High harmonics generation from excited states of atomic lithium.

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Abstract. We present a calculation of the harmonics yield from the lithium atom driven by a pulse of $\lambda = 3.5$ μm laser. Our calculation shows that a considerable increase of the yield of high harmonics can be achieved if initially the atom is prepared in an excited $2p$ state or in a superposition of the ground and excited states. Dynamic analysis shows that harmonics yield enhancement in this case is a result of mutual interaction of several excited atomic states.

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1. Introduction

High harmonic generation (HHG) is a nonlinear atomic process which manifests itself in appearance of odd-order multiple frequencies in the spectra of the atom placed in an intense electromagnetic (EM) field. From the theoretical side, this is a relatively well understood phenomenon. Many essential features of the HHG, such as the existence of the plateau in the spectrum (Krause *et al* 1992, L'Huillier and Balcou 1993), can be explained using classical concepts (the so-called 3-step or recollision model (Corkum 1993). The model describes the HHG phenomenon as a 3-step process consisting of tunneling ionization of an atomic electron at the moment of time which ensures that electron in the EM field, following a (completely classical) trajectory, will eventually return to the nucleus and recombine. This model was used as a basis for a number of semiclassical approaches (Becker *et al* 1994, Kuchiev and Ostrovsky 1999, Kuchiev and Ostrovsky 2001). Entirely quantum description of the HHG process has also been developed (Usachenko and Pazderezsky 2002). Qualitatively, these later approaches do not modify significantly the clear picture of the 3-step model. From the quantum mechanical point of view, HHG can still be regarded as release and quantum evolution of the atomic electron (described e.g., with the help of the fully quantum-mechanical strong field approach as in cited work of Usachenko and Pazderezsky (2002)) followed by emission of the HHG photon. The typical pattern of the resulting HHG spectrum consists of the first few generally quickly decreasing harmonics followed by the plateau ending with a relatively sharp cut-off.

The first works in which such a structure of the spectrum was established considered the atom initially in the ground state driven by the single colour EM field. The spectrum can be modified if these conditions are changed. Extension of the recollision model for the case when the atom is driven by a combination of two fields with different frequencies (Watanabe *et al* 1994, Protopapas *et al* 1995) shows that in this way one extends the spectrum to higher orders. Numerical study of the combined effect of two fields on the hydrogen atom (Ishikawa and Midorikawa 2002) showed that by adjusting properly the frequencies of the driving fields one can enhance the high harmonics in the spectrum.

Another approach to the problem of the modification of the HHG spectra is choosing excited atomic state as an initial state of the process. It was shown that admission of the coherent superpositions of various atomic states as initial state may considerably modify the HHG spectrum (Sanpera *et al* 1996) introducing additional plateaus in the spectrum. In work by Paul *et al* (2005), several orders of magnitude increase in harmonics yield, in particular for the 7-th and 9-th harmonics, was observed in the experiment on Rb atoms. Experimental conditions were such that few rubidium states ($5s$, $5p$, $4d$) were populated. The observed enhancement of the harmonics yield was attributed to mutual interaction of these levels.

In the present work, we perform a study of the HHG process from the lithium atom, initially in the excited $2p$ state or in a superposition of the ground and excited states. We show that a considerable increase of the harmonics yield can be achieved in this case. Our interest in lithium stems from recent experiments on the intense laser field ionization of magneto-optically trapped (MOT) Li atoms (Steinmann 2007). The strong field ionization was driven by a Ti:Sapphire laser at $\lambda = 0.795$ μm. However, the combination of the laser wavelength and the characteristic field intensities used in this experiment did not satisfy the HHG condition as the plateau of the harmonics spectrum would contain only few harmonics. To stretch the HHG plateau, we apply in the present study a driving pulse with $\lambda = 3.5$ μm, as in the experimental work by Paul *et al* (2005). In addition to the lowest $2p$ excited state, we shall also briefly consider the process of harmonics generation from higher lying Rydberg states of Li.

2. Theory.

We describe the field-free lithium atom in the ground state by solving a set of self-consistent Hartree-Fock equations (Chernysheva *et al* 1976). We adopt the single active electron approach and describe the one-electron excitations from the valence $2s$ shell in the frozen-core approximation. This approximation provides reasonably accurate description of the manifold of excited Li states. It places the excitation energies of $2p$, $3s$, $3p$, and $3d$ states at respectively 1.841, 3.333, 3.797 and 3.830 eV, which compares well with the known transition energies of Li shown in Figure 1.

