Interference Stabilization and Possibility of Amplification and Lasing in Plasma Channel Formed in Gas by Intense Femtosecond Laser Field

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Abstract: The effect of interference stabilization of Rydberg atoms in high-intensity laser field is proposed to create the plasma channel with population inversion between set of Rydberg states and the ground state for conversion of the input laser energy into the VUV and XUV frequency band. Furthermore, there is a possibility to create a population inversion between high-lying Rydberg states which can be used for lasing and amplification in the IR, mid-IR and sub-terahertz frequency band.

1 INTRODUCTION

Progress in strong-field physics and laser – matter interaction makes it possible to investigate a number of new physical processes. In particular, generation of high order harmonics of the incident laser pulse is possible as a result of recombination process of the photoelectron with the parent ion, when the electron rescatters on it (Agostini and Di Mauro, 2004; Krausz and Ivanov, 2009). On the other hand, production of quasi-dc electric current in plasma arising due to ionization of gases by a few-cycle or bi-chromatic laser pulses causes the generation of low-frequency terahertz waves (Kreß, et al, 2006; Gildenburg and Vvedenskii, 2007; Wu, et al, 2008; Silaev and Vvedenskii, 2009).

The idea of amplification of radio-frequency (RF) radiation in ionized gas with population inversion in the electronic continuum in gases characterized by a Ramsauer minimum in transport cross section was proposed long ago (Bekefi, et al, 1961; Bunkin et al, 1972).

In (Bogatskaya and Popov, 2013) it was mentioned that such an amplification can be simply realized in heavy rare gases with a Ramsauer minimum in transport cross section if the gas media is ionized by UV laser pulse with the duration less than the characteristic time of relaxation of photoelectron energy spectrum. This idea was analyzed in detail in (Bogatskaya et al, 2013; Bogatskaya et al, 2014).

On the other hand self-focusing and filamentation of high-intensity laser pulse can lead to remote lasing action from a filament (Luo, et al, 2003). In (Wang et al, 2013a; Wang et al, 2013b) lasing action from the Ti-Sa laser femtosecond laser filament in air was demonstrated experimentally. Different theoretical analyses of the filament-initiated nitrogen laser are presented in (Penãno et al, 2012; Kartashov et al, 2015). Recently, population inversion in fluorescent fragments of super-excited molecules inside an air filament and spontaneous emission from these fragments were also detected by (Chin and Hu, 2015).

In this paper we would like to pay attention that interference stabilization (IS) phenomenon predicted and studied in (Fedorov and Movsesian, 1988; Fedorov et al, 1996; Fedorov et al, 2012) or population trapping in high-lying Rydberg states (Talebpour et al, 1996; Azarm et al, 2013) in strong laser fields also results in population inversion between the set of Rydberg states and low-lying excited or ground states. It is shown that such a trapping can be used for lasing and amplification of electromagnetic radiation in a range from visible to VUV or XUV frequency band.

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2 INTERFERENCE STABILIZATION, POPULATION TRAPPING IN RYDBERG STATES AND IDEA OF AMPLIFICATION OF ELECTROMAGNETIC RADIATION

Interference stabilization (IS) phenomenon consists in the coherent repopulation of neighboring Rydberg states via the continuum by Raman Λ - type transitions, which leads to the destructive interference of photoionization transition amplitudes from neighboring Rydberg levels. Theoretical consideration of this phenomenon was first discussed in (Fedorov and Movsesian, 1988) for the case of an atom initially prepared in a Rydberg state. Nevertheless, the phenomenon of IS was also found to take place for ground states of atoms interacting with high-intensity laser field (Dubrovskii et al, 1991). In this case the multiphoton excitation via Freeman resonances (Freeman et al, 1987) appearing with ac Stark-shifted-levels and IS results in population trapping in a number of high-lying (Rydberg) states. Independently, similar idea was suggested by S L Chin to explain the experimental results obtained in his group on the strong-field ionization yields in atoms of rare gases (Talebpour et al, 1996). We believe that the set of recent experimental data (Azarm et al, 2013; Nubbemeyer et al, 2008; Eichmann et al, 2013; Eichmann, et al, 2009) can be interpreted in frames of the discussed model. Recently the effect of population trapping in Rydberg states in argon irradiated by the femtosecond Ti-Sa laser pulse was also observed experimentally by Fechner et al, 2015.

