Spontaneous emergence of pulses with constant carrier-envelope phase in femtosecond filamentation.

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Abstract: We study the possibility to obtain high-intensity pulses that maintain a constant Carrier Envelope Phase (CEP) during propagation in *dispersive* media, i.e. pulses such that the carrier-wave offset with respect to the main intensity peak remains fixed. Our numerical experiments strongly suggest that pulse splitting and X-wave formation within femtosecond laser pulse filamentation leads to the formation of "constant-CEP" within well-defined regions inside the filament. We study the creation of "constant-CEP" pulses in both gasseous and condensed media showing that this is a generic feature of filaments.

OCIS codes: (190.5940) Self-action effects; (320.2250) Femtosecond phenomena; (320.7110) Ultrafast nonlinear optics

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#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008

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1. Introduction

Recent advances in laser technology and pulse shaping have lead to wide-spread availability of extremely intense and short pulses that may contain only a few optical cycles. This in turn has opened the road to what may be called "extreme nonlinear optics" or "carrier-wave nonlinear optics" [1], i.e. nonlinear light-matter interactions that depend strongly on the carrier-wave profile rather than on the envelope profile. Well known examples of such interactions are related to coherent higher order harmonic generation in the XUV regime and generation

#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008

of attosecond pulses (see e.g. Refs. [1, 2, 3, 4]). If $A(t,z)$ represents the complex analytic signal corresponding to the real electric field intensity $E(t,z) = Re\{A(t,z)\}$, then $|A(t,z)|^2$ is the modulus squared of the optical field envelope. For a given propagation distance z, the phase of the analytic signal evaluated at the time t_c that corresponds to a maximum of $|A(t,z)|^2$, namely $\psi(z) = \mathrm{Arg}[A(t_c,z)]$, characterizes the relative shift between the maximum of the real field and that of the envelope. This phase is called the carrier-envelope phase (CEP). Due to the sensitivity of XUV or attosecond pulse generation to the CEP ψ, it is important to be able to measure and then stabilize ψ from one laser shot to the next. A number of techniques have been introduced in this sense (see e.g. Refs.[5, 6]) and are now employed in many laboratories. However, recent developments in the direction of achieving phase-matched XUV generation imply the necessity of stabilizing CEP not only from shot-to-shot but also during the propagation [7, 8]. Furthermore, recent experimental and numerical studies involving semi-infinite gas cells have been performed, leading to long interaction lengths between the pump pulse and the gas medium [9, 10, 11].

To date there is no known method to stabilize the CEP over propagation. If the pulse propagates in vacuum, clearly the carrier-wave phase velocity and the envelope group velocity are equal, and the CEP will not change during propagation. However, pulse propagation may not necessarily occur in vacuum, and nonlinear processes occurring over long distances necessarily imply propagation in a dispersive medium so that ψ will vary with the propagation distance z, dramatically altering the nonlinear interaction. For example in air at 1 atm pressure, a linearly propagating Gaussian pulse will experience a CEP shift $\delta\psi = \pi/8$ in roughly 2 cm (or in 4 cm at 0.5 atm), and any CEP-sensitive interactions occurring over longer distances will be compromised.

In this work, we study the possibility to obtain few-cycle pulses that maintain a nearly constant CEP during propagation in *dispersive* media. Our numerical experiments show that femtosecond laser pulse filamentation can lead to the formation of such pulses within well-defined regions inside the filament and the CEP may be made to vary by less than $\pi/8$ over distances that are more than one order of magnitude better than for Gaussian pulses. We study the formation and evolution of "constant-CEP" pulses in both gasseous and condensed media showing that this is a very general feature of filaments. We explain this finding as a purely kinematic effect related to the spatio-temporal reshaping of the input pulse and the consequent modification of the on-axis intensity peak group velocity.

