Spectral "Soliton" Transformation and Four-Wave Mixing under Femtosecond Laser Radiation Filamentation in Molecular Gases

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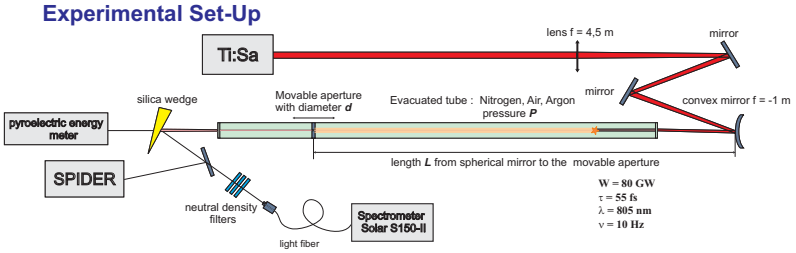
Introduction

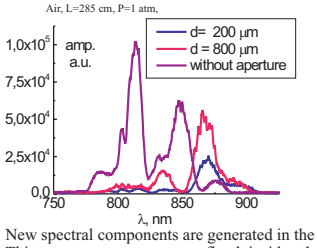
Filaments created during propagation of an ultrashort laser pulses in gases and solids combine high non-linearity with huge intensity and long interaction path. Four-wave mixing, coherent Raman scattering, third harmonic production and others non-linear processes were observed [1, 2, 3]. These studies were made with focused laser beams, when interaction length depends on the Rayleigh length. In this paper we report on thorough investigation of new spectral components formation under filamentation of pre-collimated laser beam in molecular gases. These components arose at the red and blue sides of the initial spectrum and were attributed to the generation of vibrational Raman "spectral solitons" and their cascaded four-wave mixing.

Experimental Set-Up

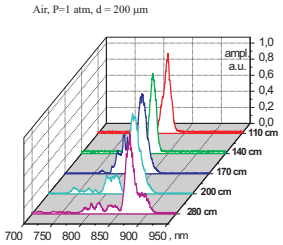
Single filament was created by 80 GW, 55 fs, 805 nm laser pulse at repetition rate of 10 Hz. To launch filamentation at the proper distance laser beam (M^2=1.8) was telescoped down to the diameter of 1.3 mm. The tube 2–4 m long filled with different gases of variable pressure (argon, nitrogen, etc.) was placed 0.5 m away from the telescope exit. We measured energy, temporal and spectral intensity distributions as well as spectral phase (using SPIDER technique) of the radiation passing through the aperture with diameter of 100-1000 μm placed at different positions along the tube.



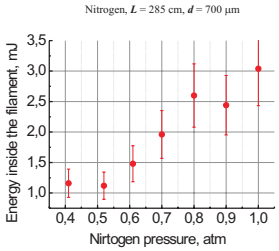
Key Findings:



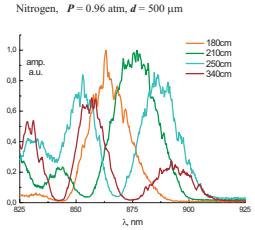
New spectral components are generated in the filament. This new components are confined inside plasma channel of the filament. Spectra measured in different paraxial areas with use of different apertures reveal new spectral components above 830 nm.



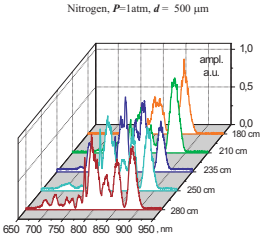
The most intense component experience red shift along the filament. This shift monotonically increases with the distance ***L***

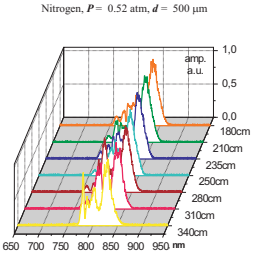


This component carries nearly half of the energy of the initial pulse

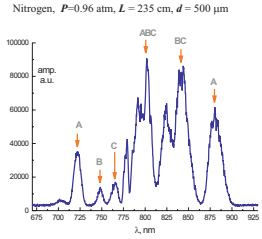


At longer distances this component decays while new component forms showing the same behavior as the first one.

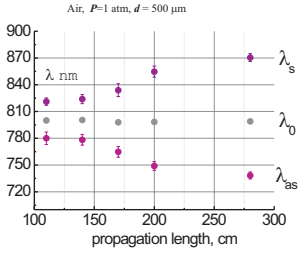




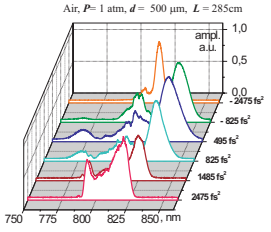
At low gas pressures those components are weak and do not experience red shirt with the propagation along the filament



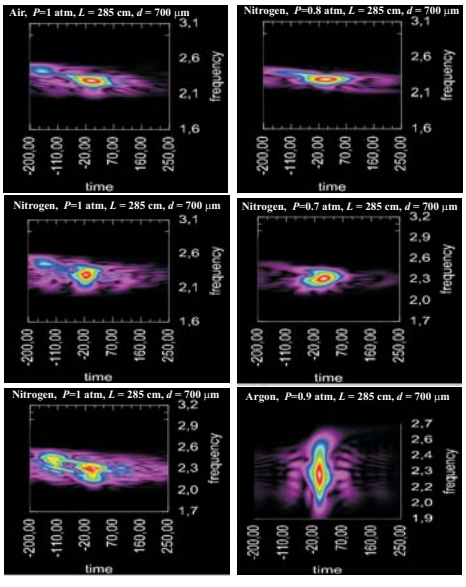
At long distances/high pressures new components appear on the blue wing of the initial spectrum.



These components are linked to the red shifted one and might appear due to four-wave mixing process in the filament core.



Initial pulse chirp greatly affects on the spectral "soliton" behavior. In particular the spectral position of these components can be easily controlled by adjusting the chirp of the initial laser pulse.



SPIDER measurement reveal that this component is confined both in spectral and temporal domains with pulse duration approximately equal to the duration of the initial laser pulse. Fivefold pulse duration compression in Argon at 0.9 atm.

Conclusions

Spectral transformation of collimated radiation undergone filamentation in molecular gases differs drastically from that in noble gases. In particular in molecular gases we did not observe prominent spectral broadening toward blue side in the filament core that is specific for filamentation in noble gases. It is this broadening that is essential for efficient self-compression of femtosecond laser pulses down to few optical cycles. By the contrast, in molecular gases new red shifted components appear due to rotational Raman scattering. Those components are confined inside the filament core while fundamental beam propagates as energy reservoir. Spectral position of those components shifted toward IR along the filament and new components start to grow if the spectral distance between the first component and fundamental frequency becomes large. In molecular gases the newly generated spectral components take part in the four-wave mixing processes. As a result additional components arise also at the blue side of the spectrum. Both the process of Raman shifted components generation and the four-wave mixing process exhibit threshold like behavior depending on gas pressure. SPIDER measurements confirm that new the red shifted component has short less than 30 fs duration thus being confined both in temporal (spatial) and spectral domains.

[1] Couairon A., Mysyrowicz A., Physics Reports. **441**, 47 (2007).
[2] Volkov R.V., Khakhulin D.V., Savel'ev A.B. , Opt. Lett. **33**, 666 (2008).
[3] Chen Y., Theberge F., Marceau C., Xu H., Akozbek N., Kosareva O., Chin S.L., Appl. Phys. B 91, 219–222 (2008).