Super high power mid-infrared femtosecond light bullet

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Supplementary material

Instability analysis and carrier wave shock expectancy

A straightforward modulation stability analysis supports the idea that mid-IR filament waists are significantly larger than their 800nm counterparts and, moreover, that such filaments should be more robust and resistant to external perturbation. The modulational instability is responsible for the initiation of hundreds of chaotic filaments across an 800nm beam although the final relative disposition of such filaments in the transverse cross-section is often dictated by other strong perturbations such as phase perturbations from the multi-stage amplifiers. Small perturbations to an infinite transverse plane wave solution of the nonlinear Schrödinger Equation (NLSE) are unstable over a finite band of transverse spatial wavenumbers. As these perturbations grow nonlinearly they can develop into multiple filaments1. The gain spectrum of the transverse instability is given by

\[
g(K_{\perp}) = \frac{\lambda_0 |K_{\perp}|}{2\pi n_0} \left[ \frac{16\pi^2 n_0 n_2 I_0}{\lambda_0^2} - K_{\perp}^2 \right]^{1/2}
\]  

(1)

where the peak gain is \( g_{\text{max}}(K_{\perp, \text{max}}) = 4\pi n_2 I_0 / \lambda_0 \). A comparison of the gain spectrum for the wavelengths of 800nm, 1.6\( \mu \)m, and 4\( \mu \)m is shown in Figure S.1. What one observes is a rapid shrinking of the unstable domain and reduction in the rate of growth of the most unstable mode (maximum of each curve) as the wavelength gets longer. The initial fastest growing transverse spatial wavelength is given by

\[
\Lambda_{\perp} = \frac{\pi}{K_{\perp, \text{max}}} = \frac{\lambda_0}{4} \sqrt{\frac{2}{n_0 n_2 I_0}}
\]  

(2)

This plane wave analysis if of course simplistic and a more sophisticated analysis using the WKB approximation taking into account the curvature of an initially finite beam (diffraction losses) shows that the unstable domain and consequently growth rate of the unstable wavenumbers shrinks even further. Consequently from inspection of the modulation growth rate curve (red) for the 4\( \mu \)m wavelength in Fig. S.1a, we can conclude that this unstable domain will further shrink making such mid-IR transverse beams very resistant to modulational instability growth and their transverse unstable wavelength even longer. The regularization of nonlinear growth into filaments at mid-IR wavelengths is limited by optical carrier wave self-steepening and shock walk-off, limiting transverse filament waist sizes to 1-2mm. It is also important to emphasize that the power in each filament is much higher at longer wavelengths, scaling with \( \Lambda_{\perp}^2 \) and
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Small perturbations to an infinite transverse plane wave solution of the nonlinear Schrödinger Equation (NLSE) are unstable over a finite band of transverse spatial wavenumbers. As these perturbations grow nonlinearly they can develop into multiple filaments. The gain spectrum of the transverse instability is given by

$$\Lambda = \frac{2}{g_{\max}} = \frac{\lambda_0}{2\pi n_2 I_0}$$

where the peak gain is

$$g_{\max} = \frac{\lambda_0^2}{2\pi n_2 I_0}$$

A comparison of the gain spectrum for the wavelengths of 800nm, 1.6 μm, and 4 μm is shown in Figure S.1. What one observes is a rapid shrinking of the unstable domain and reduction in the rate of growth of the most unstable mode (maximum of each curve) as the wavelength gets longer. The initial fastest growing transverse spatial wavelength is given by

$$k_{\max} = \frac{2\pi}{\lambda_0}$$

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Consequently from inspection of the modulation growth rate curve (red) for the 4 μm wavelength in Fig. S.1a, we can conclude that this unstable domain will further shrink making such mid-IR transverse beams very resistant to modulational instability growth and their transverse unstable wavelength even longer. The regularization of nonlinear growth into filaments at mid-IR wavelengths is limited by optical carrier wave self-steepening and shock walk-off, limiting transverse filament waist sizes to 1-2mm. It is also important to emphasize that the power in each filament is much higher at longer wavelengths, scaling with

$$\frac{\lambda_0^2}{2\pi n_2 I_0}$$

One last observation comes from the characteristic scale corresponding to the first appearance of instability growth, namely multiple filamentation takes longer to develop at longer wavelengths.

Figure S.1 | a, Modulational instability analysis of an infinitely extended transverse plane wave showing the trend towards growth of longer transverse spatial wavelengths as one moves to longer optical carrier wavelengths. b, Schematic graph of the partition between classical NLSE-like critical collapse (black region) and optical carrier shocks (blue region) as obtained from an analysis of the MKP, represented in the I-λ plane.
Fig. S.1b shows the partition between the pure blow-up (critical self-focusing – black region) and a mix of carrier shock and blow-up singularities (blue region). In the latter, the optical carrier shock development is further accelerated as a consequence of increasing intensity at the onset of blow-up. This figure is best interpreted by fixing the carrier wavelength and moving vertically upward.

