Self-focusing in air with phase-stabilized few-cycle light pulses

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We investigate the nonlinear optical phenomenon of self-focusing in air with phase-stabilized few-cycle light pulses. This investigation looks at the role of the carrier-envelope phase by observing a filament in air, a nonlinear phenomenon that can be utilized for few-cycle pulse compression [Appl. Phys. B **79**, 673 (2004)]. We were able to measure the critical power for self-focusing in air to be 18 ± 1 GW for a 6.3 fs pulse centered at 800 nm. Using this value and a basic first-order theory, we predicted that the self-focusing distance should deviate by $790 \mu\text{m}$ as the carrier-envelope phase is shifted from 0 to $\pi/2$ rad. In contrast, the experimental results showed no deviation in the focus distance with a 3σ upper limit of $180 \mu\text{m}$. These counterintuitive results show the need for further study of self-focusing dynamics in the few-cycle regime. © 2010 Optical Society of America

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The development of mode-locked lasers and chirped-pulse amplification has enabled the production of light pulses whose duration is only a few cycles of the optical field [1]. Further advances have allowed for the carrier-envelope phase (CEP) of these pulses to be controlled via the f - $2f$ interferometer technique [2]. Short pulses can have very large peak powers on the order of gigawatts and readily undergo nonlinear propagation effects such as the Kerr effect, which describes a modification in the refractive index of a medium proportional to the intensity of the light passing through it, such that $\Delta n = n_2 I$, where Δn is the change in the refractive index, n_2 is the nonlinear refractive index coefficient, and I is the intensity of the light. When n_2 is positive (e.g., air, fused silica, and sapphire) the Kerr effect will cause a light pulse to self-focus if the power is greater than the critical power P_{cr} . For a beam with a Gaussian intensity profile, the self-focusing distance is given by Marburger [3] and can be modified to include an initial focusing condition through the lens transformation formula given by Talanov [4]. Thus, the observed focus position is

$$z_{\text{sf}} = \frac{0.367ka^2}{\sqrt{\left(\sqrt{\frac{P}{P_{\text{cr}}}} - 0.852\right)^2 - 0.0219 + \theta}}, \quad (1)$$

where k is the wavenumber of the radiation, a is the initial $1/e^2$ beam radius, P is the peak power of the pulse, and $\theta = 0.367ka^2/f$, where f is the initial focus of the beam. When a self-focusing beam has sufficient intensity to ionize the medium, a plasma is formed. The free-electron density in the plasma alters the refractive index so that a self-defocusing occurs. These two effects of self-focusing and defocusing come to an equilibrium and form a filament, whose length is greater than the Rayleigh range of the linear focus [5]. Recent investigations of filamentation have

shown that it is a reliable method for pulse compression to the few-cycle regime while maintaining the CEP stability [6–8]. Hauri *et al.* [9] demonstrated that filamentation is responsible for phase stable pulse compression from 43 fs, 0.84 mJ to 5.7 fs, 0.38 mJ.

The time scale of the pulse also influences the amount of self-focusing a pulse will experience, such that longer pulses will benefit from both nuclear and electronic contributions to the nonlinear refractive index coefficient [10]. A pulse with a duration shorter than the quickest molecular vibration will not induce any nuclear motion, decreasing n_2 and increasing the critical power for self-focusing. Nuclear motion contributes to the nonlinear polarizability at the 42 fs time scale. Liu and Chin found that decreasing the pulse duration from 200 to 42 fs resulted in the critical power increasing from 5 to 10 GW [11]. We measure the critical power for self-focusing in air for 6.3 fs pulses, a time scale that to our knowledge has not been previously investigated.

