SCIENCE CHINA
Physics, Mechanics & Astronomy

• Research Paper •
Femtochemistry, Femtobiology and Femtophysics

June 2010 Vol.53 No.6: 1036–1039
doi: 10.1007/s11433-010-3217-9

Molecular wakes for ultrashort laser pulses

CAI Hua, WU Jian*, LU PeiFen, TONG YuQi, YANG Xuan, BAI XueShi, LI Hao, PAN HaiFeng & ZENG HePing*

State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, China

Received April 2, 2010; accepted April 21, 2010; published online May 14, 2010

The molecular wake-assisted interaction between two collinear femotosecond laser pulses is investigated in air, which leads to the generation of a controllable 1.8 mJ super-continuum pulse with an elongated self-guided channel due to the cross-phase modulation of the impulsively aligned diatomic molecules in air. For two parallel launched femtosecond laser pulses with a certain spatial separation, controllable attraction and repulsion of the pulses are observed due to the counter-balance among molecular wakes, Kerr and plasma effects, where the molecular wakes show a longer interaction distance than the others to control the propagation of the intense ultrashort laser pulses.

molecular wakes, ultrashort laser pulse interaction, super-continuum generation

PACS: 33.80.-b, 42.65.Jx, 42.65.Re, 42.50.Wk

1 Introduction

The interaction between ultrashort laser pulses has been extensively studied for their propagation dynamics control [1–3] to facilitate the atmosphere detection [4], lighting control [5] and remote terahertz emission [6]. The nonlinear processes in the self-guided filament channel could lead to the generation of super-continuum (SC) pulse with broadband spectrum, which has stimulated a lot of promising applications such as white-light lidar [7], few-cycle pulse generation [8] and time-resolved spectroscopy [9]. By considering the quantum wakes of the pre-aligned molecules, the propagation of ultrashort laser pulses in molecular gases could be much more interesting than that in atomic gases. Recently, the additional cross-phase modulation (XPM) of the molecular wakes [10] has been demonstrated to modulate the spectrum of ultrashort laser pulse [11,12], self-compress femtosecond laser pulses to the few-cycle oscillations [13], generate the X-waves with controllable shock edges [14], promote the SC generation [15,16], and so forth.

In this work, we experimentally demonstrate the mulecular wake assisted strong interaction between two ultrashort laser pulses ensuring a high-energy SC pulse generation with an elongated self-guided channel for the collinear scheme, and controllable attraction and repulsion of the intense ultrashort laser pulses for the parallel launched scheme.

2 Experimental results and discussion

Figure 1 shows the calculated molecular alignment signal of air, and the diatomic molecules in air were first orientated parallel ($\langle\langle\cos^2\theta_\perp\rangle\rangle > 1/3$) and then perpendicular ($\langle\langle\cos^2\theta_\perp\rangle\rangle < 1/3$) to the probe polarization, when the pump-probe delay was tuned from 8.0 ps to 9.0 ps. Here, $\theta_\perp$ represents the angle between the molecular axis and the polarization of the probe pulse. Since the molecular alignment degree is proportional to the laser intensity, the molecular alignment degree in the beam center is stronger than that in periphery by considering the Gaussian-shaped pump beam profile.

*Corresponding author (email: jwu@phy.ecnu.edu.cn, hpzeng@phy.ecnu.edu.cn)

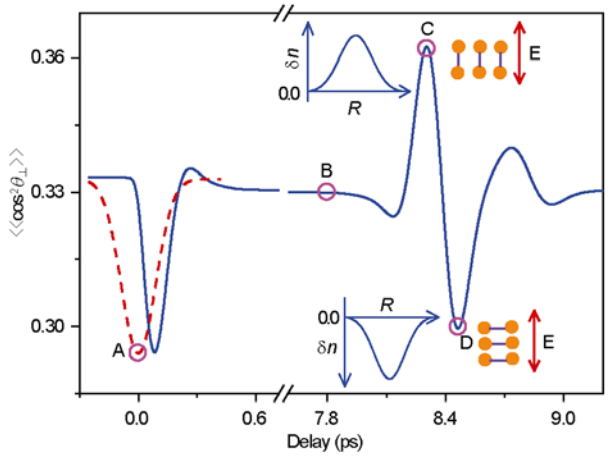

Figure 1 The calculated molecular alignment signal of air. The red dashed curve represents the pump pulse envelope. The insets show the molecular axis orientation and the spatial profile of refractive index modulation for delays C and D.

