Rotational Raman Effects in the Wake of Optical Filamentation

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(Received 11 October 2007; published 27 March 2008)

The spatiotemporal effects generated in the wake of a laser filament propagating in nitrogen are investigated. At suitable time delays, a probe light pulse propagating along the wake experiences a strong spatial confinement and a noticeable spectral broadening at the same time. Numerical simulations, well reproducing the experimental findings, show the key role of the impulsive rotational Raman response in the observed phenomena.

DOI: 10.1103/PhysRevLett.100.123006 PACS numbers: 37.10.Vz, 42.65.Jx, 42.65.Re

Propagation of light filaments in atmosphere has been a subject of intense studies for many applications, including light discharge control and atmospheric remote sensing [1]. Recently, filamentation in air has also emerged as a promising method for generating THz radiation [2]. For these reasons, several theoretical works have been focused on the propagation of intense laser pulses in molecular gases [3,4]. The majority of these studies directly addresses the (rotational) Raman excitation of the molecular medium during the driving laser pulse [5], neglecting the retarded part of the rotational response excited in the wake of the filament. This response shows periodic revivals [6] that can be thought as field-free recurrences of molecular alignment. Field-free alignment has been widely investigated and exploited in the control of high-order harmonic generation (HHG) [7,8], molecular orbital tomography [9], dynamic quasiphase matching of nonlinear optical processes [10], pulse compression [11], and phase shaping of ultrashort pulses [12]. The physics of the rotational response in the wake of a filament is crucial for many applications, like the efficient remote generation of terahertz pulses by two time-delayed overlapping filaments [13] and the filament-based remote sensing in pump-probe configuration [14]. Implications are also envisaged in the high-order harmonic generation inside molecular gas cells under impulsive alignment conditions.

In this Letter we present an experimental investigation of the spatiotemporal effects occurring in the wake of a laser filament propagating in a molecular gas. In particular, we will show that the rotational excitation of the gas affects substantially a time-delayed probe pulse propagating in the wake. Besides a strong spectral modulation, the probe experiences a tight spatial confinement in the core of the wake, which can be compared to the propagation inside a transient waveguide. Both these two aspects are ascribed to phase modulation induced by the pump beam. This unexpected experimental result is well reproduced by numerical simulations taking into account the retarded Raman response of the medium in both temporal and spatial domains.

The experiment has been performed using an amplified Ti:sapphire laser system, generating 800-nm, 60-fs pulses at 10 Hz. The laser pulses were split into two beams: the first one (energy: 0.9 mJ) was focused in a gas cell, generating a filament; the second one was frequency doubled in 1-mm thick β-barium borate (BBO) crystal followed by an IR-absorbing filter (energy: 10 μJ) and was used to probe the effects occurring in the filament wake. A half-wave plate was used to make the pump and probe polarizations parallel. The pump and the probe pulses were separately focused by $f/12$ lenses (with a 0.6 m and a 0.3 m focal length, respectively) and then collinearly combined with a dicroic beam splitter. The spot sizes at $1/e^2$ on the lenses were 1.7 mm and 3 mm for pump and probe, respectively. A translation stage on the probe arm was used to scan the pump-probe delay. The foci of the two beams were overlapped into the cell filled with N_2 at a pressure of 2.5 bar; an adjustable iris was used in order to tune the pump power for stable filament operation. The probe beam at the output of the cell was then separated from the pump using a dicroic beam splitter. The spatial shape and the spectrum of the probe were recorded as a function of pump-probe delay; probe images were acquired using a low noise CCD camera; spectra were acquired selecting the central part of the beam. Although the experiment has been performed in nitrogen, we investigated other molecular gases (CO_2, O_2), which showed similar behaviors.

