Research Article | Vol. 29, No. 7 / 29 March 2021 / *Optics Express* 10134

Optics EXPRESS

Single-cycle pulse compression in dense resonant media

Rostislav Arkhipov,[1,2,*] **Mikhail Arkhipov,**[1] **Ayhan Demircan,**[3,4] **Uwe Morgner,**[3,4] **Ihar Babushkin,**[3,4] **and Nikolay Rosanov**[2]

[1] *St. Petersburg State University, Universitetskaya nab. 7/9, St. Petersburg 199034, Russia*
[2] *Ioffe Institute, Politekhnicheskaya str. 26, St. Petersburg 194021, Russia*
[3] *Institute of Quantum Optics, Leibniz University Hannover, Welfengarten 1, 30167 Hannover, Germany*
[4] *Cluster of Excellence PhoenixD (Photonics, Optics, and Engineering—Innovation across Disciplines), 30167 Hannover, Germany*
[*] *arkhipovrostisav@gmail.com*

Abstract: We propose here a new approach for compression and frequency up-conversion of short optical pulses in the regime of extreme nonlinear optics in optically dense absorbing media, providing an alternative route to attosecond-scale pulses at high frequencies. This method is based on dynamics of self-induced transparency (SIT) pulses of nearly single cycle duration, leading to single-cycle-scale Rabi oscillations in the medium. The sub-cycle components of an incident pulse behave as separate SIT-pulses, approaching each other and self-compressing, resulting in the threefold compression in time and frequency up-conversion by the same factor. As we show, the scheme can be cascaded, staying at the subsequent stage with nearly the same compression and up-conversion ratio. In this way, as our simulations show, after only few micrometers of propagation, a 700 nm wavelength single cycle pulse can be compressed to a pulse of 200 attoseconds duration located in XUV frequency range.

© 2021 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

1. Introduction

Obtaining short pulses with a nearly single-cycle duration is an important problem since it opens up new opportunities for studying and controlling the properties of matter at extremely short time scales [1]. Most widespread and promising techniques to obtain intense pulses, containing one or two cycles, include self-phase modulation (SPM)-induced spectral broadening in a waveguide [2,3], typically with subsequent linear compression, as well as optical parametric chirped pulse amplification (OPCPA) and related techniques [4,5]. Besides, many further more exotic proposals exists [6–12]. Among them, here is to be mentioned a theoretically proposed approach to so-called SIT (or coherent) mode-locking based on coherent Rabi oscillations of atomic inversion and SIT [10,11]. Pulse compression techniques can also benefit from the SIT phenomenon [13]. Since Rabi oscillations are not limited in duration from below [14–19], SIT pulses can approach few- and even single-cycle duration [14–16,20–27]. Moreover, at a sufficiently high intensity, each half-wave can itself induce the transparency of the medium [20]. In the regime of extreme nonlinear optics [28,29], when the Rabi frequency $\Omega_R = d_{12}E_0/\hbar$ (d_{12} is the transition dipole moment, E_0 the electric field amplitude, and $\hbar$ the reduced Planck constant) becomes comparable to the resonant transition frequency ω_{12}, the transition between the excited and ground state occurs extremely fast, that is, during the time less than the carrier wave period. In such situations, sub-cycle or even unipolar (containing a constant component) were predicted to occur [20–27].

In this work, we show, using simulations of Bloch equations coupled to the wave equation, that two oscillations of a single-cycle double-humped pulse behave like independent sub-cycle solitons "attracting" to each other, at the same time decreasing their duration. This leads to a significant pulse shortening (around 3 times) accomplished by a spectral broadening and gradual shift of its

#419862 https://doi.org/10.1364/OE.419862
Journal © 2021 Received 14 Jan 2021; revised 19 Feb 2021; accepted 19 Feb 2021; published 16 Mar 2021

Optics EXPRESS

central frequency upwards, up to the 3rd harmonic. The shortening/up-conversion cascade can be repeated over and over again with other media having suitable higher resonant frequencies. Here we present simulations for a two-stage compressor which converts a single-cycle 2 fs-long pulse at 700 nm into a pulse of around 200 as duration with the central wavelength around 80 nm.

