Spectral modulation of femtosecond laser pulse induced by molecular alignment revivals

Hua Cai,[1] Jian Wu,[1,3] Arnaud Couairon,[2] and Heping Zeng[1,4]

[1]*State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai 200062, China*
[2]*Centre de Physique Théorique, CNRS, École Polytechnique, F-91128 Palaiseau, France*
[3] *jwu@phy.ecnu.edu.cn*
[4] *hpzeng@phy.ecnu.edu.cn*

Received November 17, 2008; revised February 5, 2009; accepted February 5, 2009;
posted February 18, 2009 (Doc. ID 104212); published March 12, 2009

We demonstrate experimentally and numerically that the quantum wake of molecular alignment induced by impulsive rotational Raman excitations with femtosecond pump pulses produces observable phase modulations on femtosecond probe pulses. This leads to a spectral red- or blueshift of the probe pulses when they are properly delayed around the rising or falling edge of the half-revival time of the molecular alignment, respectively.

OCIS codes: 320.7110, 260.5950, 020.2649.

The orientation of molecules can be manipulated through adiabatic (long pulse) [1,2] or nonadiabatic (ultrashort pulse) [3–7] rotational Raman excitation, and the latter leads to periodic revivals of molecular alignment after optical field excitation, at delays depending on molecular constants. Molecular alignment has been extensively studied for molecular orbital reconstruction [8], high-harmonic generation [9,10], nonlinear frequency conversion [11], filamentation control [12,13], and ultrashort laser pulse compression in gas-filled hollow fibers [14,15]. For a linear molecular gas excited by a pump pulse in free space, a probe pulse is expected to experience a modulation of the refractive index in space and time along the field polarization at various molecular alignment revivals, which correspondingly produces an additional spectral modulation. The purpose of this work is to demonstrate that molecular alignment in free space induces a spectral modulation on a probe pulse similar to that occurring in gas-filled hollow fibers [14,15] with the difference that it is assisted with cross-(de)focusing effect. In combination with filamentation control [12,13], this effect opens the way to remote probing of the quantum wake of the molecular alignment and remote control of spectral features of the probe pulse.

We investigate experimentally and numerically the spectral modulation of an ultrashort laser pulse propagating in prealigned nitrogen molecules, which shows a red- or blueshift as its temporal peak is tuned to the rising or falling edge of the molecular alignment revival along the field polarization of the probe pulse. The experimental observations are well reproduced by numerical simulations based on the nonlinear propagation equation for an ultrashort pulse in prealigned molecular gas.

As shown in Fig. 1(a), our experiments were carried out with a Ti:sapphire amplified laser system (1 kHz, 800 nm, 50 fs). Both orthogonally polarized pump and probe pulses were focused with a lens of $f=40$ cm with overlapped foci into a gas cell filled with pure N_2 gas at the pressure of 2 atm. The pump and probe pulse energies before the gas cell were 700 and 200 μJ, respectively. At the output of the cell, the pump and probe pulses were separated by a polarizer analysis, and only the central part of the probe pulse (which experienced most significantly the molecular alignment revivals) was selected for the final analysis with a spectrometer.

Figure 1(b) shows the measured molecular alignment revival signal of N_2 proportional to $(\langle\langle\cos^2\theta_{\|}\rangle\rangle - 1/3)^2$ by using the standard weak field polarization technique [4], where $\theta_{\|}$ is the angle between the molecular axis and the pump polarization. The calculated molecular alignment signal proportional to