In a first set of experiments, we measured the probe pulse spectrum as a function of the pump-probe delay. The result is shown in Fig. 1(a). The probe presents a remarkable spectral broadening at periodic delays, which is the signature of a temporal refractive index modulation occurring in the filament wake. This effect is not completely unexpected, since a similar behavior has already been observed in pump-probe experiments performed in a gas-filled hollow waveguide [11]; nevertheless, the noticeable amount of spectral broadening (about 700%) is not compatible with a free propagation of the probe, since its natural divergence would limit the interaction with the filament wake to a small distance. In order to understand

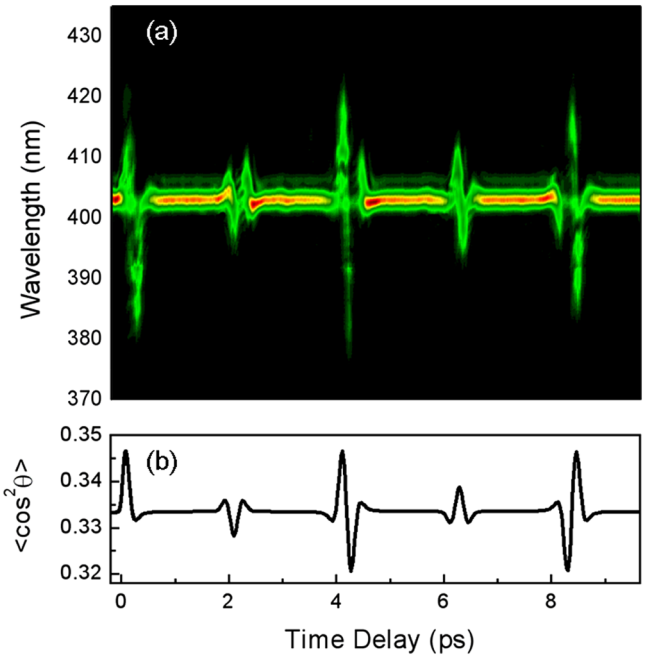

FIG. 1 (color online). Temporal evolution of (a) the experimental output probe spectrum and (b) the calculated alignment factor $\langle\cos^2\theta\rangle$ in the filament wake, as a function of the pump-probe delay.

the physics of this interaction, we must consider the nature of the filamentation process. Filamentation is a propagation effect in which dynamical equilibrium between self-focusing and plasma defocusing takes place [15]. As a result, the pump laser pulse is self-guided, therefore creating a channel in which the pulse intensity is clamped and the spatial shape is cleaned. In a molecular medium, an impulsive excitation of the rotational wave packet also takes place along all the channel in which the pump is confined [16]. It is worth noting that, in our experimental conditions, the ionization level inside the filamentation channel is lower than 1%; thus, the population depletion can be neglected. The refractive index of the medium measured along the polarization direction of the filamenting pulse is given by [11]

$$n^2(\mathbf{r}, t) = 1 + \frac{N}{\varepsilon_0}[(\alpha_{\parallel} - \alpha_{\perp})\langle\cos^2\theta\rangle(\mathbf{r}, t) + \alpha_{\perp}], \quad (1)$$

where N is the molecular density, $\alpha_{\parallel}$, $\alpha_{\perp}$ are the components of the molecular polarizability, $\langle\cos^2\theta\rangle(\mathbf{r}, t)$ is the alignment factor [17], and θ is the angle between the molecular axis and the polarization direction of the pulse.

Equation (1) implies that a probe pulse, having the same polarization direction of the pump and injected collinearly to the wake of the filament at suitable time delays, will experience a spectral broadening, occurring at a maximum or a minimum of a fractional revival [11] (revivals appear in nitrogen with a rotational period $T_r \sim 8.4$ ps), as shown in 1(a) and 1(b). Nevertheless, Eq. (1) clearly shows a coupling between the spatial and the temporal evolution of the refractive index. One can speculate that the transverse refractive index profile could affect substantially the probe propagation, thus accounting for the remarkable spectral broadening we observed.