2. Optical filamentation and pulse splitting

Intense femtosecond laser pulses that are loosely focused in dispersive nonlinear Kerr media will undergo self-focusing and will filament. The filament is characterized by a number of typical features such as the formation of an intense, spatially localized peak that propagates subdiffractively and is surrounded by a weaker photon bath, continuum generation with both axial and conical components, pulse temporal splitting and/or temporal compression and plasma generation in the Kerr medium [12, 13]. Many efforts have been devoted to optimizing the large peak intensity and short pulse durations that are obtained within the filament [15, 14, 16, 17], and potential applications have been proposed for XUV generation [18, 19]. Most interestingly, it has also been experimentally shown that the filamentation process will maintain shot-to-shot CEP offset stabilization [20, 21] and may be used for Higher-Order-Harmonic generation [17].

Recently, filament formation and the accompanying features such as conical emission and sub-diffractive propagation, were given an explanation in terms of the spontaneous formation and dynamic interaction of nonlinear X-waves [27, 29, 30]. Of particular importance to the present work is the connection between the spatio-temporal reshaping into X-waves and the pulse group velocity within the filament. Immediately after the tightest focus region of the self-

#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008

focusing input pump pulse (nonlinear focus), pulse splitting will occur with the formation of two shorter daughter pulses. Each of these may be associated to a separate X-wave, one with subluminal (for the trailing pulse) and the other with superluminal (for the leading pulse) group velocities ("luminal" being defined as the group velocity of the input Gaussian pulse). The specific X-wave group velocity is connected to the angular dispersion of the conical emission, which may be directly measured in the far-field $(k_\perp, \omega)$ spectrum [23, 24, 28] that will typically reveal conical emission tails with an angular dispersion corresponding to the X-wave in the most intense region of the filament, i.e. for propagation distances that are close to the nonlinear focus. It is important to note that in general the X-wave group velocity, v_X, is significantly different from the value $v_G = (dk/d\omega)^{-1}$ evaluated at the carrier frequency ω_0. Furthermore, the X-wave group velocity within the filament will not remain constant but will slowly drift and decelerate (for the leading superluminal pulse) or accelerate (for the trailing subluminal pulse) with increasing propagation [24, 31].

This therefore immediately implies the possibility to observe a regime in which the "drifting" group velocity of the leading X-wave starts at a value that is only slightly higher than the carrier-frequency phase-velocity, v_φ, and later, while it decelerates, the condition $v_X = v_\varphi$ is met. If this crossing-over point is achieved with a sufficiently slow drift of v_X then a significantly long propagation distance will be achieved in which the CEP offset will remain close to a constant value. We underline that the condition $v_X = v_\varphi$ is trivial in vacuum but is far from obvious in dispersive (even weakly dispersive) media. Indeed, in general with Gaussian pulses such a condition will never be met due to the fact that, in the normal group velocity dispersion regime $v_G < v_\varphi$. It is precisely the specific nature of the filament and of the spontaneously generated leading X-wave that explains the formation of on-axis intensity peaks with group velocities that are significantly different from v_G so that we may have $v_X = v_\varphi$.

The following describes some conditions in which constant CEP offset is encountered in filamentation. In particular filamentation in air at 800 nm is considered but similar conditions are shown in highly dispersive media such as fused silica.

3. Nearly-constant carrier-envelope phase pulses in air filaments

In our search for spontaneously created pulses that propagate with an almost constant carrier-envelope phase, we performed a number of simulations of optical filamentation in air. We have explored effects of focal length, pulse duration, and air pressure to find solutions that exhibit carrier-envelope phase changing slowly over a long distance.

In what follows, we present only results for a particular simulation run performed at the pressure of 0.5 atm. The Gaussian pulse duration was 35 fs, and the energy equal to 2.5 mJ. The beam with a waist of 5 mm was focused by an $f = 10$ m lens. The frequency-dependent index of refraction of air was modeled as in [22]. For the Kerr effect we used $n_2 = 1.6 \times 10^{-23}\mathrm{m}^2/\mathrm{W}$ corresponding to a pressure of 0.5 atm. The stimulated Raman effect, was parameterized by a single damped oscillator with an angular frequency of 1.6 THz, and a damping time of 77 fs. Multiphoton ionization was approximated by the rate $\sim 3.7 \times 10^{-128}(\mathrm{m}^2/\mathrm{W})^8 I^8$. It has to be noted that while the sought after effect requires suitable conditions, varying these medium parameters in reasonable bounds doesn't have significant influence.

