Intense ultrafast light kick by rotational Raman wake in atmosphere

Jian Wu, Hua Cai, Peifen Lu, Xueshi Bai, Liang'en Ding, and Heping Zeng[a)]
State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, People's Republic of China

(Received 7 September 2009; accepted 31 October 2009; published online 30 November 2009)

We report that intense ultrafast light can be particlelike kicked through the rotational Raman wake of the impulsively pre-excited diatomic molecules in atmosphere, manifested by controllable repulsion and attraction of intense pulses delayed by molecular alignment revival periods, and by mutual fusion of orthogonally polarized intense pulses nearby zero time delay. The rotational Raman wake facilitates longer ranges of interaction on the intense ultrafast light kicks than the plasma wake and Kerr effect. © *2009 American Institute of Physics*. [doi:10.1063/1.3266863]

The intense white-light filaments with facilitated light detection and ranging method are significant for atmosphere applications,[1,2] such as remote sensing for pollution analysis, ozone loss monitoring, and even weather prediction. The plasma self-channels with ionization-induced free electrons are promising for lightning control and virtual antenna.[3] Furthermore, filaments are extensively used for energetic few-cycle pulse generation,[4] remote terahertz emission,[5,6] and long-distance delivery of intense laser beams. In this context, field-free intense ultrafast light kicks by rotational Raman wake provide a robust mechanism to control filamentation dynamics and interactions for those interesting applications. In general, impulsive molecular alignment is achieved through rotational Raman excitation with a broadband ultrashort pump pulse,[7,8] which periodically revives at several picoseconds after the pump pulse extinction due to the rephrasing of the pre-excited rotational wave packets. The impulsive molecular alignment with no interference on the subsequent processes has been extensively studied for molecular orbital reconstruction,[9] high harmonic generation,[10] spectral modulation, and pulse compression,[11,12] and recently for ultrashort laser pulse propagations.[13–17]

We demonstrate here an interesting nonlinear interaction of intense femtosecond filaments which occurs even they are temporally delayed far away and brings about field-free kick of parallel self-guided light propagation with observable displacements. Our approach is based on the rotational Raman wake of the pre-excited diatomic molecules in atmosphere, which induces not only periodic kicks of intense probe filaments after the extinction of the pump pulses but also filament interaction of mutual fusion as they are temporally approached. The displacement variation can be readily controlled by tuning the time delay of probe pulse to properly match the molecular alignment revivals, which can be remotely realized without using any assisted devices and controlled in atmosphere wherever the intense filaments are created.

The intense ultrafast filament not only leaves an ionization-induced plasma wake and contributes a negative variation of the atmosphere refractive index as $-\rho(r,t)/(2\rho_{cr})$, where $\rho(r,t)$ and ρ_{cr}, respectively, account for the free electron and critical plasma density, but also impulsively aligns the diatomic molecules with periodical field-free revivals.[13–16] In prealigned molecules, a linearly polarized ultrashort probe pulse experiences a refractive index variation of $\delta n_{mol}(r,t)=2\pi(\rho_0\Delta\alpha/n_0)$ $[\langle\langle\cos^2\theta_{\|}(r,t)\rangle\rangle - 1/3]$ ($\theta_{\|}$ is the angle between the molecular axis and pump polarization), where $\Delta\alpha$, ρ_0, and n_0 denote the molecular polarizability difference, initial molecular density, and linear refractive index, respectively. It hence introduces an additional spatiotemporal cross-phase modulation in a controllable manner and determines the succeeding probe pulse propagation. As illustrated in Fig. 1(a), local increase of the refractive index induced by parallel molecular orientation counteracts on the negative refractive index variation from the plasma wake, a well-defined waveguide with a net increase of the refractive index is thus created by the pump filament, leading to attraction of the probe filament. On the other hand, local decrease of the refractive index caused by perpendicularly oriented molecules and the plasma wake sum up to produce a negative lensing effect[13–16] [Fig. 1(b)] and accordingly, the probe filament is repulsed by the pump filament wake. Additionally, closely launched pump and probe filaments can also be repulsed by the plasma wake alone [Fig. 1(c)]. In this spirit, we demonstrate that the molecular alignment in filament channel facilitate intense ultrashort light kicks in a controllable manner.

