Quantum molecular lensing of femtosecond laser optical/plasma filaments

S. Varma, a,b Y.-H. Chen, and H. M. Milchberg

Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, Maryland 20742, USA

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The long-range filamentary propagation of intense femtosecond pulses in atmosphere has been observed for the first time to be strongly effected by quantum rotational wave packets. A two-pulse experiment shows that a filamenting probe pulse can be steered and trapped in or destroyed by the rotational alignment wake following a pump filament. © 2009 American Institute of Physics. [DOI: 10.1063/1.3078105]

I. INTRODUCTION

Long-range filamentary propagation of femtosecond pulses in the atmosphere occurs due to the dynamic balance between nonlinear self-focusing of an intense optical pulse and laser plasma-induced defocusing. The effect, which is self-sustaining and can propagate in atmosphere over distances from a few centimeters to hundreds of meters, was first demonstrated in 1995 (Ref. 1) and was subsequently studied by many groups. Copropagating white light generation and a trailing plasma tail accompany these long-range filaments. The filamentary process can be unstable, and several methods have been proposed to control the formation of single and multiple filaments, which can be used for several applications including remote ionization, broadband light generation and atmospheric monitoring, remote terahertz generation, and guiding of electrical discharges. Applications of shorter-range filamentation in gas cells and jets include continuum generation, high-harmonic generation, and pulse compression.

This paper presents experimental results showing that the propagation of an intense probe pulse filamenting in air is dramatically affected by the molecular alignment quantum wake following a pump pulse filament. For slight angular pump/probe misalignment, the probe filament is transversely pulled and focused into the pump filament path or destroyed by the wake. The delay between pump and probe pulses, which determines the phase of quantum molecular alignment seen by the probe pulse, determines whether the probe is trapped and focused or destroyed. Although the air nonlinearity for single pulse filamentation is comprised of both a nearly instantaneous response due to bound electronic response and a delayed inertial molecular response, the results confirm that for pulse lengths >100 fs, the rotational component is fast and strong enough to be the overriding effect. Measurements of the spectrum of the probe pulse are consistent with quantum wake trapping. The coherent temporal and spatial response of air molecules can be used to control long-range filamentary propagation of ultrafast optical pulses.

II. ROTATIONAL WAVE PACKET EXCITATION IN ATMOSPHERIC Constituents

Nitrogen and oxygen are linear molecules that rotate in intense laser fields due to the fact that their induced dipole moments depend on the orientation of their molecular axes with respect to the laser polarization. Consequently, randomly aligned N2 and O2 molecules experience a net torque in an intense laser field toward alignment with the field. This effect is significant in molecular ensembles at room temperature up to multiple atmospheres of pressure.

The molecular alignment by the electric field can be expressed as the excitation of a quantum-mechanical wave packet \( |\phi\rangle = \sum_{j,m} e^{-\text{i} \omega j} \text{e}^{-\text{i} \omega t} |j, m\rangle \) of rotational states \( |j, m\rangle \) of energy \( E_{j,m} = h \omega j = \hbar B j (j + 1) \), where \( j \) is the quantum number for total rotational angular momentum, \( m \) is the momentum component along the laser field, \( B \) is Planck’s constant, and \( B \) is the rotational constant of the molecule. The free electrons form a negative lens that arrests self-focusing and causes refractive beam divergence. The third-order nonlinearity eventually causes sufficient nonlinear phase to accumulate, and then self-focusing and plasma-induced divergence repeat. In this way, both the laser intensity and the plasma density are limited to maximum values—the laser intensity in air is clamped at \( \sim 5 \times 10^{13} \text{ W/cm}^2 \) (Ref. 2) and the gas ionization fraction at \( \sim 10^{-4} - 10^{-3} \). These experimental measurements have been confirmed by simulations. Over the course of many meters of propagation, the plasma filament can retain a diameter of tens of microns.

bInvited speaker.
alignment by the pump. In this case, we replace \( \langle \cos^2 \theta \rangle \) with \( \frac{1}{2}(1-\langle \cos^2 \theta \rangle) \).

The difference in phase between the center and waist of a Gaussian beam a Rayleigh length \( z_0 \) after focus is of order unity. Over the same distance, an on-axis refractive index bump \( \Delta n \) would retard the phase of the center of the beam by \( k \Delta n z_0 \), where \( k \) is the vacuum wave number. A fractional increase in the index of refraction at the center of a beam \( \Delta n \) will compete with beam diffraction if \( \Delta n > (k z_0)^{-1} \). Therefore, a laser beam launched with a number of pulses can be significantly refracted by atmospheric molecular alignment, where \( f_{...,\text{molec}} \) is an effective number due to \( \Delta n \) from alignment. For example, at the peak of the air quantum revival near \( t = 8 \) ps, which is comprised of the full revival of \( N_2 \) and the 3 \( \frac{1}{2} \) revival of \( O_2 \), \( \langle \cos^2 \theta \rangle \) may be significantly longer than the molecular rotation period, giving \( n_{...,\text{molec}} \sim 3 \times 10^{-6} \) at atmospheric pressure, and giving \( f_{...,\text{molec}} \sim 200 \). A molecular effect on propagation then requires \( f_{...,\text{molec}} > 200 \).