In this paper we draw attention to the fact that IS and population trapping in a gas media pumped by the external source of energy from a physical point of view is very close to the effect of the population inversion of gas atoms. This phenomenon reveals a thought-provoking idea of new methods of amplification and lasing of electromagnetic radiation (Bogatskaya and Popov, 2015).

At fig. 1 we present some results of numerical simulations of ionization and excitation probabilities versus the peak laser intensity for argon obtained for the second harmonic ($\hbar\omega = 3.1$ eV) of the sine-squared linear-polarized Ti-Sa laser pulse of forty cycles duration (≈ 53 fs). These data were obtained by the method of the direct numerical integration of the TDSE for model single electron atoms with

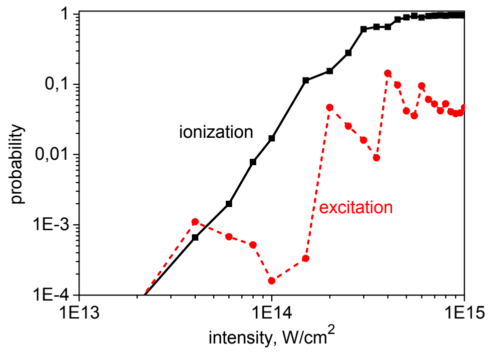

Figure 1: Probabilities of ionization and excitation of argon atoms irradiated by the 2nd harmonic of femtosecond Ti-Sa laser pulse in dependence on laser peak intensity.

energy spectra similar to that of real Ar atoms interacting with external laser field. The detailed information for chosen atomic potential can be found in (Bogatskaya et al, 2013). One can see the stabilization phenomenon leading to the substantial fraction of atoms being trapped in the Rydberg states and channel closing effect similarly to data obtained previously (Volkova et al, 2011; Popov et al, 2010; Fedorov et al, 2012) for the fundamental frequency of the Ti-Sa laser. The dependence of excitation probability on the laser intensity reveals a lot of maxima and minima changing each other. The maxima observed on the excitation curve are seen to be separated by approximately the same value of laser intensity equals to $\Delta I \approx 2\times10^{14}$ W/cm^2, which exactly corresponds to the condition that the ponderomotive shift of the ionization threshold $U_{pond} = \varepsilon_0^2 / 4\omega^2$ reaches the value of the photon energy $\hbar\omega$, $\Delta U_{pond} = \hbar\omega$. Confirmation of the aforesaid can be seen at fig. 2: photoionization peaks gradually are shifted towards the lower energies as the pumping intensity goes up. For example, the 6-photon ionization channel for argon atom is closed near the intensity value 2×10^{14} W/cm^2, next one at 4×10^{14} W/cm^2 and so on (see fig. 2). Furthermore, the indented structure of the photoelectron spectrum is related to the coherent repopulation of Rydberg states via Raman Λ-type transitions. Fig. 3 keeps more extended information about peak positions and channel closing over the intensities less than 10^{15} W/cm^2.

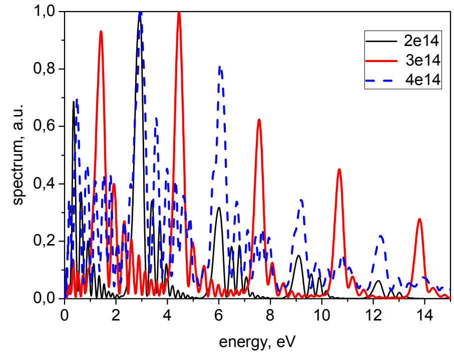

Figure.2: Photoelectron spectra for argon atoms irradiated by the 2nd harmonic of Ti-Sa laser pulse.