2. Model equations

Our theoretical analysis is based on solving the well-known system of Maxwell-Bloch equations describing interaction of radiation with a two-level resonant medium [14–25]. The medium is described using a system of equations for the off-diagonal element of the density matrix ρ_{12} and inversion n. The evolution of the electric field is determined by the nonlinear wave equation. The system of equations has therefore the form:

$$\frac{\partial \rho_{12}(z,t)}{\partial t} = -\frac{\rho_{12}(z,t)}{T_2} + i\omega_{12}\rho_{12}(z,t) - \frac{i}{\hbar} d_{12}E(z,t)n(z,t), \tag{1}$$

$$\frac{\partial n(z,t)}{\partial t} = -\frac{n(z,t)-n_0(z)}{T_1} + \frac{4}{\hbar} d_{12}E(z,t)\mathrm{Im}\rho_{12}(z,t), \tag{2}$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E(z,t)}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2} \tag{3}$$

Here n_0 is the equilibrium difference between the diagonal elements in the absence of an electric field, T_1 is the relaxation time of the population difference, T_2 is the polarization relaxation time, $P(z,t) = 2N_0 d_{12}\mathrm{Re}\rho_{12}(z,t)$ is the polarization of the medium, N_0 is the concentration of the two-level atoms. Importantly, Eqs. (1)–(3) are written without using the approximations of slowly varying amplitudes (SVEA) and rotating wave (RWA). Therefore, they describe the coherent interaction at arbitrary time scales. In our work, the equations for the density matrix (1)–(2) were solved numerically using the Runge-Kutta method, and the wave Eq. (3) by the finite difference method. The incident single-cycle pulse was given in the form of a Gaussian waveshape:

$$E(t) = E_0 e^{-\frac{t^2}{\tau_p^2}} \sin\omega_0 t. \tag{4}$$

Here E_0 is the pulse amplitude, τ_p the pulse duration, and ω_0 the central frequency.

3. Results of the numerical simulations

We sent a strong, short optical pulse into a two-level medium 1 (see inset to Fig. 1), which the parameters of the medium given in Table 1. The input pulse is described by Eq. (4) with $E_0 = 1.7 \cdot 10^6$ ESU, $\tau_p = 2.33$ fs, and the central frequency $\omega_0 = \omega_{12}$.

Table 1. Parameters of the media

Parameters	medium 1	medium 2
Medium length, μm	2.24	1.9
Atomic concentration N_0, cm^{-3}	$4 \cdot 10^{22}$	$1.6 \cdot 10^{22}$
Transition dipole moment d_{12}, Debye	5	8
Relaxation time T_1, sec	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$
Relaxation time T_2, sec	$1 \cdot 10^{-14}$	$1 \cdot 10^{-14}$
Transition frequency ω_{12}, rad/sec	$2.69 \cdot 10^{15}$ (700 nm)	$8.07 \cdot 10^{15}$ (233.5 nm)

Evolution, resulting from the solution of Eqs. (1)–(3) is shown in Fig. 1 and in Visualization 1 [30]. This evolution consists of several distinct stages which are detailed in Fig. 2. At the

Optics EXPRESS

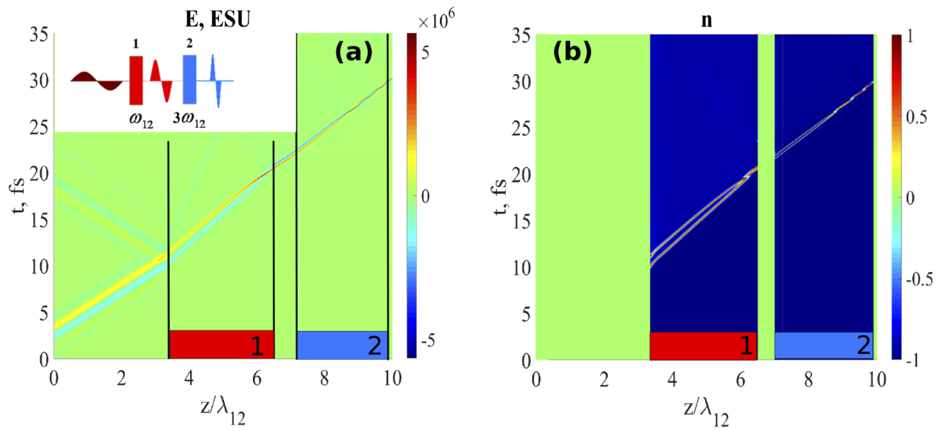

Fig. 1. A two-stage compressor containing two optically dense media 1 and 2 with transition frequencies ω_{12} and $3\omega_{12}$ (see inset). In (a), the evolution of the field and in (b) of the population difference of initially single-cycle double-humped pulse is presented. The positions of the media 1 and 2 are marked by red and blue bars in (a) and (b) and by vertical lines in (a) (see also Visualization 1 [30]).

