Controllable supercontinuum generation by the quantum wake of molecular alignment

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(Received 3 January 2009; published 29 April 2009)

We demonstrate the generation of supercontinuum with controllable cutoff extension in ultraviolet spectral region through femtosecond filamentation in parallel aligned CO_2 molecules. The additional cross-focusing effect from the quantum wake of impulsive molecular alignment and the subsequently enhanced self-phase-modulation as well as self-steepening and space-time focusing play important roles for the observed supercontinuum generation, which also result in a self-cleaned beam profile by attracting almost all the energy into a single core.

PACS number(s): 37.10.Vz, 52.38.Hb, 42.65.Jx, 33.80.−b

As one of the most fundamental nonlinear phenomena concurrent with the nonlinear propagation of ultrashort laser pulses in various transparent nonlinear optical media, supercontinuum generation with ultrabroadband spectrum covering the spectral range from near-infrared to ultraviolet (uv) has been extensively studied and stimulated various important applications [1–3]. Several nonlinear processes, including self-phase-modulation (SPM), four-wave mixing, stimulated Raman scattering, self-steepening, and third harmonic generation, are now understood to broaden the pulse spectrum and contribute the supercontinuum generation [4]. Supercontinuum generation by self-guided filament of intense ultrashort laser pulse has been investigated [5–7], producing extremely broadband spectrum covering spectral range from uv to infrared or even midinfrared accompanied with conical emissions [8,9]. The cutoff of the generated supercontinuum in the uv spectral region could be extended [10] for driving pulse of few-cycle temporal duration (sub-10 fs at 800 nm) or ultrahigh intensity, where the generated supercontinuum was restricted in pulse energy by the difficulty to have energetic driving pulse in the few-cycle range for the former case and typically multifilaments for the latter case.

In this paper, we demonstrate that supercontinuum cutoff extension in the uv spectral region can be controlled by using the quantum wake of impulsive molecular alignment of CO_2 through filamentation of a femtosecond laser pulse with a long temporal duration and moderate driving intensity. It meanwhile shows a self-cleaned beam profile owing to the suppression of beam breakup by attracting almost all the energy into a single core. The dependences of the supercontinuum generation on the pump intensity for molecular alignment and the gas pressure are investigated.

After impulsive rotational Raman excitation by an ultrashort pump pulse, the quantum beatings of the prepopulated molecular states result in periodic revivals of the molecular alignment with a typical lifetime of several hundred femtoseconds for each recurrence [11,12]. The impulsive molecular alignment has been widely studied for ultrashort pulse compression [12,13], high-order harmonic generation [14–16], and recently for the propagation of intense femtosecond pulse [12,17–19]. For a linear molecular gas,

the temporally delayed probe pulse experiences an increased or decreased refractive index when the molecules are oriented with the molecular axis parallel or perpendicular to its field polarization as $\delta n(r,t) = 0.5(\rho_0 \Delta\alpha/n_0)[\langle\langle \cos^2 \theta(r,t)\rangle\rangle - 1/3] + \delta n_{rR}(r,t)$ [12], where ρ_0 is the initial molecular density, $\Delta\alpha = \alpha_\parallel - \alpha_\perp$ is the polarizability difference, n_0 is the linear refractive index, δn_{rR} is the rotational Raman contribution of the pre-excited molecules [20] evolving as the molecular alignment revivals. The degree of molecular alignment is characterized by the averaged term $\langle\langle \cos^2 \theta\rangle\rangle$, where $\langle\langle \cos^2 \theta\rangle\rangle - 1/3 > 0$ or $\langle\langle \cos^2 \theta\rangle\rangle - 1/3 < 0$ accounts for the linear molecules orientated parallel or perpendicular to the field polarization of the probe pulse, and $\langle\langle \cos^2 \theta\rangle\rangle = 1/3$ for randomly oriented molecules. By considering the Gaussian-shaped transverse profile of the pump pulse, molecular alignment induces a larger change in the refractive index in the beam center than in its periphery. It hence introduces a cross-(de)focusing effect to the delayed probe pulse depending on the transient orientations of the prealigned linear molecules. Meanwhile, the ultrashort probe pulse accumulates a nonlinear phase shift φ_m from the time-dependent quantum wake of the molecular alignment, leading to an additional spectral modulation as $\delta\omega(t) = -\partial\varphi_m/\partial t \sim -\partial\delta n(t)/\partial t$ [12,17–19]. The succeeding filamentation of the probe pulse is therefore dominated by the interplay of the molecular alignment induced spatiotemporal phase modulation and linear diffraction or dispersion as well as nonlinearities of the gas medium, allowing us to control the generation and extension of the resulted supercontinuum.