The experiments were performed with an output from an amplified Ti:sapphire laser system (35 fs, 800 nm, 1 kHz), which was split in two by a beam splitter with one of them as the pump pulse and the other as the probe pulse. The time delay between them was tuned by using a motorizing translation stage in the pump arm, where the negative and positive delays, respectively, account for the advancing of the probe and pump pulses. The polarization of the pump pulse was rotated orthogonal to the probe pulse using a half-wave plate. The pump and probe pulses were, respectively, focused by using two fused-silica lenses ($f=100$ cm) before they were parallel recombined by means of a thin film polarizer. The final energies of the pump and probe pulses were, respectively, measured to be 1.6 and 0.8 mJ. Two femtosecond filaments of pump and probe parallel to each other (~5.0 cm in length) were first launched as shown in Fig. 2(a). The kick of the probe filament by the pump was determined by imaging the plasma fluorescence to a fluorescence sensitive charge-coupled device after a microscope objective (10×). The pump and probe filament separation was measured according to the distance between the filament cores. The di-

a)Electronic mail: hpzeng@phy.ecnu.edu.cn.

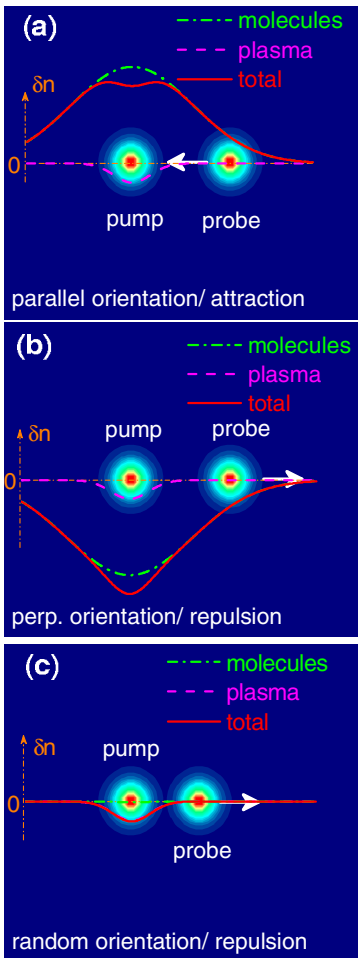

FIG. 1. (Color online) Schematic illustration of light kick by prealigned molecules. The probe filament is, respectively, tuned to match the (a) parallel revival, (b) perpendicular revival, and (c) random orientation of the prealigned molecules excited by an advancing pump filament. The relative refractive index variation induced by the prealigned neutral molecules, plasma, and the sum of them (total) are plotted vs the displacement from the pump filament.

ameters of the pump and probe filaments were, respectively, extracted to be ~108 and 50 μm with an initial separation of ~106 μm. Figure 2(b) shows the measured revival signal ($\sim|\langle\langle\cos^2\theta_{\|}\rangle\rangle-1/3|^2$) of the impulsively excited atmospheric molecules by using the well-established weak field polarization spectroscopy technique,[8] where the quantum wake of the prealigned molecular nitrogen and oxygen are clearly identified with periods of 8.3 and 11.6 ps, respectively. The calculated molecular alignment revivals relative to the probe polarization $\langle\langle\cos^2\theta_{\perp}\rangle\rangle=(1-\langle\langle\cos^2\theta_{\|}\rangle\rangle)/2$ is correspondingly plotted in Fig. 2(c), where $\theta_{\perp}$ is the angle between the molecular axis and probe polarization.

We begin by demonstrating the field-free kick of the probe filament around the half-revivals of molecular nitrogen and oxygen in atmosphere, where the diatomic molecules were first orientated perpendicularly and then parallel to the probe polarization, as shown in Fig. 2(c). Accordingly, as shown in Fig. 3(a), the probe filament was first kicked away and then anear the pump filament with a modulated displacement. In comparing with the case out of the quantum revivals after the pump excitation, a maximum positive (negative) displacement of 15 (−12) and 14(−9) μm are observed nearby the half-revivals of molecular nitrogen and oxygen, respectively. This field-free kick of the intense probe filament was further confirmed by analyzing the induced fluorescence intensity of molecular ions inside the filament. The electron in a diatomic molecule parallel oriented to the filed polarization experiences a double-well potential of the ionic cores and is thus much more easily ionized[10] than that in a perpendicularly oriented diatomic molecule which experiences a single-well potential. Consequently, as plotted in Fig. 3(b), a modulation of the integrated fluorescence intensity of the probe filament was observed by following the rotational Raman wake of the atmosphere. The fluorescence intensity of the probe filament was, respectively, increased and de-