first stage, the double-humped input pulse [blue line in Fig. 2(a)] is separated into two nearly independent sub-cycle single-hump pulses (orange line), each of them behaving almost like a separate SIT-soliton, that is, the almost full Rabi cycle of energy conversion into the medium and back takes place [see dotted lines showing the dynamics of population in Fig. 2(a)]. Note, that a tail of remaining population remain after every of single-hump subpulses. At this stage, the spectrum is noticeably broadened [see Fig. 2(b)] At the next stage, the sub-cycle single-humped sub-pulses decrease their durations and simultaneously approach to each other (green line). During this process, the double-humped character of the whole pulse is preserved. At the same time, the spectrum is continuously shifting to the higher frequencies [Fig. 2(b)]. On the third stage, the two sub-pulses join themselves to a single entity [red line in Fig. 2(a)]. The shape of this pulse changes quickly from double- to single-humped and back, and at the position where the shape is double-humped, every sub-pulse does not behave like a single SIT pulse; instead, the whole pulse behaves like a SIT pulse, as the population dynamics indicate [red dotted line in Fig. 2(a)]. At this stage, the compression does not take place anymore. As a result, at the end of the first compression stage (z=6.5λ_0), a threefold compression is obtained, and the maximum of the spectrum is located at around $3\omega_{12}$. The origin of this dynamics is discussed in the next section.

Thus, as we see, the pulse keeps nearly the same shape after the compression stage, despite of shortening and up-conversion. This opens a possibility to repeat the compression cycle, by placing a second layer with a medium having 3 times larger transition frequency. In Fig. 1, the dynamics in such second compression cascade is presented as well. The resulting pulse after the compression is shown in Fig. 2 (purple line). The pulse dynamics follows roughly the same scenario as in the first stage, resulting in a further 3-fold compression, that is, the obtained pulse is 9 times shorter than the initial pulse. The spectrum also experiences further 3 times up-conversion [see Fig. 2(b)], with the final wavelength centered at around 80 nm. After the second stage, the pulse loses its double-humped shape. We remark however that the output pulse shape strongly depends on the length of the cascade and the shape of the input pulse (see the next section for more discussion). Therefore, further cascades, might be not impossible if the input pulses are fine tuned.

Optics EXPRESS

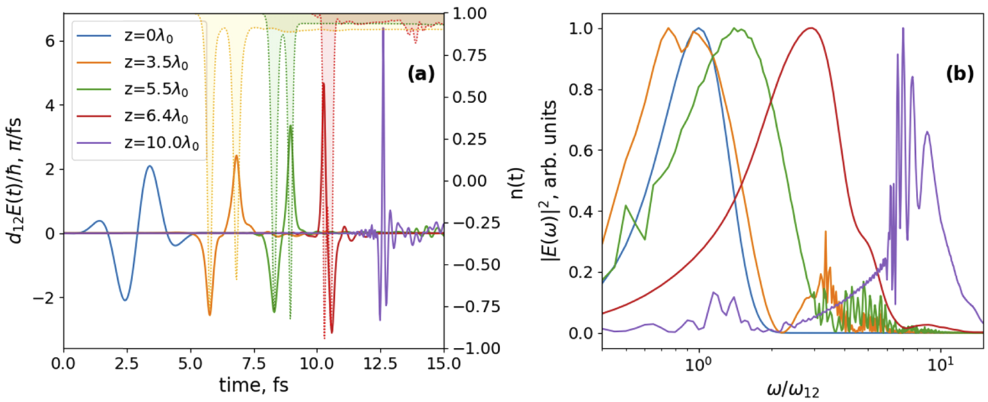

Fig. 2. The pulse shapes (a) at different propagation distances in Fig. 1 and their spectra (b). The dotted lines and shading show the population dynamics corresponding to the pulses with the same color. For the incident pulse and the pulses in medium 1, the tails of the pulses resulting from reflection from surfaces have been manually removed, to reduce spectral interference in (b).

4. Discussion and conclusions

Here we showed that a single-cycle pulse with a special double-humped shape (equivalent to the π/2 carrier-envelope offset phase) can be compressed and its frequency increased almost one order of magnitude, via a cascadable compression/up-conversion process involving SIT-like dynamics in every cascade.

The gradual transformation of the spectrum and shortening of intense radiation in resonant media was observed in [21], although for longer pulses. This process is interpreted, according to [21], as a result of intrapulse four-wave mixing process $\omega+\omega\rightarrow\omega'+\omega''$, producing, for every frequency ω, frequencies ω′and ω′′ and thus leading to a spectral reshaping. In our case, every oscillation behaves as a separate sub-pulse. The sub-pulses approach each other because of different velocities, since the residual population created by the first sub-pulse lead to different propagation conditions on the sub-pulses. This process is dependent on the initial amplitudes and shapes of the pulses, as well as on the length of the medium, so that these parameters must be good tuned to achieve the optimal result. The compression and up-conversion process can be cascaded giving rise to even shorter pulses at higher frequencies.