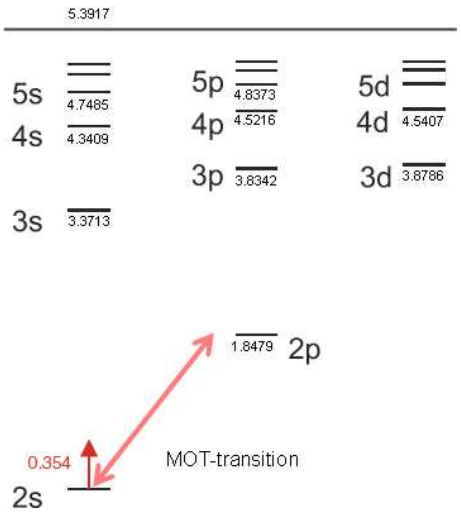

Figure 1. Energy levels of Li. The excitation energies (in eV) are from the NIST database (Sansonetti *et al* 2005). The arrows indicate the photon energy of the driving 3.5 μm laser and the MOT $2s \rightarrow 2p$ transition

The time dependent Schrödinger equation (TDSE) describing the motion of the valence electron in the presence of the external EM field can be written as:

$$i\frac{\partial \Psi}{\partial t} = \hat{H}\Psi, \tag{1}$$

with the Hamiltonian

$$\hat{H} = \hat{H}_{\rm atom} + \hat{H}_{\rm int}(t), \tag{2}$$

where $\hat{H}_{\rm atom}$ is the Hamiltonian of the field-free lithium atom. To describe interaction of the atom and the external electromagnetic field we use the length gauge. The EM field is chosen to be linearly polarized along the z-axis. The interacting part of the Hamiltonian can be written therefore as (atomic units are used):

$$\hat{H}_{\rm int}(t) = f(t) z F \cos\omega t \tag{3}$$

Here $f(t)$ is a switching function which is smoothly growing as: $f(t) = \dfrac{3t^2}{T_1^2} - \dfrac{2t^3}{T_1^3}$ for $0 < t < T_1$, and is constant for $t > T_1$. The switching time $T_1 = 5T$ where T is a cycle of the laser field. The whole interval of time on which the time-evolution was computed was 30 cycles of EM field.

To solve the TDSE we follow the strategy similar to that we have applied before for two-electron systems (Ivanov and Kheifets 2006, Ivanov and Kheifets 2007). The solution of the TDSE is sought in the form of an expansion on a square-integrable basis

$$\Psi(\boldsymbol{r}, t) = \sum_j a_j(t) f_j(\boldsymbol{r}). \tag{4}$$

Here the basis states $f_j(\boldsymbol{r})$ are pseudostates obtained by diagonalizing the field-free lithium Hamiltonian in a suitable square integrable (Laguerre in our case) basis (Bray 1994):

$$\langle f^N_{nl}|\hat{H}_{\rm atom}|f^N_{n'l'}\rangle = E_{nl}\delta_{nn'}\delta_{ll'} \ . \tag{5}$$

Here E_{nl} is the energy of a pseudostate and N is the size of the basis.

We shall be interested below in the harmonics of the order not higher than 9. Since we consider modestly strong electric fields of the order of 0.01 a.u. corresponding to 3.5×10^{12} W/cm^2 intensity, we can retain in expansion (4) only the basis states with angular momenta $l = 0 - 9$. The total number of pseudostates for each l was 40.

In order to solve TDSE (1), we rewrite it as a system of differential equations for the coefficients $a_j(t)$ in Equation (4). The pseudostates as defined in Equation (5) are computed within a box (the box size of 200 a.u. was used in this work). Since we are interested in sufficiently long pulses (thirty periods of the EM radiation) we must ensure that no artificial effects due to possible reflections of the wavepackets from the boundaries of the box are present. This was achieved by means of the complex absorbing potential $-iW(\boldsymbol{r})$ which was chosen as a smooth function, zero for $r \leq 120$ a.u. and continuously growing to a constant $-iW_0$ with $W_0 = 40$ a.u. outside this region. Introduction of the absorbing potential distorts, of course, the wave function in the region $r > 120$ a.u. But that is not important if we are interested only in calculation of the HHG process to which electrons reaching distances of 100 a.u. from the nucleus cannot contribute. With the complex absorbing potential thus defined, the TDSE was propagated for the time interval $(0, T_1)$, $T_1 = 30T$. The harmonics spectrum was then calculated as prescribed by Krause *et al* (1992):

$$|d(\omega)|^2 = \left| \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} e^{-i\omega t} d(t)\ dt \right|^2 \ . \tag{6}$$