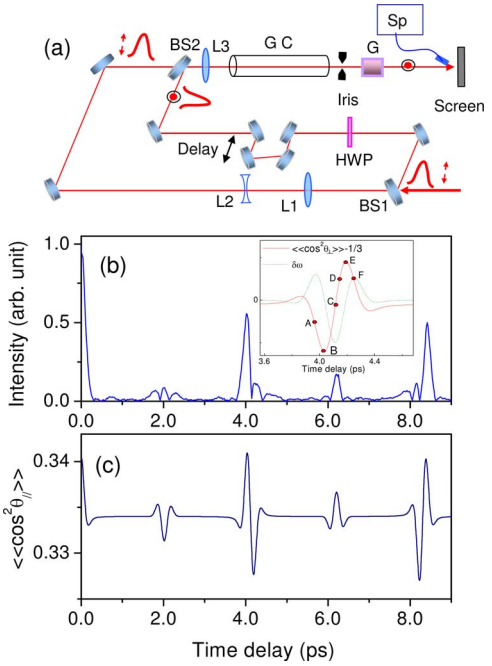

Fig. 1. (Color online) (a) Experimental setup. BS1 and BS2, beam splitter; L1–L3, lens; HWP, half-wave plate; GC, gas cell; G, Glan laser prism; Sp, spectrometer. (b) Measurement of the molecular alignment signal and (c) the calculated signal proportional to $\langle\langle\cos^2\theta_{\|}\rangle\rangle$ versus the pump–probe delay. The time-dependent molecular alignment metric and the corresponding frequency shift are presented in the inset of Fig. 1(b).

$\langle\langle\cos^2\theta_{\parallel}\rangle\rangle$ is presented in Fig. 1(c). For the orthogonally polarized pump and probe pulses in our experiments, the additional refractive index variation originated from the molecular alignment reads $\delta n_{\perp}=0.5(\rho_0\Delta\alpha/n_0)\quad(\langle\langle\cos^2\theta_{\perp}\rangle\rangle-1/3)=0.25(\rho_0\Delta\alpha/n_0)(1/3-\langle\langle\cos^2\theta_{\parallel}\rangle\rangle)$, where $\theta_{\perp}$ is the angle between the molecular axis and probe polarization, ρ_0 is the initial molecular density, $\Delta\alpha=\alpha_{\parallel}-\alpha_{\perp}$ is the polarizability difference [5], and n_0 is the linear refractive index. This results in a spectral modulation and a frequency shift induced on the probe pulse from the accumulated phase modulation Φ_m owing to the molecular alignment $\delta\omega=-\partial\Phi_m/\partial t\sim-\partial\delta n_{\perp}/\partial t$. The modulated frequency shift of the probe pulse following the wake of the molecular alignment is therefore proportional to the slope of the refractive index profile. As revealed in the inset of Fig. 1(b) around the half-revival time of the molecular alignment, the probe pulse experiences a red- or blueshift during the rising or falling edge of the revivals of the molecular alignment, respectively. In the following discussion, the temporal peak of the probe pulse is tuned to various delays (labeled as A, B, C, D, E, and F) around the half-revival time of the molecular alignment [inset of Fig. 1(b)].

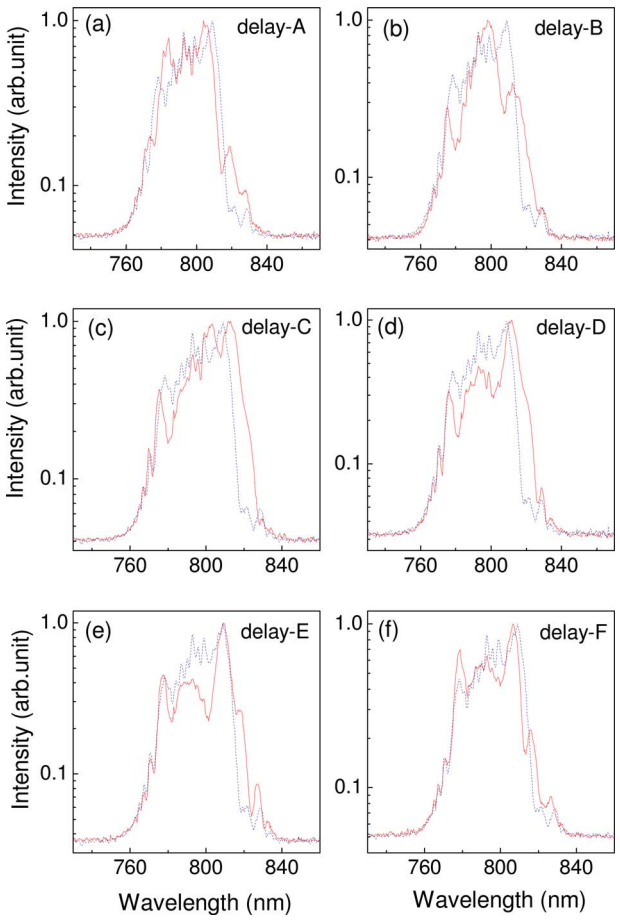

Fig. 2. (Color online) Measured spectra of the probe pulse at various delays of the molecular alignment revival as indicated in the inset of Fig. 1(b). The solid (red online) and dashed (blue online) curves represent the spectra of the probe pulse with and without molecular alignment, respectively.

