Water vapor based dispersion management in a high-pressure gas cell

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Abstract. We use a mixture of a high pressure gas with a minor (about 1 percent) addition of variable density water vapor to control group velocity dispersion of a near-infrared femtosecond pulse and switch it from normal to anomalous regime in the short-wavelength vicinity of the absorption band.

We have shown numerically the formation of typical for anomalous dispersion O-wave frequency-angular spectrum with full suppression of conical emission on the short-wavelength side for 1.3-\(\mu\)m femtosecond pulse filamenting in the mixture of 30-bar nitrogen and 0.4-bar water vapor.

1. Introduction

Femtosecond filament [1] is a source of broadband coherent supercontinuum [2–7]. Simultaneously with frequency spectrum broadening the angular distribution is enriched as well. Filament in air formed by Ti:Sapphire laser pulse emits the short-wavelength supercontinuum in the form of the conical emission [8, 9], i.e. bright rings from blue to red color surrounding a filament. The shorter is the wavelength of the spectrum, the larger is the angle at which this spectral component propagates with respect to the optical axis thus forming the so-called X-wave in the frequency-angular distribution [10,11].

In theoretical works [12, 13], the formation of X-wave was associated with the normal dispersion of transparent media (dry air is the medium with normal dispersion throughout the transparency window). The maxima in the frequency-angular distribution correspond to the condition of quasi-phase matching between emitted continuum and ionization front within the high-power pulse. In the case of anomalous dispersion the frequency-angular distribution remains confined, or O-shaped, as the phase-matching condition is not satisfied [13, 14]. This was experimentally demonstrated in such media as water [15] or sapphire [16] by comparing frequency-angular distributions in the filament produced at different pump wavelengths. These works confirm the role of the material dispersion role in the shape of the frequency-angular spectrum. However, different filament pump wavelengths used in experiments (e.g. from 0.527 to 1.055 \(\mu\)m [15]) alter not only dispersion, but also self-focusing, the order of multiphoton processes, plasma density, filament length etc.
Could we control just the dispersion and switch it from normal to anomalous regime while preserving as many other parameters as possible? One way to control dispersion is to add a resonantly absorbing component to transparent medium with normal dispersion in the wavelength range studied. Close to the absorption band, the dependence of the second-order dispersion coefficient is discontinuous and its value can be varied from the one corresponding to the normal dispersion to the one corresponding to the anomalous dispersion by means of either a little change in pump wavelength or the change in the resonant component concentration. In that case, the changes in the beam diffraction will be negligible and ionization will be similar as the resonant component (i) has a low density and (ii) does not change the order of multiphoton ionization dramatically. For the implementation of this scheme, one can prefer a gaseous media (for precise and fine control of impurity density) and mid-infrared (MIR) laser with the central wavelength being close to vibrational resonances of lightweight molecules (0.1–1 eV, 12–1.2 μm). The lower nonlinearity of gases (as compared to liquid/solid state) and the absence of widely available high-peak-power laser sources in MIR can be compensated by means of the gas pressure. The influence of resonant absorption on MIR filamentation in molecular gases was studied in several experimental and theoretical works [17–26]. However the shape of frequency-angular distribution of supercontinuum during filamentation of femtosecond pulse with the central wavelength in the vicinity of resonance has not been studied yet.

In this work we study the frequency-angular distributions of filamenting pulse in the conditions close to the experiment [26] on MIR high-power femtosecond pulse propagation in the pressurized up to 30 bar nitrogen (N₂) with the addition of water vapor (H₂O) with the pressure smaller than 1 bar. We simulate the filamentation of the pulse with the central wavelength of 1.3 μm close to the resonance at 1.35 μm [27] at different densities of gaseous H₂O. The transition from normal to anomalous dispersion in N₂–H₂O mixture at 1.3 μm with the increase in the water vapor density results in conical emission degradation in the short-wavelength part of spectrum (shorter than 1.3 μm) and formation of O-wave frequency-angular spectral distribution. So, we have shown, that such a typical marker of filamentation in gases as conical emission could be unobservable for the pulse with the central wavelength in the short-wavelength vicinity of the absorption band.

2. Model
We have performed numerical simulations based on the Forward Maxwell Equation (FME) [28, 29] for the time-domain Fourier harmonics \( \hat{E}(\omega,r,z) \) of the electric field \( E(t,r,z) \):

\[
\frac{\partial \hat{E}(\omega)}{\partial z} = -i \left( k(\omega) + \frac{\Delta_{\perp}}{2k(\omega)} \right) \hat{E}(\omega) - \frac{2\pi}{c} \hat{J}(\omega),
\]

where \( \Delta_{\perp} = r^{-1}\partial/\partial r(r \partial/\partial r) \), \( r \) is the transverse coordinate, \( z \) is the propagation distance, and \( c \) is the speed of light in vacuum. The paraxial equation (1) is adequate for the description of the experiment [26] due to loose focusing conditions (numerical aperture of about \( 3 \times 10^{-3} \)). The spatio-temporal grid of our FME solver has \( 2^{16} \) equidistant temporal and \( 1 \times 426 \) nonequidistant spatial nodes with the best resolution of 107 as and 1 μm, respectively. The frequency resolution is 143 GHz.

The first term on the right-hand side of Eq. (1) describes diffraction, dispersion, and resonant absorption through the complex-valued wave number \( k(\omega) = \omega [ n'(\omega) + in''(\omega) ]/c \). The absorption \( n''(\omega) \) and refractive \( n'(\omega) \) indices are connected through the Kramers-Kröning relations [21]:

\[
n'(\omega) = 1 + \mathcal{P}_{N_2} \times (A + B\omega^2) + \mathcal{P}_{H_2O} \times \mathcal{F} \left[ \text{sgn}(t) \times \mathcal{F}^{-1} \left[ in''(\omega) \right] \right].
\]

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Figure 1. Dependence of the (a) absorption coefficient $n''(\lambda)$, (b) refractive index $n'(\lambda)$ and (c) second-order dispersion coefficient $k_2$ on the wavelength $\lambda$ obtained from HITRAN database (red curves) and fitted onto our numerical grid (black curves).