In Fig. S.2 we can see a comparison between the full 3D+1D simulation with initial random perturbations of the beam phase-front (blue curve) and an identical run in radial symmetry without the perturbation (red curve), for the 177.2mJ – 24fs (FWHM) – 1.5cm beam waist wavepacket that is the main focus in this work. While both models accurately describe the self-focusing process and the 30m long filament, there is an obvious difference at z=~70m. The radial symmetric run exhibits a large intensity spike, which is not predicted by the 3D+1D case. This spike, while not a numerical artifact, is created by the intermixing of out-of-phase harmonics that are forced on axis by the radial symmetry of the numerical grid, creating the illusion of a linear rogue wave. Similar type of behavior has been observed in highly multi-moded glass fibers by Arecchi et.al.3. In our case however, the intensity spike at 70m is a result of the radially symmetric enforced spatial grid and the on-axis temporal energy leakage native to the mid-IR filamentation regime. Therefore we strongly believe that the numerical study of mid-IR filaments in the regime where temporal walk-of is the main collapse arresting mechanism should always be conducted in 3D+1D geometry, as is the case in this work.

![Graph showing peak intensities along propagation distance](image_url)
Physical origin of critical collapse and shock regularization

The question of what physics contributes to regularizing the critical collapse singularity, most commonly identified with the famous nonlinear Schrödinger equation (NLSE) has a long history and remains mostly unresolved. What we do know is that a short, intense laser pulse at near-IR wavelengths undergoing critical self-focusing in a transparent solid or liquid will split into two separated pulses in a normally dispersive medium. This can lead to the spectacular development of so-called X-waves. In an anomalously dispersive medium, the pulse simultaneously compresses in space and time. A higher power pulse propagating in a gas will experience a much weaker dispersion but the limit of dispersion being stronger than nonlinearity still holds. Here, ionization (plasma) acts to defocus the pulse by creating a negative transverse spatial lens and one observes a sequence of short focusing bursts typical of filamentation in air. There are significant gaps between individual pulses and they die out rapidly over meter length distances. In such media a small parameter asymptotic analysis of the Maxwell equations leads the usual envelope NLSE-like equations. We should stress here, that in the ideal NLSE model describing a CW beam, regularization of the critical collapse singularity only requires an infinitesimally small decrement in power of the collapsing waveform.

In the long wavelength case under consideration here, the NLSE envelope description fails and we are now in the limit where the nonlinearity dominates over dispersion – the appropriate model derived from an asymptotic analysis of Maxwell’s equations in this limit is now the modified Kadomtsev Petviashvili (MKP) equation. Now we have two types of singularities, blow-up and optical carrier wave shock. This is the case considered in the present paper.

As carrier shock waves dominate, we digress a little here to discuss what these are and how they are regularized through weak dispersion. As discussed in the Methods section, shocks are easiest to describe in terms of the solution of the Inviscid Burger’s equation

\[ \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} = 0 \]
\[ u(x,0) = 1 - \cos(x) \]  \hspace{1cm} (4)

This is a nonlinear wave equation where the velocity is now inversely proportional to the amplitude of the solution. The initial data \(u(x,0)\) describes a simple truncated cosine waveform at time \(t=0\) - any shape can be considered. If propagated in \(x\) this shape exhibits the classic self-steepened shock-like structure before folding over and becoming multivalued as shown in Figure S.3. The lower amplitude part of the solution at the back of the pulse propagates faster than the central high amplitude part and piles up on it, creating the steep front.
This multi-valuedness is clearly nonphysical and drove Korteweg and DeVries in 1895 into adding a small linear dispersive term on the right hand side. The KdV extension to Burger’s equation takes the form.

\[ u_t + uu_x = \varepsilon u_{xxx} \]  

(5)

The added dispersion via the triple derivative of \( u(x,t) \) in \( x \), is weighted by a small term \( \varepsilon \). If \( \varepsilon \) is very small, the shock front will become very steep but not fold over and will eject high frequency oscillations at its back end – this is dispersive regularization. A further simple modification of the KdV equation involves replacing the nonlinear function \( u(x,t) \) by \( u^2(x,t) \) and leads to the modified KdV equation.

\[ u_t + u^2 u_x = \varepsilon u_{xxx} \]  

(6)