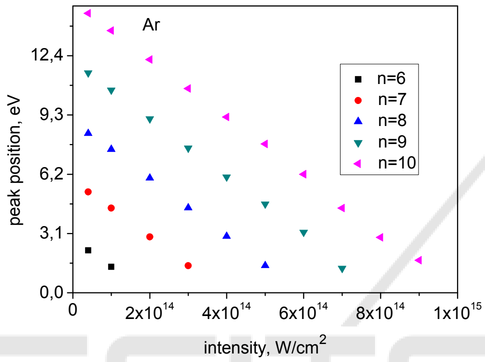

Figure 3: Peak positions in photoelectron spectra for argon in dependence on laser intensity.

More detailed information about the distribution of trapped population in dependence on Rydberg states with different angular momentum and principal quantum number is performed at fig. 4 for different laser peak intensities in the case of argon atoms. One can point out that for the intensity $2 \cdot 10^{14}$ W/cm^2 the states with even angular momentum are dominantly populated (see fig. 4a). This results from the six-photon resonance between the ground state and the set of Rydberg states accompanied by the Λ - type transitions repopulating the Rydberg states with even angular momentum. As laser intensity grows (4×10^{14} W/cm^2), seven-photon ionization channel is found to be closed and hence we observe seven-photon excitation which effectively populates states with odd value of angular momentum (see fig. 4b).

Thus, we have a variety of emission lines which could be obtained after the irradiation of the atom by intense laser pulse. Using transitions between neighboring Rydberg states it is possible to obtain quanta from THz to IR frequency range. Cite data enable us to estimate gain factors for different

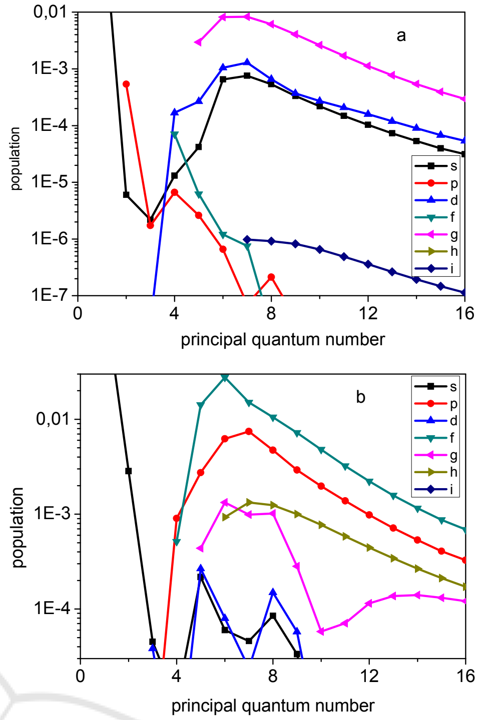

Figure 4: Population distribution of Rydberg states with different angular momentum in dependence on principal quantum number for argon atoms. Laser intensities are 2×10^{14} W/cm^2 (a) and 4×10^{14} W/cm^2 (b).

types of radiation. For example, the transition $n_1, l \to n_2, l \pm 1$ ($n_1 >> 1, \Delta n = n_1 - n_2 = 1$) with the quanta $\Omega \approx 1/n_1^3\big|_{n_1=6-7} \approx 0.1$ eV (see fig. 4) represents the down conversion of the 2nd harmonic of Ti-Sa laser radiation into the far IR frequency band. Similar, for $n_1 = 4-5$, $n_1 \approx 10$ ($\Delta n = 1$) we obtain mid IR and THz frequency band respectively.

On the other hand transitions from populated Rydberg to the low-lying excited states ($n_2 = 2,3$) give rise to the emission of visible light.

There is also a possibility to generate high energy quanta using the transition $np \to 1s$, which is available at higher laser intensities when the ground state is depleted. Data on fig. 5 depict the difference between population of *np* and 1*s* states in argon-like atom depending on principal quantum number. Thus, the set of lines from the Layman series $np \to 1s$ with the quanta $\Omega \approx Ry = 1/2$ may be emitted effectively after the irradiation of atom by the intense laser radiation of intensities above 7×10^{14} W/cm^2.

