Extremely Short Pulses via Stark Modulation of the Atomic Transition Frequencies

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We propose a universal method to produce extremely short pulses of electromagnetic radiation in various spectral ranges. The essence of the method is a resonant interaction of radiation with atoms under the conditions of adiabatic periodic modulation of atomic transition frequencies by a far-off-resonant control laser field via dynamic Stark shift of the atomic levels and proper adjustment of the control field intensity and frequency, as well as the optical depth of the medium. The potential of the method is illustrated by an example in a hydrogenlike atomic system.

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Investigations of extremely short pulses of electromagnetic radiation in various spectral ranges from terahertz up to x rays have been drawing great attention ([1–4] and recent review [5]). The major goals in this field are shortening of pulses through the attosecond to the zeptosecond and yoctosecond scales [2,3], enhancement of the yield [5], and formation of subcycle pulses [4,5]. The current methods to generate extremely short pulses are specific for each narrow spectral range. At present, attosecond pulses are produced only by high-harmonic generation (HHG). They have quite a small energy ($\sim$ nanojoule per pulse) and low conversion efficiency ($\sim 10^{-4}$–10^{-6}) [5,6]. The recently proposed method of harmonic generation via relativistically oscillating plasma surfaces [2] requires extremely high intensities of the laser pulse. The generation of subcycle optical pulses is still an open problem.

We propose a universal method for production of extremely short pulses up to subcycle duration which can be used in various spectral ranges from far-infrared up to the x-ray domain. It may have high conversion efficiency and requires laser field intensities, much below the atomic ionization threshold. The method is based on a combination of strong-field physics with the physics of resonant processes. It utilizes drastic spectral transformation of a high-frequency (HF) near-resonant field in a medium, where atomic frequency is adiabatically modulated by a strong low-frequency (LF) far-off-resonant control field via dynamic Stark or Zeeman shift of the energy levels. Modulation of the atomic frequency results in a multifrequency atomic response even in the case of the monochromatic incident HF field. This leads to multiple parametric scattering and generation of a series of sidebands during propagation of the HF field through a medium [7]. The proper adjustment of intensities and carrier frequencies of the HF and LF fields as well as the optical depth of a medium produces a broad phase-matched output spectrum, which corresponds to a train of extremely short pulses with pulse duration and repetition rate determined by intensity and frequency of the LF field, respectively.

To illustrate the potential of the method let us consider a hydrogenlike atomic system irradiated with a HF monochromatic field,

$$\vec{E}_{\text{in}} = \vec{x}_0 E_0 \cos(\omega t - kz), \tag{1}$$

and LF monochromatic control field

$$\vec{E}_C = \vec{x}_0 E_M \sin(\Omega t - Kz), \tag{2}$$

where $E_0 \ll E_M$. The HF field is near resonant to the transition $|n = 1\rangle \leftrightarrow |n = 2\rangle$ (where n is the principal quantum number) with frequency ω_{21}^0, $|\omega - \omega_{21}^0| \ll \omega_{21}^0$. The LF control field is very far off resonance, $\Omega \ll \omega_{21}^0$ such that the perturbation of atoms by the LF field is quasistatic. We consider the case where the LF control field is not sufficiently strong to induce the multiphoton transitions to the excited atomic states and direct ionization from the ground state to the continuum (intensity of the LF control field is much below the atomic ionization threshold). This means that the LF control field alone does not interact at all with atoms in the ground state. The LF control field can interact with atoms exclusively due to the presence of the HF field, which selects the proper near-resonant energy level, $|n = 2\rangle$, and excites atoms into the corresponding states with $n = 2$. At the same time the LF control field does interact with atoms in the exited energy states. It aligns the dipole moments and produces spatiotemporal-dependent splitting of the excited energy levels via the linear Stark effect. Because of the selection rules two sublevels of the split fourfold degenerate level $|n = 2\rangle$, namely $|2\rangle = (|200\rangle + |210\rangle)/\sqrt{2}$ and $|3\rangle = (|200\rangle - |210\rangle)/\sqrt{2}$ (where numerals $|nlm\rangle$ label principal, orbit, and magnetic quantum numbers respectively [8]), are coupled with the HF field. As a result, only three bound states including these states and the ground state, $|1\rangle = |100\rangle$, can be considered instead of the full wave function of the electron (Fig. 1). The corresponding three-level system is described by the following density matrix equations:

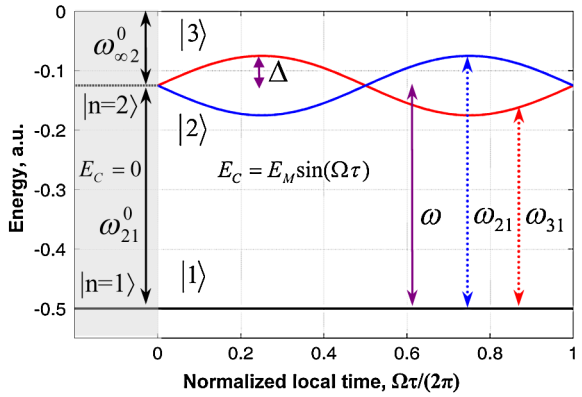

FIG. 1 (color online). Generic scheme of interaction of the HF and LF fields with hydrogenlike system, $\tau = t - z/C$. The LF field produces dynamic Stark splitting of the first excited energy level. The HF field induces quantum transitions from the ground state to the Stark levels.

$$\frac{d\rho_{21}}{dt} + i\left(\omega_{21}^0 - \frac{\vec{d}_{22}\vec{E}_C}{\hbar}\right)\rho_{21} - i\frac{\vec{d}_{21}\vec{E}}{\hbar}(\rho_{11} - \rho_{22}) + i\frac{\vec{d}_{31}\vec{E}}{\hbar}\rho_{23} = -\gamma_{21}\rho_{21},$$
$$\frac{d\rho_{31}}{dt} + i\left(\omega_{21}^0 - \frac{\vec{d}_{33}\vec{E}_C}{\hbar}\right)\rho_{31} - i\frac{\vec{d}_{31}\vec{E}}{\hbar}(\rho_{11} - \rho_{33}) + i\frac{\vec{d}_{21}\vec{E}}{\hbar}\rho_{32} = -\gamma_{31}\rho_{31}, \quad (3)$$

where $\rho_{mn} \equiv \langle m|\rho|n\rangle$ and $\vec{d}_{mn} \equiv \langle m|\vec{d}|n\rangle$ are the matrix elements of the density operator and the dipole moment, respectively, $E = E(z,t)$ is the HF field for $z > 0$, ($E(z \le 0) = E_{\text{in}}$). Decoherence rates γ_{31} and γ_{21} ($\gamma_{31} = \gamma_{32} = \gamma$) are determined by the collisional broadening, γ_c, and the population decay rate, W, of the upper levels, $\gamma = \gamma_c + W/2$. The latter takes into account ionization via tunneling from the upper levels under the action of the control field, $W = A + w$, where A and $w = w(E_C)$ are the radiative decay rate and the field-dependent ionization rate from the upper levels (ionization from the ground state is negligible). The maximum value of the ionization rate is supposed to be much smaller then the frequency of the control field. In this case the ionization rate averaged over the cycle of the control field $\bar{w} = \bar{w}(E_M)$ (calculated according to [9]) can be used such that $\gamma = \gamma_c + (A + \bar{w})/2$. The HF field is chosen to meet the condition $|\vec{d}_{mn}\vec{E}_0|/\hbar \ll \sqrt{\gamma W}$. It allows considering the level populations as unperturbed, $\rho_{11} = 1$, $\rho_{22} = \rho_{33} = 0$, and neglecting the coherence between the Stark levels, ρ_{32}. Under these conditions the set of Eqs. (3) after substitution of $\vec{E}_C$ from (2) takes the form

$$\frac{d\rho_{21}}{dt} + \{\gamma + i[\omega_{21}^0 + \Delta\sin(\Omega(t - z/C))]\}\rho_{21} = i\frac{d_0 E}{\hbar},$$
$$\frac{d\rho_{31}}{dt} + \{\gamma + i[\omega_{21}^0 - \Delta\sin(\Omega(t - z/C))]\}\rho_{31} = -i\frac{d_0 E}{\hbar}, \quad (4)$$

where $\Delta = 3ea_0E_M/(Z\hbar) \ll \omega_{21}^0$ is the modulation depth of the Stark levels (e is the electron charge, a_0 is the Bohr radius, and Z is the atomic number), $d_0 = -2^7 ea_0/(3^5 Z)$, $C = c/\sqrt{\varepsilon(\Omega)}$ is the phase velocity of the control field in a medium, c and $\varepsilon(\Omega)$ are the light velocity in free space and the background dielectric permittivity at the frequency of the control field, respectively. It is worth noting that the modulation depth, Δ, may essentially exceed the frequency of the control field, Ω, and is fundamentally limited by the ionization frequency, $\omega_{\infty 2}^0$, from the excited state, $\Delta < \omega_{\infty 2}^0$ (Fig. 1).