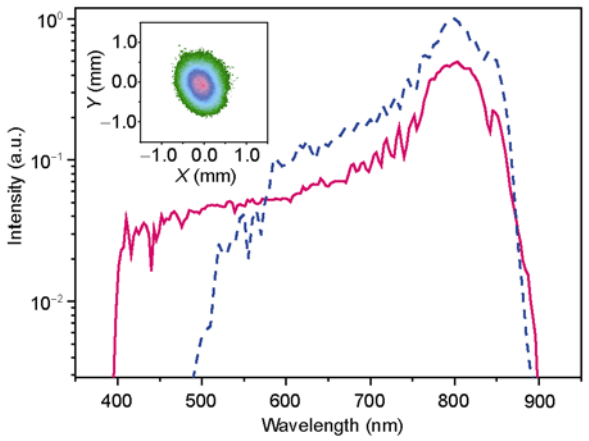

Figure 2 The measured output spectra of the femtosecond laser pulse when its time delay is tuned to delay-D (solid curve) and random orientation (dashed curve) of the molecules in air. The inset is the output spatial profile of the high-energy SC pulse for delay-D.

Therefore, as shown in the inset of Figure 1, the parallel or perpendicular orientated molecules induce the profile of increased or decreased refractive index, respectively, which lead to an additional focusing or defocusing effect. The succeeding propagation dynamics of the probe pulse would be dominated by the combination of the molecular alignment and linear/nonlinear processes in air.

We performed our experiment [17] with high–energy femtosecond laser pulses of 2.7 mJ for controllable SC generation, which showed fairly different features as compared with that of low-energy pulses [15,16]. A self-guided channel of 24 cm in length was observed for the high-energy probe pulse, which collinearly propagated with the pump pulse of 1.4 mJ. The output spectra and spatial profiles of the probe pulse at various pump-probe delays in Figure 1 were respectively measured with a PMT-based spectrometer and a CCD. The broadest and narrowest spectra of the probe pulse were observed for the perpendicularly (delay-D) and parallel (delay-C) orientated molecules, respectively. Figure 2 shows the extended spectrum of the probe pulse for delay-D, where the multi-filaments were suppressed to a single core. The perpendicular revival of the molecular wake induced defocusing effect to loosen the focusing condition of the high-energy probe pulse, which decreased the threshold of SC generation while increased the threshold of other high-order nonlinear processes such as optical breakdown. Meanwhile, the ionization losses were also decreased for the reduced ionization cross-section at the perpendicular revival of the molecular alignment [18]. Eventually, a 1.8 mJ SC pulse with a broadband spectrum ranging from 400 nm to 900 nm was produced in a single spatial core at delay-D for the perpendicularly orientated diatomic molecules in air.

We also measured the self-guided channel length of the probe pulse by detecting the transverse fluorescence intensity of the plasma column versus the propagation distance [19]. The self-guided channel length of the probe pulse was nearly doubled (43 cm) for delay-D as compared with that in randomly orientated molecules (delay-B). It is also the molecular alignment-induced defocusing effect that caused the elongation of the self-guided channel length, which was consistent with the generation of energetic SC pulse by the molecular wakes. As shown in Figure 3, the self-guided channel length of the probe pulse for delay-D decreased with the decreasing pump pulse energy, resulting from the decreased degree of the molecular wake, which was actually proportional to the pump intensity.

