Molecular-alignment-assisted high-energy supercontinuum pulse generation in air

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We demonstrate controllable generation of a high-energy supercontinuum pulse of 1.8 mJ through femtosecond filamentation in prealigned diatomic molecules in air. For high-energy femtosecond laser pulses of linear polarization, the focusing condition is loosened by the cross-defocusing effect from the perpendicularly orientated molecules, which promotes the high-energy supercontinuum generation with suppressed multifilamentation and decreased multiphoton ionization loss due to the reduced ionization probability.

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High-energy ultrashort laser pulses propagating in transparent nonlinear optical media can lead to the generation of supercontinua (SC) [1–3] and have stimulated a lot of promising applications [4–6]. By transmitting femtosecond laser pulses in gas-filled hollow fibers [7] or the free space of noble gases assisted with self-guided filaments [4], the pulse energy of the generated SC was typically limited to submillijoules. Recently, SC with pulse energy of 1.2 mJ was demonstrated by using elliptically polarized femtosecond filaments in high-pressure gases [8]. Similar SC pulse energy was achieved in a single filament core by using prealigned molecules at a gas pressure of two atmospheres [9,10], where the cross-focusing effect induced by parallel orientated molecules and the subsequently enhanced self-steepening effect played important roles to broaden the pulse spectrum. The self-guided nonlinear propagation of intense ultrashort pulses in impulsively aligned molecules [11] could be dramatically influenced [12–16].

In this Letter, we demonstrate that the interplay of alignment-induced cross-(de)focusing, Kerr self-focusing, and plasma defocusing in high-energy ultrashort filaments exhibits quite different features as compared with that in low-energy filaments. We experimentally show that the broadband SC pulse generation with an output pulse energy up to 1.8 mJ and an spectrum extension from 400 to 900 nm can be assisted by the cross-defocusing effect from the perpendicularly prealigned molecules, where the multifilamentation is meanwhile suppressed. We emphasize that the underlying physics of the energetic broadband SC generation relies on loosening the focusing condition of high-energy pulses by cross defocusing from perpendicularly prealigned molecules, rather than enhancing the focusing of low-energy pulses by cross focusing from parallel prealigned molecules [9].

Experimentally, the output of an amplified Ti:sapphire laser system (1 kHz/35 fs/800 nm) was first split in two parts, with one of them as the pump for molecular alignment and the other as the probe for the SC generation, whose polarizations were orthogonal to each other. Different from the previous experimental setup in [9], here our experiments were performed in ambient air, and only the pump pulse was down collimated with a minification of 2:1. The pump and probe beams were focused with lenses of $f=1$ and 2 m, respectively. The final pulse energies for the pump and probe pulses were measured to be 1.4 and 2.7 mJ, which led to a probe filament about 20 cm in air. At the end of the filament, the *s*-polarized probe pulse was separated from the *p*-polarized pump pulse by sequentially reflecting them with three substrates at grazing angles, which eliminated the *p*-polarized pump dramatically as compared with the *s*-polarized probe with an intensity ratio of ~0.03. The spatial profiles of the probe beam were measured by using a digital CCD, and the spectrum of the filament core of the probe beam (where the degree of the spatial spectral chirp was minimum) was measured by using a photomultiplier-based spectrometer (SpectraPro 750).

Figures 1(a) and 1(b) show the measured and calculated molecular alignment signals of air, which are proportional to $(\langle\langle\cos^2\theta_{\parallel}\rangle\rangle-1/3)^2$ and $\langle\langle\cos^2\theta_{\perp}\rangle\rangle=(1-\langle\langle\cos^2\theta_{\parallel}\rangle\rangle)/2$, with $\theta_{\parallel}$ and $\theta_{\perp}$ being the angles between the molecular axis and the field polarizations of the pump and probe pulses, respectively. The molecular alignment signal measured by using the conventional weak-field polarization technique [17,18] was consistent with the previous demonstration on the orientation dependent nonlinear refractive index of air [19]. Molecules orientated randomly and parallel (or perpendicular) to the probe polarization are characterized by $\langle\langle\cos^2\theta_{\perp}\rangle\rangle$ equal to one-third and larger (or smaller) than one-third, respectively.

