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The effect of propagation in air on the filament spectrum

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Abstract: Filamentation studies traditionally start from letting a beam focus in air. We present filament studies with control over the preparation propagation, in air or vacuum, using an aerodynamic window. The spectral content of the filament strongly depends on its preparation medium.

OCIS codes: (190.5530) Pulse propagation and temporal solitons; (190.7110) Ultrafast nonlinear optics.

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1. Introduction

Filamentation in air refers to a phenomenon by which self-focusing [1] of an intense pulse in air is followed by an extended region where the light remains concentrated in a diameter of the order of 100 μm. The propagation can be divided into three regions of preparation, filamentation and diffraction. Of the three, the second one had been the major concern in filamentation studies, where the theory of balance between Kerr focusing and plasma defocusing [2, 3] is applied. The third region is used for observation of far field diffraction of the surviving optical beam and conical emissions [4, 5]. Nonlinear interaction between the pulse and the air in the preparation phase supported the theory of the moving focus by Shen [6], in which the temporal intensity distribution of a large beam creates a moving focal point. Another theory considering the first region of propagation is the reshaping by nonlinear absorption and Kerr effect of the beam profile into a non-diffracting Bessel beam [7]. All these theories and interpretations are based on one common concept of nonlinear interaction of light and matter, in a regime where light intensity is sufficiently high to ionize matter, and the matter is dense enough to affect the light. In this paper we report on experimentally eliminating the nonlinear interaction in the preparation phase, by focusing the beam through vacuum and launching the filament at a well defined position determined by the placement of an aerodynamic window. The spectrum of this filament is compared to the conventionally created filament, with a preparation phase in air (rather than in vacuum). Our study shows that most spectral broadening and frequency generation take place during the preparation in air. Moreover, for the filaments prepared in vacuum, the filamentation can only be due to a balance between self-focusing and de-focusing, excluding other mechanisms.

The concept of self-trapping of intense pulses or filamentation has been the object of controversy since the early 60's. The first observations were hair like traces (hence the name "filaments") of shattered glass inside a window or prism, after irradiation by an intense pulse. Akhmanov [8] assumed a nonlinearity of the type $n = n_0 + n_2 I + n_4 I^2$, ($n_0$ and n_2 being the linear and nonlinear indices of refraction, respectively, and I the intensity) where the higher order term $n_4 I^2$ has a de-focusing effect (n_2 being positive, n_4 negative). For a Gaussian laser beam of wavelength λ and power that exceeds a "critical power" [9] $P_{cr} = \lambda^2/(8\pi n_0 n_2)$, the focusing power due to n_2 exceeds the natural diffraction, resulting in a beam collapse. Once the focal point is approached, higher order effects dominate and arrest the collapse. With a parabolic approximation applied to the beam profile, the propagation equations lead to an analytic solution for the beam diameter oscillating between a minimum and maximum value [8, 10]. This first interpretation of a filament was soon contested by Shen [6], arguing that the filament-like trace of shattered glass created by an intense Q-switched pulse was the manifestation of a "moving focus". Starting from a beam waist w_0 at the entrance of the nonlinear medium, as the power increases along the leading edge of the pulse above the critical power P_{cr}, the "self-focus" re-

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cedes from $-\infty$ to a focal point f_{sf} at a distance $f_{sf} \approx \rho/\sqrt{P/P_{cr}-1}$ (where $\rho = \pi n w_0^2/\lambda$ is the Rayleigh range, $P = 2I/(\pi w_0^2)$ is the laser power [9]). The filament like laser damage results from the high peak intensity at the focus. The controversy has only intensified since, with the partisans of "moving focus" denying the existence of self-trapped filaments, despite clear examples of self-trapping such as the observation of CW filaments in micro-emulsions [11]. The controversy further intensified with the observation of filamentation in air [12]. After more than one and a half decades of research with 800 nm filaments in air, the following interpretations are being debated as to the nature or definition of filaments:

1. Moving focus [13]
2. Balance between self-focusing and plasma defocusing [3]
3. Reshaping of the beam profile by nonlinear effects, resulting in "conical self-focusing" and a Bessel beam [7]
4. The filaments are continuously created from a surrounding reservoir [14].

