \frac{1}{r_0(t)^{3/2}} e^{\int_{t_0}^{t} ds r_0(s)^2} \Phi(y, t).
\]

Doing such transformation and introducing of the new time-variable defined as [51, 54, 61]

\[
\tau = \int_{t_0}^{t} ds \frac{1}{r_0(s)^2},
\]

we reduce Eq. (1) into the Hermitian for which can be written as

\[
i \frac{\partial \Phi}{\partial \tau} = -\frac{1}{2} \frac{\partial^2 \Phi}{\partial y^2} + \left( \frac{1}{2} r_0^2 y^2 + \frac{l(l+1)}{2y^2} - \frac{Zr_0}{y} \right) \Phi.
\]

The boundary condition for \( \Phi \) is imposed as

\[
\Phi(y, t)|_{y=1} = 0.
\]

We note that Eq. (6) can be obtained from Eq. (1) by using following unitary transformation for the Hamiltonian \( H \) [51, 54, 61]:

\[
\Phi(y, t) = \frac{1}{r_0(t)^{3/2}} e^{\int_{t_0}^{t} ds r_0(s)^2} \Phi(y, t).
\]
\[ \hat{H} = e^{-iV} e^{-iU} (\hat{H} - i \frac{\partial}{\partial t}) e^{iU} e^{iV}, \]

where

\[ U = i (r \frac{\partial}{\partial r} + \frac{3}{2} \ln r_0(t)), \]

and

\[ V = -\frac{1}{2} r_0 \frac{d r_0}{d t} y^2. \]

Eq. (6) is the Schrödinger equation for an electron moving in the field of Coulomb and time-dependent harmonic oscillator potentials. The charge of the Coulomb field is time-dependent due to the factor \( r_0(t) \). The whole system is confined in a spherical box with unit radius. Time and coordinate variables cannot be separated in Eq. (6) and one needs to solve it numerically. To do this we expand \( R(y, t) \) in terms of the complete set of eigenfunctions of a spherical box with unit radius:

\[ \Phi(y, t) = \sum_{n l} C_{n l}(t) \phi_{n l}(y), \tag{7} \]

where \( \phi_{n l}(y) = N_{n l} y j_l(\lambda_{n l} y) \) are the eigenfunctions of the stationary Schrödinger equation for a spherical box of unit radius, \( j_l \) are the spherical Bessel functions. Inserting this expansion into Eq. (6) we get the system of first order differential equations with respect to \( C_{n l}(t) \):

\[ i C_{n l}(t) = r_0^{-2} \sum_{n' l'} C_{n' l'}(t) V_{n n' l l'}(t) + \varepsilon_{n l} r_0^{-2} C_{n l}, \tag{8} \]

where \( \varepsilon_{n l} \) are the eigenvalues of the Schrödinger equation for spherical box and

\[ V_{n n' l l'}(t) = < \phi_{n l'} | - \frac{Z r_0}{y} + \frac{1}{2} \frac{3 \varepsilon_{n l} y^2}{r_0^2} | \phi_{n l} > . \]

In solving Eqs. (8) numerically one should take into account the normalization condition for the expansion coefficients, \( C_{n l} \):

\[ \sum_{n l} |C_{n l}(t)|^2 = 1, \]

which follows from the normalization condition for the wave function:

\[ \int_0^{r_0(t)} |\Psi(r, t)|^2 r^2 \, dr = 1. \]

Initial conditions for Eq. (5) are imposed as an atomic state with a given set of quantum numbers, \((n_1, l_1)\), i.e., all coefficients, \( C_{n l} = 0 \), except \( C_{n_1 l_1} \).

Having found the wave function \( \Psi(r, t) \), one can compute the main characteristics of high harmonic generation, i.e., the average dipole moment, which is given by

\[ \bar{d}(t) = - \langle R(r, t) | r | R(r, t) \rangle, \]

where the wave function \( R(r, t) \) is related to \( \Phi(y, t) \) via Eq. (4).

We are interested in the study of optical harmonic generation in the system described by Eqs. (1) and (2). The spectrum of harmonic generation is characterized by the power spectrum, i.e., absolute square of the Fourier transform of the average dipole moment, which is given by

\[ |\bar{d}(\omega)|^2 = \frac{1}{T} \int_0^T e^{-i \omega t} \bar{d}(t) dt|^2, \tag{9} \]

where \( T \) is the interaction time.