We used the Unidirectional Pulse Propagation Equation solver ([25, 26]) to simulate the pulse evolution and filament creation. Since the UPPE is formulated in terms of the real field (as opposed to envelope), it gave us direct access to the carrier-envelope phase.

The first indication that constant carrier-envelope phase pulses might occur in air filaments came from the experiment in which a far-field spectrum was measured after the filament [29]. It was realized that an X-wave spectral feature present in the experimental spectrum corresponds to the intensity-peak group velocity which was very close to the phase velocity at the given

#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008

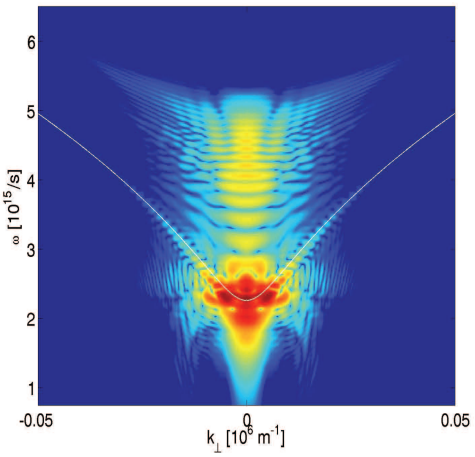

Fig. 1. Far-field spectrum generated in a femtosecond filament in air. In the filament, the incident pulse undergoes temporal splitting giving rise to two daughter (or split-off) pulses. The white line marks the spectral structure that is generated by the leading split-off pulse. The location of this structure is essentially determined by the group velocity of the peak which in this case is very close to the phase velocity of the carrier. This indicates the possibility of constant carrier-envelope phase in the leading split-off peak.

wavelength. A similar situation, only numerically modelled, is illustrated in Fig. 1. It shows the (logarithmic) spectral power as a function of the angular frequency and of the transverse wavenumber (i.e. far-field spectrum). The white line marks the locus which is "populated" due to a nonlinear-response peak moving with a group velocity that is equal to that of the phase velocity at 820 nm wavelength. One can see that the line traces closely a distinct X-wave spectral feature, and this suggests that there is an intensity peak in which phase and group velocities are equal.

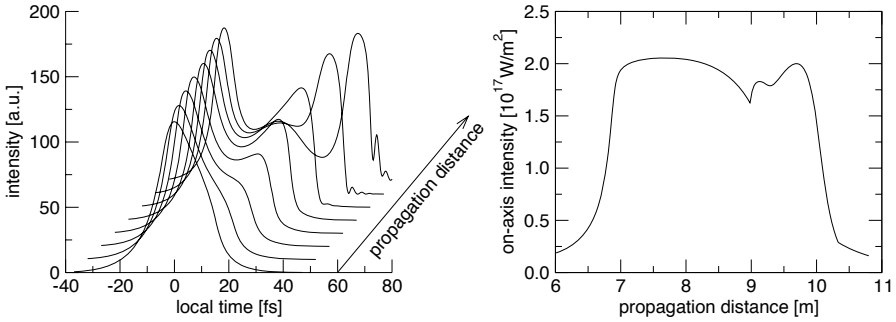

Fig. 2. Left: Pulse splitting in the air filament as seen in the on-axis intensity vs local time for several propagation distances. The leading peak can exhibit slowly changing carrier-envelope phase. Right: On-axis maximal intensity in the filament. The nearly constant carrier envelope phase occurs just beyond the first maximum and before the second pulse splitting event.

