Experimental Tests of the New Paradigm for Laser Filamentation in Gases

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Since their discovery in the mid-1990s, ultrafast laser filaments in gases have been described as products of a dynamic balance between Kerr self-focusing and defocusing by free electric charges that are generated via multiphoton ionization on the beam axis. This established paradigm has been recently challenged by a suggestion that the Kerr effect saturates and even changes sign at high intensity of light and that this sign reversal, not free-charge defocusing, is the dominant mechanism responsible for the extended propagation of laser filaments. We report qualitative tests of the new theory based on electrical and optical measurements of plasma density in femtosecond laser filaments. Our results consistently support the established paradigm.

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Since the first experimental observations of filament propagation in air in the mid-1990s [1], the field of ultrafast laser filamentation in gases has matured and enabled various technologically important applications including few-cycle optical pulse generation [2], terahertz generation [3], and remote sensing [4]. In parallel with experimental progress, the theory and simulation of laser filamentation has advanced greatly, allowing, for example, for modeling of the supercontinuum generation and third-harmonic generation that accompany filament propagation [5]. Given this tremendous progress, it is still fair to say that the basic paradigm of filament formation has remained unchanged from the mid-1990s. Namely, the self-focusing collapse of a high peak power input pulse in a gas due to the nonlinear Kerr effect is arrested by the defocusing action of free electrons that are generated on the beam axis via multiphoton ionization [6,7].

The established ''old'' paradigm has been recently challenged by a suggestion that Kerr self-focusing in various gases can saturate and even become defocusing as the optical intensity is increased [8]. This suggestion represents a radical departure from our past understanding of filament formation. Although the saturation of the Kerr effect has been suggested to be a factor in laser filamentation [9–11], it was never assumed to be the dominant one. If proven valid, the ''new'' paradigm would have profound implications in filamentation science. On one hand, the sign reversal of the Kerr effect would enable plasma-free filament propagation, which could be much longer ranged, owing to the much reduced energy losses into ionization compared to that in the established filamentation scenario [12]. In addition, various nonlinear optic conversion processes inside filaments, such as third- and fifth-harmonic generation, could become very efficient [13]. On the other hand, applications that rely on significant plasma generation inside filaments would be severely hampered if the new theory prevails.

The goal of this Letter is to present comprehensive experimental tests of the new paradigm. Our results show that defocusing by free electric charges is certainly strong enough to balance Kerr self-focusing in femtosecond laser filaments, thus supporting the established paradigm. Since our experiments are designed such that they measure qualitative, not quantitative, differences between the predictions of the established and the new theories, our conclusions should not be affected by the variability of the material parameters. We stress that our findings do not imply that the higher-order nonlinear terms advocated in Ref. [8] are nonexistent. Our conclusion is that, even if the higher-order nonlinear terms do exist, the free-charge generation and the associated defocusing in a filament occur early enough to mask their effect, thus rendering them inoperative.

Our first test is based on the electrical conductivity of filaments. Direct and quantitative measurements of the density of charges generated through laser filamentation are problematic owing to both the very high intensity inside filaments and the short lifetime of the generated plasma. Accordingly, the reported values of plasma density inside filaments vary by several orders of magnitude (see, for example, [6]). Furthermore, plasma density inside filaments has been shown to be strongly dependent on the external focusing conditions [14]. An attempt to validate or disprove the new filamentation paradigm based on a quantitative measurement of electric charges generated through filamentation would be very challenging. Luckily, for a particular pair of gases, namely, air and argon, the old and new filamentation theories predict vastly different values of the generated free-charge density, thus offering an opportunity to validate the new theory based on a semiqualitative measurement.

Indeed, by examining the experimental data on the nonlinear refractive index as a function of the intensity of light that has been reported for various gases in Ref. [8], one notices that the curves for air and argon are essentially identical. This close similarity extends well into the intensity range in which, according to the new filamentation theory based on these very data, the Kerr effect changes sign and becomes defocusing. At the same time, the ionization potential of argon is considerably higher than that of air. For laser pulses at 800 nm that we use in our experiments, it takes 11 photons to ionize argon, while ionizing oxygen, one of the major constituents of air, requires only 8 photons.

According to the above considerations, if the filament is indeed stabilized by sign reversal of the Kerr effect (new theory), then filaments generated in air and argon under otherwise identical conditions would have very similar spatial intensity distributions. Plasma, which is a bystander in this scenario, would be generated in proportion to the multiphoton ionization rate. In that case, one would expect substantially more plasma to be generated through filamentation in air than in argon.

On the other hand, if plasma defocusing is the major stabilizing mechanism in filaments and the Kerr effect is always positive (old theory), then filamentation in air would generate less plasma than that in argon. In this scenario, the higher ionization potential of argon would cause plasma generation to be initiated later in the focusing cycle. The filament in argon would be thinner than in air. It would take more plasma to defocus this thinner filament because it has higher peak intensity and associated stronger self-focusing.