The general expression for the photoabsorption cross section for transitions $n_1\, l \rightarrow n_2\, l \pm 1$ reads

$$\sigma_{ph} = \frac{\lambda^2}{2\pi} \frac{A_{n_1\, l \rightarrow n_2\, l \pm 1}}{\Delta \nu} \quad (1)$$

where λ is the transition wavelength, $A_{n_1\, l \rightarrow n_2\, l \pm 1}$ is the Einstein coefficient for the spontaneous emission and $\Delta \nu$ is the width of the emitted line

$$A_{n_1, l \rightarrow n_2, l \pm 1} = \frac{4\Omega^3}{3c^3} \left| d_{n_1, l \rightarrow n_2, l \pm 1} \right|^2 \quad (2)$$

with speed of light $c = 137$. The matrix element for the dipole moment in WKB approximation (Delone et al, 1983) can be estimated as $d_{np \rightarrow 1s} \approx 1/n_1^{3/2} n_2^{3/2}$ ($l \sim 1$). Then, for example, for visible radiation one obtains

$$A_{n_1 l \rightarrow n_2 l \pm 1} \approx \frac{\Omega^3}{c^3} \left| d_{n_1 l \rightarrow n_2 l \pm 1} \right|^2_{\Omega = \frac{1}{2n_2^2}} \approx \frac{3 \cdot 10^{-10}}{n_1^3}. \quad (3)$$

The width $\Delta \nu$ of the Rydberg state arising from the collisional broadening and for $N = 10^{18}$ cm^{-3} can be estimated as $\Delta \nu \approx 3 \times 10^{-6}$ (in atomic units). Then the cross section can be estimated as $\sigma_{ph} \approx 10^{-17} \div 10^{-15}$ cm^2. If one assumes, that the population inversion is $\Delta N \approx 10^{15}$ cm^{-3} the gain factor is calculated as $g = \sigma_{ph} \times \Delta N \approx 0.01 - 1$ cm^{-1}. It means that if $g = 0.5$ cm^{-1} for the propagation length of 10 cm the intensity of the visible electromagnetic radiation will be increased 100 times. For longer wavelengths corresponding to IR, mid IR, THz radiation the gain factor is supposed be smaller due to lower value of the Einstein coefficient of the spontaneous emission. Furthermore, for high values of Rydberg levels it is necessary to consider that the size of atoms increases as n^2: for example, for $n = 10$ we have gas atoms 100 times larger. Thus, one should diminish concentration meaning that the gain factor will decrease as well.

In view of the above argumentations one can conclude that processes of amplification and lasing in visible and VUV frequency bands seems to be more beneficial therefore we are going to dwell upon them in the next section.

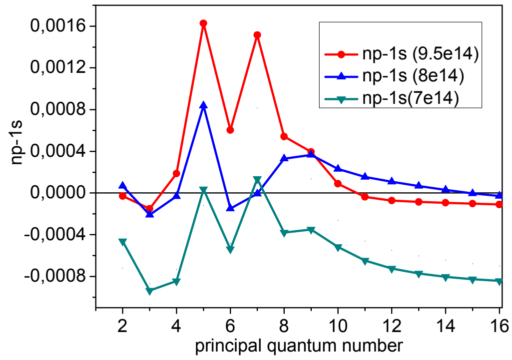

Figure 5: The difference between population of np and 1s states in argon-like atom in dependence on principal quantum number for different laser peak intensities.

