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Field-free molecular alignment control of filamentation

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Abstract: With an approach of controlling the nonlinearity of medium rather than the light field, the effect of field-free molecular alignment on filamentation and resulting white-light generation is studied. This is done by measuring the rotational wavepacket evolution of nitrogen molecules after passing of a femtosecond laser pump pulse by observing the nonlinear propagation dynamics of a variably delayed filament-producing probe pulse.

Key words: Atomic and molecular physics, ultrafast nonlinear optics, femtosecond phenomena, ultrafast measurements

1. Introduction

Intense pulse propagating in a Kerr medium can experience self-focusing [1,2] with modification of its spectrum due to nonlinear optical processes [3,4]. The increasing utilization of femtosecond laser pulses renewed interest in self-focusing phenomena [5] and development of beam instabilities [6] and small-scale self-focusing, related to filamentation [7,8], white light generation [9–11], and conical emission [12]. White light generation is usually accounted for by self-phase modulation [13,14]; however, stimulated Raman scattering [15], parametric four-wave mixing [16,17], and cascading of these effects as the beam propagates can also contribute to the generation of a supercontinuum.

There are several possibilities investigated to control the spatial and temporal characteristics of the filamentation process and resulting white-light generation [11,18–21]. By changing the transverse spatial phase of an initial Gaussian beam with a computer-generated hologram technique and a spatial light modulator [11] beams with phase discontinuities and steeper intensity gradients were created. Consequently, transverse intensity profiles, development of self-focusing, and resulting locations of filaments producing white-light generation in water were controlled. An intense femtosecond pulse filamentation and propagation were experimentally studied in water for Bessel–Gaussian beams with different numbers of radial modal lobes .[21]. The filament propagation length increased with increasing number of lobes under the conditions of the same peak intensity, pulse duration, and the size of the central peak of the incident beam, suggesting that the radial modal lobes feed energy to the filaments formed by the central intensity peak. It was shown experimentally that two beams interacting in a gas [18,22] or in a liquid [19] can exchange their energy and mutually transform their spectra. Then, in a scheme with two crossing beams, it has been shown that depending on the relative delay between two interacting intense femtosecond laser pulses, white-light generation can be enhanced or suppressed, and the output of white light generation closely correlates with filament formation, which in turn depends on the energetic characteristics of

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the pump beams [20]. In the pump-probe technique, spectral modulation of a probe pulse transmission was used to determine the molecular dynamics of a nonionized impulsively aligned medium. The induced rotational wave packet was mapped as a function of the angular difference between polarization directions of femtosecond pump and probe pulses as well as their relative delay, i.e. "quantum carpet" was measured [23,24].

Filaments have remarkable properties such as pulse self-compression in the time domain, spectrum broadening in the frequency domain, and mode improvement and controllable multifilamentation patterns in the spatial domain, which make them a unique source for many applications. In particular, it is important to study the filamentation dynamics of a femtosecond laser pulse in the quantum wake of molecular alignment, produced by a pump pulse, which opens a new way of its control. In this study, our approach is to use field-free molecular alignment as a means of modification of the ionizing medium rather than the light field, thus achieving control of a filament and resulting white-light generation.

2. Experimental setup

The experiments were carried out with a Ti:sapphire amplified laser system with pulse duration of ∼50 fs, central wavelength of 800 nm, and output energy of 1 mJ per pulse at a 1 kHz repetition rate. The time delay between the linearly polarized pump and probe pulses was precisely adjusted (0.67 fs) using an optical variable time delay with a translational stage controlled by a stepping motor (GTS150, ESP300, Newport). The recombined pump and probe beams were then focused using a lens with a focal length of f = 60 cm into a gas cell of 85 cm, which was filled with pure nitrogen gas with the pressure 4 bar. Experimentally, the nature of the signal of aligned molecules depends on the time delay between the pump and probe laser pulses, and so at the exit of the gas cell the white light signals, spectral profiles, and durations of the pulses were measured simultaneously after the cell as seen in the experimental setup in Figure 1.

