

Elongation of femtosecond filament by molecular alignment in air

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Abstract: We study the influence of the molecular alignment on the plasma channel length of femtosecond filament at 800 nm in air. The filament length is observed to be nearly doubled when the high-energy femtosecond laser pulse is properly tuned to match the perpendicular revivals of the molecular alignment, which is different from the low-energy femtosecond laser pulse where the filament is promoted for parallel molecular alignment revivals. These are understood to be the loosened or tightened focusing condition of the propagation of the femtosecond laser pulse by the cross-(de)focusing effect from the prealigned molecules.

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1. Introduction

The self-guided propagation of infrared femtosecond laser pulses in transparent optical nonlinear media has sparked many interests and important applications [1–7] such as atmospheric remote sensing [4,5], trigger and guide electric discharges [6], and remote terahertz source [7]. For most applications, a long-distance filament is desired, and it was shown that, for constant input pulse energy, the length of the filament could be longest for an optimum pulse duration [8], which was also shown to be sensitive to the temporal chirp [9] of femtosecond laser pulses. For molecular gas, we have recently numerically demonstrated [10] that the length of the filament could be significantly increased by properly tuning the femtosecond laser pulse to match the molecular alignment revivals after an impulsive excitation of an ultrashort pump pulse. The controllable orientation of molecules with respect to the field polarization [11] provides us an additional degree of freedom to control the filament propagation and interaction, such as filament formation [12,13], filament induced supercontinuum generation [14,15], couplings of crossly overlapped filaments [16], and kicking of intense filaments [17].

In this paper, we experimentally demonstrate that the length of the filament channel could be significantly increased by propagating the energetic femtosecond laser pulse in prealigned diatomic molecules in air. When the high-energy femtosecond pulse was properly tuned to match the perpendicular revivals of the molecular alignment, the filament length was nearly doubled with respect to the randomly orientated molecules. The basic underlying physics is that the cross-(de)focusing effect of the prealigned molecules modulated the focusing condition and hence the filament dynamics.

2. Experiment setup

Our experiments, as schematically shown in Fig. 1, were carried out with an amplified Ti:Sapphire laser system (1 kHz, 800 nm, 35 fs). The original beam diameter of the pump and probe pulses were 12 mm ($1/e^2$), and only the p-polarized pump beam was down-collimated with a minification of 2:1 to achieve a larger interaction volume, which were recombined collinearly with the s-polarized probe pulse by a thin film polarizer (TFP). The time delay between the pump and probe pulses was tuned by using a motorizing translation in the probe arm. The final pulse-energies after the TFP for the pump and probe pulses were measured to be 1.43 and 2.26 mJ, which were focused with two separated lenses of $f = 1$ m and $f = 2$ m, respectively. These two lenses were positioned properly with respect to the TFP so that the pump filament was located at the onset of the probe one to control the onset of the probe filament through the molecular alignment. By using a half wave plate just before the TFP in the pump arm, the pulse energy of the pump could be adjusted from 0.1 to 1.43 mJ. We determined the filament channel length by measuring the transverse fluorescence emission perpendicular to the filament propagation. Our detection setup comprised of a fused-silica lens, a slit, and a photomultiplier tube (PMT). In order to eliminate the influence of the input pulse and supercontinuum emission, a dielectric mirror (HR 800 nm @ 0°) and a bandpass filter (UG11, transmission 200–400 nm) were placed just in front of the PMT. The whole detection setup was installed on a breadboard mounted on a motorizing translation stage, which could be moved along the filament propagation direction. The chopper frequency signal was used to provide the reference input for the lock-in amplifier, and only the output signal from the PMT at the chopping frequency was processed by the lock-in amplifier. Hence, by setting a chopper in the pump or probe arm, only the fluorescence emission from the pump or probe pulse was measured, respectively.