In order to investigate the role of spatial effects, we focused our study on the evolution of the probe beam profile as a function of the pump-probe delay. Figure 2(a) shows the probe beam pattern at the output of the gas cell without the pump beam, whereas Fig. 2(c) displays the same pattern for some pump-probe delays around the first half revival (~4.2 ps). The calculated difference in refractive index between the central region of the filament wake (aligned molecules) and the not excited region (randomly oriented molecules) is shown in Fig. 2(b) as a function of

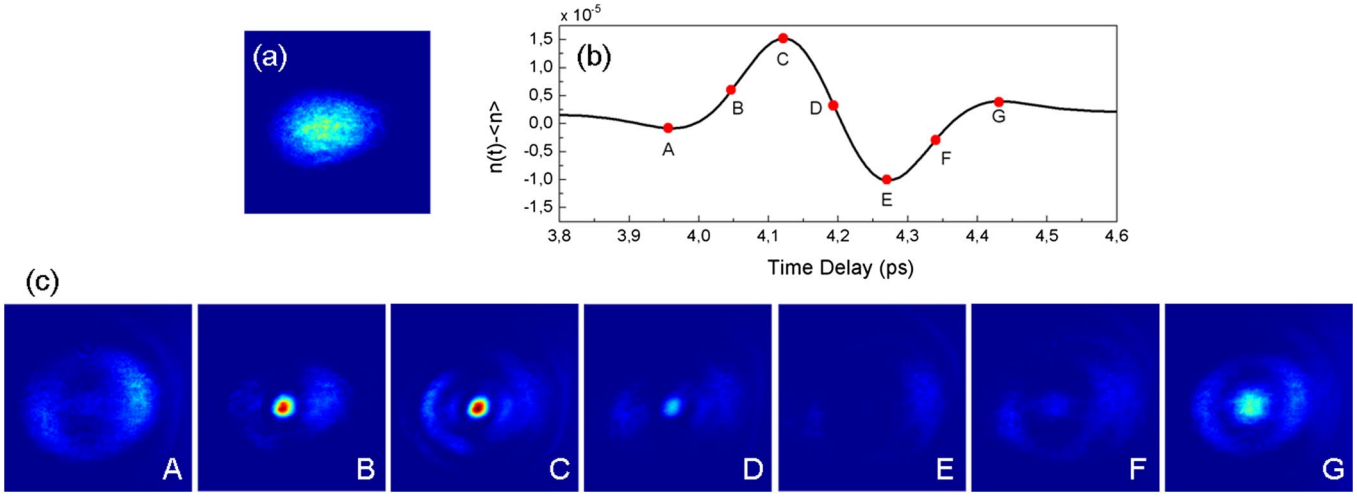

FIG. 2 (color online). (a) Recorded probe beam pattern without the pump beam. (b) Calculated evolution of the refractive index difference between aligned and randomly oriented molecular regions as a function of the pump-probe delay. (c) Measured spatial shape of the probe beam for some pump-probe delays (A–G) marked as red dots in (b).

delay. For a delay coincident with the maximum of the half revival [point marked as C in Fig. 2(b)], the refractive index in the wake reaches its maximum value. Therefore the probe pulse experiences a convex refractive index profile and, as a result, it is guided inside the filament wake. This effect is clearly visible in the recorded pattern displayed in Fig. 2(c), panel C: about 30% of the input energy is coupled in a quite circular and smooth mode. For a pump-probe delay corresponding to the minimum of the half revival [point marked as E in Fig. 2(b)], the refractive index assumes its minimum value. In this case the difference in refractive index between aligned and unaligned regions becomes negative, resulting in an antiguiding profile. As a result, the probe beam is deviated away from the center of the filament wake, and its spatial pattern assumes an annular shape as displayed in Fig. 2(c), panel E. As shown in Fig. 2(c), smaller antiguiding and guiding effects are also present at time delays corresponding to the local minimum and the local maximum of the half revival [marked, respectively, as A and G in Fig. 2(b)]. It is worth noting that the probe pulse spectrum reported in Fig. 1(a) shows a lack of signal at the minima of the $\langle\cos^2\theta\rangle$ curve. This behavior confirms the antiguiding effect: the spectrometer, aligned on the center of the beam, does not detect the deviated light. On the other hand, at the maxima of the $\langle\cos^2\theta\rangle$ curve, the guiding effect combined with the temporal refractive index modulation accounts for the enhanced spectral broadening we observed.