In the experiments, a femtosecond pulse from an amplified Ti:sapphire laser system (35 fs, 800 nm, 1 kHz) was first split in two by a beam splitter with one of them as pump pulse for molecular alignment and another one as probe for filamentation. In order to have a larger interaction volume, both beams were down-collimated with two separated telescope systems and then recombined collinearly. The delay between the s-polarized pump pulse and the p-polarized probe pulse was tuned by using a motorized translation stage in the pump arm. The collinearly recombined pump and probe pulses were then focused by using a lens with the focal length $f = 60$ cm into a gas cell of 85 cm in length, which was filled with pure molecular CO_2 with a maximum gas pressure of 2 atm. The pulse energies for the pump and probe pulses before the gas cell were 0.5 and 1.5 mJ, respectively.
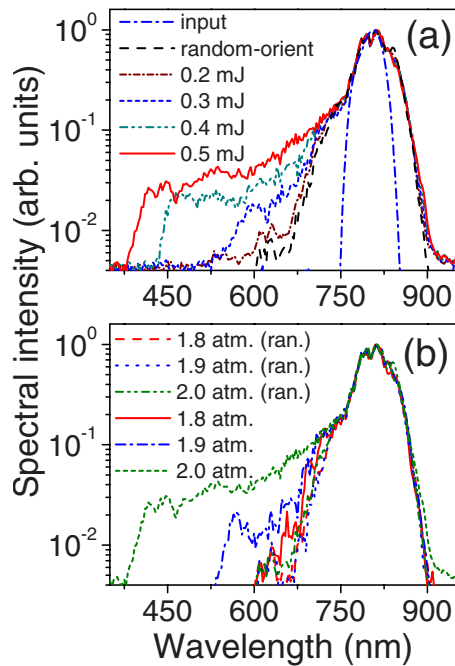
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FIG. 1. (Color online) Measured output spectra of the probe pulse at various (a) pump energies and (b) gas pressures when its temporal peak is tuned to match the molecular alignment revival at delay-*p* with a maximum value of $\langle\langle\cos^2\theta\rangle\rangle$. The gas pressure is fixed at 2 atm for (a) and the pump energy is set to be 0.5 mJ for (b).
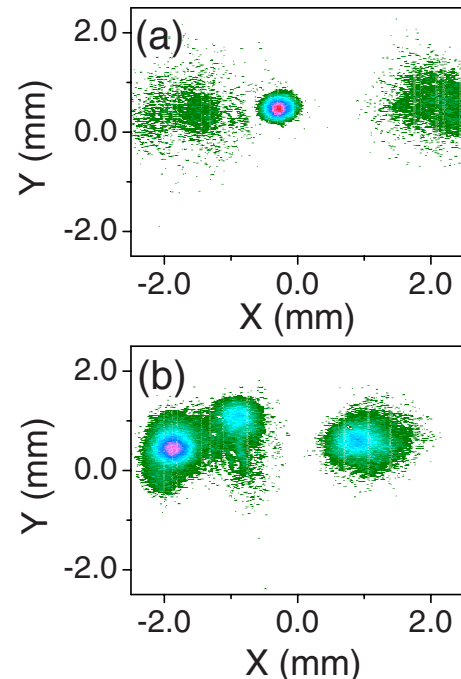

FIG. 2. (Color online) Measured output spatial profiles of the probe pulse when the molecular alignment is (a) turned on and (b) turned off. The gas pressure and pump energy are, respectively, set to be 2 atm and 0.5 mJ for (a), and the probe pulse is tuned to experience molecular alignment revival at the delay-*p*.

At the exit of the gas cell, the spectral and spatial profiles of the probe pulse were measured simultaneously by using a photomultiplier (PMT) based spectrometer (SpectraPro 750) and a charge-coupled device (CCD) camera after a polarization analyzer, since we concerned the influence of molecular alignment on the both pulse spectral extension and bream profile.

As shown in Fig. 1(a), for randomly oriented molecules, the output spectrum of the intense probe pulse was broadened with respect to the input one due to the SPM within the filament channel, covering a spectral range from 600 to 900 nm. We then turned on the molecular alignment gradually by increasing the pulse energy of the pump from 0.2 to 0.5 mJ. The corresponding pump intensity around the geometric focus was varied from 1.5×10^{13} to 3.7×10^{13} W/cm². As we can see from Fig. 1(a), for the molecular alignment revival with the maximum value of $\langle\langle\cos^2\theta\rangle\rangle$ (or maximum parallel orientation degree) around the half-revival time (we labeled this molecular alignment revival as delay-*p* hereafter), the output spectrum of the probe pulse extended continuously in the uv spectral region as the pump intensity increased. It eventually led to a uv cutoff of the generated supercontinuum around 380 nm with a broadband plateau for a pump energy of 0.5 mJ. The output energy of the probe pulse was measured to be 1.3 mJ. As shown in Fig. 1(a), the spectral extension of the probe pulse at the short wavelengths was almost negligible for a relatively low molecular alignment degree with a pump energy of 0.2 mJ. In addition to the supercontinuum generation around the molecular alignment revival at delay-*p*, the generated supercontinuum pulse also

shows a self-cleaned beam profile [Fig. 2(a); no noticeable spatial chirp was observed] rather than multifilaments along the long axis of the incident beam [Fig. 2(b); a similar effect was reported in Ref. [21]] for an input spatial ellipticity of $e \sim 1.6$ with randomly oriented molecules.