The interaction between two parallel launched ultrashort laser pulses were also studied by considering the molecular wakes as well as the Kerr and plasma effects [20–22]. The refractive index modulations from the self-phase modulation (SPM) and XPM of the orthogonally polarized pump

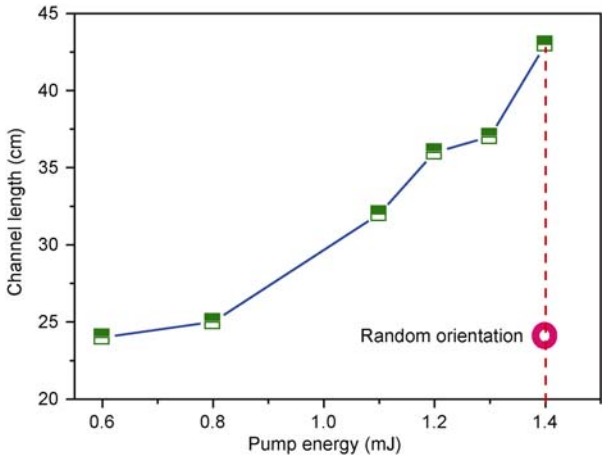

Figure 3 The self-guided channel length of the probe pulse for delay-D with different pump intensities. The pink round point represents the self-guided channel length of the probe pulse for randomly orientated molecules.

and probe pulses were calculated as shown in Figure 4(a). The overlapped profiles of the refractive index (Figure 4(a)) would induce the attraction between the two parallel launched pulses each other. The plasma effect would decrease the refractive index (blue dashed curve in Figure 4(b)) and influenced the propagation of probe pulse (color mapping curve in Figure 4(b)). The Kerr effect could be observed only during the pulse duration, and the plasma effect could exist with a typical lifetime up to several nanoseconds after the pulse excitation, while the molecular wakes could be observed periodically during the specific revival durations of the impulsively excited rotational molecular wave-packets.

Experimentally, we recorded the ionization-induced fluorescence of the two self-guided femtosecond laser pulses with various initial separations and at different pump-probe delays to distinguish various interaction mechanisms. The fluorescence images and intensity profiles of the pump and probe filaments for different pump-probe delays (Figure 1) were shown in Figure 5, and the initial separation of the two filaments was 70 μm. As shown in Figure 5(a), the two self-guided ultrashort laser pulses fused together when they were temporally synchronized at delay-A. The polarizations of the pump and probe pulses were orthogonal to each other, so this significant filament fusion should be induced by the Kerr effect of the SPM and XPM of the two pulses. Since the core of the self-guided pump pulse has the maximum degree of molecular wakes, which decreased along the transverse distance from the core, the probe filament experienced a gradual modification of the refractive index along the transverse direction. The refractive index change in the pump core was positive and negative for the parallel and perpendicularly orientated molecules, respectively. Therefore, when the molecules in the pump waveguide were orientated parallel to the probe polarization for delay-C, the probe pulse was attracted with a decreased separation (Figure 5(c)) as compared with the randomly orientated case for delay-B in Figure 5(b). The fluorescence intensity of the probe pulse was also increased for delay-C as shown in Figure 5(c) due to the orientation-dependent ionization-section [18]. However, when the time delay of the probe pulse was tuned to delay-D matching

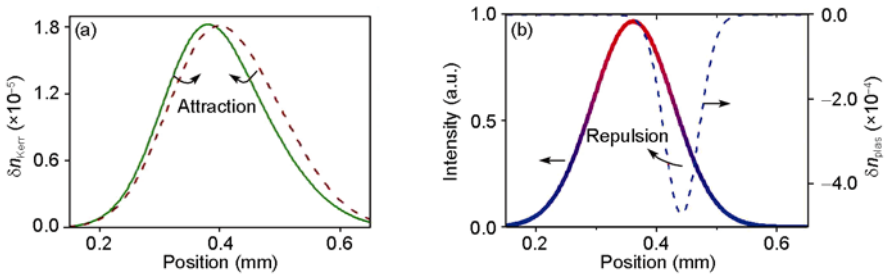

Figure 4 The calculated profile of refractive index modulation due to Kerr effect (δn_{Kerr}) (a) and the calculated profile of refractive index modulation due to plasma (δn_{plas}) produced by the pump pulse (blue dashed curve in (b)) with an initial pump-probe separation of 70 μm. The color mapping curve in (b) represents the intensity profile of the probe filament deduced from the measured fluorescence profile.

