Non-linear space-time dynamics of ultrashort wave-packets in water

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We have monitored the space-time transformation of 150-fs pulse, undergoing self-focusing and filamentation in water, by means of the nonlinear gating technique. We have observed that pulse splitting and subsequent recombination apply to axial temporal intensity only, whereas space-integrated pulse profile preserves its original shape.

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Spatial and temporal transformations of wave packets which undergo self-focusing in transparent media with Kerr nonlinearity attract a great deal of interest from the point of view both of fundamental and applied science. Self-focusing of ultrashort light pulses with power well exceeding that for continuous-wave (CW) beam collapse gives rise to a variety of spatial, temporal and interrelated (spatio-temporal, ST) effects and has been a topic of intense theoretical and experimental research\textsuperscript{1} [2, 3, 4, 6, 8]. Discovery of a long range propagation (filamentation) of ultrashort laser pulses in air boosted an interest in ST transformation through filamentation dynamics in gasses\textsuperscript{2} [9, 10], solids\textsuperscript{1} [11] and liquids\textsuperscript{1} [12, 13].

Numerical models of different complexity have been elaborated to study temporal, and, more generally, ST dynamics. Propagation of intense light pulses in media with rather distinct optical properties (amount of normal GVD, nonlinearity, etc; namely gasses, solids and liquids) under various initial conditions (pulse duration, beam width, power, wavelength) has been examined. In spite of different regimes, temporal pulse splitting emerged as a common feature resulting from the interplay between self-focusing, self-phase-modulation and normal dispersion. The open question is then what happens after the pulse splitting occurs. A scenario leading to subsequent multiple splitting has been predicted, but not observed directly (see the discussion in Ref\textsuperscript{14} and references therein). In Ref.\textsuperscript{15} pulse recombination after splitting has been observed and attributed to possible underlined multiple splitting.

More recently, two interpretations have been proposed for understanding filament formation in condensed matter. Some of the present Authors, after having experimentally demonstrated that filaments cannot be treated as a self guided beams, have outlined the key role played by non-linear losses in ruling a spontaneous transformation from gaussian to conical (Bessel-like) wave, in the frame of a CW model\textsuperscript{15}. In this context, the conical wave provides the beam with a large (and not absorbed) power reservoir that keeps “refuelling” the hot central spot and so ensures stationarity, even in presence of non-linear losses. Independently from this work, a second approach has been proposed\textsuperscript{10} sharing with the first the genuine idea of filaments as non-soliton, but conical waves. The important difference is that here the Authors have outlined the key role of chromatic dispersion, fully neglected in previous case, thus interpreting the filament on the basis the dynamics of non-linear X waves\textsuperscript{17, 18, 19}.

The major problem concerning the experimental characterization of the wave packet dynamics is that most of the measurements have been limited either to the pure temporal domain, by on-axis auto or cross correlation technique, or to the pure spatial one, by time integrated CCD-based detection. An example of ST experimental characterization has been that reported in Ref\textsuperscript{20}. The used technique, based on optical polarigraphy, has shown indeed a quite poor resolution, and did not allowed the fine details of the ST structure to be recovered. In a recent work related to the investigation of the nonlinear dynamics of X-waves in quadratic non-linear media some of the present authors have demonstrated a very powerful, high ST resolution 3D-mapping technique, based on the use of ultrafast $\chi(2)$ (sum-frequency, SF) gate\textsuperscript{16, 21}. The approach resembles the cross-correlation measurement to some extent, but instead of recording the space-integrated (or simply on-axis) signal, here the entire space-resolved SF profile is captured. The whole wave-packet is then reconstructed from the assembly of time slices recorded at different delay times.

In this work we apply the same technique to provide a complete characterization of the nonlinear ST dynamics of ultrashort wave-packets in water. The results confirm the occurrence of the pulse recombination after splitting. However the feature of the peripheral part of the beam seems to indicate that observed recombination has to be linked not to multiple splitting, but in contrast to refoc-
FIG. 1: Intensity spatial profiles of different temporal slices of the wave packet, $I(x, y, t, z=22\text{mm})$; $t=0$ at pulse leading edge.