The output spectral and spatial profiles of the probe pulse at various molecular alignment revivals were further studied by tuning its temporal delay with respect to the pump pulse, resulting in decreased spectral extensions at short wavelengths and recurrence of beam breakup. For molecular alignment revivals with $\langle\langle\cos^2\theta\rangle\rangle - 1/3 < 0$, owing to the additional cross-defocusing effect from the perpendicularly orientated molecules, the output spectrum of the probe pulse was even narrowed as compared with the case of random orientation. And the narrowest spectrum was observed for molecular alignment revival with the minimal value of $\langle\langle\cos^2\theta\rangle\rangle$, where the cross-defocusing effect was most significant.

Based on the above analysis of the time-dependent molecular alignment revivals, the maximum spectral modulation is expected around the revival position with the largest value of $|\partial\delta n(t)/\partial t|$, which occurs at revival transition from parallel to perpendicular (or from perpendicular to parallel) orientation. Note that $\partial\delta n(t)/\partial t \sim 0$ at the delay-*p*. It therefore suggested that the molecular alignment revival induced spatial cross-focusing effect, which reached its maximum value at the delay-*p*, played an important role in the observed supercontinuum generation. It further enhanced the SPM, self-steepening as well as space-time focusing effects during the probe pulse filamentation, and eventually extended the supercontinuum continuously into the uv spectral region. The self-

steepening and space-time focusing have been demonstrated [10,22] to be significant for supercontinuum generation and especially spectral extension at short wavelengths. During the supercontinuum generation, the probe pulse energy was redistributed to a much broader spectral range, and decreased the peak intensity around the central wavelength of the incident one, which therefore suppressed the beam breakup and led to the observed energy attraction into a single core [Fig. 2(a)].

The dependence of the supercontinuum generation on the gas pressure of molecular CO_2 was studied for a given pump energy of 0.5 mJ at the delay-p. Figure 1(b) presents the output spectra of the probe pulse when the molecular alignment was turned on or turned off for various gas pressures. Here, the molecular alignment degree was fixed for a given pump intensity. The additional cross-focusing effect or refractive index variation was changed as the molecular density. As shown in Fig. 1(b), for randomly oriented molecules, the spectral broadening of the probe pulse was almost unchanged when the gas pressure decreased from 2.0 to 1.8 atm. It means that the influence from the change in the material nonlinearity (estimated to be ~10% by considering a linear dependence of the molecular density) for general broadening of the probe pulse was minor in our experiments. However, the spectral extension of the generated supercontinuum decreased rapidly as the gas pressure decreased, and almost no additional uv extension was observed for a gas pressure of 1.8 atm. The cross-focusing effect, proportional to the molecular alignment degree and number density of the molecular gas, decreased gradually with the decreasing gas pressure. It further weakened the SPM as well as self-steepening and space-time focusing effects, and therefore decreased the spectral broadening of the probe pulse rapidly. It clearly indicates the important role of the molecular alignment for the supercontinuum generation and extension by filamentation in our experiments.

The observed supercontinuum generation by molecular alignment assisted filamentation is then modeled as [12,23]

$$\frac{\partial E}{\partial z} = \frac{i}{2k_0}U^{-1}\left(\frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r}\right)E - i\frac{k_0^{(2)}}{2}\frac{\partial^2 E}{\partial t^2} + \frac{k_0^{(3)}}{6}\frac{\partial^3 E}{\partial t^3}$$
$$+ U^{-1}\left[T^2 i k_0(n_2|E|^2 + \delta n)E - \frac{\sigma}{2}(1 + i\omega_0\tau_c)\rho E\right.$$
$$\left. - T\frac{\beta_K}{2}|E|^{2K-2}\left(1 - \frac{\rho}{\rho_0}\right)E\right]. \tag{1}$$

As an extension of the general model presented in Ref. [23], the contributions from molecular alignment induced spatiotemporal modulation of the refractive index $\delta n(r,t)$, and orientation-dependent ionization probability σ_K due to the molecular multiwell potential [24] were included in our model. The multiphoton ionization induced electron density ρ was calculated by resolving
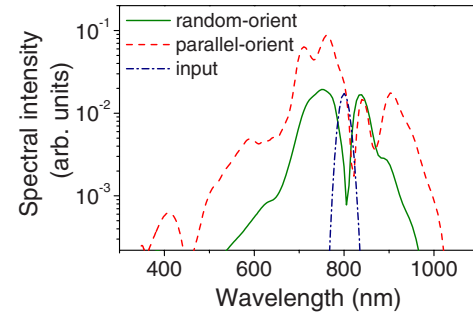

FIG. 3. (Color online) Simulated on-axis spectral profiles of the probe pulse when the CO_2 molecules are parallel or randomly orientated.