Figure 2 shows typical results of the measured spectra of the probe pulse averaged over 100 shots for different pump–probe delays around the half-revival time of the molecular alignment, which reproduced well for our measurements. For delay-A as shown in Fig. 2(a), the spectrum was slightly narrowed, and new frequency components around 780 nm appeared with respect to the case of randomly oriented molecules, indicating a spectral blueshift during the falling edge of the molecular alignment revival. Spectral narrowing is due to the weaker phase modulation resulting from the cross-defocusing effect of the perpendicularly oriented molecules in the negative side of the molecular alignment revival [13]. The spectrum was noticeably narrowed when the probe pulse was tuned to the negative dip of the molecular alignment revival at delay-B [Fig. 2(b)], for which the cross-defocusing effect is most significant. In contrast, as shown in Figs. 2(c) and 2(d), the spectra were obviously redshifted, and the frequency components around 780 nm disappeared when the probe pulse was tuned to delay-C and delay-D during the rising edge of the molecular alignment revival. Meanwhile, the spectra were broadened owing to the enhanced phase modulation associated with the cross-focusing effect from the parallel orientated molecules in the positive side of the molecular alignment revival. For delay-F as shown in Fig. 2(f), the frequency components around 780 nm became more significant, and the spectrum was slightly broadened with respect to the case of delay-A, indicating a blueshift and enhanced cross-focusing effect during the falling edge but positive side of the revival of the molecular alignment. Moreover, as shown in Fig. 2(e), the prominent frequency components around 780 nm and the noticeable redshifted frequency components as compared with other molecular alignment revivals indicate the underwent spectral red- and blueshifting of the probe pulse, whose leading and trailing parts are located in the rising and falling revival edges of the molecular alignment for this delay, respectively.

The experimental observations are thus modeled by the propagation equation of the linearly polarized probe pulse in prealigned molecules [13] with dispersion described up to the third order. The evolution of the electron density ρ is calculated by resolving $\partial\rho/\partial t=\sigma_K|E|^{2K}(\rho_0-\rho)$ [16], where σ_K is the orientation-dependent cross section for multiphoton ionization as modeled in [9]. As shown in Fig. 3, the numerical results reproduce well our experimental measurements. With respect to the reference case of randomly oriented molecules (dashed curves), blue- or redshifted spectra (solid curves) of the probe pulse are obtained when its temporal peak is tuned to the falling [Figs. 3(a) and 3(f)] or rising edge [Figs. 3(c) and 3(d)] of the molecular alignment revival, respectively. The spectrum becomes narrower when the probe pulse is tuned to the negative dip of the molecular alignment revival at delay-B [Fig. 3(b)]. Both red- and blueshifted spectral components are generated when the probe pulse is tuned to the positive peak of the molecular alignment revival at delay-E [Fig. 3(e)], as observed in our experiments.

