Long-Wavelength Spectral Shift in Ultraviolet Filament

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The intensity of ∼100 TW/cm^2 [1] in infrared femtosecond filaments in air provides a significant broadening of the pulse spectrum, reaching several microns [2-4]. The intensity in the ultraviolet (UV) filament is at least one order of magnitude less than in the case of the infrared one, and its spectral broadening is insignificant. Experiments on UV filamentation reveal a long-wavelength shift of the spectrum as a whole to the long-wavelength range by several nanometers [5]. This long-wavelength spectral shift was experimentally reproduced in the wide range of pulse durations 450 fs–5 ps and energies 2–7.5 mJ. However, simulations [5] demonstrated the symmetrical spectral broadening. In this work we study experimentally and theoretically the long-wavelength spectral shift under UV filamentation.

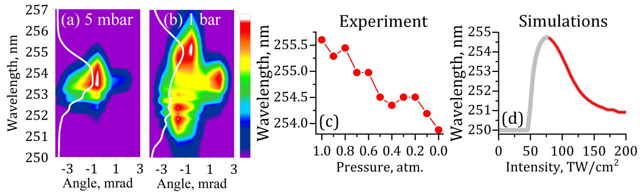

Figure 1: Measured angle-wavelength spectra of femtosecond UV pulse with the energy of 0.2 mJ focused into air-filled cuvette at a pressure of 0.005 atm (a) and 1 atm (b). (d) Spectral shift obtained numerically by filamentation of a pulse with a central wavelength of 250 nm and a duration of 40 fs depending on its intensity. (c) The same shift obtained from experiment depending on the gas pressure in the cuvette

We carried out an experiment in which the pulses centered at ∼250 nm with a duration of ∼100 fs and energy up to 0.2 mJ were focused into the cuvette filled with argon of various pressures. We have measured angle-wavelength distribution for different pressures of gas in cuvette. The increase in pressure results in the monotonic shift of the UV pulse spectrum mass center towards the long wavelengths [Fig. 1(a, b)].

In order to reveal the physical explanation for the long-wavelength shift of the spectrum of ultraviolet filament we calculate the nonlinear response of a gas medium. We numerically solve time-dependent Schrödinger equation with one-dimentional potential well with 3 bound states corresponding to lowest energy levels of argon and laser pulses at duration of 10–80 fs, a central wavelength of 250 nm in a wide range of intensities. In our simulations for the intensities lower than ∼80 TW/cm^2 the spectrum shifts towards the long wavelengths as whole. For higher intensities central wavelength of the spectrum decreases due to plasma effects and returns closer to 250 nm, but stays larger than 250 nm [Fig. 1(d)]. Nonlinear polarization obtained from our simulations delays on the intraperiod timescale relative to the cube of the pump electric field and induces the long-wavelength spectral shift [6]. The increase in pressure leads to the decrease in the pulse intensity in our experimental conditions. Thus, we believe that the increase in the shift of the central wavelength with increasing pressure obtained in the experiment lies in the part of the curve in Fig. 1(c), highlighted in red.

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