$\partial\rho/\partial t = \sigma_K|E|^{2K}(\rho_0-\rho)$, where σ_K was modeled as Ref. [14] with $\sigma_{K0}=3.98\times10^{-111}$ s^{-1} cm^{18}/W^9 accounted for the average ionization probability of randomly oriented CO_2 molecules. The other parameters were set as: $\rho_0=5.0\times10^{19}$ cm^{-3}, $k_0^{(2)}=0.67$ fs^2/cm, $k_0^{(3)}=0.44$ fs^3/cm, $n_2=6.3\times10^{-19}$ cm^2/W, $\sigma=1.1\times10^{-19}$ cm^2, $\tau_c=175$ fs, $\beta_K=4.39\times10^{-109}$ cm^{15}/W^8, $K=9$. The pump pulse with polarization orthogonal to the probe one was set to have an intensity of 3.0×10^{13} W/cm^2 around the geometric focus. The input power and beam diameter of the probe pulse were set as $P_{in}=2.4$ GW and 1.5 mm [full width at half maximum (FWHM)]. Figure 3 shows the simulated output spectra of the probe pulse when the molecular alignment recurrence at the delay-p was turned off and turned on, indicating the generation of supercontinuum assisted by molecular alignment as observed in our experiments.

In summary, we demonstrate that supercontinuum could be generated with controllable extension in the uv spectral region by using molecular alignment revival assisted femtosecond filamentation. The additional cross-focusing effects from the parallel molecular alignment revival enhanced the succeeding SPM as well as self-steepening and space-time focusing effects to broaden the spectrum of the probe pulse, which simultaneously suppressed the beam breakup and attracted almost all the energy into a spatially self-cleaned single core. This provide us an efficient approach to control the intense filamentation dynamics by using molecular alignment revival, and to produce tunable supercontinuum pulse of high pulse energy and self-cleaned single core for practical use in various fields.

[1] C. P. Hauri *et al.*, Appl. Phys. B: Lasers Opt. **79**, 673 (2004).

[2] G. Stibenz, N. Zhavoronkov, and G. Steinmeyer, Opt. Lett. **31**, 274 (2006).

[3] G. Méjean *et al.*, Appl. Phys. B: Lasers Opt. **77**, 357 (2003).

[4] S. L. Chin *et al.*, Can. J. Phys. **83**, 863 (2005).

[5] J. Yu *et al.*, Opt. Lett. **26**, 533 (2001).

[6] V. P. Kandidov *et al.*, Appl. Phys. B: Lasers Opt. **77**, 149 (2003).

[7] J. Liu *et al.*, Opt. Express **13**, 10248 (2005).

[8] D. Faccio *et al.*, Phys. Rev. A **78**, 033825 (2008).

[9] F. Théberge *et al.*, Opt. Lett. **33**, 2515 (2008).

[10] N. Aközbek *et al.*, New J. Phys. **8**, 177 (2006).

[11] H. Stapelfeldt and T. Seideman, Rev. Mod. Phys. **75**, 543 (2003).

[12] J. Wu *et al.*, Opt. Lett. **33**, 2593 (2008).

[13] R. Bartels *et al.*, Phys. Rev. Lett. **88**, 013903 (2001).

[14] T. Kanai, S. Minemoto, and H. Sakai, Nature (London) **435**, 470 (2005).

[15] G. H. Lee *et al.*, Opt. Lett. **33**, 2083 (2008).

[16] J. Wu, H. Qi, and H. Zeng, Phys. Rev. A **77**, 053412 (2008).

[17] F. Calegari *et al.*, Phys. Rev. Lett. **100**, 123006 (2008).

[18] S. Varma, Y.-H. Chen, and H. M. Milchberg, Phys. Rev. Lett. **101**, 205001 (2008).

[19] H. Cai *et al.*, Opt. Lett. **34**, 827 (2009).

[20] E. Nibbering *et al.*, J. Opt. Soc. Am. B **14**, 650 (1997).

[21] A. Dubietis *et al.*, Opt. Lett. **29**, 1126 (2004).

[22] A. Gaeta, Phys. Rev. Lett. **84**, 3582 (2000).

[23] A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47 (2007).

[24] J. Wu, H. Zeng, and C. Guo, Phys. Rev. A **74**, 065403 (2006); **75**, 043402 (2007).