The molecular-alignment-induced controllable SC generation in air was studied by tuning the pump–probe time delay. As shown in Fig. 1(b), the molecules were first orientated parallel, then perpendicular,

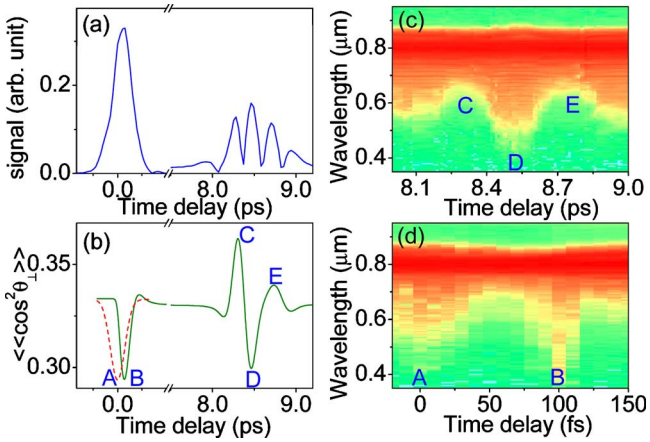

Fig. 1. (Color online) (a) Measured and (b) calculated molecular alignment signals of air. The red dashed curve in (b) represents the pump-pulse envelope. The measured output spectra of the probe pulse when it is tuned to (c) the molecular alignment revivals from 8.0 to 9.0 ps and (d) around their zero time delay.

then parallel again to the probe polarization as the time delay increased from 8.0 to 9.0 ps. Accordingly, as shown in Fig. 1(c), the spectrum of the probe pulse was modulated by following the molecular-alignment revivals, which were narrowed and broadened for parallel and perpendicular revivals of the molecular alignment, respectively. The broadest spectrum was observed at delay D, and the narrowest spectra were obtained at delays C and E. For randomly orientated molecules [Fig. 2(a)], the output spectrum of the probe pulse covered a spectral range from 520 to 880 nm with a spatial profile of three filaments [Fig. 3(a)]. Figure 2(a) also presents the output spectrum of the pump pulse, which exhibits a narrow bandwidth around 800 nm. However, as shown in Fig. 2(b), SC with a significantly extended spectrum at short wavelengths was generated. The intensity sum of the extended spectrum below 750 nm occu-

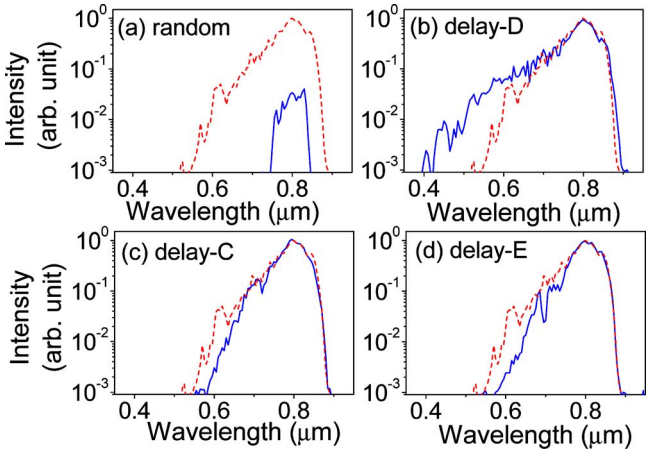

Fig. 2. (Color online) Measured output spectra of the probe pulse when it is tuned to experience (a) randomly orientated molecules and (b)–(d) molecular alignment revivals at various delays, as labeled in Fig. 1(b). The dashed and solid curves stand for the output spectra of the probe pulse when the molecules are randomly oriented and prealigned, respectively. The solid curve in (a) is the recorded output spectrum of the pump pulse.