We performed our calculations for the parameters corresponding to a "general solid". For the first cascade, semiconductors like GaAs (bandgap 1.43 eV) can be used whereas for the second cascade dielectrics such as Si_3N_4 (bandgap 5 eV) are suitable. More generally, there is a broad class of semiconductors and dielectrics with a tunable bandgap [31]. Besides, nanostructures with tunable resonances [32,33] are also available.

In our study, we focused on a two-level medium, despite of short durations and broad spectra. In this connection we note that there is no fundamental limit on the duration of Rabi oscillations. It was theoretically [14,16–18] and experimentally shown [15] that such Rabi-floppings can approach single cycle duration, even in complex media such as semiconductors. Besides, previous theoretical studies show that short SIT solitons are preserved in three- and multilevel media [23–27], so the dynamics presented here have all chances to survive if more complex level structure, or even bandgaps are considered.

Results presented here open a new route to obtain intense attosecond XUV pulses on the micrometer propagation scales, based on extreme optics regime in resonant media. The conversion is cascadable, promising further shortening and up-conversion.

Optics EXPRESS

Funding. Cluster Excellence PhoenixD (EXC 2122) (390833453); Deutsche Forschungsgemeinschaft (BA 4156/4-2, MO 850-19/2, MO 850-23/1); Russian Foundation for Basic Research (20-32-70049).

Disclosures. The authors declare no conflicts of interest.