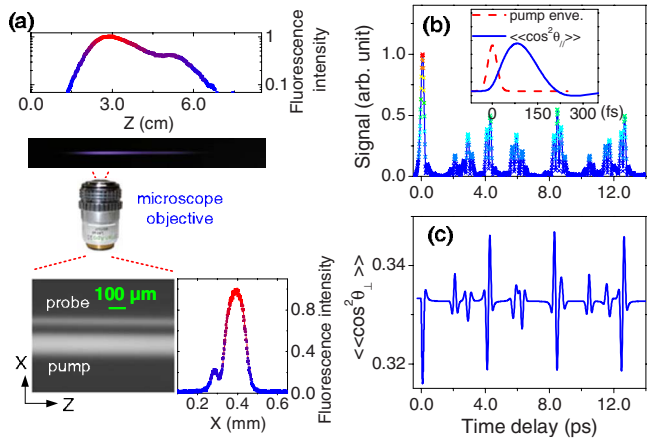

FIG. 2. (Color online) (a) The measured fluorescence wake of the pump and probe filaments in atmosphere. The (b) measured and (c) simulated molecular alignment revivals of the diatomic molecules in atmosphere. The inset of (b) shows the delayed rotational Raman response of the atmosphere with respect to the pump pulse envelope.

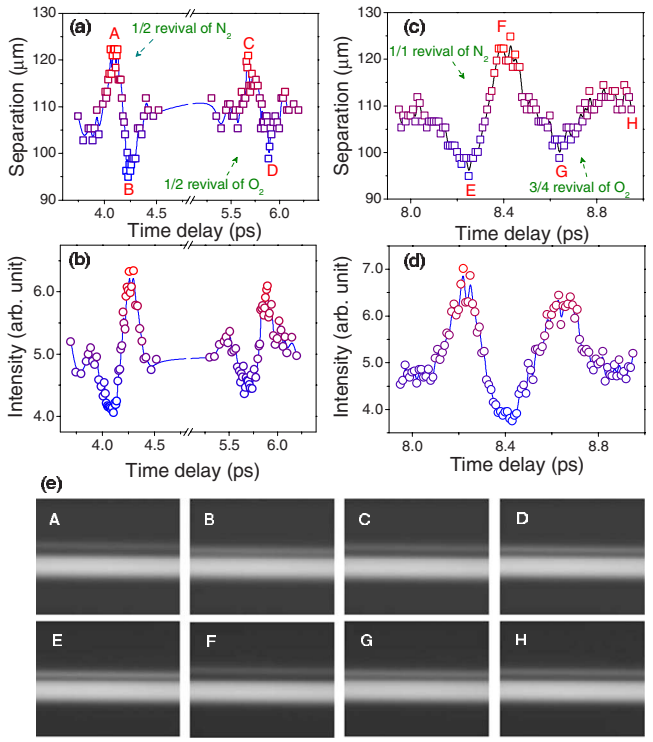

FIG. 3. (Color online) Measured displacement and fluorescence intensity of the probe filament vs rotational Raman wake of atmosphere. The pump-probe filaments separation and fluorescence intensity of the probe filament as it is tuned to match the (a) and (b) half-revivals and (c) and (d) full- and three-quarter revivals of the prealigned molecular nitrogen and oxygen molecules in atmosphere. (e) The corresponding fluorescence intensity profiles of the pump and probe filaments.