Here $d(t) = \langle \Psi(t)|z|\Psi(t)\rangle$ is expectation value of the dipole momentum, t_1, t_2 are chosen to be large enough to minimize the transient effects (we used $t_1 = 20T$, $t_2 = 30T$, i.e., last 10 cycles of the pulse duration).

3. Results.

3.1. HHG from excited 2p lithium state.

If Figure 2 we present the harmonics spectrum obtained from the Li atom prepared initially in the $2p$ state. In the same figure, we also show an analogous spectrum of the Li atom prepared initially in the ground state. As is seen from the figure, the excited $2p$ state turns out to be much more efficient generator of high harmonics.

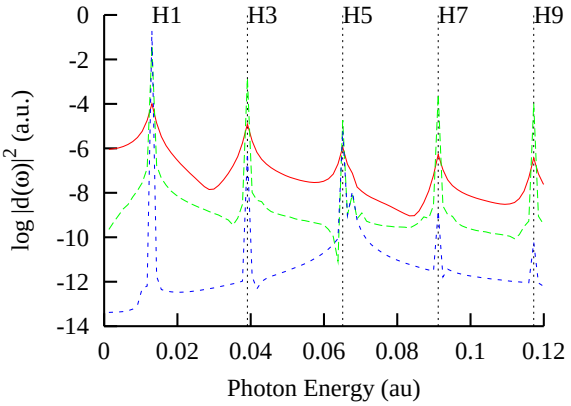

Figure 2. Harmonics spectrum of Li from $2p$ state, peak strength of the EM field $F = 0.005$ a.u. (red) solid line, $F = 0.0025$ a.u. (green) dashed line and from $2s$ state, $F = 0.005$ a.u. (blue) dots.

At the peak strength of EM field $F = 0.005$ a.u., the harmonics H7 and H9 are 3 to four orders of magnitude more intensive if evolution starts from the $2p$ state as compared to the process starting from the ground state. Actually, at this field strength, the $2p$ state depopulates very rapidly and the harmonics generation is saturated. It would be physically more appropriate to compare generation from the ground state at $F = 0.005$ a.u. with generation from the $2p$ state at a weaker field, e.g. $F = 0.0025$ a.u. which will give approximately equal values of the ionization rates and Keldych parameters (1.62 for $2s$ and 2.63 for $2p$) for these states. Figure 2 shows that for this field strength the intensity of harmonics H7 and H9 gains yet two more orders of magnitude, comparing to the generation from the $2p$ state at $F = 0.005$ a.u.

To get a closer insight into the origin of this phenomenon, we performed a wavelet analysis of the function $d(t)$ in Equation (6). The wavelet transform of the square integrable function $d(t)$ is defined according to Tang *et al* (2000) as

$$T_\Psi(\omega, \tau) = \int d(t)\sqrt{\omega}\Psi(\omega t - \omega\tau)\, dt\ . \tag{7}$$

This transform is generated by a function $\Psi(x)$ (the mother wavelet). As a mother wavelet we choose here the Morlet wavelet $\Psi(x) = x_0^{-1}\exp(ix)\exp[-x^2/(2x_0^2)]$ with the parameter $x_0 = 10$. Inverse wavelet transform allows to represent the signal (function $d(t)$ in our case) as a superposition (with weights proportional to $T_\Psi(\omega,\tau)$) of the so-called daughter wavelets $\Psi(\omega t - \omega\tau)$ with various ω, τ, which can be interpreted as a superposition of Gaussian pulses with the carrier frequency ω emitted at $t = \tau$. For the Morlet wavelet, $T_\Psi(\omega,\tau) = |T_\Psi(\omega,\tau)|e^{i\delta}$ is a complex quantity. For a given frequency ω, the values of τ at which maxima of $|T_\Psi(\omega,\tau)|$ occur can be interpreted as moments of a peak emission of pulses constituting the harmonic of this frequency (Chu and Chu 2001). Values of δ corresponding to these times τ provide information about the phases of these pulses.