To complicate things further, some of these models partially overlap with each other. The objective of this paper is not to contradict of discredit any of these models. Rather, we present the result of experiments that demonstrate filamentation in conditions in which the mechanisms (1), (3) and (4) cannot contribute. These mechanisms are eliminated by focusing all the energy of the beam in a 200 μm diameter beam waist in vacuum, prior to launching the pulse into the atmosphere. Since the models (1), (3), and (4) interpret the filaments as resulting from the nonlinear effects in a "preparation phase" in air, their contribution is automatically eliminated by putting the preparation phase as a focal spot in vacuum. The scope of applicability of mechanism (3) will be investigated in a future experiment in which the filament prepared in air will be launched into vacuum. The filaments that will be produced by launching from a vacuum beam waist into the atmosphere, result from a balance of self-focusing and de-focusing, consistent with the description of Akhmanov [8], where the pulse energy is trapped in a self-induced waveguide. In our study we measure the spectral content of the filament as a function of the propagation distances in filaments (a few selected positions are shown in Fig. 1). We do not observe any generation of spectral component around 700 nm, when the filament undergoes a preparation phase in vacuum. This is in contrast with the numerical simulation of Kosareva*et al* [15] which do not include a preparation phase in air. The measurements of Fig. 1 show unequivocally that although spectral broadening, both on-axis and in the form of conical emission, develops continuously over the whole filament length, it is actually observed only if *initiated* in the "preparation phase".

2. Experiments

Filaments have generally been created by letting a collimated high power laser pulse self-focus in air. Besides the spectral redistribution of energy during that propagation process, the starting point of the filament is undefined. Furthermore, the phenomena associated with the "preparation phase" cannot be distinguished from the true "filamentation phase". Instead of letting the filament develop from a macroscopic beam of $\approx$ 1 cm diameter, we focus the pulse inside a vacuum cell, to a minimum beam waist that is placed close to a transition of vacuum to air. The intensity at the focal spot in vacuum exceeds 1 TW/cm^2. At this intensity any window material is either damaged or shows significant nonlinear effects. The only element that offers a gradual transition from vacuum to atmosphere with a minimum of distortion and absorption is an aerodynamic window. It provides a pressure gradient across a supersonic air stream, in an expansion chamber profiled in such a way that the pressure on one side is atmospheric, and on

#162318 - $15.00 USD Received 31 Jan 2012; accepted 7 Mar 2012; published 26 Mar 2012

the other side less than 10 torr (the transition from a few mm of Hg to atmosphere takes place over approximately 2 mm). The supersonic gas flow enters the diffuser, recovers its pressure back to atmospheric conditions, and ejects into the atmosphere. The supply pressure upstream (approximately 4 kg/cm^2) of the supersonic nozzle can be varied to achieve optimum performance. The compressor associated with our window supplies a continuous flow of 10 m^3/min (150 dm^3/s) at a pressure of 8 kg/cm^2.

A standard technique to attenuate linearly the filament before detection is to use a thin plate of fused silica at grazing incidence. The small fraction of the beam that is transmitted through the plate at a grazing incidence of θ_i=89 degrees is 6%. For a 10 mJ filament of 200 fs duration, that represents a power of 3 GW). The critical power for self-focusing in fused silica is about 2.5 MW. A simple back of the envelope calculation shows that the self-focusing length [9] for a $w_0 = 100\mu$m beam is 1 mm, which is thus the maximum thickness that a grazing incidence attenuator can have. It has not been possible to have the required flatness of $\lambda/10$ of such a thin plate over the 10 cm diameter required for the grazing incidence, and therefore the filament profiles that we obtained by this method were distorted. The solution we adopt is to use a large rigid mirror with a maximum reflectivity coating at grazing incidence. We have found that a standard Nd:YAG high reflector coating (maximum reflectivity at normal incidence for 1.06 μm) has, at 795 nm, a transmission of less than $0.9 \cdot 10^{-6}$ at 89^o incidence (*s*-polarization). The transmitted power for a filament is reduced to 25 kW, far below the critical power, and a thick (12.5 mm) substrate of good optical flatness can be used. At the grazing incidence of $\theta \geq 89^o$, the filament covers a sufficiently large area on the plate, to have the intensity below damage threshold for the coating. This technique is limited to *linear* polarization: near the wavelength of 795 nm, the transmission is increased by a factor 70,000 in *p*-polarization as compared to *s*-polarization.