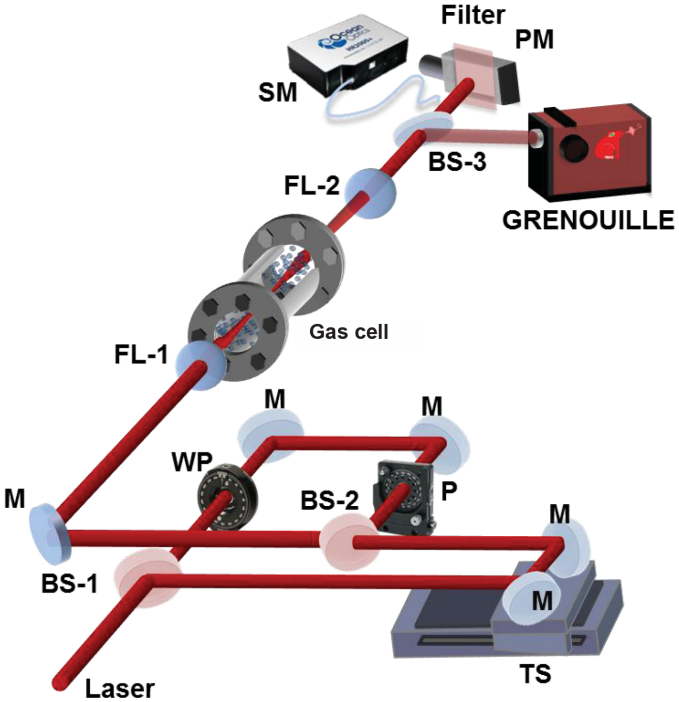

Figure 1. The schematic of the experimental setup. BS: beam-splitters, TS: translational stage, WP: wave plate, P: polarizer, M: flat mirrors, FL: focusing lens, SM: spectrometer, and PM: power meter.

3. Theory

In principle, when any anisotropic molecule is kicked by an intense and sufficiently short laser pulse (its duration is less than the characteristic molecular rotational time), the molecules can be aligned in field-free conditions [25]. In the field-free alignment, the short laser pulse creates a rotational wave packet (coherent superposition of the rotational states) in each molecule that will rephase and dephase in time after the laser pulse has ended. This is due to the quantum beatings of the prepopulated rotational states $|\Phi\rangle$, where J is an integer of the orbital momentum quantum number, and for each J the projection quantum number M takes the values $-J$, $-(J-1)$, ..., 0, ...$(J-1)$, J.

The effective Hamiltonian of a linear symmetric top molecule subjected to linearly polarized laser field of the field of the pump pulse is $H = H_0 - 1/2\left(\Delta\alpha\cos^2\theta + \alpha_\perp\right)\left\langle\varepsilon^2(t)\right\rangle$ [26,27]; $H_0 = B_0 J(J+1) - D_0 J^2(J+1)^2$ is the field-free Hamiltonian with B_0 and D_0 the rovibrational molecular constants, which determine the revival period, $\Delta\alpha$ is the difference between parallel $\alpha_{||}$ and perpendicular $\alpha_\perp$ polarizability components with respect to the molecular axis, $\varepsilon(t)$ is the field of the pump pulse, and θ is the angle between the molecular axis and the pump laser field polarization. During the excitation by the pump pulse, the evolution of the molecular rotational state is $|\Phi\rangle = \sum_{JM} d_{JM}|JM\rangle$, where the expansion coefficients d_{JM} are determined by solving the set of differential equations [28] stemming from the time-dependent Schrödinger equation $i\hbar\frac{\partial}{\partial t}|\Phi\rangle = H|\Phi\rangle$. After extinction of the pump, the population of each rotational state continues to evolve, following the equation with the field-free Hamiltonian.