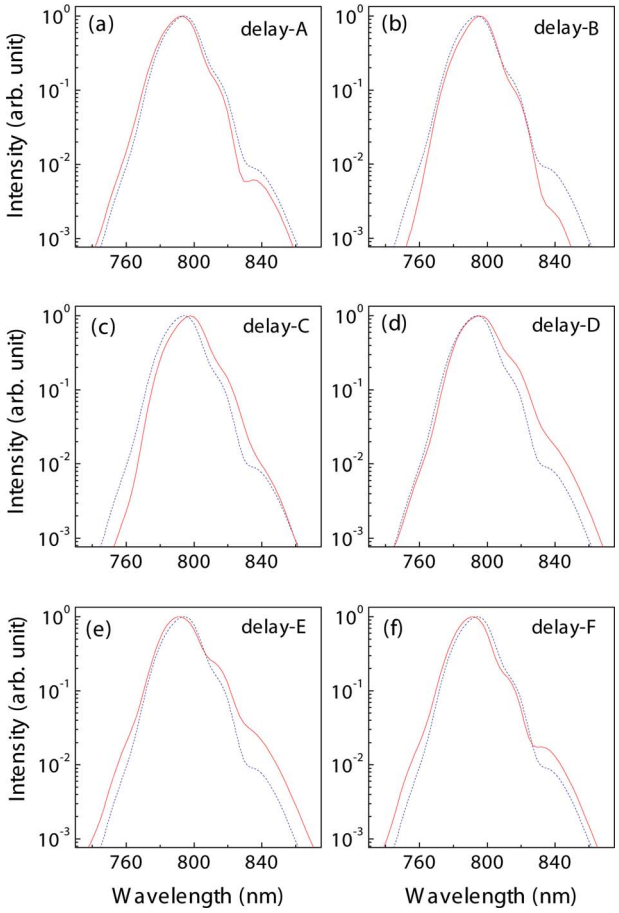

Fig. 3. (Color online) Simulated spectra of the probe pulse with same presentation as Fig. 2.

In summary, we demonstrated both experimentally and numerically that the spectrum of an ultrashort probe pulse can be modulated to be red- or blue-shifted by means of field-free molecular alignment, which is expected to be even more significant for larger pump intensities or interaction distances. In view of potential atmospheric applications with pump pulses generating a filament [16], this effect allows us to remotely probe the quantum wake of the molecular alignment and can act as a flexible method to tune the spectral broadening and central wavelengths of ultrashort pulses.

This work was funded in part by National Natural Science Fund (grants 10525416 and 10804032), National Key Project for Basic Research (grant 2006CB806005), projects from Shanghai Science and Technology Commission (grants 06JC14025 and 08ZR1407100), Program for Changjiang Scholars and Innovative Research Team in University, and Shanghai Educational Development Foundation (grant 2008CG29).

References

1. B. Friedrich and D. Herschbach, Phys. Rev. Lett. **74**, 4623 (1995).
2. H. Sakai, C. P. Safvan, J. J. Larsen, K. M. Hilligsøe, K. Hald, and H. Stapelfeldt, J. Chem. Phys. **110**, 10235 (1999).
3. F. Rosca-Pruna and M. J. J. Vrakking, Phys. Rev. Lett. **87**, 153902 (2001).
4. V. Renard, M. Renard, S. Guérin, Y. T. Pashayan, B. Lavorel, O. Faucher, and H. R. Jauslin, Phys. Rev. Lett. **90**, 153601 (2003).
5. H. Stapelfeldt and T. Seideman, Rev. Mod. Phys. **75**, 543 (2003).
6. E. Hertz, D. Daems, S. Guérin, H. R. Jauslin, B. Lavorel, and O. Faucher, Phys. Rev. A **76**, 043423 (2007).
7. C. Z. Bisgaard, M. D. Poulsen, E. Péronne, S. S. Viftrup, and H. Stapelfeldt, Phys. Rev. Lett. **92**, 173004 (2004).
8. J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J. Kieffer, P. Corkum, and D. Villeneuve, Nature **432**, 867 (2004).
9. T. Kanai, S. Minemoto, and H. Sakai, Nature **435**, 470 (2005).
10. C. Vozzi, F. Calegari, E. Benedetti, J.-P. Caumes, G. Sansone, S. Stagira, M. Nisoli, R. Torres, E. Heesel, N. Kajumba, J. P. Marangos, C. Altucci, and R. Velotta, Phys. Rev. Lett. **95**, 153902 (2005).
11. R. A. Bartels, N. L. Wagner, M. D. Baertschy, J. Wyss, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **28**, 346 (2003).
12. S. Varma, Y.-H. Chen, and H. M. Milchberg, Phys. Rev. Lett. **101**, 205001 (2008).
13. J. Wu, H. Cai, H. Zeng, and A. Couairon, Opt. Lett. **33**, 2593 (2008).
14. R. Bartels, T. Weinacht, N. Wagner, M. Baertschy, C. Greene, M. Murnane, and H. Kapteyn, Phys. Rev. Lett. **88**, 013903 (2001).
15. N. Zhavoronkov and G. Korn, Phys. Rev. Lett. **88**, 203901 (2002).
16. A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47 (2007).