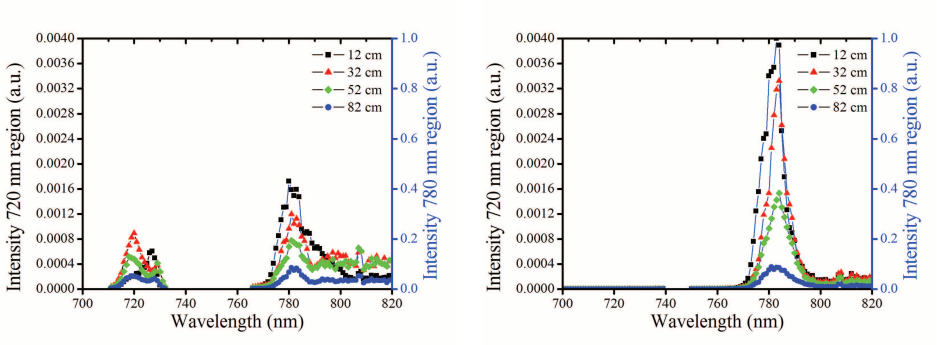

Fig. 1. Spectra recorded at various distances from the geometrical focus of the 3 m lens are displayed for the case of (right) focusing in vacuum (aerodynamic window on) and (left) focusing in air (aerodynamic window off). New wavelengths are generated in the blue region of the spectrum when (left) focusing in air (aerodynamic window off).

A beam of 4 mm 1/e^2 radius (or FWHM/1.177) is focused by a 3 m focal distance lens at the entrance of an aerodynamic window, creating a beam waist of 200 μm (1/e^2 radius). The combination of a grazing incidence plate and a simple prism spectrometer are positioned at various distances from the geometrical focus of the lens (located at the entrance of the aerodynamic window). The spectra shown are corrected for the transmission function of the grazing incidence plate. The calculated transmission curve of the grazing incidence plate was matched by measurements at 8 wavelengths in the range of 750 to 800 nm. The recorded spectra for the

#162318 - $15.00 USD
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wavelength range at and around the central laser wavelength are shown in Fig. 1. One notices that, for the spectra corresponding to the filaments launched from vacuum, there are very few new lower frequencies generated in the IR portion of the spectrum, and little distortion of the original spectrum [Fig. 1(right)]. Profile measurements show the beam to remain transversally confined up to a distances of 72 cm, corresponding to diffraction-free propagation over $\sim 5\times$ the Rayleigh length.

The contrast between the two spectral measurements is striking for the wavelength region shorter than that covered by the pulse spectrum. For the case of the spectrum generated in air, new wavelengths are created [Fig 1(left)]. This spectrum is in agreement with other experimental observations (for example [15]). When the filaments are generated from a focal spot in vacuum, no light is detected in the wavelength region between 700 and 740 nm. One would be led to believe from these measurements that filaments are not generated from a focal spot in vacuum. Measurements of filament profiles, plasma formation and conductivity have confirmed that filaments exist for preparation in vacuum, as "self-induced waveguides". It should be noted that the theoretical model of ref. [15], with a small diameter pulse as initial condition, corresponds to our experimental situation when focusing *in vacuum*. However, their results show emission between 700 and 740 nm, which is not consistent with our data presented in Fig. 1 (right).