The wave equation for propagation of the HF field $\vec{E}$ through a medium is

$$\frac{\partial^2\vec{E}}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2\vec{D}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\vec{P}}{\partial t^2}, \quad (5)$$

where D takes into account all effects of nonresonant field-matter interaction and $\vec{P} = \vec{x}_0 N d_0(\rho_{21} - \rho_{31} + \text{c.c.})$, N is the atomic density. The boundary condition at the input edge of a medium is $\vec{E}|_{z=0} = \vec{E}_{\text{in}}|_{z=0}$.

We are looking for a solution of the set of Eqs. (4) and (5) for $z > 0$ in the form of a superposition of spectral components including the non-RWA terms,

$$\rho_{21} = \sum_{m=-\infty}^{\infty}\beta_{(1)m}^{(-)}(z)e^{-i(\omega_m t - k_m z)} + \sum_{m=-\infty}^{\infty}\beta_{(1)m}^{(+)}(z)e^{i(\omega_m t - k_m z)},$$
$$\rho_{31} = \sum_{m=-\infty}^{\infty}\beta_{(2)m}^{(-)}(z)e^{-i(\omega_m t - k_m z)} + \sum_{m=-\infty}^{\infty}\beta_{(2)m}^{(+)}(z)e^{i(\omega_m t - k_m z)}, \quad (6)$$

where

$$\vec{E} = \frac{1}{2}\vec{x}_0\sum_{m=-\infty}^{\infty}e_m(z)e^{-i(\omega_m t - k_m z)} + \text{c.c.}, \quad (7)$$

$$\vec{D} = \frac{1}{2}\vec{x}_0\sum_{m=-\infty}^{\infty}\varepsilon_m e_m(z)e^{-i(\omega_m t - k_m z)} + \text{c.c.}, \quad (8)$$

where $\varepsilon_m \equiv \varepsilon(\omega_m)$, $\omega_m \equiv \omega + m\Omega$, $m = 0, \pm 1, \pm 2, \ldots$ Substitution of (6)–(8) into (4) and (5) leads to an infinite set of linear equations for complex amplitudes of the spectral components of the HF field, e_m, and coherences, $\beta_{(1)m}^{(\pm)}$, $\beta_{(2)m}^{(\pm)}$, which can be solved by the regular method. The solution is searched for in the form $e_m(z) \sim e^{-gz}$ (where $g = g' + ig''$) resulting in an infinite sum of multifrequency eigenmodes. Each qth eigenmode consists of even spectral components only, $e_{2m}^{(q)}$, due to mutual annihilation of the odd spectral components resulting from antiphase modulation of the frequencies ω_{21} and ω_{31} (Fig. 1).

The propagation of the qth eigenmode through the medium is characterized by the resonant absorption with the rate g'_q ($g'_q > 0$) and the resonant phase incursion with the rate g''_q controlled by the transition linewidth γ as well as by intensity and frequency of the LF field and frequency of the HF field. The phase incursion $g''_q h$ and amplitude reduction $e^{-g'_q h}$ of the qth eigenmode can be adjusted via variation of the thickness, h, of the medium.

The output HF field, $\vec{E}_{\text{out}}$, for $z > h$ ($\vec{E}_{\text{out}}|_{z=h} = \vec{E}|_{z=h}$) can be represented as a comb of the spectral components, which are the result of collecting the terms oscillating at the same frequencies from different eigenmodes,

$$\vec{E}_{\text{out}} = -\frac{1}{2}\vec{x}_0 E_0 \sum_{m=-\infty}^{\infty} |E_{2m}| e^{-i(\omega_{2m}t - k_{2m}z + \phi_{2m})} + \text{c.c.}, \quad (9)$$

where $n = 2m$, $|E_n| e^{-i\phi_n} = \sum_{q=-\infty}^{\infty} u_q e_n^{(q)} e^{-g'_q h} e^{-ig''_q h}$ and coefficients u_q are determined from the boundary conditions. The output field, $\vec{E}_{\text{out}}$ can have a form of a sequence of short pulses with pulse duration of the order of the inverse spectral width of the comb and pulse repetition rate equaled to the double frequency of the LF field if the total phases ϕ_n and amplitudes $|E_n|$ are properly adjusted via optimization of h, $e_n^{(q)}$, u_q, g'_q, and g''_q. Such optimization was implemented numerically for the case of phase matching between the adjacent HF spectral components and the LF field, $|(k_m - k_{m-1}) - \Omega/C|h \ll 1$ (where $k_m = \frac{\omega_m}{c}\sqrt{\varepsilon_m}$). Several optimal combinations of the parameters were found. The choice can be based on the available experimental facilities.