References

1. F. Krausz and M. Ivanov, "Attosecond physics," Rev. Mod. Phys. **81**(1), 163–234 (2009).
2. M. T. Hassan, T. T. Luu, A. Moulet, O. Raskazovskaya, P. Zhokhov, M. Garg, N. Karpowicz, A. M. Zheltikov, V. Pervak, F. Krausz, and E. Goulielmakis, "Optical attosecond pulses and tracking the nonlinear response of bound electrons," Nature **530**(7588), 66–70 (2016).
3. J. C. Travers, T. F. Grigorova, C. Brahms, and F. Belli, "High-energy pulse self-compression and ultraviolet generation through soliton dynamics in hollow capillary fibres," Nat. Photonics **13**(8), 547–554 (2019).
4. M. Nisoli, S. D. Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, Ch. Spielmann, S. Sartania, and F. Kraus, "Compression of high-energy laser pulses below 5 fs," Opt. Lett. **22**(8), 522–524 (1997).
5. G. Mourou, "Nobel Lecture: Extreme light physics and application," Rev. Mod. Phys. **91**(3), 030501 (2019).
6. G. Mourou, S. Mironov, E. Khazanov, and A. Sergeev, "Single cycle thin film compressor opening the door to Zeptosecond-Exawatt physics," Eur. Phys. J. Spec. Top. **223**(6), 1181–1188 (2014).
7. S. V. Bulanov, T. Zh. Esirkepov, M. Kando, A. S. Pirozhkov, and N. N. Rosanov, "Relativistic mirrors in plasmas — novel results and perspectives," Phys.-Usp. **56**(5), 429–464 (2013).
8. I Babushkin, A Tajalli, H Sayinc, U Morgner, G Steinmeyer, and A Demircan, "Simple route toward efficient frequency conversion for generation of fully coherent supercontinua in the mid-IR and UV range," Light: Sci. Appl. **6**(2), e16218 (2017).
9. P. A. Zhokhov and A. M. Zheltikov, "Attosecond shock waves," Phys. Rev. Lett. **110**(18), 183903 (2013).
10. V. V. Kozlov, N. N. Rosanov, and S. Wabnitz, "Obtaining single-cycle pulses from a mode-locked laser," Phys. Rev. A **84**(5), 053810 (2011).
11. V. V. Kozlov and N. N. Rosanov, "Single-cycle-pulse passively-mode-locked laser with inhomogeneously broadened active medium," Phys. Rev. A **87**(4), 043836 (2013).
12. M. V. Arkhipov, A. A. Shimko, N. N. Rosanov, I. Babushkin, and R. M. Arkhipov, "Self-induced-transparency mode locking in a Ti: sapphire laser with an intracavity rubidium cell," Phys. Rev. A **101**(1), 013803 (2020).
13. H. M. Gibbs and R. E. Slusher, "Optical pulse compression by focusing in a resonant absorber," Appl. Phys. Lett. **18**(11), 505–507 (1971).
14. S. Hughes, "Breakdown of the area theorem: carrier-wave Rabi flopping of femtosecond optical pulses," Phys. Rev. Lett. **81**(16), 3363–3366 (1998).
15. O. D. Mücke, T. Tritschler, M. Wegener, U. Morgner, and F. X. Kärtner, "Signatures of Carrier-Wave Rabi Flopping in GaAs," Phys. Rev. Lett. **87**(5), 057401 (2001).
16. D. V. Novitsky, "Propagation of subcycle pulses in a two-level medium: Area-theorem breakdown and pulse shape," Phys. Rev. A **86**(6), 063835 (2012).
17. R. M. Arkhipov, A. V. Pakhomov, M. V. Arkhipov, I. Babushkin, and A. Demircan, "Population density gratings induced by few-cycle optical pulses in a resonant medium," Sci. Rep. **7**(1), 12467 (2017).
18. R. Arkhipov, A. Pakhomov, M. Arkhipov, I. Babushkin, A. Demircan, U. Morgner, and N. Rosanov, "Population difference gratings created on vibrational transitions by nonoverlapping subcycle THz pulses," Sci. Rep. **11**(1), 1961 (2021).
19. R. Arkhipov, A. Pakhomov, M. Arkhipov, A. Demircan, U. Morgner, N. N. Rosanov, and I. Babushkin, "Selective ultrafast control of multi-level quantum systems by subcycle and unipolar pulses," Opt. Express **28**(11), 17020–17034 (2020).
20. R. K. Bullough and F. Ahmad, "Exact solutions of the self-induced transparency equations," Phys. Rev. Lett. **27**(6), 330–333 (1971).
21. V. P. Kalosha and J. Herrmann, "Formation of Optical Subcycle Pulses and Full Maxwell-Bloch Solitary Waves by Coherent Propagation Effects," Phys. Rev. Lett. **83**(3), 544–547 (1999).
22. X. Song, W. Yang, Z. Zeng, R. Li, and Z. Xu, "Unipolar half-cycle pulse generation in asymmetrical media with a periodic subwavelength structure," Phys. Rev. A **82**(5), 053821 (2010).
23. N. V. Vysotina, N. N. Rozanov, and V. E. Semenov, "Extremely short pulses of amplified self-induced transparency," JETP Lett. **83**(7), 279–282 (2006).
24. N. N. Rosanov, V. E. Semenov, and N. V. Vysotina, "Few-cycle dissipative solitons in active nonlinear optical fibres," Quantum Electron. **38**(2), 137–143 (2008).
25. N. V. Vysotina, N. N. Rosanov, and V. E. Semenov, "Extremely short dissipative solitons in an active nonlinear medium with quantum dots," Opt. Spectrosc. **106**(5), 713–717 (2009).
26. A. Y. Parkhomenko and S. V. Sazonov, "Self-induced transparency for ultrashort pulses propagating in a multilevel quantum medium," J. Exp. Theor. Phys. **87**(5), 864–874 (1998).
27. R. M. Arkhipov, M. V. Arkhipov, and N. N. Rosanov, "Unipolar light: existence, generation, propagation, and impact on microobjects," Quantum Electron. **50**(9), 801–815 (2020).
28. T. Tritschler, O. D. Mücke, and M. Wegener, "Extreme nonlinear optics of two-level systems," Phys. Rev. A: At., Mol., Opt. Phys. **68**(3), 033404 (2003).
29. M. Wegener, "Extreme nonlinear optics: an introduction". Springer Science & Business Media (2005).

Optics EXPRESS

30. See **Visualization 1** for the visualization of electric field and inversion dynamics.
31. J. Ryou, Y. S. Kim, K. C. Santosh, and K. Cho, "Monolayer MoS2 bandgap modulation by dielectric environments and tunable bandgap transistors," Sci. Rep. **6**(1), 29184 (2016).
32. L. Yuan, W. Hu, H. Zhang, L. Chen, and Q. Wang, "Cu5FeS4 nanoparticles with tunable plasmon resonances for efficient photothermal therapy of cancers," Front. Bioeng. Biotechnol. **8**, 21 (2020).
33. P. C. Wu, T. H. Kim, A. Suvorova, M. Giangregorio, M. Saunders, G. Bruno, A. S. Brown, and M. Losurdo, "GaMg alloy nanoparticles for broadly tunable plasmonics," Small **7**(6), 751–756 (2011).

194.95.159.37 {ts '2023-10-19 08:13:22'}