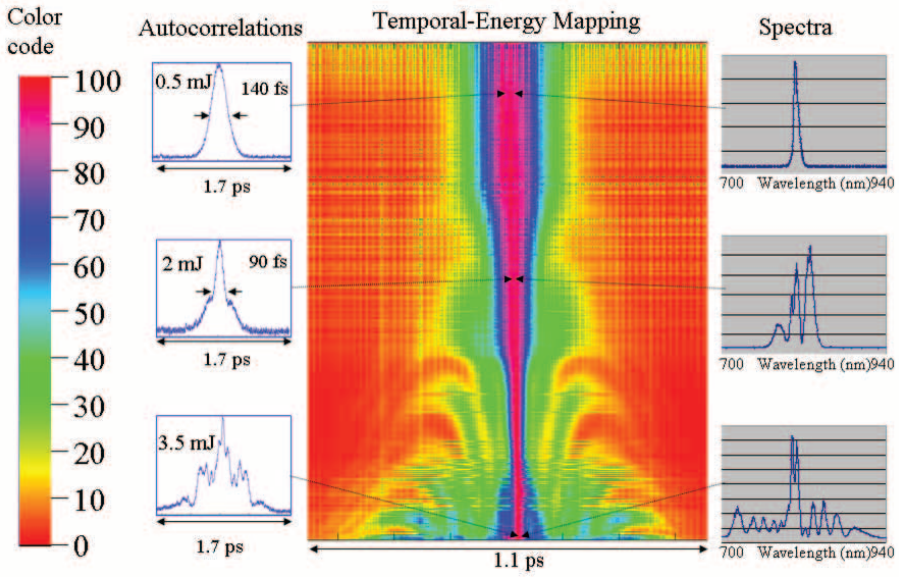

Fig. 2. Autocorrelations and pulse spectra measured at 23 m from the source (compressor). From left to right: false color coding for the autocorrelation intensity; sample traces of the autocorrelation at specific pulse energies; detailed mapping of the autocorrelations versus pulse energy in the range of 0.1 to 3.5 mJ; selected pulse spectra between 700 and 940 nm, for the same 3 representative pulse energies selected for the sample autocorrelations.

Even in the absence of filamentation, we have observed [16] the generation of new wavelengths and pulse splitting in a collimated high power laser beam propagating in air. A collimated beam of 140 fs autocorrelation FWHM with a $1/e^2$ radius of w = 4 mm is let to self-focus over a distance of up to 23 m. The beam profile, pulse autocorrelation, pulse spectrum were measured at fixed distances from the source, as the pulse energy was ramped from 0.1 mJ to 3.5 mJ. This is a study of the *preparation* phase, as a filament was not produced, even

#162318 - $15.00 USD
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at the maximum distance of 23 m. An example of data is presented in Fig. 2, where the autocorrelation is displayed in false colors as a function of increasing energy at a distance of 23 m from the source. On the left and right of the central figure, selected autocorrelation and spectral traces are presented. The spectra generated in this preparation phase are consistent with the measurements presented in Fig. 1.

A series of FROG traces taken after 10 m propagation in air for a 140 fs laser pulse monitors the spectra-temporal evolution of the pulse [17, 16]. Representative temporal pulse amplitude and phase as well as spectra taken at that location for a 0.3 mJ pulse and a 2.2 mJ pulse are shown in Fig. 3 (See supplementary material for more data).