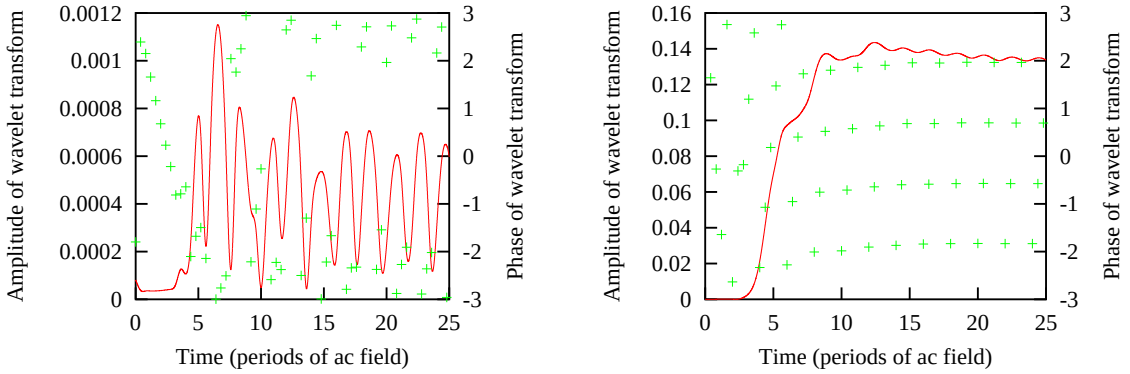

Figure 3. Wavelet transform (amplitudes - solid red curves and phases - ditted green curves) for the frequency corresponding to the 7-th harmonic generated from the $2s$ state (F=0.005 a.u.) (left panel) and $2p$ state (F=0.0025 a.u.) (right panel).

Figure 3 shows amplitudes and phases of the wavelet transform $|T_\Psi(\omega,\tau)|$ for the fixed frequency ω corresponding to the 7-th harmonic. As one can see from the figure, generation of the 7-th harmonic from the ground and excited $2p$ states seem to be completely different processes. Maxima of generation from the ground state occur at a sequence of points. The phases δ at these points assume more or less random values. This generally leads to destructive interference of these pulses and, correspondingly, to a weak output of the 7-th harmonic. On the contrary, generation from the $2p$ excited state seem to be a process occurring more uniformly in time, except for the interval of $(0, 5T)$ corresponding to the switching of of the EM interaction. The phases lie on

smooth curves, which not vary much allowing for the constructive interference of the pulses.

The wavelet analysis does not tell us which mechanism is responsible for the enhancement of the harmonics yield for the case of the generation from the $2p$ state. We can speculate, however, that similarly to the case of Rb studied by Paul *et al* (2005), this enhancement may be due to mutual interaction of several excited states. That this is indeed might be the case for Li can be seen in Figure 4 where we present harmonic spectrum generated by the $2p$ state if in the trancate expansion Equation (4), thereby removing all low-lying excited states of Li from the description of the time-evolution of the system. More exactly, for each angular momentum l the first 5 pseudostates have been removed (except of course, the $2p$ state itself).

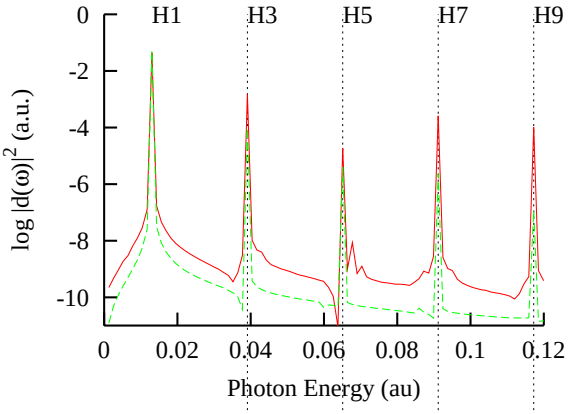

Figure 4. Harmonics spectrum of Li from $2p$ state, (red) solid line, and the spectrum obtained if low lying excited states of Li are excluded from the expansion (4) (green) dashed line. Peak strength of the EM field $F = 0.0025$ a.u.

As one can see from Figure 4 the removal of the manifold of low-lying excited Li states does produce considerable changes in the intensities of higher order harmonics (2 and 3 orders of magnitude for 7-th and 9-th harmonics respectively). Intensities of the lower order harmonics do not change appreciably.