Figure 2 illustrates the temporal and spatial evolution of the on-axis light intensity for several propagation distances. The pulse splitting shown in the left panel is a generic dynamic feature

#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008

in femtosecond filaments. It is the leading split-off peak that propagates with the group velocity that is higher than the group velocity v_G corresponding to the central pulse frequency, and thus has a potential for its velocity being close to the pulse-carrier phase velocity. In the right panel, we show the maximal on-axis intensity versus the propagation distance. The propagation regime we are interested in will be shown to occur after the first maximum, and before the onset of the second pulse splitting, i.e. in region between 7.8 and 8.8 m.

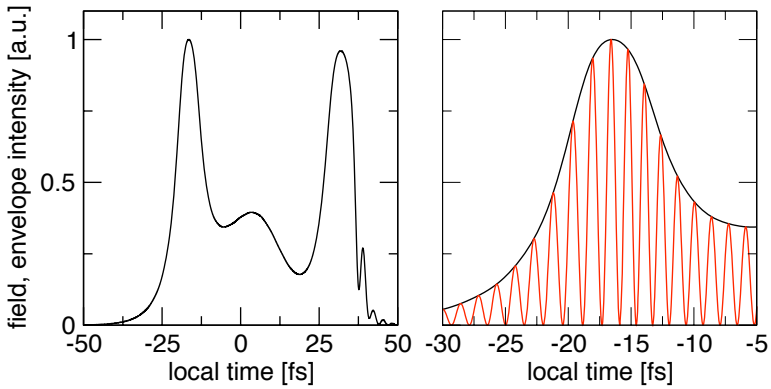

Fig. 3. Snapshot of the on-axis intensity profile (left) after pulse splitting in the filament. The leading split-off pulse is shown in detail in the right-hand-side panel together with the squared electric field intensity (red curve). The maxima of the envelope and carrier are aligned (CEP=0).

Figure 3 shows the detail of the leading pulse at the propagation distance when the maximum of the envelope happens to be aligned with the maximum of the field. Note that despite the fact that the original incident pulse duration was relatively long (35 fs), the sub-pulses created in the splitting event are considerably shorter, and may contain only several field oscillations. Next we will look closer at how the maxima of the envelope and of the field propagate, and examine the corresponding carrier-envelope phase $\psi(z)$ as a function of the propagation distance.

In general, neither of the split-off peaks exhibits a constant or even a slowly evolving carrier-envelope phase behavior. However, for suitable conditions, the evolution of the group velocity of the leading peak may be such that at certain propagation distance it equals the phase velocity corresponding to its central frequency. If this "crossing" occurs sufficiently slowly, we obtain a propagating, nearly constant carrier-envelope phase pulse. The relevant scale to quantify to what degree is the carrier-envelope phase constant is given by the difference of the group and phase velocities at a given frequency:

$$\phi(z) = z\left[k(\omega) - \omega\frac{\partial k(\omega)}{\partial \omega}\right] \tag{1}$$

Up to a constant, this describes the carrier-envelope phase evolution of a freely propagating Gaussian pulse at the central frequency ω, and $k(\omega) = \omega n(\omega)/c$ with $n(\omega)$ standing for the frequency-dependent index of refraction.

Figure 4 compares the carrier-envelope phase of the filament leading split-off pulse to that of a linearly propagating Gaussian pulse given by the above formula. Clearly, while the observed carrier-envelope phase is not strictly constant, it approaches a zero value (corresponding to the aligned envelope and field maxima) and stays small for a distance more than an order of magnitude longer than in a freely propagating pulse.

An equivalent, but more visual way to depict the behavior of the carrier-envelope-phase is in Fig. 5 where we plot the location in time, in the delayed reference frame of the input Gaussian

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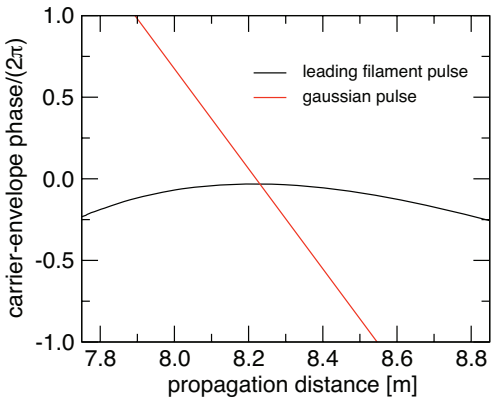

Fig. 4. Carrier-envelope phase for the leading peak vs propagation distance (black line) compared to the carrier-envelope phase in the free-propagating Gaussian pulse given by (1).