The dominant ultrastiff contribution of rotational quantum states to the refractive index is often overlooked. A wave packet is a coherent sum of rotational states up to the maximum rotational quantum number \( J_{\text{max}} \). At room temperature and \( I = 4 \times 10^{13} \) W/cm\(^2\), \( J_{\text{max}} \sim 20 \). The refractive index response time due to the alignment effects of such a wave packet is \( \Delta t \sim 2T/J_{\text{max}} \), giving \( \Delta t \sim 40 \) fs. This time scale is confirmed by the inset of Fig. 3(a), which shows a center lineout of the air response from Fig. 1. Molecular alignment dominates the nonlinear response of air unless a pulse is appreciably shorter than \( \Delta t \), in contrast to earlier work, in which the effective response time is typically shorter than the beam filamentation time.

### III. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 2. A dual polarization interferometer (DPI) is used to produce two orthogonally polarized pulses with tunable energy from a single input pulse. The delay between the two pulses is adjusted using a computer controlled stepper motor on the retroreflecting arm. The alignment of the two arms of the DPI can be individually tuned for collinear propagation, though in some experiments the two pulses were deliberately angularly separated. The two pulses, each 800 nm, 130 fs, and with a few millijoules of energy, are apertured to 1 cm diameter and focused by a 3 m lens at \( f_{\text{L}} \sim 300 \). The dual pulse beam then propagates 6 m across the laboratory and enters a filament profile imaging system (not pictured). This system uses two glass wedges to attenuate the beam intensity, a cube polarizer to remove the pump pulse, and a lens which images the probe filament onto a charge coupled device camera. The filament is typically \( 2 \) m long and the imaging system images a plane 25 cm after the end of the filament. Figure 2 also shows a sequence of single beam images on a far-field screen, taken by a digital camera. These images show the
onset of filamentation and white light generation up to $2.8P_{cr}$ (using $P_{cr}=10$ GW).

### IV. RESULTS AND DISCUSSION

The strong effect of molecular alignment on the filamentary propagation of the probe pulse is first demonstrated by an experiment in which the probe pulse copropagates with the pump pulse at delays near $t \sim 0$, where the pump and probe can be temporally overlapped. The vertically polarized pump pulse ($1.4 \text{ mJ}, 1.1P_{cr}$) generates a filament which is preceded or followed by the horizontally polarized probe filament ($2 \text{ mJ}, 1.5P_{cr}$). The results are shown in Fig. 3(a), where slices of the image of the probe filament are plotted versus delay. Thirty images were taken at each 30 fs intervals, averaged and converted into slices by integrating the two-dimensional (2D) profiles along their vertical axis. Sample 2D images are shown at several delays. A strong probe pulse filament is seen, as expected, well before $t=0$, but within approximately 50 fs of overlap, the probe filament is extinguished [location A in Fig. 3(a)] and remain so until $\sim 50$ fs beyond overlap, at which point the strong probe filament returns. This is proof that ionization-induced defocusing is not the cause of probe filament destruction, since the filament recovers in $\sim 100$ fs, whereas the plasma lifetime is many nanoseconds.\textsuperscript{13} The temporary destruction is a direct result of the orientational contribution to the nonlinearity of the neutral gas. If the probe was polarized parallel to the pump, the molecular alignment from the pump would focus the probe near $t=0$. However, in this experiment, the probe is perpendicularly polarized and would defocus. The $\sim 100$ fs interval over which the probe filament is destroyed is consistent with the molecular defocusing interval. Figure 3(b) repeats the format of Fig. 3(a) for 330 fs pulses with pump/probe energies of $5.85 \text{ mJ}/2.75 \text{ mJ}$ ($3.7P_{cr}/4.6P_{cr}$) using 85 fs delay steps. The probe filament disruption interval is slightly broader, which is expected due to the longer pump pulse.