The role played by these states can be further clarified if we consider probabilities to find the Li atom in various states as function of time. Figure 5 presents these probabilities for two cases: evolution starting from the $2s$ state ($F = 0.005$ a.u.) and evolution starting from the $2p$ state ($F = 0.0025$ a.u.).

One can observe that the probability curves on the right panel (especially one for the $3s$ state) are modulated in a rather complicated way. This modulation is also present in the amplitudes in Equation (4), and will translate into modulation of the

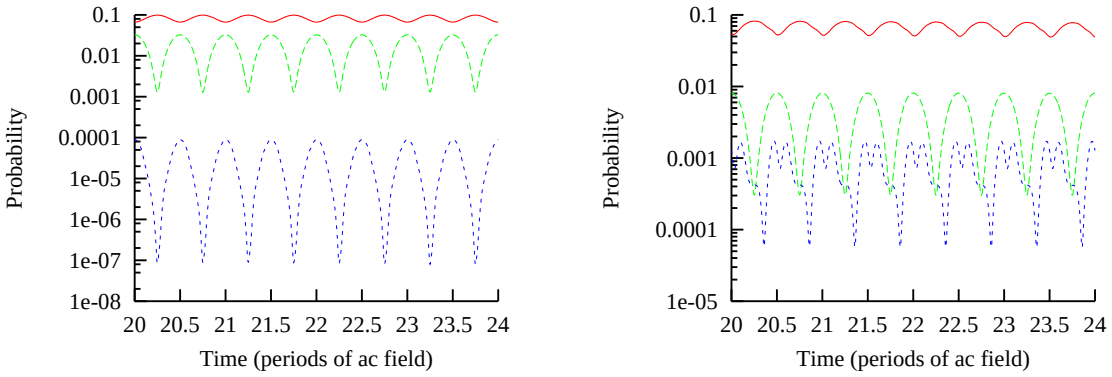

Figure 5. Probabilities to find Li atom in the states: left panel: $P_{2s} - 0.9$ (red) solid line, P_{2p} (green) dashed line, P_{3p} (blue) dots, $F = 0.005$ a.u., $2s$-initial state; right panel: $P_{2p} - 0.9$ (red) solid line, P_{2s} (green) dashed line, P_{3s} (blue) dots, $F = 0.0025$ a.u., $2p$-initial state.

dipole momentum.

This may lead to the considerable modification of the Fourier transform of the function $d(t)$ and hence, the HHG yield. Formula often used for the evaluation of $d(t)$ is (Lewenstein *et al* 1994, Sanpera *et al* 1996):

$$d(t) = -i \int_{t_0}^{t} dt' \int d\boldsymbol{q} D^*(\boldsymbol{q} + \boldsymbol{A}(t)) E(t') D(\boldsymbol{q} + \boldsymbol{A}(t')) e^{iS(\boldsymbol{q};t,t')} + c.c., \quad (8)$$

where $S(\boldsymbol{q};t,t') = \int_{t}^{t'} d\tau \left((\boldsymbol{q} + \boldsymbol{A}(\tau))^2/2 + I_p \right)$ is the action, $D(\boldsymbol{v})$ is the z-component of the dipole transition matrix element between bound and continuum state in which electron has a velocity $\boldsymbol{v} = \boldsymbol{q} + \boldsymbol{A}(t)$, $\boldsymbol{A}(t)$-the vector potential. Product of three terms on the r.h.s. of Equation (8) corresponds to ionization of an electron into a continuum state with the velocity $\boldsymbol{v}$ at the moment of time t' , propagation in the laser field from t' to t with constant canonic momentum $\boldsymbol{q}$ and recombination at the moment of time t (the three-step model). Expression (8) is obtained if, in the spirit of the Keldych theory, propagation of electron in the EM field is described by means of the Volkov states, which neglect any atomic potential. If we wish to include influence of the atomic potential on the electronic motion, it is the description of this propagation which should be modified. Instead of the Volkov propagator $e^{iS(\boldsymbol{q};t,t')}$ we should use the exact propagator $G(\boldsymbol{q},t,;\boldsymbol{q}'t')$, and introduce additional integration over $\boldsymbol{q}'$ (canonic momentum $\boldsymbol{q}$ is no longer conserved quantity if we take into account atomic potential). This makes the problem hardly tractable. One feature of $d(t)$ obtained in this way,

can be however guessed. Exact propagator $G(\boldsymbol{q}, t, ; \boldsymbol{q}'t')$ is to be built form the exact time-dependent state of Li in the laser field. As we saw, such states may vary in time with various, generally incommensurate frequencies. That will lead to presence of such terms in the expression for $d(t)$. If any of those incommesurate frequencies is close to a frequency of a given harmonic, that may lead to the enhancement of the harmonic intensity.