In order to demonstrate that the experimental finding can be assigned to the Raman response excited in the filament wake, we developed a numerical model in cylindrical symmetry for the propagation of pump and probe pulses. The pump filamentation process can be modeled by solving the nonlinear propagation equation for the envelope of the pump electric field $\mathcal{E}_p(r, z, t)$ [18]:

$$\partial_z\mathcal{E}_p + \frac{i}{2k_p}\hat{T}^{-1}(\nabla_\perp^2\mathcal{E}_p) + i\hat{D}\mathcal{E}_p + i\frac{k_p n_2}{2\mu_0 c}\hat{T}(|\mathcal{E}_p|^2\mathcal{E}_p) - \frac{i}{2k_p}\hat{T}^{-1}\left(\frac{\omega_p^2(t)}{c^2}\mathcal{E}_p\right) = 0, \quad (2)$$

where k_p is the pump wave number, z is the propagation distance, t is the local time, n_2 is the nonlinear Kerr index, $\hat{D}$ is the dispersive operator, $\hat{T} = 1 - (i/\omega_0)\partial_t$, $\hat{T}^{-1}$ is the inverse operator of $\hat{T}$, and $\omega_p(t)$ is the instantaneous plasma frequency, which takes into account multiphoton ionization effects. The Raman contribution to the pump propagation was considered negligible for the pulse duration we used. We integrated Eq. (2) using the same parameters of the experiment; the resulting pump pulse distribution was used to determine the alignment factor evolution, according to standard procedures [16,17]. The refractive index modulation was then calculated from Eq. (1) and used to model the propagation of the probe beam in the filament wake. Ionization, self-focusing, and dispersion of the probe were considered negligible. The evolution of the probe pulse envelope $\mathcal{E}_{pr}(r, z, t)$ was modeled as

$$\partial_z\mathcal{E}_{pr} + \frac{i}{2k_{pr}}\nabla_\perp^2\mathcal{E}_{pr} - ik_{pr}[n(r, z, t) - n_0]\mathcal{E}_{pr} = 0, \quad (3)$$

where n_0 is the refractive index of the randomly oriented gas and k_{pr} is the probe wave number. Note that the third term in Eq. (3) takes into account the molecular alignment through the space and time dependent refractive index $n(r, z, t)$.

Figure 3 shows the measured [Fig. 3(a)] and the calculated [Fig. 3(b)] tridimensional beam profile for a pump-probe delay corresponding to the maximum of $\langle\cos^2\theta\rangle$, around the first half revival. The corresponding radial profiles, shown in Fig. 3(c), are in excellent agreement. It is worth noting that the guiding effect allows one to obtain a clean beam profile, as previously mentioned. We show in Fig. 3(d) the comparison between experimental (solid lines) and calculated (dashed lines) probe spectra at the input [light gray (green) and black, respectively] and the output [gray (blue) and light gray (red)] of the gas cell. Although the model is not comprehensive, a good agreement between experimental and calculated spectra is observed. According to our simulations, all the spectral components of the probe beam show almost the same spatial distribution.

The guiding effect is clearly evidenced by the calculated change in refractive index induced by the pump beam at the maximum of the half revival, as shown in Fig. 4(a). The transient profile at this pump-probe delay has a positive Gaussian radial distribution (FWHM 120 μm), and it ex-