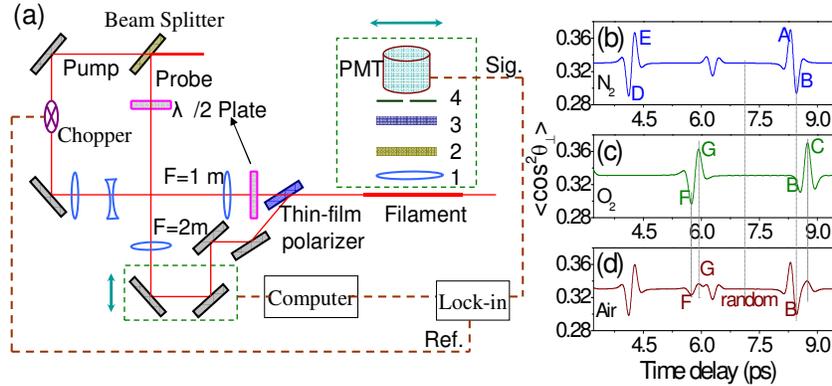


Fig. 1. (a) Schematic illustration of the experimental setup, 1: lens ($f = 5$ cm, $D = 5$ cm), 2: dielectric mirror, 3: UG11, 4: slit. The calculated molecular alignment metric $\langle\langle\cos^2\theta_{\perp}\rangle\rangle$ versus pump-probe delay in (b) N_2 , (c) O_2 , and (d) air.

3. Results and discussions

Figure 2 shows the measured filament channels of the probe pulse when it was tuned to experience various molecular alignment revivals. The zero position of the propagation axis in the figure was defined to be at the distance of 80 cm after the TFP and the termination of the filament [3] was defined to be the position with fluorescence intensity smaller than 2.9% of the plateau region in this paper. The filament channels for the pump and probe pulses when they propagated alone in the air are also shown in Fig. 2.

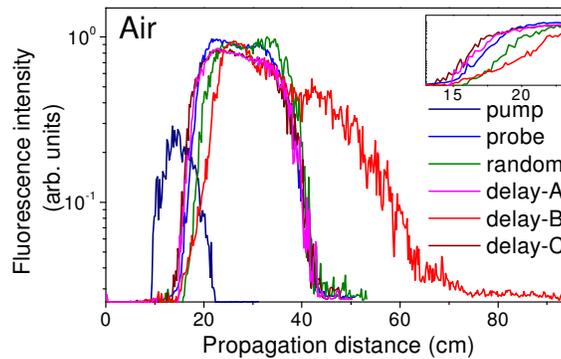


Fig. 2. The measured filament channels of the probe pulse when it is turned to various molecular alignment revivals as labeled in Fig. 1. The filament channels of the pump (navy curve) and probe (blue curve) pulses when they propagate alone are also shown.

We calculated the corresponding molecular alignment revival $\langle\langle\cos^2\theta_{\perp}\rangle\rangle$ in N_2 , O_2 and air by considering the coherent superposition of the molecular rotational wave packets excited by the pump pulse as described in Ref [18], which were respectively shown in Figs. 1(b), 1(c) and 1(d), and θ_{\perp} is the angle between the molecular axis and the probe polarization. The revival signals $\langle\langle\cos^2\theta_{\perp}\rangle\rangle$ greater and smaller than $1/3$ account for, respectively, the transient molecular orientations parallel and perpendicular to the field polarization of the probe pulse, which equals to $1/3$ for randomly orientated molecules.

We then turned on both the pump and probe pulses simultaneously and investigated the influence of the molecular alignment induced by the pump pulse on the probe filament. The plasma defocusing effect by the pump filament was firstly observed for the case of randomly orientated molecules. By considering the Gaussian-shaped transverse profile of the pump beam, the decrease of the refractive index originated from the ionization-induced plasma was

more significant at the beam center than that at its periphery, which acted as a negative lens to defocus the probe beam. As shown in the inset of Fig. 2, comparing to the case of probe pulse alone, the onset position of the probe filament moved forward (along the beam propagation direction) even for the randomly orientated molecules by the pump pulse. The molecular alignment further modulated the probe filament when it was tuned to the molecular alignment revivals. The alignment-induced change of the refractive index increased or decreased when the molecules were orientated parallel or perpendicular to the field polarization [12–15], respectively. Since the molecular alignment degree was proportional to the pump intensity, similar to the plasma effect by considering the Gaussian-shaped transverse distribution of the pump beam, an additional cross-(de)focusing effect was introduced to the probe pulse and hence determined the probe filament. As shown in the inset of Fig. 2, as compared to the case of probe pulse alone or randomly orientated molecules, the onset positions of the probe filament were moved forward and backward for the perpendicular and parallel revivals of the molecular alignment as labeled in Fig. 1. It also indicates a more significant influence of the molecular alignment than the plasma effect on the onset of filament. The molecular alignment not only influenced the onset of the filament, but also dominated the succeeding filament dynamics of the probe pulse. As we can see from Fig. 2, the filament of the probe pulse was terminated at the position of 44 cm for randomly orientated molecules. However, at delay-B, the fluorescence intensity of the probe pulse at the position of 44 cm was increased about 15 times as compared with that of random orientation and the filament of the probe pulse was terminated at the position of 73 cm, which indicated that the filament length of the probe pulse was nearly doubled. For tightly focused high-energy femtosecond laser pulse, the multi-photon ionization of the molecular gas competed with the long-distance filament propagation in a way similar to the competition between the laser breakdown and supercontinuum generation [19], where the supercontinuum generation was promoted with suppressed laser breakdown when a lens of longer focusing length was used. The additional cross-defocusing from the perpendicular alignment revival weakened the high-order nonlinear processes and ionization losses. Actually, in our experiments, supercontinuum generation was observed to be significantly improved at delay-B, which was consistent with previous work [19]. The filament elongation was also observed at multiple revivals determined by the molecular constants, which attenuated with decreased molecular alignment degree along the time delays.