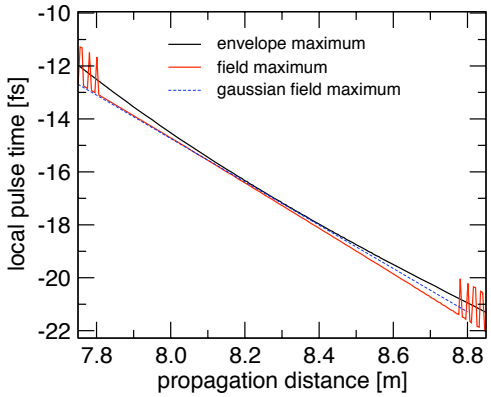

Fig. 5. Temporal locations, in the local frame of reference co-moving with the pulse, of the envelope maximum of the leading split-off peak (black line), and of the nearest real-field peak (red line). For comparison, the dashed line represents the "movement" of the real-field peak as it would be observed in a freely propagating Gaussian pulse (i.e. this line represents the phase velocity at 800nm).

pulse, of both envelope and field maxima. They co-propagate together over a significant distance while we emphasize that in this picture a freely-propagating, Gaussian envelope maximum would be represented by a horizontal line. In the same figure we also show the carrier field maximum evolution within a linearly propagating Gaussian pulse. The filament and linear Gaussian field maxima temporal locations are nearly identical, indicating that the nonlinear filament regime produces negligible effects on the carrier-wave velocity. This in turn implies that the constant-CEP arises solely as a consequence of the group-velocity properties of the envelope.

Thus, the field and envelope maxima survive aligned, with a relative phase shift less than $\pi/8$ for about one meter propagation distance. This should be compared to the four centimeters required for the same carrier envelope phase slip in a freely propagating pulse (at 0.5 atm pressure).

In the above example, the field and envelope maxima became almost exactly aligned at a certain propagation distance (CEP=0). This is the outcome of an suitable choice of the input

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phase. We have verified that the resulting carrier-envelope phase offset can be controlled by the carrier-envelope phase of the initial pulse so that the actual offset value observed in the constant CEP region may therefore be chosen to take any desired value between 0 and 2π. This behavior further corroborates and gives a deeper insight into recent experiments [20, 21], that showed that shot-to-shot CEP stabilization is maintained at the filament output.

To conclude this Section, we have observed a long-lived, almost-constant carrier-envelope phase few-cycle pulse generated spontaneously in a femtosecond filament in air. The length of constant CEP propagation can be optimized by a suitable choice of focusing, pulse duration, and gas pressure. The optimal pressure in our settings was ~ 0.5 atm. Decreasing the gas pressure below this value did not lead to a significant increase of the constant-CEP region. Moreover the constant CEP regime appears later after the pulse splitting and, consequently, the achieved peak intensity becomes lower. On the other hand, at higher pressures, e.g. 1 atm the CEP varies by less than $\pi/8$ for a shorter propagation distance of about 40 cm but in any case still an order of magnitude larger with respect to a linear Gaussian pulse. In the following Section, we address the question if this effect is special to air (or gases in general), and provide indication that constant CEP waveforms can be observed also in highly dispersive (condensed) media.

4. Nearly constant carrier-envelope phase pulses in silica

Though from the potential application point of view the constant CEP pulses are most interesting in gases, an important question regards the origin of the effect and the possibility that they are made possible by the relatively weak dispersion and nonlinearity of the gaseous medium. To show that this is not the case, we give a demonstration of this effect in a silica sample.