3 RATE EQUATIONS AND PROCESSES OF GENERATION AND AMPLIFICATION OF VISIBLE AND VUV PULSES IN PLASMA CHANNEL

To study the phenomena of generation and amplification of electromagnetic pulses in a plasma channel with population inversion the approach of rate equations was applied (Kartashov et al, 2015). Based on this approach the following system for the forward generation (amplification) can be written:

$$\frac{\partial I(z,\tau)}{\partial z} = g(\tau) I(z,\tau) + \frac{\hbar\omega}{2\tau_s} \left(\frac{d}{2l} \right)^2 \Delta N, \quad (4)$$

$$\frac{d\Delta N}{d\tau} = -\sigma_{ph} \frac{I(z,\tau)}{\hbar\omega} \Delta N - \frac{\Delta N}{\tau_{ion}}. \quad (5)$$

Here $g(\tau)$ is the gain factor, $\tau = t - z/c$ is the retarded time, τ_s and τ_{ion} is the time of spontaneous decay and of ionization of Rydberg levels by electron impact respectively, ΔN is population inversion for the certain transition. The factor $d/2l$ (d is the plasma channel diameter, and l is the plasma channel length) means the part of spontaneous emission that lies in the direction of ionizing pulse propagation and can be used for amplification.

It is possible to provide estimation for the intensities of generated (amplified) pulse when we are able to neglect its impact on the population inversion decay:

$$I << \frac{\hbar\omega}{\tau_{ion}\sigma_{ph}} \sim 10^{10}\ \mathrm{W/cm^2}. \qquad (6)$$

This estimation was performed in the assumption that the electronic density is ~10^{17} cm^{-3}, and that the electron energy in continuum is significantly larger than the binding energy of Rydberg atoms. Provided the condition (6) is accomplished at propagation distance L one obtains the following expression for the population inversion decay:

$$\Delta N = \Delta N_0 \exp(-\tau/\tau_{ion}). \qquad (7)$$

Formulas (4) and (7) give us the following dependence of the generated signal intensity:

$$I(z,\tau) = \frac{\hbar\omega}{2\tau_s\sigma_{ph}}\left(\frac{d}{2l}\right)^2 \times \left[\exp\left(-\sigma_{ph}\Delta N_0 \exp(-\tau/\tau_{ion})z\right)-1\right] \qquad (8)$$

For the Rydberg states with principal quantum number $n = 6-10$ τ_{ion} can be estimated as 100 fs.

Fig. 6 and 7 show the spatio-temporal development of generation of the VUV and visible radiation respectively in a plasma channel with population inversion. The data obtained for the visible radiation correspond to the laser intensity 4×10^{14} W/cm^2 while for VUV radiation generation the laser intensity was chosen significantly higher (9.5×10^{14} W/cm^2) in order to reach the regime of the depletion of the ground state and higher values of population inversion. The quantity of population inversion for transition $7p \to 1s$ (VUV radiation) was taken from fig. 5 and from fig. 4 for transitions $np \to 2s$ (visible light). For VUV radiation we observe significant lasing process based on the spontaneous decay of the Rydberg state, in particular generated intensity could reach 10^5 W/cm^2 at a distance of 5 cm. However in the case of visible radiation mainly due to the less value of generated frequency it is possible to attain generation much less than for the VUV (see fig. 7).

The last question we would like to discuss in this section is the process of amplification of a seed pulse in such a plasma channel. We assume the input pulse ($z = 0$) to be characterized by the sine-squared envelope