### III. HIGH HARMONIC GENERATION BY HYDROGEN-LIKE ATOM CONFINED IN A BREATHING SPHERICAL BOX

Atom-in=-box system described by Eqs. (1) and (2) have been studied in the Ref. [46] by focusing on the atomic electron’s dynamics. Here we will consider high harmonic generation in this system caused by the interaction of atomic electron with the oscillating wall. In atom optic experiments such a ”wall” can be created e.g., by an optical field. In the following we will focus on the regime, when the wall of the box is harmonically oscillating, i.e., in harmonically breathing regime, where the radius if given by \( r_0(t) = a + b \cos \omega_0 t \), where \( \omega_0 \) and \( b \) are the oscillating frequency and amplitude, respectively.
For the "virtual" system described by Eq. (9) the oscillating potential term can be written in the form of multichromatic field including 4 different frequencies as

\[ V(y, t) = \frac{1}{2} \alpha_0^3 v y^2 = \frac{1}{2} (A + B \cos \omega_0 t + C \cos 2\omega_0 t + D \cos 3\omega_0 t + E \cos 4\omega_0 t) y^2 \]

where

\[ A = -\frac{3b^2\omega_0^2}{8} (4a^2 + b^2), \quad B = -\frac{3ab\omega_0^2}{4} (4a^2 + 9b^2), \]

\[ C = -\frac{b^2\omega_0^2}{2} (3a^2 + b^2), \quad D = -\frac{3ab\omega_0^2}{4}, \quad E = -\frac{b^4\omega_0^2}{8}. \]

Thus the virtual system can be considered as a quantum box with unit size and driven by nonlinearly polarized multichromatic field. In Fig. 1 spectra of high harmonic generation are plotted at different values of the atomic nucleus charges. By analyzing the high harmonic generation spectra in the dynamical confinement regime, one may achieve the regime, the "cur-off" frequency can be increased. Possible increasing the cut-off frequency has been discussed earlier in [3].

IV. CONCLUSIONS

In this work we studied high harmonic generation by hydrogen-like atom under the dynamical confinement created by impenetrable spherical box with time-dependent radius. The main focus of the study is given to the dependence of high harmonic generation intensity on the box initial size, oscillation amplitude and atomic nucleus charge. The analysis of high harmonic generation spectra is done using the numerical solutions of time-dependent Schrodinger equation for Coulomb potential, for which the time-dependent box boundary conditions are imposed. Behavior of of the Fourier transform (power spectrum) of the average dipole moment is studied for different box breathing regimes, as well as for for different atomic nucleus charges. By analyzing the high harmonic generation spectra in the dynamical confinement regime we found that the intensity of generation is much higher than that for unconfined (free) atom. This implies that the above model of high harmonic generation can be very effective proposal for attosecond pulse generation.

ACKNOWLEDGEMENT

This work is partially supported by the grant of the Ministry of Innovation Development of Uzbekistan (Ref. No. BF-2-022).

[1] Xiao-Min Tong, Shih-I Chu, Chem. Phys. B 217 119 (1997).
[2] T. Brabec and F. Krausz, Rev. Mod. Phys., 72 545 (2000).
[3] I. Yousef, et.al, Phys. Rep., 427 41 (2006).
[4] C. Winterfeldt, C. Spielmann, and G. Gerber, Rev. Mod. Phys., 80 117 (2008).
[5] F. Krausz, M. Ivanov, Rev. Mod. Phys., 80 117 (2008).
[6] M. Nisoli, G.Sansone, Prog. Quant. Electr. 33 17 (2009).
[7] M.C.Kohler, T.Pfeiler, K.Z.Hatsagortsyan, C.H.Keitel, Advances In Atomic, Molecular, and Optical Physics, 62 159 (2012).
[8] V.V. Strelkov, V.T. Platonenko, A.F. Sterzhantov and M.Yu. Ryabikin, Phys. Uspekhi 59 (2016).
[9] C. F. de Morisson Faria and J-M. Rost, Phys. Rev. A 62 051402(R) (2000).
[10] R.W. Boyd, Nonlinear Optics.3rd ed., Academic Press.(2007).
[11] Theory of Confined Quantum Systems-Part One, J.Sabin and E. Brandas (Eds.), Advances in Quantum Chemistry Academic, 57 (2009).
[12] A.N. Sila, S. Canutob and P.K. Mukherjee, Advances in Quantum Chemistry Academic, 58 115 (2009).
[13] K.D. Sen (Eds), Electronic Structure of Quantum Confined Atoms and Molecules. Springer (2014).