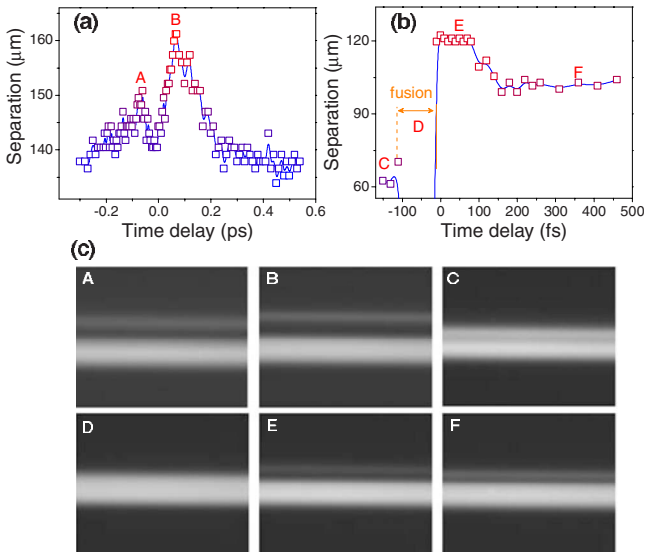

FIG. 4. (Color online) Measured displacement and fluorescence intensity of the probe filament nearby the zero time delay. (a) and (b) The pump-probe filaments separation vs their time delay nearby the zero delay for various initial separations. (c) The corresponding fluorescence intensity profiles of the pump and probe filaments.

creased when it was kicked anear and away by the parallel and perpendicular revivals of the prealigned atmospheric molecules.

The field-free kick of the probe filament was also confirmed by tuning it to the full-revival of nitrogen molecules around 8.3 ps and the three-quarter revival of oxygen molecules around 8.7 ps. As presented in Figs. 3(c) and 3(d), a modulation of the displacement and fluorescence intensity of the probe filament were observed similar to the cases nearby the half-revivals, indicating a noticeable kick of the intense probe filament by the rotational Raman wake of the atmosphere. The periodic repulsion and attraction of the intense probe filament are clearly illustrated in Fig. 3(e) for various time delays [as labeled A–H in Figs. 3(a) and 3(c)].

As we moved to the zero time delay, rather than the rotational Raman and plasma responses, the Kerr effect of the atmospheric molecules[18] also participated in the filament kicks. These effects were examined for different filament separations. The rotational Raman wake was confirmed to have a relatively longer range of interaction than the others. For an initial separation of ~140 μm, Fig. 4(a) shows that the rotational Raman effect with perpendicularly oriented molecules kicks the filaments away at delay-A (−60 fs) and delay-B (65 fs) for the advancing probe and pump pulses, respectively. Such noninstantaneous filament kicks are originated from the time delayed responses of molecular alignment (~80 fs with respect to the pump pulse excitation) as numerically revealed in the inset of Fig. 2(b). We then decreased the initial filament separation to ~60 μm, and Fig. 4(b) presents the kick dynamics. Except for kick away of the probe filament by the perpendicularly oriented molecules around delay-E, filament fusion was observed when they walked together nearby delay-D, where the nonlinear response was dominated by the Kerr effects. The Kerr effects experienced by the pump and probe pulses can be written as $\delta n_{\mathrm{Kerr\_pump}}(r)=n_{2\_\mathrm{paral}}\mathrm{I}_{\mathrm{pump}}(r)+n_{2\_\mathrm{orthog}}\mathrm{I}_{\mathrm{probe}}(r)$ and $\delta n_{\mathrm{Kerr\_probe}}(r)=n_{2\_\mathrm{paral}}\mathrm{I}_{\mathrm{probe}}(r)+n_{2\_\mathrm{orthog}}\mathrm{I}_{\mathrm{pump}}(r)$, where n_{2_paral} and $n_{2\_\mathrm{orthog}}=2n_{2\_\mathrm{paral}}/3$ are the Kerr nonlinear coefficients of air for the parallel and orthogonally polarized field components, respectively. These strong Kerr effects from the orthogonally polarized filaments with increased refractive index led to the observed fusion at delay-D when they temporally approached. The repulsion of the probe filament induced by the plasma wake alone [as illustrated in Fig. 1(c)] was observed when the pump filament was ahead, leading to an increased separation around delay-F after the molecular alignment. However, this plasma wake effect on the filament kick at delay-C was negligible when the probe one was ahead for its weak ionization. The plasma and Kerr responses were significant only for closely launched filaments, in contrast with the relatively long interaction range of the rotational Raman wake. Figure 4(c) illustrates the recorded plasma fluorescence and clearly indicates repulsion, attraction, and fusion of the pump and probe filaments nearby the zero time delay.