In our experiment, a light filament was excited by launching a focused 0.9-µJ ($P = 5P_{cr}$, where $P_{cr} = 1.15\text{MW}$ is the critical power for CW self-focusing), 150-fs, 527-nm pulses into a syringe shaped water cell, using focusing geometry as described in Ref. [12] (note that the actual focal length is 50cm and not 50mm, as written in Ref. [12] owing to typing mistake). The input pulses were generated by a frequency up-converted optical parametric amplifier (TOPAS, Light Conversion Ltd.), pumped by 100-fs, 800-nm pulses delivered by Ti:Sapphire laser system (Spitfire, Spectra Physics) at 1 kHz repetition rate. A high contrast, 20-fs and 13-µJ probe pulse with wavelength centered at 710 nm was provided by a non-collinear optical parametric amplifier (TOPAS White, Light Conversion Ltd.) pumped by frequency doubled Ti:Sapphire laser pulses. The non-linear gating was performed via SF mixing the probe and the image of the output wave-packet in a thin (20 µm) type I phase-matching β-barium borate crystal under slightly noncollinear geometry. SF signal centered at 302 nm was then imaged onto high dynamic range (16-bit) CCD camera (Andor Technology). We employed a probe pulse with constant intensity over large area (note 1 mm FWHM diameter) and ensured that SF process is performed in the low conversion limit. Then by spanning the delay $t$ using 12 fs steps (see Fig. 1), the complete intensity map $I(x, y, t, z)$ was retrieved, the ultimate temporal resolution being defined by the probe pulse width, by its front steepness and by its intensity contrast.

Fig. 2 illustrates the full characterization of the wave-packet at different $z$, as obtained by changing the length of the water cell. In the right panel we show the time-integrated energy density (fluence) distribution at the output facet of the cell, as recorded by the CCD detector using suitable imaging telescope. In the left panel we plot the reconstructed ST intensity profiles (3D maps). We note that the launched pulse has asymmetric temporal profile (see 3D map at $z=0\text{mm}$ in Fig. 2 with a shoulder on its trailing edge). We expect that this shoulder, because of its low intensity, does not play a major role in the nonlinear dynamics. The results show that a single filament of $\sim 45\mu\text{m}$ FWHM diameter is formed, being surrounded by oscillating annular ring structure. The ST maps show evident pulse splitting on axis at $z=22\text{mm}$, followed by pulse recombination at $z=30\text{mm}$. Out of the beam axis intense modulated wings appear, whose intensity remain peaked at the pulse central temporal
FIG. 3: (a) Normalized intensity (temporal) profiles at the beam center, $I(x=0, y=0, t, z)$ and (b) normalized power profiles, $P(t, z)$, for different propagation length, $z$.

coordinate. This last feature is particularly important if one accounts that the major part of the wave-packet energy is indeed contained into the outer part of the beam. In fact, direct measurements by means of pinholes and stoppers has shown that the filament contains only 20% of the pulse total energy. This feature is clarified also by the results in Fig. 3 where the temporal distribution of the on-axis intensity and of the total power (the space-integrated intensity) are compared. These results show that the widely used term "pulse splitting" is, at least in this case, incorrect. In fact, not the entire wave-packet gets split in the time scale, but only the very central (on-axis) portion of it. Indeed the power distribution remains peaked at the pulse center at any value of $z$. These results let us foresee that pulse recombination could appear because of refocusing of a portion of the power distributed in these intense outer rings. To our opinion, these findings provide an indication of the fact that the wave-packet is attempting a reshaping toward a supported eigenmode of the system, either with dominant Bessel type or X-type profile, owing to the interplay among self-focusing, dispersion and nonlinear losses.

In conclusion, by using high-resolution, high dynamic range detection of the ST intensity profile, we have demonstrated that pulse splitting and recombination of intense, ultra-short pulses in water characterize just the temporal intensity profile at the beam center, but not the pulse as a whole. The entire wave-packet dynamic seems ruled by relevant energy exchange between the beam center and the self-built, slowly decaying, oscillating tails.

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