The effect of the pump filament rotational wave packet excitation on the probe filament can be decoupled from the pump pulse’s bound electronic effect by examining the probe filament at delays much later than $t=0$ that coincide with recurrences of the molecular alignment. Figure 4(a) shows results near $t=8$ ps, near which the probe pulse experiences the summed effects of the full revival of $N_2$ and the $\frac{3}{4}$ revival of $O_2$. The pump/probe energies are $1.4 \text{ mJ} (1.1P_{cr}/2.5 \text{ mJ}$)

![FIG. 2. (Color online) Setup showing the DPI. QWP=quarter-wave-plate; HWP=half-wave-plate; TFP=thin film polarizer. The right panel shows beam images of filamentation and white light generation for increasing pulse power up to $\sim 2.8P_{cr}$.](image1)

![FIG. 3. (Color online) (a) One-dimensional (1D) integrated images of the probe filament as a function of delay (30 fs steps) through the pump-probe temporal overlap ($t=0$). The laser pulse width is 130 fs. Pump/probe energies are $1.4 \text{ mJ}/2 \text{ mJ} (1.1P_{cr}/1.6P_{cr})$. Each image slice is a 30-shot average. Selected 2D images are shown as insets, as is a beam center lineout of the air alignment response (black line) and the pump pulse (red squares) in the $t=0$ region. (b) Scan as in (a) but with 330 fs laser pulses, 85 fs delay steps, and pump/probe energies of $5.85 \text{ mJ}/7.25 \text{ mJ} (3.7P_{cr}/4.6P_{cr})$.](image2)

![FIG. 4. (Color online) (a) 1D integrated images of probe filament as a function of delay (30 fs steps) through the air revival near $t=8$ ps, comprised of the $N_2$ full revival ($t=T_{21}=8.3$ ps) and the $O_2$ three-quarter revival ($t=2T_{21}=8.7$ ps). Pump/probe energies are $1.4 \text{ mJ}/2.5 \text{ mJ} (1.1P_{cr}/2P_{cr})$. The gray dashed line shows the position of the center of the pump filament. Selected 2D images are shown as insets, as is a lineout of the air alignment response in the $t=8$ ps region. (b) Pump pulse energy scan ($0.7P_{cr}$ to $2.5P_{cr}$), holding probe energy constant at $4.35 \text{ mJ} (3.4P_{cr})$. The bottom panel has pump/probe energies of $4.65 \text{ mJ}/3.05 \text{ mJ} (3.6P_{cr}/2.4P_{cr})$.](image3)
(1.9P_cr), and the two pulses are angularly separated by ∼0.1 mrad. The gray dashed line shows the transverse position of the center of the pump filament, and therefore also highlights the regions of maximally aligned air molecules. Here we see, depending on delay, that the probe filament cannot only be extinguished but it can be trapped and enhanced. The inset in Fig. 4(a) shows the molecular alignment near t =8 ps. From this, it is clear that the temporal evolution of the molecular alignment maps exactly onto the focusing and defocusing intervals of the probe filament. The delays at which the probe is steered into the pump beam path and enhanced (labeled A and B in the figure) correspond precisely with antialignment peaks in the inset. The inset shows alignment as it would be seen by a parallel polarized probe, so for the perpendicularly polarized probe experiment, as expected, we see the opposite effect. The filament destruction at locations C and D are coincident with molecular alignment. The probe filament returns to its original position after the revival. Full images of the probe filament at selected delays are shown as additional insets. Depending on pump-probe alignment and on a shot-by-shot basis, the probe filament “destruction” is seen as either wild swings of the filament position or as filament breakup. The steering effect on the probe filament is analyzed by examining off-axis ∆n from molecular alignment. Trapping can occur if k∆nL ~1, where L is the filament length and is approximately 2 m. The off-peak ∆n required for trapping is then ~6 × 10^{-8} (0.002 of the measured ∆n_peak value at r=0). If the pump and probe are misaligned by 0.1 mrad at their source, the DPI, they are separated by ∼350 µm at the onset of filamentation. If the molecular lens profile corresponds to a filament full width at half maximum diameter of ∼100 µm diameter, then ∆n/∆n_peak ~0.002 at r~200 µm, and the probe filament will have some spatial overlap with enough molecular alignment to be steered and trapped in the wake of the pump pulse.

Figure 4(b) shows the result from a repeat of the delay scan of the probe pulse near t=8 ps seen in Fig. 4(a), with the probe energy fixed (4.4 mJ, 3.4P_cr) and varying pump energy. At the lowest pump energy (0.85 mJ, 0.7P_cr), the molecular effect is not evident. This is reasonable, since a nonfilamenting pump pulse will only be intense enough to significantly align air molecules over its Rayleigh range, which is ∼10 cm and much shorter than the filament length. Notably, as the pump energy is increased to values greater than that of the probe, the steering, enhancement, and destruction of the probe filament remain evident. The visible robustness of the molecular effect over such a wide range of pump and probe powers is discussed below.