We show results for a 1.25 J, 100 fs pulse at 1.05 m wavelength, collimated to a 25 m beam waist at the entrance into a fused silica sample. Linear chromatic dispersion was modeled by a Sellmeier formula. The nonlinear index value used in these simulations was $2.7 \times 10^{-20}\mathrm{m}^2/\mathrm{W}$, including an 18% fraction of the stimulated Raman response approximated by a single oscillator with the angular eigen-frequency of $8.3 \times 10^{13}\mathrm{s}^{-1}$ and a decay time of 32 fs. Multi-photon ionization was described by a simple power-law rate $\approx 10^{-130}(\mathrm{m}^2/\mathrm{W})^8 I^8$. Besides the 100 fs pulse duration, we explored a range of pulse duration values. At both longer (above 200 fs) and shorter (down to 50-25 fs) pulse durations the carrier-envelope phase becomes less stable, as will be explained in the following.

In condensed media filaments, the pulse splitting phenomenon is even more pronounced than in gases. Typically, one can observe one or more splitting events that produce rather symmetric split-off pulses. The relative velocity of these pulses can vary with the initial pulse duration, which can be used in an attempt to "tune" the group velocity of the leading pulse to that of its phase velocity.

Figure 6 shows the evolution of the carrier-envelope phase (compare to Fig. 4 above). Compared to the CEP z-dependence of a freely propagating pulse, the carrier phase is rather flat. Similarly, the location in time of the envelope and field maxima shown in Fig. 7 indicate that they move with very similar velocities that in turn are very close to the phase velocity corresponding to the central wavelength.

Thus, our numerical observation suggests that the effect is quite similar to that in air. Naturally, due to stronger dispersion and nonlinearity, all scales are proportionally smaller/shorter, but there is no qualitative difference. We therefore believe that the constant CEP phase in propagating filament pulses is indeed a generic effect observable both in gaseous and condensed media.

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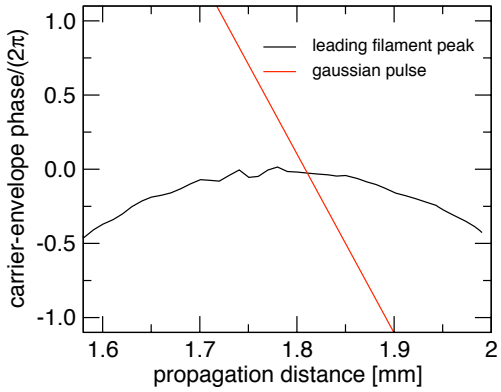

Fig. 6. Carrier-envelope phase for the leading peak vs propagation distance (black line) compared to the carrier-envelope phase in the free-propagating Gaussian pulse for a filament created in silica. Due to the condensed medium stronger dispersion and nonlinearity, the co-propagation effect is scaled down, but qualitatively similar to that observed in the air filament.

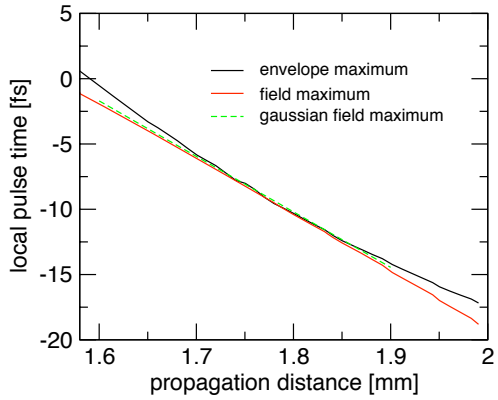

Fig. 7. Envelope and field maximum co-propagation in silica. Temporal locations, in the local frame of reference co-moving with the pulse, of the envelope maximum of the leading split-off peak (black line), and of the nearest real-field peak (red line). For comparison, the dashed line represents the "movement" of the real-field peak as it would be observed in a freely propagating Gaussian pulse (i.e. this line represents the phase velocity at 1050 nm).