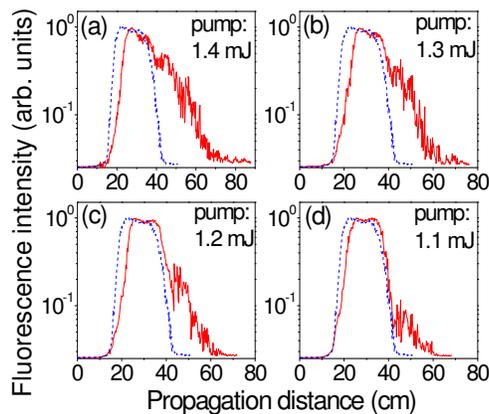


Fig. 3. The measured probe filament channels with different pump intensities in air for molecular alignment revival at delay-B. The dashed and solid curves stand for the cases when the pump pulse is turned off and turned on, respectively.

The cross-(de)focusing effect from the molecular alignment could be adjusted by changing the intensity of the pump pulse, which could be used to continuously control the probe filament. As shown in Fig. 3, for the perpendicular alignment revival at delay-B, the length of the probe filament channel decreased gradually as the decreasing pump energy

from 1.4 to 1.1 mJ. According to the decreased cross-defocusing effect from the perpendicularly orientated molecules, the onset positions of the probe filament moved backward as the pump intensity decreased. No noticeable elongation of the probe filament was observed when the pump energy was lower than 1.0 mJ for the negligible influence of the molecular alignment.

In the above discussion, the observed filament elongation was a combined contribution of molecular N_2 and O_2 in air, especially at delay-B where the longest filament channel was observed. In order to individually show the molecular alignment effect of N_2 and O_2 on the filament length control, we then tuned the probe pulse to the half revivals of molecular N_2 and O_2 in air around 4.2 and 5.8 ps, respectively. The measured probe filaments were shown in Fig. 4. Similar to the above observations around 8.0~9.0 ps, the movement of the onset positions of the probe filament due to the cross-(de)focusing effect from the plasma and molecular alignment revivals were also observed. As compared to molecular O_2 , the change of the onset position of the probe filament was more significant around the molecular alignment of N_2 . This was consistent with the fact that much more N_2 (~80%) are contained than O_2 (~20%) in air, which therefore influenced the probe filament more drastically. Correspondingly, the probe filament elongation was more significant around the perpendicular alignment revival of N_2 at delay-D than that of O_2 at delay-F, which both were shorter than that at delay-B (as shown in Fig. 2) where the molecular alignment revival of N_2 and O_2 cooperated together to dominate the probe filament.

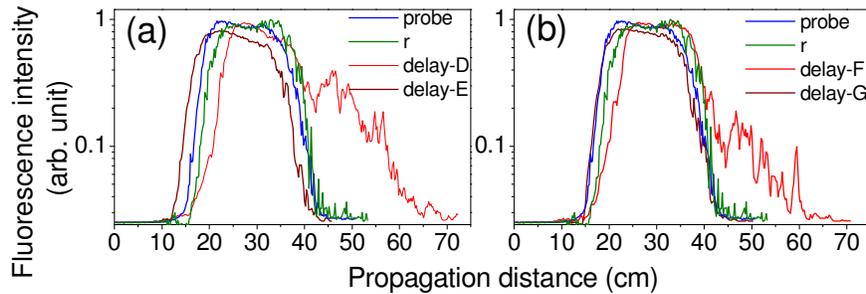


Fig. 4. The measured probe filament channel when it is tuned to match the individual revivals of molecular (a) N_2 and (b) O_2 in air, and random orientation (labeled as r). The energies of the pump and probe pulses were 1.43 and 2.26 mJ, respectively