Not surprisingly, this equation also exhibits shock structures and shock regularization. Simply adding one or more transverse second derivatives to this equation leads to the MKP equation referenced above. The latter’s solutions, which now represent the real electric field, mimic the behavior of the full UPPE simulations presented in this paper and can simultaneously exhibit shock and blow-up singularities. The figure on the right above shows that an identical Burger’s-like behavior, where now the optical carrier wave tries to fold over, happens if we set GVD to zero in the UPPE propagator. This folding over happens long before the intensity saturates from self-focusing.
Although dispersion is low, it is finite and it prevents rollover of the carrier wave shown above. This allows harmonic components, initiated primarily by carrier shocks, with associated broad spectral content to slowly separate from the fundamental at a finite but slow group velocity difference. We actually solve the full UPPE (essentially unidirectional Maxwell) here from which MKP can be derived as an asymptotic limit. Our observation of the regularization of the critical collapse in the present paper is in stark contrast to the established nonlinear envelope scenarios. We observe instead, a soft transition into a main single pulse that retains an invariant quasi-solitonic leading edge, an almost fixed extended beam waist and a tail that recurrently emits bursts of much weaker spectrally broad harmonic components. Once the shock dominates on the main pulse, otherwise well separated higher harmonic spectral components merge into an almost featureless broad supercontinuum spectrum. This merging of the multiple higher harmonic peaks happens gradually as the pulse propagates in space, and can be seen in Figure S.4 where the spectrum at 40, 50, 60, and 80 meters is shown.

While the whole supercontinuum is essentially walking-off the main pulse, the process first starts with the shock initiated formation and walk-off of individual odd harmonics. The spectral broadening of those harmonics does not change the physics in essence, but rather complicates the phenomenon. The above observation justifies our choice of neglecting the dispersion poles of air located in the spectral region that lies in between the odd harmonics. It was shown in a previous work that when using the UPPE spectral propagator, finite poles well away from the driving frequencies will modify the supercontinuum spectrum only locally, but will not terminate it like in the case of the NEE model. Furthermore, additional simulations that were conducted in the mid-IR in radial geometry show that indeed our assumption is valid. Fig. S.5 shows the results of the comparative simulations with (red curve) and without (blue curve) the dispersion poles added to the susceptibility of dry air around 2.7μm and at and beyond 4.1μm (left panel). As we can see in the right panel, while the electric fields are slightly shifted with respect to each other, the physics are qualitatively the same and result in the formation of long optical structure. In both cases the electric field undergoes steepening and new higher frequencies are generated that are left behind due to the different propagation velocity (walk-off).
The role of plasma generation in mid-IR filamentation in air

The main point of this work is the presentation of a new paradigm for filamentation in the mid-IR, where the main collapse arresting mechanism is shock driven spectrally broadened harmonic walk-off instead of plasma generation. This claim is well supported by comparing the ionization losses to the energy losses due to walk-off, where the latter was found to be about 5 times higher.

In order to further support this claim we conducted simulations where plasma generation is switched off. In this scenario, where the classical collapse arresting mechanism is absent, simulations are in general not expected to converge to a finite solution. Fig. S.6a shows the peak intensity build-up for plasma on and off and Fig. S.6b shows the electric fields at their peak value along z (z = 63m), for simulations with (red curve) and without (blue curve) plasma generation. The input pulse carries 60mJ of energy, has 1cm beam waist and 117fs duration, (same as the one depicted in the red curve of Fig. 6). While it is clear that plasma generation does play a role in the process, it is not needed to arrest the collapse. The difference between the two cases is attributed to a weak plasma induced spectral blue shift that further adds to the already very small intrinsic GVD. Thus, the blue shift acts against shock formation and in effect helps to soften it. The inset in Fig. S.6b contrasts the slope of the shock for both cases showing that the “ionization off” shock is much steeper. One can view this ionization-mediated process as an additional channel for dispersive shock arrest. The full evolution of the fields with ionization on and off are shown in detail in the accompanying movie 3. If, additionally, one switches GVD off, the shark fin shaped shock front is already well established at 6m as in Fig. S.3 above and will break the numerics. The GVD off situation is shown as the black dotted curve in the Fig. S6. The consequence of this is shown in the inset of this figure and in more detail, in Fig. S.3 above.

Most importantly the physics, with ionization off, remains qualitatively the same, with shock driven harmonic walk-off being the main collapse arresting mechanism for both cases.
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Figure S.5 | Comparative simulations with and without the dispersion poles. Left: resonant part of the susceptibility added to the dry air dispersion. Right: Snapshot of electric fields when maximal intensities are reached at z = 65m and z = 62m, respectively. Simulation in (r,z,t).

References