5. Kinematic or nonlinear effect?

Next we address the question of the physical origin of the spontaneous, nearly-constant carrier-envelope phase pulses in optical filaments. We would like to note that at present, full understanding of the observed effect is still missing. Nevertheless, it should be useful to discuss some open possibilities even if only on a rather speculative level. As noted in the introduction, we believe that this is a kinematic, rather than a nonlinear dynamic effect. Below we offer several observations that support this opinion.

First, if one looks at the pulse splitting in filaments, one that produces a constant-CEP pulse and one that does not, there is no apparent difference in the pulse evolution, as observed in numerical simulations. Only a closer inspection of the carrier wave may reveal the presence of the constant carrier-envelope phase.

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Second, up to now we have only observed the effect further down after the initial pulse splitting. This is when the relative velocities and decelerations of the two daughter pulses decreased, and the crossing with the phase velocity thus occured on a slower scale. However, it is possible to construct examples when this happens very quickly after the splitting. In such a case, the evolution of the carrier envelope phase still exhibits the characteristic shape shown in Fig. 4, but the curvature of the $\phi(z)$ curve is significantly larger due to higher rate of change of the subpulse velocity. A similar behavior can be observed when one changes parameters that affect the pulse splitting rate, such as pressure, linear focus, and pulse duration.

In order to test the role of the nonlinear interactions in the formation of these peculiar pulses, we performed a numerical experiment in which we study the evolution of the pulse after we switch off all nonlinear interactions after a certain chosen propagation distance.

Figure 8 shows the carrier-envelope phase vs the propagation distance for three cases. The black line represents the unperturbed evolution, while the colored curves are for cases when the nonlinearity was switched off at the respective distances where the curves depart from the reference. Immediately after entering the linear propagation regime, the carrier-envelope phase rapidly deviates from the reference. However, only shortly after, the curve still displays once again a region with a relatively slow CEP evolution although only over a significantly shorter propagation distance, and with a perturbed extremal CEP value. This is fully in line with the X-wave picture of the filament formation and long-distance propagation [27, 29], more precisely with the fact that the nonlinear interactions create X-waves in which the linear propagation properties closely match the propagation of the nonlinear response in the filament core.

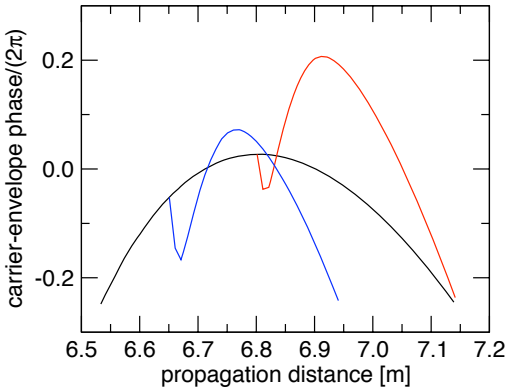

Fig. 8. Carrier-envelope phase vs distance for a normal filament (black) and two cases of linear evolution beyond certain chosen distance. The curves immediately separate, but still exhibit relatively slow CEP evolution. The latter is restored very quickly after the sudden switch-off of the nonlinearity.

The carrier-envelope phase behavior following the cross-over into a fully linear regime in some sense resembles the self-healing in femtosecond filaments [32]. In both cases, it is the conical nature of the waveform that makes it robust with respect to perturbations that mainly affect the narrow on-axis region. One can say that the carrier-envelope evolution is encoded, in a non-linear way, into the wavepacket during the early stages of the pulse splitting. As a consequence of its conical nature, the pulse can survive the loss of nonlinearity and maintain intact the peak group velocities and CEP properties.

Finally we note that one may relatively easily guess or estimate the parameters of the regime suitable to generate constant CEP pulses. Namely, one can plot, as a function of wavelength, the required group velocity change that would result in a constant CEP wavepacket (see Fig. 9).