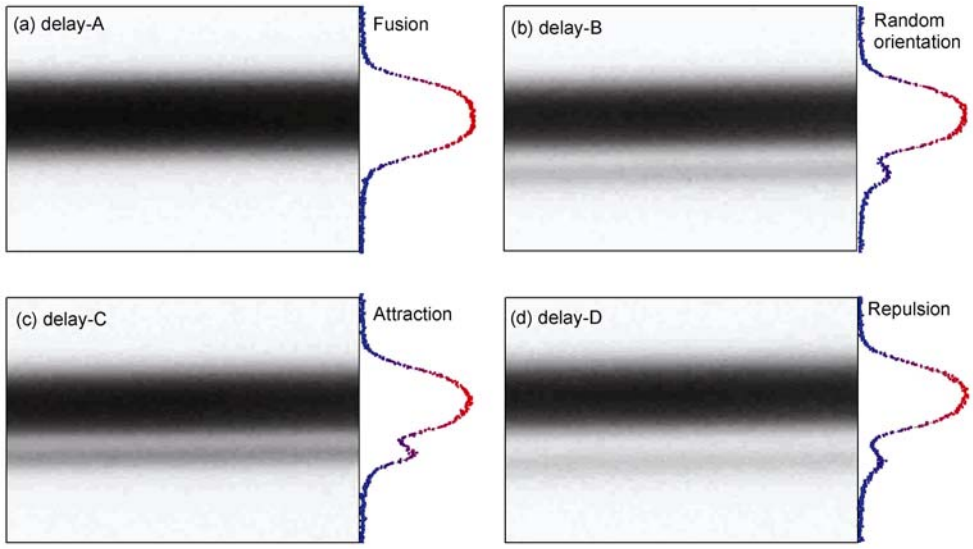

Figure 5 The measured fluorescence images and intensity profiles of the pump and probe pulses at various pump-probe delays with an initial separation of 70 μm.

the perpendicular revival of the molecular wake, it was correspondingly repulsed (Figure 5(d)), where the fluorescence intensity of the probe pulse also decreased by the reduced ionization probability for the perpendicularly orientated molecules [18].

The influence of Kerr nonlinearity and molecular wakes were also investigated with an initial separation of 140 μm. No fusion of the ultrashort laser pulses was observed around zero time delay. However, the attraction and repulsion by molecular wakes could be still observed similar to that of the initial separation of 70 μm. This indicates that the interaction range of the molecular wakes was longer than that of the Kerr effect. The plasma defocusing also led to the repulsion of the ultrashort laser pulses due to the reduction of the refractive index. A larger separation between the ultrashort laser pulses was observed when the more intense pump pulse propagated ahead with a larger plasma density as compared with that of the probe pulse with modest field intensity propagated ahead. Similar to the Kerr effect, the plasma defocusing showed a shorter interaction range than that of the molecular wakes. Finally, we emphasize that the molecular wake assisted interaction between ultrashort laser pulses could also be achieved for pulses at different wavelengths [22], and the modulation of the laser pulse spectrum was clearly observed.

3 Conclusions

In summary, we have demonstrated that the propagation dynamics of intense ultrashort laser pulses in molecular gases can be manipulated by using the molecular wakes, which is actually field-free achievable and expected to stimulate promising applications in various fields.

This work was partly funded by the National Natural Science Foundation of China (Grant Nos. 10525416, 10804032, and 10990101), the National Basic Research Program of China (Grant No. 2006CB806005), Projects from Shanghai Science and Technology Commission (Grant Nos. 08ZR1407100 and 09QA1402000), and Shanghai Educational Development Foundation (Grant No. 2008CG29).

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