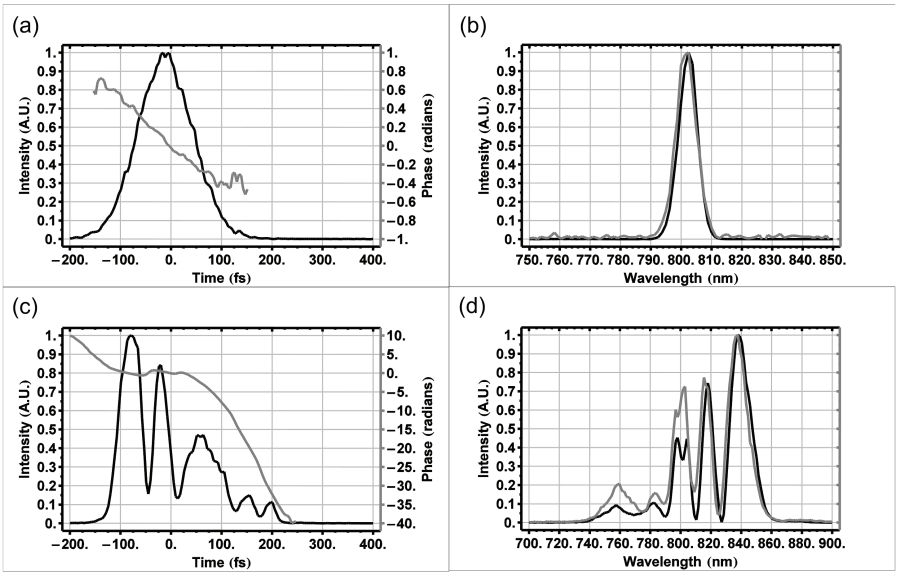

Fig. 3. Comparison between a frog-retrieved pulse shape measured at 10 m from the source, for an initial energy of 0.3 mJ and 2.2 mJ. (a) and (c) are the pulse amplitude and phase in time. (b) and (d) are their Fourier transforms superimposed on independently measured on-axis spectra of the same shots.

3. Discussion

The preceding measurements taken with different focusing conditions are all consistent in showing that filament-induced spectral broadening is linked to the existence of a *nonlinear* preparation phase. This finding is in agreement with computer simulations of Gaeta [18], in which turning off the plasma still led to blue-shifted emission. Measurements by Faccio *et al.* [4] and Maioli *et al.* [19] have found the spectral-angular distribution of the conical emission to be consistent with "X-waves" [20]. X-waves are a manifestation of dispersion, and do not presuppose any mechanism for creating the additional wavelengths. However, for these dispersion relations to apply, all k vectors and frequencies ω need to be present in the beam.

In the case of filaments prepared in air, nonlinear absorption results in beam reshaping, resulting in axicon-like focusing which creates a distribution of k-vectors. This modification of the k-vector spectrum is in general responsible for the spontaneous formation of conical or Bessel-like pulses or beams, as originally proposed by Dubietis et al. [7]. The filaments created in air have been shown in simulations to persist even in vacuum, which is a clear manifestation of the Bessel-like nature of the filament [Fig. 2(b) in reference [4]].

#162318 - $15.00 USD
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Thus, as the pulse starts to self-focus in air the *k*-vector spectrum expands. Not only does the angular spread of the pulse increase in the preparation phase in air, but also the pulse splitting and compression seen in Figs. 2 and 3 leads to the generation of new frequencies. The new waves that are generated and propagated have to add in phase along the various *k*-vector directions, i.e. fulfill the requirements of stationarity. This is the driving mechanism that leads to X-wave formation [21]. Non-stationary k–ω points will just lead to an overall cancellation of light in those other regions. Stationarity also depends on the dispersion landscape, which is why changing medium (air, water, alcohol) changes the dispersion to lead to different X-waves, O-waves *etc*.

By contrast, in the case of the self-trapped filament generated from a focal spot in vacuum, the absence of a "gentle" preparation phase that is now substituted by a very abrupt and violent transition from linear to highly nonlinear propagation, does not allow the pulse to gradually reshape itself following stationary phase trajectories. This results in a severe reduction of spectral reshaping or broadening, features typically considered as a clear signature of filamentation. Nevertheless, even in the absence of significant spectral broadening, the pulse focused in vacuum does still form a filament that is maintained through a dynamical interplay between Kerr-induced self-focusing and plasma defocusing. Our results indicate that significant nonlinear propagation effects take place during the preparation phase in air. This nonlinear propagation is crucial for initiating spectral broadening and pulse reshaping that occur within the actual filament itself. Removal of the nonlinear preparation phase by focusing in vacuum suppresses spectral reshaping but does not eliminate the filament itself.

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