Here $\mathcal{F}$ and $\mathcal{F}^{-1}$ are the operators of direct and inverse Fourier transform respectively; nitrogen $P_{N_2}$ and water vapor $P_{H_2O}$ pressures are given in bars; constants $A = 1.993 \times 10^{-4}$ and $B = 0.558$ as$^2$ determine the Cauchy equation for nitrogen at the pressure 1 bar. The data on absorption index of water vapor $n''(\omega)$ are given from HITRAN database [27]. The absorption spectrum of $H_2O$ in HITRAN has the frequency resolution 300 MHz, and we fit HITRAN data onto our numerical grid with the resolution of 143 GHz so as to preserve the total absorption inside the band, see Fig. 1(a). Therefore, despite the three-order of magnitude difference between HITRAN and our resolution, Eq. (2) ensures adequate representation of the dependence $n'(\omega)$ on our grid at least outside the resonances, see Fig. 1(b). The second-order dispersion coefficient $k_2(\omega) = \text{Re}(\partial^2 k/\partial \omega^2)$, which determines the normal or anomalous regime of dispersion, was calculated both from HITRAN and from our discrete data. The agreement is reasonable as seen in Fig. 1(c).

The nonlinear current in Eq. (1) is $J(t) = J_{\text{free}} + J_{\text{abs}} + \partial P_{\text{inst}}/\partial t + \partial P_{\text{rot}}/\partial t$, where $J_{\text{free}}(t)$ and $J_{\text{abs}}(t)$ are the free electron and absorption currents, respectively, $P_{\text{inst}}(t)$ is the third-order instantaneous polarization of bound electrons, and $P_{\text{rot}}$ is delayed nonlinear response of $N_2$ molecules, see the details in Ref. [25].

3. Results and discussion

The parametric amplifier used in the experiment [26] provides the power higher than 2.5 GW (energy of $\sim 250 \mu J$ and duration lesser than 100 fs) at the wavelength $\sim 1.3 \mu m$. Filamentation of the pulses with the central wavelength of about 1.3 $\mu m$ in dry and moist nitrogen was realized in the 60-cm high-pressure cuvette (up to 100 bar) [26], i.e. under at least twice excess of the pulse peak power over the critical power for self-focusing. In the simulations the nitrogen pressure
was $P_{N_2} = 30 \text{ bar}$, which corresponds to the critical power of $\sim 1 \text{ GW}$. So, the pulse with the energy of 120 $\mu$J and FWHM duration of 60 fs experiences filamentation in such high-pressure gas. These parameters are chosen as initial conditions for numerical integration of Eq. (1). The beam diameter and focusing in the simulations coincide with the experimental ones [26]: 2 mm and 30 cm, respectively. We compare the frequency-angular distributions of the pulse after filamentation for dry ($P_{H_2O} = 0 \text{ bar}$) and moist ($P_{H_2O} = 0.4 \text{ bar}$) nitrogen. The addition of 0.4-bar $H_2O$ to nitrogen ensures relatively strong anomalous dispersion at 1.3$\mu m$ [$k_2 = -44 \text{ fs}^2/\text{cm}$, see Fig. 1(c); cf. with $k_2 = 5 \text{ fs}^2/\text{cm}$ for dry $N_2$]. This value was chosen as a central wavelength in our simulations.

Figure 2 shows the frequency-angular distributions of the pulse at the end of the filament $z = 31 \text{ cm}$ (bottom part under the black line) and at the end of the cuvette $z = 60 \text{ cm}$ (top part above the black line) for (a) dry ($P_{H_2O} = 0 \text{ bar}$) and (b) moist ($P_{H_2O} = 0.4 \text{ bar}$) nitrogen. In both cases the frequency-angular distribution is established quite after the end of filamentation (cf. bottom and top parts in Fig. 2). The strong linear absorption in the moist nitrogen results in deep minimum at the wavelengths of $H_2O$ bands, see the top part in Fig. 2(b). Nevertheless, the resonant absorption does not affect the propagation of the radiation outside the band, and the frequency-angular distributions at the end of the filament and at the end of the cuvette are the same, except in the vicinity of the bands. Therefore, the dispersion does determine their X-[Fig. 2(a)] or O-shape [Fig. 2(b)]. X-shape is typical for filamentation in air and atmospheric density transparent gases. It was observed for 0.8 $\mu m$ radiation in [10,11]. In our simulations we obtained full suppression of the conical emission and transformation of X-shape in dry nitrogen to O-shape in moist nitrogen. Thus, the variation of water vapor density allows one to control the dispersion and, as the result, the frequency-angular distribution during filamentation in gases.

4. Conclusion
We have performed numerical simulations of femtosecond mid-infrared laser pulse filamentation in the high pressure gaseous medium (30 bar $N_2$) with the addition of water vapor (0.4 bar $H_2O$). The central wavelength of our pulse 1.3$\mu m$ is in the short-wavelength vicinity of 1.35-$\mu m$ resonance of water vapor, i.e. in the spectral region where one can control the group velocity dispersion and switch it from normal to anomalous regime by varying $H_2O$ density. For gaseous medium X-wave is a common frequency-angular distribution. We show for the first time that...
the increase in partial water vapor pressure leads to gradual suppression of conical emission and eventually formation of O-wave frequency-angular distribution typical for filamentation in solids and liquids with anomalous dispersion.

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