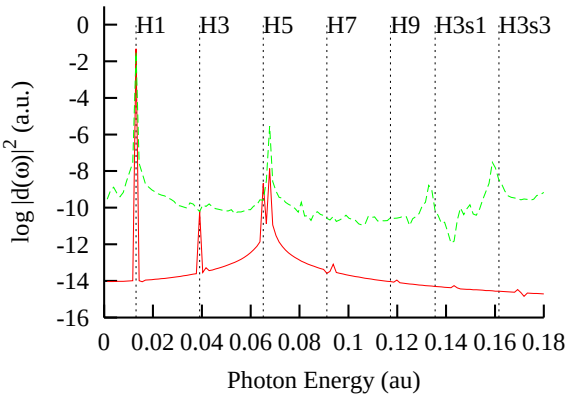

Figure 6. Harmonics spectrum of Li from the $2s$ state ((red) solid line and a superposition ((green) dashed line), $a|2s> +\sqrt{1-a^2}|3s>$ ($a = 0.9$), $F = 0.0025$ a.u.

If, as the starting point of the process we choose a coherent superposition of the ground and an excited state then, due to a much larger ionization rate, presence of the excited state modifies considerably the HHG spectrum. In Figure 6 we show the harmonic spectra obtained for the initial state $2s$, and a superposition $a|2s> +\sqrt{1-a^2}|3s>$ with $a = 0.9$ and the field strength $F = 0.0025$ a.u.

For such a field strength the harmonics yield from the ground state is virtually zero since the ground state hardly ionizes. Therefore, even a relatively small contribution of the $3s$ state completely changes the spectrum. In Figure 6 one can see the harmonics H1, H3, H5 present in the spectrum as well as an additional peak marked H3s . This structure can be explained with the help of a recently developed theory of HHG from coherent superpositions of several states (Milošević 2006). According to this theory, harmonics in this case can be produced in two ways. For the harmonics with frequencies smaller than excitation energy (in our case $E_{3s} - E_{2s} = 0.1225$ a.u.), the conventionally mechanism of the single-state harmonic generation is at work producing odd multiples of laser frequency. For harmonics with frequencies in the vicinity of the excitation energy, the resonance mechanism produces harmonics with the frequencies $E_{3s} - E_{2s} \pm (2k+1)\omega$.

To produce such harmonics, the electron ionizes from the $3s$ state and recombines later into the ground state. Figure 6 shows presence of two harmonics of this kind (marked H3s1 and H3s3 on the figure).

3.2. Harmonic generation from Rydberg states.

In this section we consider briefly the process of harmonics generation from the Rydberg states of lithium. Harmonics generation from such states is of less interest since they are ionized very rapidly and we should either use very small field strengths or larger frequencies of the source of EM radiation. In both cases we leave the domain of the HHG proper. For the typical laser parameters used above ($F = 0.005$ a.u., $\omega = 0.3544$ eV), the HHG cutoff defined as $3.17U_p + I$, where U_p is ponderomotive potential, I- ionization potential, is of the order of 0.2 a.u. The plateu of the harmonics spectrum contains then approximately 20 harmonics, which allows us to speak about HHG. However, for these laser parameters, Rydberg states are ionized extremely fast. To take a look at the harmonics generation from Rydberg states we consider below a laser source with larger frequency, viz. the Ti:sapphire laser with the main frequency of 1.56 eV. If we use EM fields of the order of 0.005 a.u. as above, we cannot talk about HHG of course (the "plateu" in this case contains 3 harmonics only). The harmonics, however, can be produced, and, as we shall see, the Rydberg states produce them more efficiently than the ground state.