We also investigate the effect of the CEP on self-focusing using few-cycle phase-locked light pulses. Few-cycle light pulses with a locked CEP are interesting, because they allow for the peak of the electric field to be altered independently of all other pulse characteristics. Figure 1 shows a 6 fs pulse at a central wavelength of 800 nm (period of 2.6 fs) as the CEP is shifted from 0 to $\pi/2$. It is clear that the electric field, and hence the intensity, is largest when the phase is 0 rad and smallest at $\pi/2$ rad. At these few-cycle time scales there are no nuclear vibrations in the constituents of air, so the only contribution to the Kerr effect is electronic. Assuming that the electronic Kerr response is instantaneous, the modulation of the peak electric field should behave similarly to a modulation of the peak power. This physical picture is consistent with previous experiments on the phase stability of pulse compression in filamentation. If the measurable effect of self-focusing is power dependent

Fig. 1. (Color online) A 6 fs pulse with a central wavelength of 800 nm will have a smaller peak E -field when the CEP is $\pi/2$ compared with zero. The dashed curve represents the envelope of the pulse, the solid curves represent the carriers, and the dotted line shows the decrease in the E -field for a phase of $\pi/2$ rad.

and if filamentation exhibits phase preserving qualities, then it seems reasonable to assume that the filament will be affected in some way. The development of a substantive theory for this subject is still ongoing; however, a very simple method to estimate the change in the self-focusing distance is to take a first-order Taylor expansion of Eq. (1) and substitute the phase-dependent $P(\phi) = |E_{\text{peak}}(\phi_{\text{CEO}})|^2$ for the standard long-pulse peak power P_{peak} , yielding

$$\frac{\Delta z_{\text{sf}}}{z_{\text{sf}}^2} \approx \frac{1.362 \left(\sqrt{\frac{P}{P_{\text{cr}}}} - 0.852 \right) \Delta P}{ka^2 \sqrt{\left(\sqrt{\frac{P}{P_{\text{cr}}}} - 0.852 \right)^2 - 0.0219 \sqrt{\frac{P}{P_{\text{cr}}}} P_{\text{cr}}}}, \quad (2)$$

where $\Delta P = \delta_{\text{CEP}} P_{\text{peak}}$ with $\delta_{\text{CEP}} = 3.0\%$ for a 6.3 fs pulse centered at 800 nm as the CEP is changed from 0 to $\pi/2$.

The experimental investigation utilized Griffith University's new Attosecond Science Facility, which contains a commercially available Femtolasar CE Compact Pro consisting of a self-mode-locked 80 MHz Ti:sapphire oscillator coupled with a multipass Ti:sapphire amplifier and a hollow-core fiber to provide sub-7-fs pulses of 200 μJ energy at a repetition rate of 1 kHz. The $1/e^2$ beam radius was 2.2 mm for these experiments. The CEP stabilization is provided by a f -2f interferometer system, consisting of a fast phase lock to stabilize the oscillator and a slow phase lock before the hollow-core fiber to account for any long-term drifts that are caused by the amplifier stage as demonstrated by Baltuska *et al.* [12]. The pulses are focused initially by an off-axis parabolic reflector with a focal length of 750 mm. Fused silica wedges are used to compensate for the excess negative dispersion given to the pulse by the dispersion compensating mirror set and are optimized by maximizing the bandwidth of the self-phase modulation (SPM) spectrum that is generated beyond the focus; longer pulses will result in less SPM broadening. A charge-coupled device (CCD) camera is used to image

the fluorescence of the plasma that is formed at the focus. Figure 2 depicts a typical image that the camera captures as well as the intensity distribution of the plasma. The critical power for these few-cycle pulses was measured by attenuating the energy of the pulse with a set of pellicle beam splitters. The method devised by Liu and Chin [11] was then utilized to interpret P_{cr} from these data. Investigating the self-focusing distance with the CEP consisted of varying the phase with the slow phase-lock control system and capturing a series of images with the camera. We used a MATLAB routine to locate the brightest row of pixels in the images, and then the onset position was defined as the point where the intensity reached a given fraction of the maximum intensity (see below). This position was monitored so that any periodic deviation could be detected.