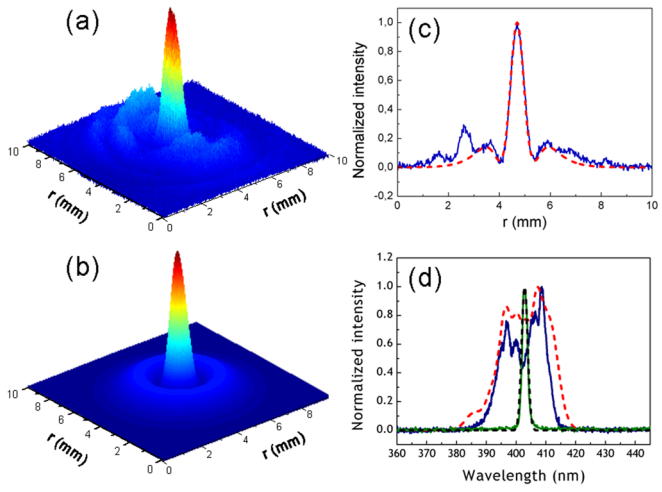

FIG. 3 (color online). Recorded (a) and calculated (b) tridimensional probe beam profile for the pump-probe delay of 4.12 ps. (c) Experimental (solid line) and calculated (dashed line) radial section of the beam. (d) Experimental (solid lines) and calculated (dashed lines) probe spectra around the maximum of the first half revival at the input [light gray (green) and black, respectively] and the output [gray (blue) and light gray (red)] of the gas cell.

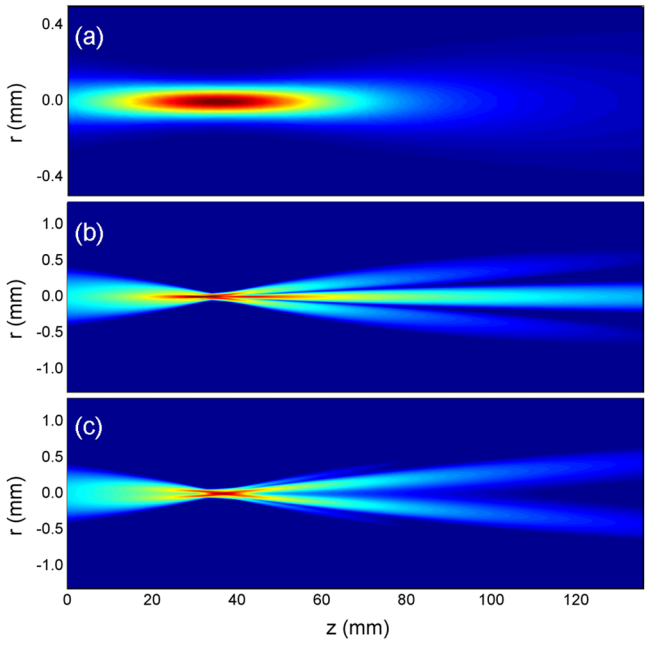

FIG. 4 (color online). (a) Calculated change in the refractive index along the propagation axis at the maximum of the first half revival (linear color map). (b),(c) Calculated radial profile of the probe beam along the propagation axis at the maximum and at the minimum of the half revival, respectively, (logarithmic color map). Note the different vertical scale in (a) with respect to (b)–(c).

tends for about 5 cm along the propagation axis. The change in refractive index at the minimum of the half revival (not shown) is inverted with respect to the previous case. In order to further emphasize the guiding or antiguiding nature of the process, we show in Figs. 4(b) and 4(c) the calculated radial profile of the probe beam along the propagation axis at the maximum and the minimum of the first half revival, respectively. Note that quantities shown in Fig. 4 have been calculated in the reference frame of the probe pulse at a fixed pump-probe delay as a function of the propagation coordinate. As already predicted, a guided propagation is observed in coincidence to the maximum molecular alignment, whereas an antiguiding behavior occurs for the minimum alignment. We can thus exclude that the measured spatial patterns are due to simple lensing effects, since the spatial confinement of the probe, as shown in Fig. 4(b), occurs in the region of the guiding channel reported in Fig. 4(a).

In conclusion, we demonstrated that the retarded Raman response of nitrogen in the wake of a light filament can dramatically affect the spectral bandwidth and spatial pattern of a time-delayed probe pulse. In particular, we showed that light confinement and a simultaneous significant spectral broadening can be achieved in the filament wake at maxima of rotational revivals. Numerical simulations give a deeper insight into this process and well reproduce the experimental results.

We envisage that transient optical guiding by retarded Raman response in molecular materials could be applied to many areas of nonlinear optics, for instance, guideless optical pulse compression and nonlinear generation processes.

We acknowledge the financial support from the Italian MIUR PRIN Project No. 2006027381 and from the Marie Curie research training network XTRA (Grant No. MRTN-CT-2003-505138).

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