Transient Waveguiding in a Rotationally Excited Molecular Gas

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Abstract. Transient waveguiding and spectral broadening of a delayed probe pulse were observed in the wake of a laser filament propagating in Nitrogen and Oxygen. The observed effects are ascribed to the excitation of rotational wavepackets.

Introduction

Propagation of light filaments in atmosphere has been a subject of intense studies [1, 2] for many applications including light discharge control, atmospheric remote sensing and THz waves generation. For these reasons, several theoretical works have been developed on filamentation in molecular gases. However, the majority of these studies neglects the retarded part of the rotational response excited in the wake of the filament. This response shows periodic revivals [3], that can be thought of as field-free recurrences of molecular alignment.

In this work we report on the experimental investigation of the spatio-temporal effects occurring in the wake of a filamenting pulse propagating in a molecular gas. In particular, we demonstrate that the evolution of the rotational wave packet excited by the filamenting pump affects substantially a time-delayed probe pulse propagating in the wake of the filament. 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 [4].

Experimental Methods

The experiment has been performed using an amplified Ti:sapphire laser system, which generates 60-fs pulses at 800 nm with a repetition rate of 10 Hz. The laser pulses were split in two beams: the first pulse (pump, maximum energy 0.9 mJ) was focused in a gas cell, generating a filament; the second pulse was frequency doubled in 1-mm thick BBO crystal followed by an IR-absorbing filter (energy 10 J) and was used to probe the effects occurring in the filament wake. The two beams were collinearly recombined by a dichroic beam splitter and their foci were overlapped into the gas cell. The probe was extracted after the cell by a second dichroic beam splitter; its spatial shape and spectrum were recorded as a function of pump-probe delay. We performed the experiments both in N_2, at a pressure of 2.5 bar, and in O_2 at 2 bar.

Results and Discussion

Figure 1(a) shows the simulated molecular alignment factor induced by the pump pulse in the core of the filament generated in Nitrogen; only the evolution around the first half revival is displayed. A maximum (point A) and a minimum (point B) in the alignment factor are present. Figure 1(b) shows the spectrum of the probe pulse propagating in the wake of the filament at a time delay corresponding to point A (solid line); as can be seen, the probe presents a remarkable broadening with respect to its original spectrum (dashed line) in correspondence of the maximum alignment; this effect is ascribed to modulation of the refractive index due to excitation of a rotational wavepacket in the gaseous medium [5].

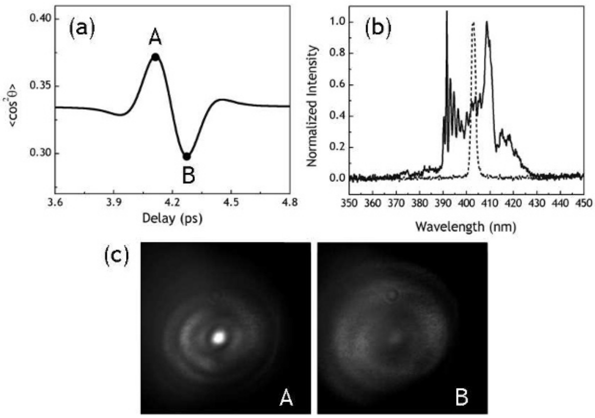

Fig. 1. (a) Simulated alignment factor around the first half revival in N_2 after excitation with the filamenting pump. (b) Probe spectrum with (solid line) and without (dashed line) rotational excitation at time delay A in panel (a). (c) Spatial profiles of the probe beam at delays A and B of panel (a).

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 excited region to a small distance. Nevertheless, as shown on the left of Figure 1(c), the probe beam undergoes a strong spatial confinement in correspondence of the maximum alignment (delay A) since it experiences a convex refractive index profile and, as a result, it is guided inside the filament wake. This spatial confinement extends the interaction of the probe with the rotationally excited gas, thus accounting for the observed spectral broadening. On the other hand, at the minimum of the half revival (delay B), an antiguiding refractive index profile is present. As a result, in this case the probe beam is deviated away from the center of the filament wake and its spatial pattern assumes an annular shape, as displayed on the right of Figure 1(c).

We further explored these effects in Oxygen, where quarter revivals are stronger owing to a different nuclear spin statistic with respect to Nitrogen. Figure 2(a) shows the simulated alignment factor induced by the filamenting pump pulse in O_2 around the first quarter revival; in this case a maximum (delay B) and two minima (delays A and C) are observed. A huge spectral broadening of the probe pulse was observed for a pump-probe delay corresponding to point B, as shown in Figure 2(b). The probe spatial profiles recorded in Oxygen are reported in Figure 2(c); as can be seen, the probe beam is guided in correspondence of the maximum alignment (delay B) and antiguided in correspondence of the minimum alignment (delays A and C).

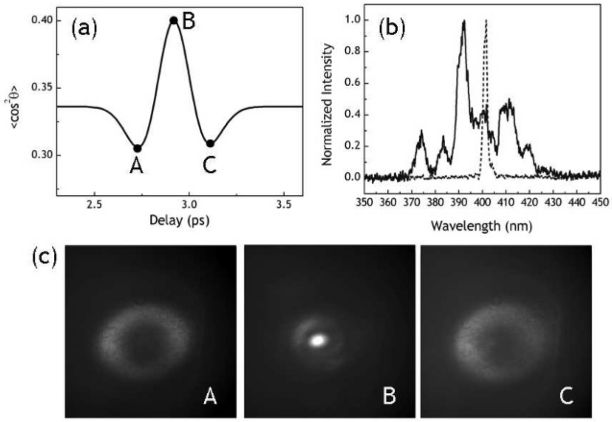

Fig. 2. (a) Simulated alignment factor around the first quarter revival in O_2 after excitation with the filamenting pump. (b) Probe spectrum with (solid line) and without (dashed line) rotational excitation at time delay B in panel (a). (c) Spatial profiles of the probe beam at delays A, B and C of panel (a).

In order to demonstrate that the experimental finding can be assigned to the rotational Raman response excited in the filament wake, we developed a numerical model in cylindrical symmetry for the propagation of pump and probe pulses. Calculated probe beam evolution, performed for different pump-probe delays, is in very good agreement with experimental findings and shows that a guided propagation in the filament wake is obtained at maxima of rotational revivals.

Conclusions

In conclusion, we demonstrated that the retarded Raman response of a molecular gas in the wake of a light filament can dramatically affect the spectral bandwidth and spatial pattern of a collinear 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 for Nitrogen and Oxygen.

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