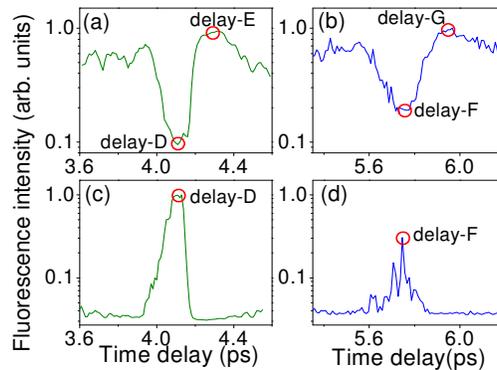


Fig. 5. The measured fluorescence intensity variations of the probe filament in air at the propagation distances of (a, b) 20, and (c, d) 50 cm as a function of the time delay.

Figure 5 shows the measured fluorescence intensity variations of the probe filament at different propagation distances as a function of the pump-probe time delay. As shown in Figs. 5(a) and 5(b) around the onset positions of the probe filament at a propagation distance of 20 cm, the fluorescence intensity decreased at delays D and F, and increased at delays E

and G, which were in accordance with the forward and backward movements of the onset positions of the probe filament (Fig. 4). As shown in Figs. 5(c) and 5(d) at a propagation distance of 50 cm, the fluorescence intensity of the probe filament channel was increased only at delays D and F, indicating that the probe filament channels were significantly elongated only at these time delays with perpendicular orientated N_2/O_2 molecules.

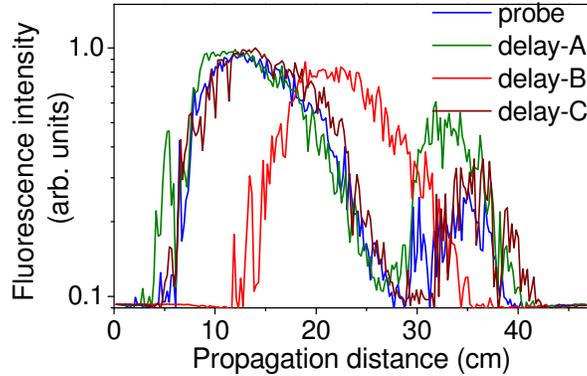


Fig. 6. The measured probe filament channels in air at various molecular alignment revivals when the energy of the probe pulse was reduced to 1.3 mJ.

For high-energy femtosecond laser pulse, it is clear that the filament can be promoted by using the cross-defocusing effect from the perpendicular revivals of the molecular alignment. However, when we reduced the energy of the probe pulse, as shown in Fig. 6 for a probe pulse of 1.3 mJ, the filament was promoted only for the parallel revivals of the molecular alignment. The onset positions of the probe filament were moved forward (delay-B) and backward (delay-A). Differing from the high-energy case, for the perpendicular revival of the prealigned molecules at delay-B, the length of the probe filament was dramatically decreased for the low-energy probe pulse, which was consistent with the prediction in Ref [10]. For the low-energy probe pulse, the self-focusing is not so tight as compared to the high-energy case where the filamentation drastically competed with other high-order nonlinear processes [19]. As demonstrated for controllable supercontinuum generation by filament [14,15], the cross focusing effect from the parallel orientated molecules enhanced the self-phase modulation and self-steepening effect to promote the probe filament, which were correspondingly suppressed with a shortened filament length for the cross-defocusing effect when it was tuned to match the perpendicular revivals of the molecular alignment.

4. Conclusion

In summary, we have demonstrated that the femtosecond filament can be controlled by using the impulsive molecular alignment of air. The filaments of the high-energy and low-energy femtosecond pulses were promoted respectively by the perpendicular and parallel revivals of the prealigned molecules, which correspondingly modulated the focusing condition of the probe pulse and the succeeding filamentation dynamics. This provides us an efficient approach to control the filament length, and is therefore significant for filamentation based atmospheric applications [2–7].

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