First of all, let us consider the conditions that could be realized with commercially available facilities for Li^{2+} ions at the transition 13.51 nm and for atomic hydrogen at the transition 121.6 nm. In the former case, emission from the Li^{2+} source normally used for EUV lithography [10,11] could serve as the incident HF field. In the latter case, radiation of the Ar_2^* laser at 126.1 nm or H_2-laser at 121.6 nm is suitable. Emission from the Nd:YAG-laser at 1.064 μm and the CO_2 laser at 10.65 μm serves as the LF control field for the case of Li^{2+} and hydrogen, respectively. Obviously, pulsed lasers with radiation spectral widths much smaller than Ω and γ are appropriate.

The parameters of the output field are illustrated in Figs. 2 and 3 for the case of Li^{2+} ions with the density $N \sim 2 \times 10^{18}$ cm^{-3} [11] in a cell of the length $h = 100$ μm irradiated by the Nd:YAG-laser with intensity 1.2×10^{14} W/cm^2 (beam diameter $\sim 30\lambda$ in focal waist, pulse duration $\sim$0.1 ns, and pulse energy $\sim$0.1 J [12]). Radiation from a Li^{2+} source is converted into $\sim$9 spectral components separated by 2×10^4 cm^{-1} and extending within spectral domain $\sim 3 \times 10^{16}$ s^{-1}. The central output component is two times externally attenuated which can be done using a Mo:Si mirror [13]. The resulting spectrum shown in Fig. 2 corresponds to a few-cycle pulses in Fig. 3 with duration $\sim$300 as, repetition period $\sim$1.8 fs, and peak

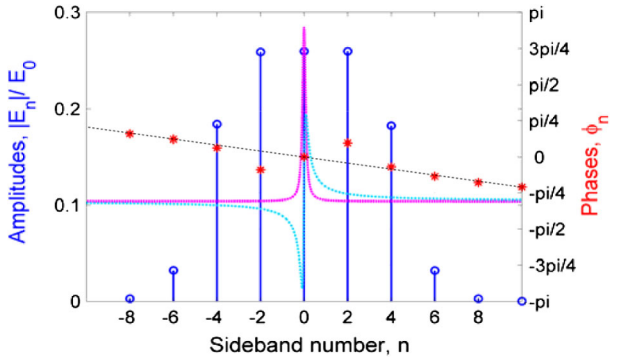

FIG. 2 (color online). Amplitudes (marked by circles) and phases (marked by stars) of the HF-output-field spectral components $\omega_n = \omega + n\Omega$ in the case of Li^{2+}. The dashed contours correspond to spectral location and average width of atomic resonance broadened by ionization.

intensity 1.4 of the incident HF radiation intensity. The parameter values are within validity of the model. Namely, $\Omega \approx 10^{-2}\omega_{21}^0$ and the Rabi frequency of the LF field is $|\vec{d}_0\vec{E}_M|/\hbar \approx 9 \times 10^{-3}\omega_{21}^0$ (that means all multiphoton transitions from the ground state due to LF field are negligible), $\Delta \approx 7 \times 10^{15}$ s^{-1} $\approx 5 \times 10^{-2}\omega_{21}^0$, the quadratic correction to the amplitude of the Stark shift is $\approx 0.16\Delta$, and $\bar{w} \approx 2.6 \times 10^{14}$ s^{-1}. (It is worth noting that $\bar{w} \gg A, \gamma_c, \delta_D$ (where δ_D determines the Doppler broadening) since $A \approx 4.9 \times 10^{10}$ s^{-1}, $\gamma_c \approx 2.2 \times 10^7$ s^{-1}, and $\delta_D \approx 1.4 \times 10^{13}$ s^{-1} [11,14].) The ionization rate from the ground state is negligible indeed ($\sim 10^{-24}$ s^{-1}) [9]. The HF incident field of intensity 1.8×10^{12} W/cm^2 corresponds to the relation $|\vec{d}_{mn}\bar{E}_0|/\hbar = 0.1\sqrt{\gamma W}$. The efficiency of conversion introduced as a ratio of the output peak pulse intensity to the intensity of the total (HF plus LF) input fields is then $\sim$2%, the ratio of the average HF output intensity to the total input intensity is $\sim$0.4%. It is worth noting that the spot size of the incident HF field can be chosen of the order of the beam diameter of the LF field (~ 30 μm). In this case the beam can be focused up to 10^3