$$I(z=0,\tau) = \sin^2(\pi t/2\tau_p) \qquad (9)$$

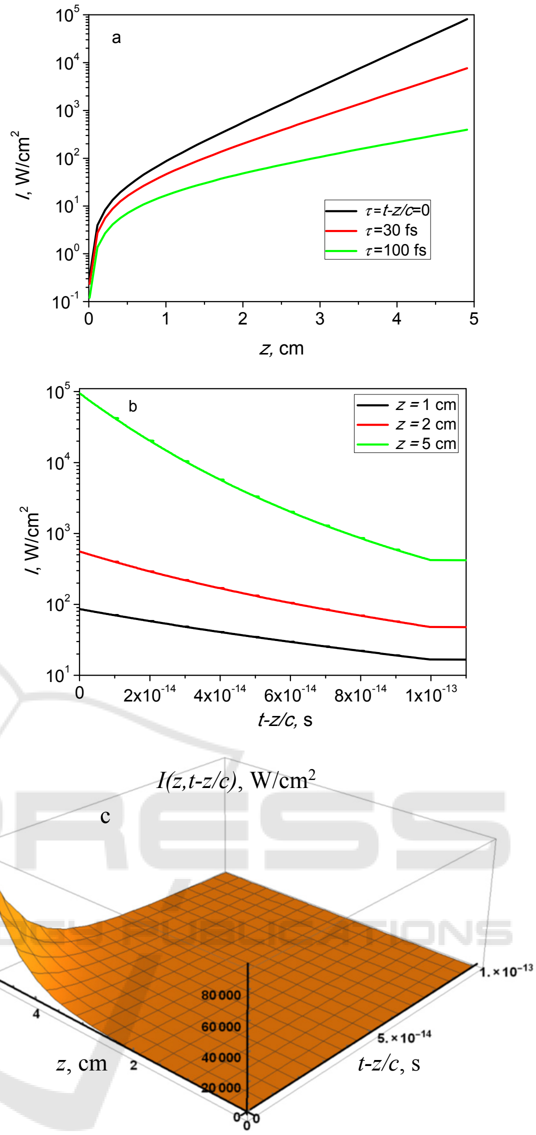

Figure 6: Spatial dependence of generated VUV pulse at different instants of retarded time (a), temporal dependence of generated VUV pulse at different propagation distances (b) and two-dimensional distribution of propagated pulse (c).

with $\tau_p = 20$ fs. Then general solution of (4) can be written in a form

$$I(z,\tau) = \sin^2(\pi\tau/2\tau_p)\times \exp\left[\sigma_{ph}\Delta N_0 \exp(-\tau/\tau_{ion})z\right]. \qquad (10)$$

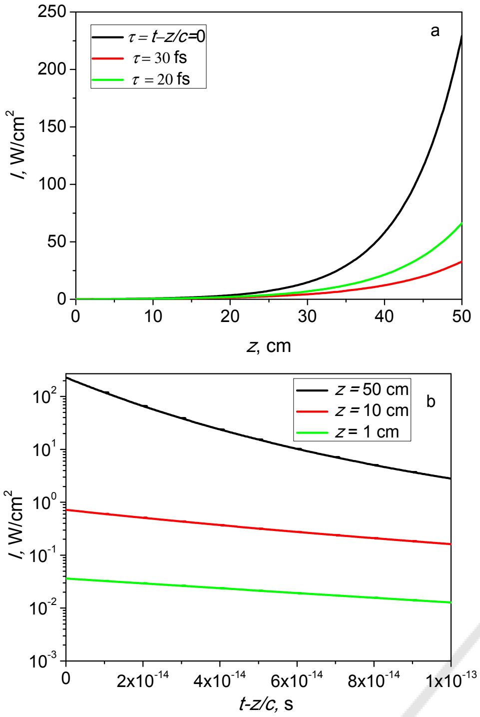

Figure 7: Spatial dependence of generated visible radiation at different instants of retarded time (a), temporal dependence of generated visible radiation at different propagation distances (b).

Typical dependences obtained from (10) for propagation length of $L = 25\,\mathrm{cm}$ for a number of transitions $np \rightarrow 2s$ corresponding to visible radiation are presented at fig.8. One can see considerable enhancement of a seed pulse up to 25 times at a given distance.

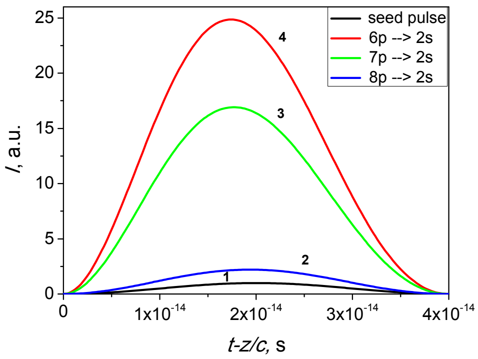

Figure 8: Amplification of visible radiation in a plasma channel. Curve (1) is the input pulse. Curves 2-4 correspond to the transitions $np \rightarrow 2s$, n=8 (2), 7 (3), 6 (4). Propagation length L=25 cm.