The degree of molecular alignment is characterized by the quantity $<<\cos^2\theta>>$, which is calculated by a double averaging procedure. Firstly, the Schrödinger equation is solved for each initial molecular state $|\Phi(t=0)\rangle = |J_0M_0\rangle$, and the degree of alignment evolving from a given initial state is characterized by $<\cos^2\theta>_{J_0M_0}(t) = \langle\Phi_{J_0M_0}(t)|\cos^2\theta|\Phi_{J_0M_0}(t)\rangle$. Secondly, an averaging should be performed taking into account that before the interaction the gas ensemble was in thermal equilibrium with Boltzmann distribution at temperature T. In the quantum-mechanical approach, the ensemble is described by a statistical mixture of states $|J_0M_0\rangle$ with different angular momenta, where again $J_0 = 0, 1, 2, ...$ and $M = -J_0, -(J_0-1), ..., 0, ...(J_0-1), J_0$. The probabilities of $|J_0M_0\rangle$ states follow Boltzmann distribution $P_{J_0} \sim (2J_0+1)\exp(-E_{J_0}/kT)$, where E_{J_0} is the rotational energy of $|J_0M_0\rangle$ state and k is the Boltzmann constant. The $(2J_0+1)$ term accounts for the degeneracy within a given J_0 state.

For a complete description, the influence of nuclear spin of atoms constituting the molecule must be taken into account. Consequently, an additional factor g_{J_0} appears, which originates from the nuclear spin statistics in the Boltzmann distribution: $P_{J_0} \sim g_{J_0}(2J_0+1)\exp(-E_{J_0}/kT)$. Finally, the degree of alignment of an ensemble at temperature T can be found by averaging the different states over the Boltzmann distribution including nuclear spin statistics [29,30]:

$$<<\cos^2\theta>>(t) = \frac{\sum_{J_0}\sum_{M_0=-J_0}^{J_0} <\cos^2\theta>_{J_0M_0}(t)\, g_{J_0}\exp\left(-BJ_0(J_0+1)/kT\right)}{\sum_{J_0}\sum_{M_0=-J_0}^{J_0} g_{J_0}\exp\left(-BJ_0(J_0+1)/kT\right)}.$$

For the nitrogen molecule, $g_{J_0} = 6$ for even J's and $g_{J_0} = 3$ for odd J's [31]. The ratio of even to odd states is therefore 2:1 for nitrogen gas.

When molecules with anisotropic polarizability experience the aligning (pump) pulse in a nonadiabatic alignment regime for the case without ionization, nonadiabatic molecular alignment induces a periodic modulation of the refractive index written as $\Delta n(\mathbf{r},t) = \frac{3\pi N}{n_0}\Delta\alpha\left(<<\cos^2\theta>>(\mathbf{r},t) - \frac{1}{3}\right)$ [32], where $<<\cos^2\theta>>(\mathbf{r},t)$ is the thermally averaged alignment expectation value, N is the molecular density, and n_0 is the linear refractive index. The real part of the expressions determines the changes in the speed of light and the imaginary part determines the absorption of light. We note that polarizabilities can be modified when the field becomes sufficiently strong; in particular, absorption can include also the contribution from multiphoton processes. Since $n_0 - 1 = \alpha N/2$, where $n_0 = n_0' + in_0'', \alpha$ is the average polarizability of the gas, changes in both the real and imaginary parts of the refractive index are proportional to the alignment factor, $\Delta n(\mathbf{r},t) = (6\pi)\frac{(n_0-1)}{n_0}\left(\frac{\Delta\alpha}{\alpha}\right)\left(<<\cos^2\theta>>(\mathbf{r},t) - \frac{1}{3}\right)$, and can be used for the alignment monitoring [24].

4. Results and discussion

Alignment produces a noticeable change in filament formation and white-light generation when the amplitude of the change in the refractive index due to alignment $\Delta n_{align} = \Delta n(\mathbf{r},t)$ produced by the pump beam becomes comparable to the refractive index change owing to the optical Kerr effect, $\Delta n_{Kerr} = n_2 I_{probe}$, induced by the probe beam with intensity I_{probe}. With $n_2(N_2) = 2.3\times10^{-19}\mathrm{cm}^2/W$ and $I_{probe} = 5\times10^{13}W/\mathrm{cm}^2$, we obtain $\Delta n = 4.4\times10^{-5}$ for a typical gas pressure of 4 bar. A similar value of the magnitude of the refractive index variations due to alignment Δn_{align} follows from calculations of the pump pulse with intensity $I_{pump} = 10^{13}W/\mathrm{cm}^2$ and duration $\tau \approx 250fs$, which agrees well with experimental parameters. The additional contribution to the refractive index caused by the alignment changes the self-focusing length: it decreases for alignment and increases for antialignment; thus the dynamics of the rotational wave packet changes the filament formation and modulates the white-light generation.