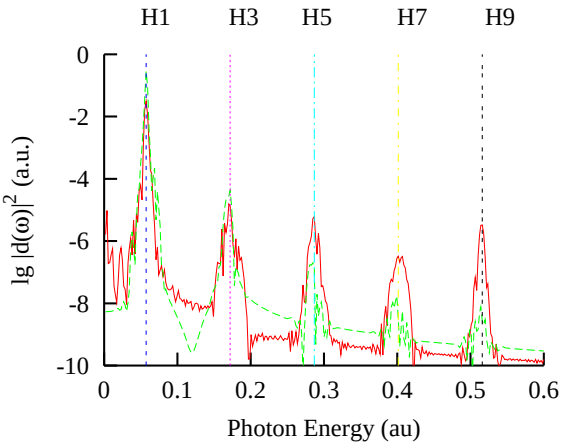

Figure 7. Harmonics spectrum of Li for the peak strength of the EM field $F = 0.005$ a.u., ω=1.56 eV; the $5p$ initial state: (red) solid line, the $2s$ initial state: (green) dashed line.

In Figure 7 we show the harmonics spectrum from the $5p$ initial state at peak strength of the EM field $F = 0.005$ a.u. In the same figure we also show the harmonics

spectrum obtained from the ground $2s$ state. A considerable enhancement of the seventh and ninth harmonics can be observed. The origin of this enhancement is probably due to the presence of the manifold of the closely lying Rydberg states. As was observed by Zeng *et al* (2002) for the process of two-colour harmonics generation of a model 1D atom, the atomic structure can greatly modify the yield of some harmonics. Some indication that the Rydberg levels do indeed interact strongly with each other can be gained from Figure 8. In this figure we blow up the time interval of $(14T, 16T)$ where the EM field attains its peak strength.

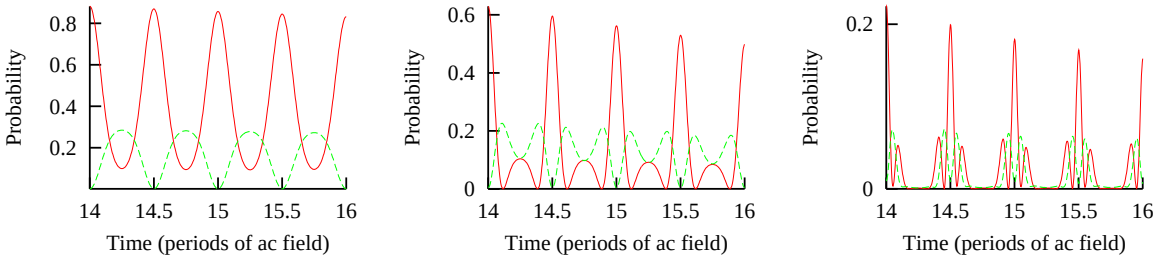

Figure 8. Probability to find the lithium atom in the $5p$: (red) solid line, and $5s$: (green) dashed line, states as a function of time for $t \in (14T, 16T)$ and peak strengths of the EM field 0.0025, 0.005, 0.01 a.u. (from left to right).

4. Conclusion.

We presented a series of calculations of the HHG from the lithium atom initially in an excited state or in a superposition of the ground and excited states. We found that excited states, in particular the $2p$ state is extremely efficient high harmonics generator. Gain of several orders of magnitude in harmonics intensity can be achieved if the Li atom is initially in the $2p$ state. For an atom prepared initially in a coherent superposition of the ground and excited states, we also see a considerable increase of the harmonics yield and appearance of additional harmonics due to the resonant mechanism predicted by Milošević (2006) and studied in that work for a model two-level atom. We demonstrated these effects for a real atomic system.

Wavelet analysis shows, that harmonics from the ground and excited states are produced in a markedly different way. In particular, the distribution of phases of the wavelet transform for the $2p$ state suggests that the 7-th harmonic in this case

is produced by a superposition of pulses belonging to several (four) families. Pulses of the same family have nearly identical phases leading to constructive interference and large harmonic intensity. This is in strike contrast to the results of the wavelet analysis for the ground state. The obvious difference between the ground and excited states is the different extent to which they are influenced by outher states. Dynamic of the system in an excited state is considerably more complex, involving mutual interaction of several states. This is even more so for the Rydberg states.

The consideration of these effects in the paper concerned only the single-atom aspect of the problem. To produce experimentally observable effect of the N^2 enhancement (where N is a number of Li atoms in the target), one needs to be able to prepare a coherent superpositions of Li atoms in a given state. Availbale techniques, such as *pi*-pulse technique or stimulated Raman adiabatic passage (Bergmann *et al* 1998, Vitanov *et al* 2001) allow to solve this problem.

5. Acknowledgements.

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