In summary, we have demonstrated remotely controllable and field-free kicking of intense ultrafast pulses by virtue of the rotational Raman wake of the atmosphere without requirements of any additional devices, which opens varies of possibilities for atmosphere applications,[1,2] remote intense light control,[3] and light bullet interactions.[19] It is a further step toward field-free curving or bending of intense ultrafast light through manipulating a gradually varied rotational Raman response of the atmosphere remotely.

This work was funded in part by National Natural Science Fund (Grant Nos. 10525416 and 10804032), National Key Project for Basic Research (Grant No. 2006CB806005), and Projects from Shanghai Science and Technology Commission (Grant Nos. 08ZR1407100 and 09QA1402000).

[1] J. Kasparian, M. Rodriguez, G. Méjean, J. Yu, E. Salmon, H. Wille, R. Bourayou, S. Frey, Y. B. André, A. Mysyrowicz, R. Sauerbrey, J. P. Wolf, and L. Wöste, Science **301**, 61 (2003).
[2] J. Yu, D. Mondelain, G. Ange, R. Volk, S. Niedermeier, J.-P. Wolf, J. Kasparian, and R. Sauerbrey, Opt. Lett. **26**, 533 (2001).
[3] M. Rodriguez, R. Sauerbrey, H. Wille, L. Wöste, T. Fujii, Y.-B. André, A. Mysyrowicz, L. Klingbeil, K. Rethmeier, W. Kalkner, J. Kasparian, E. Salmon, J. Yu, and J. P. Wolf, Opt. Lett. **27**, 772 (2002).
[4] C. P. Hauri, W. Kornelis, F. W. Helbing, A. Heinrich, A. Couairon, A. Mysyrowicz, J. Biegert, and U. Keller, Appl. Phys. B: Lasers Opt. **79**, 673 (2004).
[5] J. Dai, X. Xie, and X.-C. Zhang, Phys. Rev. Lett. **97**, 103903 (2006).
[6] Y. Liu, A. Houard, B. Prade, S. Akturk, A. Mysyrowicz, and V. T. Tikhonchuk, Phys. Rev. Lett. **99**, 135002 (2007).
[7] H. Stapelfeldt and T. Seideman, Rev. Mod. Phys. **75**, 543 (2003).
[8] V. Renard, M. Renard, S. Guérin, Y. T. Pashayan, B. Lavorel, O. Faucher, and H. R. Jauslin, Phys. Rev. Lett. **90**, 153601 (2003).
[9] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. Kieffer, P. Corkum, and D. Villeneuve, Nature (London) **432**, 867 (2004).
[10] T. Kanai, S. Minemoto, and H. Sakai, Nature (London) **435**, 470 (2005).
[11] R. A. Bartels, T. C. Weinacht, N. Wagner, M. Baertschy, and C. H. Greene, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **88**, 013903 (2001).
[12] H. Cai, J. Wu, A. Couairon, and H. Zeng, Opt. Lett. **34**, 827 (2009).
[13] F. Calegari, C. Vozzi, S. Gasilov, E. Benedetti, G. Sansone, M. Nisoli, S. De Silvestri, and S. Stagira, Phys. Rev. Lett. **100**, 123006 (2008).
[14] J. Wu, H. Cai, H. Zeng, and A. Couairon, Opt. Lett. **33**, 2593 (2008).
[15] S. Varma, Y.-H. Chen, and H. M. Milchberg, Phys. Rev. Lett. **101**, 205001 (2008).
[16] J. Wu, H. Cai, Y. Peng, and H. Zeng, Phys. Rev. A **79**, 041404(R) (2009).
[17] A. C. Bernstein, M. McCormick, G. M. Dyer, J. C. Sanders, and T. Ditmire, Phys. Rev. Lett. **102**, 123902 (2009).
[18] G. I. Stegeman and M. Segev, Science **286**, 1518 (1999).
[19] M. Shih, M. Segev, and G. Salamo, Phys. Rev. Lett. **78**, 2551 (1997).