Spectral “soliton” transformation and four-wave mixing under femtosecond laser radiation filamentation in molecular gases

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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]. 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.

The initial laser beam (55 fs, 5 mJ) had the diameter of 1.3 mm. The filament started after ~1.5 m propagation in air or nitrogen and lasted for 5–6 m. The radiation spectrum inside the filament underwent impressive changes along the propagation path. In particular we observed new spectral component – “spectral soliton” on the red side of the initial spectrum. Spectral shift of this “soliton” increases along the filament up to 100 nm from the fundamental wavelength of 805 nm at the distance of 3–4 m. The same spectral shift was observed recently in [3]. In the filament core (which was extracted by placing small 100–700 μm in diameter diaphragm into the filament) the spectral intensity of this new component was much higher than the one at the fundamental frequency. Energy of red-shifted component amounts to 2–3 mJ in 700 μm aperture, while its duration was estimated as 50–70 fs from the SPIDER measurements. In the Fig.1 one can see how the red side of the spectral intensity change along the filament. With increasing propagation distance the spectral shift becomes larger and the next “soliton” comes into play, while the first one gradually decays. Even more, up to four “solitons” were detected at the filament end. The spectral position of these components can be easily controlled by adjusting the chirp of the initial laser pulse. The gas pressure also drastically affects the observed picture: at nitrogen pressures of 0.6 atm and below spectral soliton has negligible spectral amplitude.

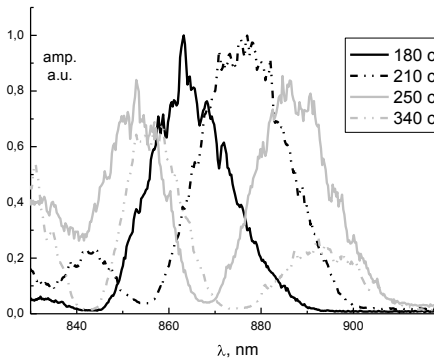

Fig.1. Transformation of the spectral intensity distribution with propagation distance (molecular nitrogen at 0.96 atm, aperture size – 500 μm).

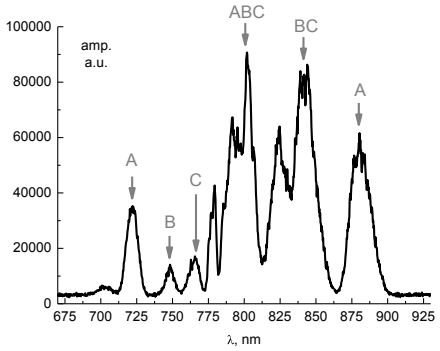

Fig.2. Example of the pulse spectrum inside the filament (molecular nitrogen at 0.96 atm, aperture size – 500 μm, propagation length – 235 cm).

Hence, the 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. Instead in molecular gases the above mentioned 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. This is shown in the Fig.2. Letters mark spectral components linked through the four wave mixing processes. These links preserve with increasing propagation distance.

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[2] Volkov R.V., Khakhulin D.V., Savel’ev A.B. “Four-wave parametric conversion of femtosecond laser pulse in a filament induced in a solid target” Opt. Lett. **33**, 666 (2008).
[3] Chen Y., Theberge F., Marceau C., Xu H., Akozbek N., Kosareva O., Chin S.L., “Observation of filamentation-induced continuous self-frequency down shift in air”, Appl. Phys. B 91, 219–222 (2008).