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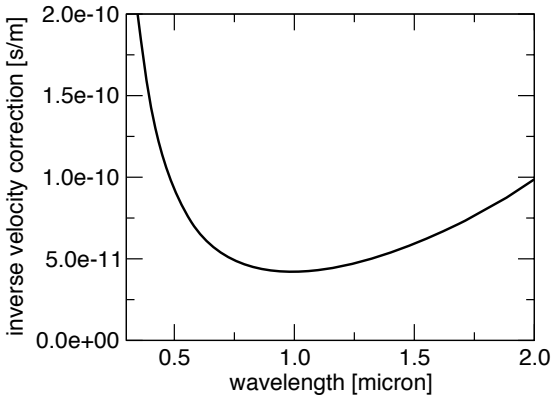

Fig. 9. Leading split-off peak inverse velocity difference $1/v - 1/v_G(\lambda)$ w.r.t. the group velocity at a given wavelength, that is required to achieve equal peak and phase velocities, $v = v_\varphi(\lambda)$, in silica. As the typical splitting rates in femtosecond filaments go, values in the range of 5×10^{-11}s/m are rather high. Therefore, constant CEP pulses are most likely to occur in the vicinity of $\lambda \approx 1$ m.

If the gap is not too large for a chosen wavelength, one can then investigate how the splitting rate, and thus also the group velocity modification, depends on other parameters such as pulse duration or linear beam focus. In a condensed medium, the chromatic dispersion may be large enough to require a significant modification of the pulse group velocity. However, there may exist a wavelength region where this difference has a minimum that is sufficiently deep. In silica, this situation occurs for the wavelength in the vicinity of one micron as illustrated in Fig. 9. Here, the required velocity of the leading splitt-off peak is in the range achievable for certain initial pulse durations, as demonstrated in Fig. 6 for a 100 fs input pulse.

6. Conclusion

In conclusion we have shown that pulses with constant CEP $\psi(z)$ during propagation may be created spontaneously in optical filaments. Clearly, the condition $\psi(z) \simeq$ constant is not achievable with standard Gaussian-like pulses in dispersive media due to the necessarily different phase and group velocities. On the contrary, the X-wave nature of the filament pulses accounts for the varying group velocities that may in certain cases be equal or very close to the carrier-wave phase velocity. The conditions for the observation of constant CEP pulses have been found both in gases and highly-dispersive condensed media, thus highlighting the generality of this effect. The propagation distance over which the CEP varies by less than $\pi/8$ is typically an order of magnitude greater than for a Gaussian pulse. In view of possible applications, it will be important to understand if it is possible to generate the constant-CEP, propagating waveforms without pedestals and satelite pulses. In particular, ways to eliminate the trailing peak and/or reduce the pulse durations should be investigated in the future.

Our numerical simulations indicate the kinematic nature of the effect: the initial group velocity of the filament pulse should be sufficiently close to the carrier-wave phase velocity and the group velocity drift should be sufficiently slow. These are the settings in which nearly constant CEP is observed. Finally the filament dynamics are such that these two conditions may be reached by tuning the input pulse parameters, e.g. wavelength and duration, and improved by tuning the material parameters such as the gas pressure. This property is also expected to hold in noble gases and may thus be advantageous e.g. for high harmonic generation generated by self-compressed filaments or the synthesis of isolated attosecond pulses which are highly

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sensitive to the CEP.

Acknowledgments

The results have been obtained in the context of the STELLA-2008 school devoted to ”training in research” (http://www.vino-stella.eu), held at IESL-FORTH in Heraklion, from April 7 to April 19 2008, as a collaboration between the MC Chair project STELLA MEXC-CT-2005-025710 and the Marie Curie Excellence Grant MULTIRAD MEXT-CT-2006-042683 (http://unis.iesl.forth.gr). This work has been partly supported by the Air Force Office for Scientific Research under grant no.FA9550-07-1-0010, the CNISM-INNESCO project ”Extreme phase matching by conical waves”, and the MIUR projects RBIN04NYLH and VINO.

#97053 - $15.00 USD Received 4 Jun 2008; revised 20 Jun 2008; accepted 5 Jul 2008; published 9 Jul 2008