The obtained data demonstrate that the amplification and generation of rather short pulses with duration less than the ionization time of Rydberg states looks fairly effective. Such a method can compete with the conventional method of HHG having been widely explored since pioneer papers (L'Huillier et al, 1993; Lewenstein et al, 1994; Becker et al, 1994).

4 LASING AND AMPLIFICATION OF ELECTROMAGNETIC RADIATION DURING THE LASER PULSE

In this section we would like to draw attention to another way of obtaining lasing from population trapped in Rydberg states with higher frequency that differs dramatically from the above described one. It is known that during the interaction of high-intensity Ti-Sa laser pulse with trapped atoms their Rydberg levels are shifted significantly that results in channel closing and essential increment of the ionization potential (see fig.9). In this case the position of Rydberg levels can be estimated as

$$E_n \rightarrow E_n + \varepsilon_0^2 / 4\omega^2 \qquad (11)$$

For example, if four channels of ionization are closed (in argon such situation appears to exist for the intensity $\sim 8\times10^{14}$ W/cm^2 – see fig.3), the energy of the quanta $\hbar\Omega$ for the transition $np \rightarrow 1s$ will be of order of 30 eV. It means that lasing in XUV frequency band is also possible. The width of the stabilized ac-Stark-shifted state np is determined by the ionization process and in the intensity range near the threshold of the IS can be written as

$$\Delta\nu = \Gamma_{IS} = \frac{(E_n - E_{n-1})^2}{2\Gamma}. \qquad (12)$$

As the threshold for the IS stabilization for the 2nd harmonic of Ti-Sa laser radiation is close to 10^{14} W/cm^2, for the intensity 8×10^{14} W/cm^2 the cross section σ_{ph} for $n \sim 6-8$ can be estimated as $\sigma_{ph} \approx 10^{-17}$ cm^2. If one assumes that the inverse population is $\Delta N \approx 10^{16}$ cm^{-3} the gain factor is estimated to be 0.1 cm^{-1} that is very large value for XUV radiation. In comparison with the emission in the after-pulse regime pulses of XUV radiation can be emitted only during rather short time interval near the peak intensity of the laser pumping. Probably, it is one more way to generate the pulses of subfemtosecond duration. As the Rydberg wave packet is continually located in the vicinity of the atomic nucleus and doesn't spread in time, the proposed effect of the up conversion should be much more efficient than the emission of the XUV radiation during the re-scattering of an electron on the parent ion (Corkum, 1993).

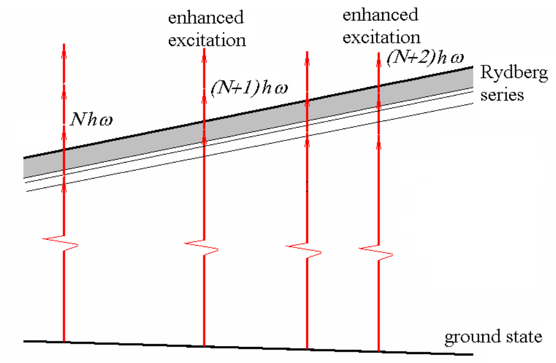

Figure 9: Schematic illustration of the ponderomotive shift of Rydberg states and the continuum boundary.

5 CONCLUSIONS

In this paper we propose to use the population inversion between set of Rydberg states and low-lying excited or even ground states appearing as a result of the interference stabilization phenomenon in strong laser fields for the amplification and generation of visible, VUV and XUV frequency pulses. Estimations for the possible gain factor are performed at it is demonstrated that for typical conditions of IS gain factor for the visible and VUV radiation can reach ~ 0.1 - 1 cm^{-1} which is very large value implementing the possibility to obtain effective emission in plasma channels produced by high intensity laser pulse. We explore the process of generation and amplification of different pulses using the approach of rate equations. The problem of generation of XUV pulses during the interaction with high intensity pumping laser pulses is also observed. We would also like to note that a given lasing effect based on the IS stabilization phenomenon can probably be observed in filaments at rather far away distances from the source of pumping laser radiation.

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