The calculated periodic modulation of the refractive index induced by nonadiabatic molecular alignment and the measured variations in the white-light signal resulting from the nonlinear pulse propagation dynamics as a function of the probe pulse delay are presented in Figure 2. The onset of a balance between self-focusing and defocusing due to plasma formation results in extended filament propagation at high intensities, which is sensitive to rotational modulation of the refractive index. The pump pulse induces the rotational response periodically in time, and so due to quantum-mechanical discreteness of the rotational eigenfrequencies of the molecules the probe laser pulse delayed at the recurrence period experiences a propagating wake of index modification created by the pump pulse. This results in the modification of the filamentation process and the white-light signal. During the experiments, we have the probe pulse field polarization oriented parallel with molecular axes (parallel molecular alignment), and so it is expected that a positive change in the refractive index is induced with molecular alignment, whereas a negative change in the refractive index is induced with molecular antialignment. Due to the beam mode distribution close to Gaussian, it is expected that the alignment is stronger in the beam center. Accordingly, a spatial focusing of the probe beam induced molecular alignment revivals with preferential orientation parallel to the probe polarization direction. Consequently, for additionally focused by parallel alignment probe pulse peak intensity increased and self-focusing developed faster, which resulted in enhanced white-light generation. Indeed, in accordance with the calculations we observed the enhancement of the white-light generation for molecular alignment parallel to the probe pulse polarization direction as seen in Figure 2.