Figure 5 again shows the probe filament profile near t =8 ps but in the case where the pump pulse (3.6 mJ, 2.8P_cr) and the probe (4.6 mJ, 3.6P_cr) are not misaligned. The significant aspect of these data is the reservoir of laser energy surrounding the filament. It is evident that the enhancement [labeled A in the data and corresponding to the Fig. 4(a) inset] of the probe filament comes at the expense of the reservoir. Conversely, molecular antialignment (labeled C) clearly expels light into the reservoir.

Figure 6 shows the result of a second experiment, where we copropagate two 130 fs pulses with the same polarization. This is done by inserting a Mach–Zender interferometer between the stretcher and the regenerative amplifier of the Ti:sapphire chirped-pulse amplification (CPA) system. The system output is two perfectly collinear pulses, each 3.3 mJ (2.6P_cr) with adjustable delay. Due to their parallel polarization, the pump cannot be passively filtered to extract the probe response. However, the pump filament little affects the probe filament beam profile at delays detuned from the molecular alignment revivals, so a combined image of pump and probe can be taken at one of these delays and halved in intensity to construct the image of the pump alone. This can then be subtracted from all combined pump-probe images to yield the probe image. The effective molecular alignment seen by the probe pulse in this case should be double that from the first experiment, because alignment is symmetric only around the input pulse polarization. In the first experiment, the “aligned” molecules are actually equally likely to be oriented in the horizontal direction and the direction of propagation. The enhancement effect at position A is greater than that from the experiment with perpendicularly polarized filaments, although shot to shot fluctuations prevented observation of a stronger enhancement.

White light generation is a feature of filamentation that is also enhanced when the probe filament is trapped in a quantum molecular wake. This effect is demonstrated in Fig.
is a time-dependent refractive index shift and therefore contributes phase modulation in the form of linear and quadratic frequency shifts: $$\Delta \omega_{\text{lin}} \sim -\frac{\partial \Phi}{\partial t}$$ and $$\Delta \omega_{\text{quad}} \sim -\left(t-t_0\right) \times \left(\frac{\partial^2 \Phi}{\partial t^2}\right)_{\text{ref.}}$$ where $$t=t_0$$ is the peak of the local alignment at a point such as location A in Fig. 4(a). The probe pulse propagating alongside the revival in the vicinity of this delay will experience both $$\Delta \omega_{\text{lin}}$$ and $$\Delta \omega_{\text{quad}}$$. If the probe is positioned at the alignment inflection points, the revival temporal profile gives both positive and negative $$\Delta \omega_{\text{lin}}$$. When the probe pulse is nested precisely in the alignment waveform at intervals A or B, it experiences $$\frac{\partial^2 \Phi}{\partial t^2} > 0$$, which explains the redshift (for $$t > t_0$$) following the blueshift (for $$t < t_0$$) at A and B.

To complete the physical picture of the molecular effect on filament propagation, we must also consider plasma generation. The lens and molecular alignment focusing effects are additive, so $$f_{\#} = f_{\#}^{\text{lens}} + f_{\#}^{\text{molec}} \sim 200$$ combine to give $$f_{\#}^{\text{eff}} = f_{\#}^{\text{lens}} + f_{\#}^{\text{molec}} \sim 1/120$$. Plasma-induced beam defocusing only competes with molecular and lens focusing for $$\frac{1}{2}(2\Delta \omega_{\text{plasma}})^{-1/2}$$, where $$\Delta \omega_{\text{plasma}} = N_e/2N_{\text{cr}}$$, $$N_e$$ is the electron density, and $$N_{\text{cr}} = 1.7 \times 10^{21} \text{ cm}^{-3}$$ is the critical density for an 800 nm wavelength. For our $$f_{\#}^{\text{eff}}$$, $$N_e/N_{\text{cr}} < 1.7 \times 10^{-5}$$. This is a fractional ionization of $$< 10^{-3}$$ at atmospheric pressure, consistent with earlier measurements.2,13

So even at the highest laser powers, essentially all molecules survive, explaining the robustness of the molecular effect.

V. SUMMARY

We have demonstrated that ultrafast rotational quantum wave packet excitation of $N_2$ and $O_2$ significantly affects the long-range filamentary propagation of a femtosecond optical pulse in atmosphere. The initial alignment of air molecules is prompt enough to dominate the nonlinear response and filamentation of pulses with duration $$> 100$$ fs. Additionally, the molecular alignment revivals following a pump filament persist over long time durations, are robust over a wide range of pulse energies, and can be effectively used to manipulate the intensity, position, and spectral content of an intense probe filament. The enhancement of the probe filament intensity may result in an increase in filament length and electron density.

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