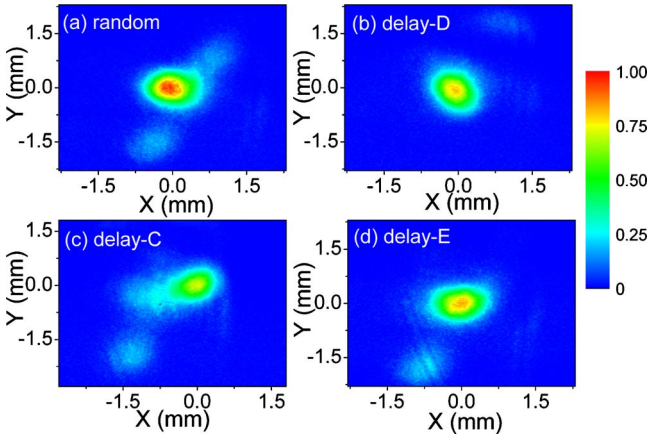

Fig. 3. (Color online) Output spatial profiles of the probe beam at various time delays.

pied 30% of the whole spectral intensity at delay D. At a distance of 100 cm after the probe filament, a small aperture with a diameter of 5.0 mm was used to select the center white core of the generated SC, and the energy in this single core was measured to be 1.8 mJ. This differed completely from the previous demonstrations for a relatively low pulse energy [9,10], where SC generation was promoted with parallel molecular orientation.

The high-energy ultrashort laser pulse made much more drastic competition among the SC generation, multifilamentation [20], multiphoton ionization losses [21], and other nonlinear processes than the low-energy probe pulse [9]. By tuning the probe pulse to the perpendicular revivals of the molecular alignment, an additional cross-defocusing effect was induced to loosen the focusing condition of the high-energy probe pulse. Accordingly, the SC generation occurred at a decreased energy threshold, while high-order nonlinear processes such as optical breakdown at increased energy thresholds [22]. Meanwhile, the multiphoton ionization losses were decreased because of the reduced ionization probability of the perpendicularly orientated diatomic molecules [21]. The SC generation was therefore promoted at delay D with an extended spectrum from 400 to 900 nm [Fig. 2(b)] by suppressing multifilaments [Fig. 3(b)]. Enhanced white-light generation was previously observed at revivals of parallel molecular alignment with 130 fs pulses of 3.2 mJ pulse energy [13], where the focused peak intensity was much lower than that used in the current experiment and thus still worked under the experimental circumstance of little competition between the SC generation and other nonlinear processes. This was quite different from the current experiment with higher-energy probe pulses experiencing a strong competition, where alignment-loosened focusing with perpendicularly oriented molecules were facilitated to broaden the SC generation. Figure 3 shows the far-field beam profiles of the probe pulse with random and prealigned molecules, which reflects the properties of filament core [23–25]. Owing to the suppressed multifilamentation resulting from the counterbalance among the Kerr self-focusing, plasma defocusing, beam diffraction, and cross-defocusing, the high-energy probe pulse exhib-

ited a well-confined single filament core [Fig. 3(b)] with only about 3% of the beam energy contained in the satellite. The probe filament length was observed to be significantly elongated at this delay [26]. For the parallel molecular alignment revivals at delays C and E, the alignment-tightened focusing increased the SC generation threshold and thus weakened the SC generation with increased multiphoton ionization losses. The spectra were correspondingly narrowed at delays C and E [Figs. 2(c) and 2(d)]. Figures 3(c) and 3(d) show the measured far-field spatial profiles of the probe pulse, where the energies contained in the central core were 59% and 63% at delays C and E, respectively. Since the molecular alignment degree was proportional to the pump intensity, for the parallel molecular alignment, the output spectra of the high-energy probe pulse became narrower as the pump intensity increased, which was also quite different from the low-energy probe-pulse case [27].

The alignment-assisted SC generation was also observed around zero time delay, where molecules were only perpendicularly oriented to the probe polarization [Figs. 1(a) and 1(b)]. Accordingly, as shown in Fig. 1(d), broadened SC generation was observed at delay B (delayed ~100 fs after the pump). Such a delayed response agreed with the impulsive alignment of the diatomic molecules in air, which was delayed about ~80 fs after the pump. Furthermore, spectral broadening of the probe pulse was observed within the pulse duration at delay A [Fig. 1(d)], which was originated from cross-phase modulation between synchronized pump and probe pulses.

In summary, we demonstrate that cross-defocusing effect induced by the perpendicularly orientated molecules could loosen the focusing condition of the high-energy probe pulse and hence promoted the SC generation with an output energy of 1.8 mJ. In contrast with the low-energy case [9] demonstrated in a gas cell, the field-free controllable and remotely achievable broadband high-energy SC generation demonstrated here offers the obvious advantage of experimental convenience in ambient air. This is very important for many applications in various areas, such as remote sensing in atmosphere.

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