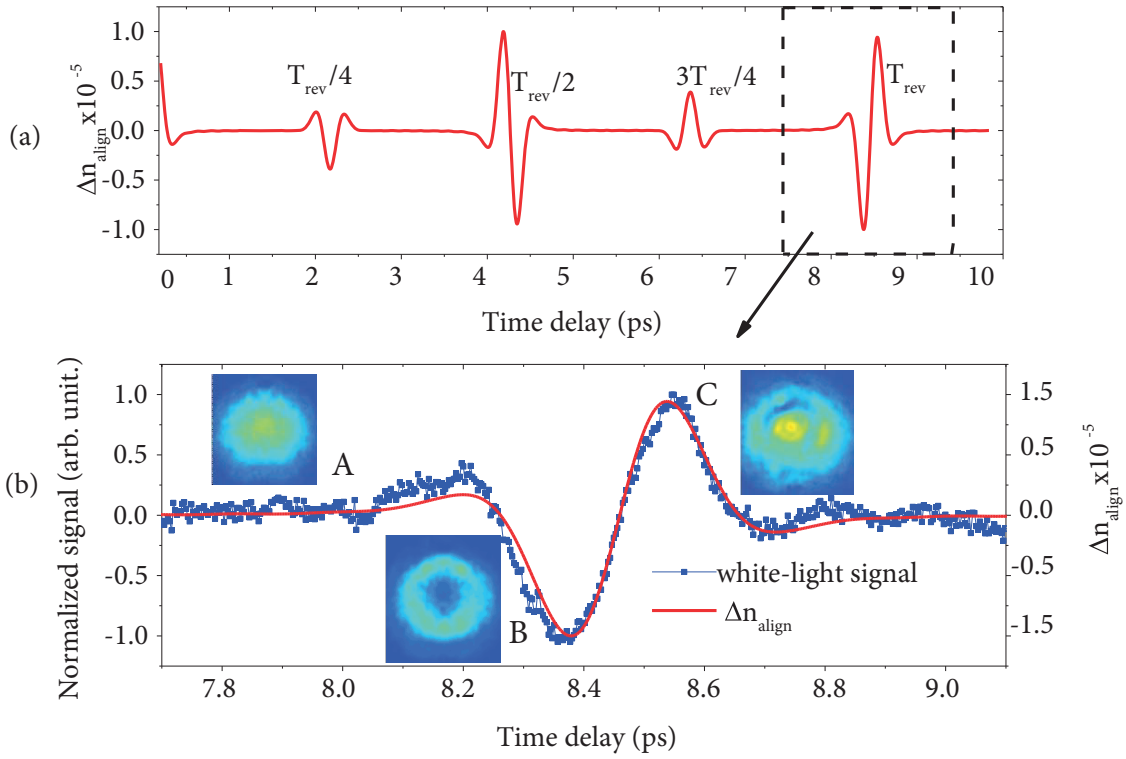

Figure 2. (a) The calculated periodic modulation of the refractive index induced by nonadiabatic molecular alignment, (b) the measured white-light signal around the full revival time of molecular nitrogen. The insets show the probe beam patterns (beam cross section) recorded by using the CCD camera at delay-A (randomly oriented molecules), delay-B (molecular antialignment), and delay-C (molecular alignment).

In Figure 3, the measured spectral modulation is presented around the full revival of molecular nitrogen. The spectral changes are dependent on the revival events induced by molecular alignment for different relative delays between the pump and probe pulses. After the aligning pump pulse propagates in nitrogen gas, the molecules in the gas experience revival events in the nonadiabatic alignment condition. The following filamenting probe pulse undergoes spectral broadening at the maxima of the rotational revival of the molecules, which can be interpreted as a positive change in the refractive index induced by the molecular alignment.

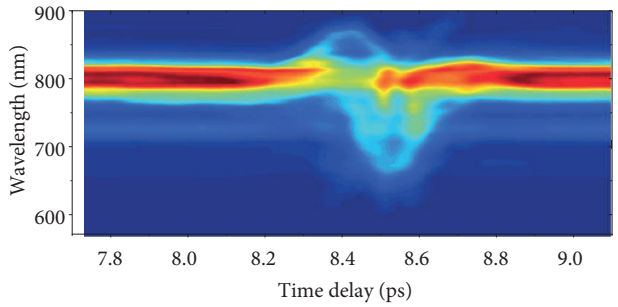

Figure 3. The measured spectral modulation around the full revival time of molecular nitrogen.

As presented in Figure 4, the pulse duration anticorrelates with the variations in white-light generation, doing so in phase with the refractive index modulation, which means that the probe pulse after the interaction

with molecules subjected to the pump pulse is the shortest for aligned molecules and the longest for antialigned molecules. Thus, at our experimental conditions the reshaping of the pulse due to variations in the self-focusing effect is more likely. The change in the pulse duration should be due to the fact that the index of refraction increases the pulse compression and vice versa. It could also be understood that a pulse with a self-modulated phase causes the fields with different phases to overlap, thus resulting in destructive interference that shortens the pulse [33]. Another explanation could be that we had a negatively chirped pulse to begin with (to increase white light signal), and, as the index of refraction increases, the pulse compresses, leading to an intensity increase above the nonaligned case, which results in a greater refractive index.

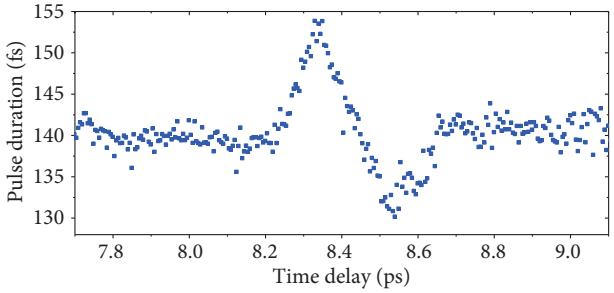

Figure 4. The measured pulse duration induced by the full revival of molecular alignment for nitrogen.

5. Conclusion

The femtosecond filamentation dynamics can be readily controlled via the quantum wake of an aligning (pump) pulse. By delaying a probe pulse, we controlled the nonlinear optical Kerr effect induced by the field-free revivals of the molecular alignment, which in turn affected filamentation and resulting white-light generation. The filamentation dynamics created by the probe pulse then becomes strongly dependent on the revivals experienced by the probe. In this way, several features of filamentation can be directly or indirectly controlled by the field-free revivals of the molecular alignment. Beyond the observations discussed here, the quantum wake-based control of the filamentation length, multiple filamentation dynamics, and white light emission can be employed for atmospheric applications with intense femtosecond filaments.

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