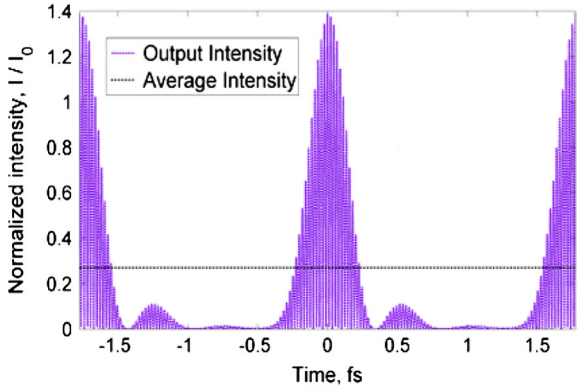

FIG. 3 (color online). HF-output-field intensity in the case of Li^{2+}.

times behind the cell that implies up to 10^6 increase of both the average output and peak pulse intensities.

Similar pulses (with slightly different optimal parameter values) can be obtained in Li^{2+} using CO_2 or Ti:sapphire-laser radiation as the control LF field as well as x-ray Ni-like Ag-laser (13.9 nm) or Ni-like Cd-laser (13.2 nm) radiation [15] as the near-resonant HF field. Using the CO_2 laser with intensity 8.2×10^{13} W/cm^2 along with an external adjustment of phases of the output, HF spectral components allows producing 520 as pulses with repetition period 17.8 fs, and peak pulse intensity 11.6 of the incident one; utilization of the Ti:sapphire laser with intensity 1.4×10^{14} W/cm^2 allows obtaining 290 as pulses with repetition period 1.34 fs, and peak pulse intensity 0.9 of the incident HF radiation intensity.

The output field qualitatively similar to that in Figs. 2 and 3, but with pulse duration $Z^2 = 9$ times longer than in Li^{2+}, could be obtained in the case of hydrogen with the density $N \sim 5 \times 10^{16}$ cm^{-3} in a cell of the length $h \sim 1$ mm irradiated by the CO_2 laser with intensity 1.1×10^{11} W/cm^2 (beam diameter $\sim 10\lambda$ in focal waist, pulse duration $\sim$10 ns, and pulse energy $\sim$130 mJ [16]). Radiation from an Ar_2^* or H_2 laser of intensity up to 10^9 W/cm^2 can be converted into the pulses with duration $\sim$2.5 fs, repetition period $\sim$17.8 fs, and peak intensity up to 2.7 of the incident intensity. The scaling parameter Z^2 determines the difference of the spectroscopic characteristics of the hydrogenlike atoms.

In conclusion, we have proposed a universal method to create extremely short pulses of radiation, which is based on adiabatic periodic Stark modulation of atomic transition frequencies and utilizes advantages of resonant field-matter interaction. The possibility to produce a few-cycle pulses with duration $\sim$300 as in Li^{2+} and $\sim$2.5 fs in atomic hydrogen and with the peak intensity exceeding an incident intensity is shown within the simplest model of the linear Stark effect and average field-dependent ionization rate in a hydrogenlike system. We emphasize the high efficiency of this method (as compared to other techniques such as HHG or stimulated Raman scattering [5]), which is due to the resonant character of interaction. Pulse duration, τ, is determined by the modulation depth, Δ, which may be larger than considered above and is fundamentally limited by ionization frequency $\omega^0_{\infty 2}$. In the case of a hydrogenlike system $\tau \sim 1/(2\Delta) > 1/(2\omega^0_{\infty 2}) \approx 10^{-16}/Z^2 s$. This means that more intense LF control field could produce several times shorter pulses. The analysis of such a regime (which is beyond validity of the model and will be presented elsewhere [17]) requires taking into account nonlinear spatiotemporal dependence of both the Stark shift and ionization rate of the excited states on instantaneous strength of the control field. On the other hand, using heavier ions (for instance, Ne^{9+}) could allow for production of much shorter few-cycle pulses (< 10 as) in the x-ray domain due to possibility of larger modulation depth scaled as Z^2. A deep modulation, $\Delta > \omega^0_{21}/4$, may result in formation of pulses of subcycle duration that however is limited by the ionization frequency, $\Delta < \omega^0_{\infty 2}$. This restriction is appreciably softer in the case of atomic transitions between higher lying states making this mechanism promising for generation of the subcycle pulses in the infrared range in hydrogen and in optical range in heavier ions. This method could be promising to create extremely short pulses in other types of the resonant media such as molecules, liquids, and solids.

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