Figure 3 depicts the data that were obtained to measure the critical power for self-focusing in air with a 6.3 fs pulse centered at 800 nm, as a function of power. This measurement does not include any CEP effects as the locking systems were turned off, effectively randomizing the pulse to the pulse phase. We measure the critical power of self-focusing in air to be 18 ± 1 GW. The uncertainty in this measurement is due to the signal-to-noise ratio for powers less than 13 GW being too low to give accurate position data. This causes a large error in the curve fit to the flat section of the data and is the dominant uncertainty in P_{cr} . This value is larger than P_{cr} measured by Liu and Chin [11], demonstrating that n_2 is not entirely instantaneous at 42 fs. We use our measurement in the theoretical calculation as P_{cr} in Eq. (2).

Figure 4 shows a typical set of plasma onset deviations for the onset defined as 1/2 of the maximum intensity as a function of the CEP with the amplitude of a best-fit sinusoid indicated by gray bands. The analysis involved the images being converted to intensity distributions, and the onset of the plasma was then defined as the point when the intensity reached a fraction of the maximum. Varying the definition of the onset between 1/2 and 1/6 of the maximum intensity did not appreciably affect the results. This onset is then graphed as a function of the CEP and a sinusoidal curve with a period of π is fit where the gray band represents its amplitude. We then ob-

Fig. 2. Image of the plasma formed at the focus as captured by the CCD camera. The intensity distribution of the plasma is found from graphing the intensity of the pixels on the brightest horizontal row.

Fig. 3. (Color online) Measuring the critical power for self-focusing in air to be 18 ± 1 GW for a 6.3 fs pulse. The graph scale is log-log with crosses representing data, and the line is a fit to show the deviation point. The vertical axis was not calibrated to length, but a smaller pixel number implies that the center of the plasma is closer to the focusing mirror.

tain an upper limit of the deviation with a 3σ confidence level from this curve fit. It is this upper limit that is then an indicator of the shift of the self-focusing distance with the CEP. We find an upper limit of $180 \mu\text{m}$ for the total deviation, or twice the amplitude of the sine fit, in the plasma position as a function of the CEP. From our basic theory in Eq. (2) and substituting in a “worst-case” critical power of 19 GW, we estimate the focus shift to be $790 \mu\text{m}$. The largest source of technical noise arises from the measurement of the peak power which has an uncertainty of $\pm 0.34\%$, corresponding to an uncertainty in

Fig. 4. (Color online) Deviation in the onset of the plasma as the CEP is shifted through a 2π rad range. Crosses are data, the gray band represents the amplitude of the sin fit, and the phase axis is not absolute. The total observed shift in focus position, with a 3σ confidence level, is $180 \mu\text{m}$, considerably smaller than the calculated $790 \mu\text{m}$.

Δz_{sf} of $90 \mu\text{m}$. This noise is small compared to the 3.0% modulation of the peak power due to the CEP.

A statistical analysis of a sinusoidal curve fit reveals that there is no significant deviation in the onset of the plasma as a function of the CEP. The amplitude of the sine fit with 3σ confidence intervals has a maximum upper limit of $89 \mu\text{m}$, meaning the total observed shift in focus position was $180 \mu\text{m}$. This observed shift is in strong disagreement with our conservatively calculated shift of $790 \mu\text{m}$. Clearly, the simple model of Eq. (2) is unable to account for the data. More sophisticated analytical and numerical studies are needed to explain our results.

This work is, to the best of our knowledge, the first to explicitly explore the role of the CEP with the nonlinear phenomenon of self-focusing in the few-cycle regime. We were able to establish the critical power for self-focusing in air to be 18 ± 1 GW for a 6.3 fs pulse centered at 800 nm, and this value was used in the prediction for Δz_{sf} . A simple first-order theory is unable to account for the effect of the CEP in self-focusing, with a discrepancy between the calculated $790 \mu\text{m}$ and the measured upper bound with a 3σ confidence interval of $180 \mu\text{m}$ in the deviation of the plasma. This counterintuitive result shows that much more study of this phenomenon is required—how can the CEP be maintained when the filament is used for pulse compression but has no effect on the self-focusing dynamics?

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