The qualitative differences highlighted above between plasma generation according to the new and old theories are supported by the simulations involving 30 fs pulses in air and argon reported in Ref. [12]. In particular, for the new theory these simulations predict about 10 times as much plasma in air as in argon, whereas for the old theory these simulations predict about twice as much plasma in argon as in air.

To experimentally verify the validity of the above predictions, we initiate filaments in air and argon under atmospheric pressure, using 35-fs-long laser pulses with 800 nm center wavelength and various pulse energies. The laser beam with a 1 cm diameter is weakly focused with a lens of 190 cm focal length, making the focusing conditions similar to those used in the simulations reported in Ref. [12]. To measure linear plasma density, in arbitrary units, we use a capacitive plasma probe schematically shown in the top portion of Fig. 1 and described in detail elsewhere [15]. In this particular case, the probe has 1 cm $\times$ 1 cm square electrodes separated by 1.5 mm. The dc voltage applied to the electrodes is 200 V. The impulse voltage response of the probe has the peak value proportional to the local plasma density between the electrodes. The results of the plasma density measurements in air and in argon are shown in the bottom part of Fig. 1. It is evident that, in all cases, plasma density generated in argon is higher than in air, in agreement with the established filamentation theory.

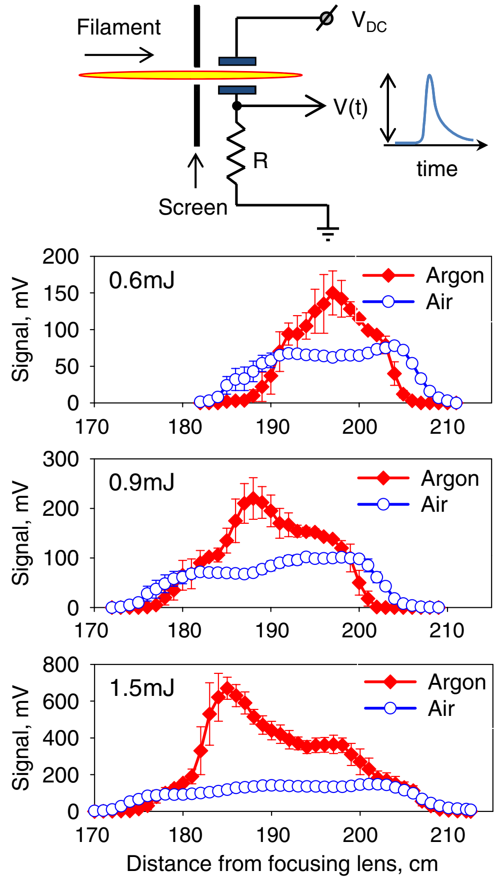

FIG. 1 (color online). Top panel: Schematic of the capacitive plasma probe. Bottom panels: Experimental data for plasma density generated through filamentation in air and argon under identical conditions, for three different values of pulse energy.

Owing to the semiqualitative nature of the above test, the end result confirming the validity of the established theory appears sufficiently conclusive. However, the weakness of this test is related to the fact that the plasma probe that we use is gas-specific. Only a small fraction of the generated charges is captured by the probe for each individual laser pulse. The measured electrical signal is related not only to the probe geometry and plasma density in the filament but also, in some nontrivial way, to the free-electron mobility and recombination rate in a particular gas. Although these parameters are similar for air and argon, they are certainly not identical for the two gases [16]. Thus strictly speaking, using this plasma probe for quantitative comparison of filament properties in different gases is not entirely justified.

Our alternative test of the new theory is based on the diffraction of a collimated probe beam on a plasma channel generated through filamentation in argon. Argon is an atomic gas, and its nonlinear optical response is free from the rotational Raman component inherent to molecular gases such as air [17,18]. Thus the refractive-index perturbation left after the passage of the intense femtosecond laser pulse in argon is exclusively due to the free charges generated through filamentation.

The experimental setup is similar to the one reported in Ref. [19]. Filaments are generated in a 1-m-long tube filled with argon at atmospheric pressure. The filamentation conditions are the same as those in the filament conductivity experiments described above: 35-fs-long pulse, 1 cm input beam diameter, weak focusing with a 190 cm focal-length lens. The pulse energy is fixed at 0.9 mJ.

A fraction of the incident 800 nm beam is split off, frequency doubled in a 200-μm-thick, $1\ \mathrm{cm} \times 1\ \mathrm{cm}$ β-barium borate crystal, and used as a collimated probe beam. We estimate that the probe pulse has a duration of less than 50 fs. The energy of the probe pulse is about 10 μJ; thus, the probe propagates in a linear regime. In the experiments reported here, polarizations of the pump and probe pulses are parallel.

The collimated probe beam, after being combined with the pump in a dichroic mirror, propagates collinearly with the pump. The time delay between the pump and the probe pulses is controlled via a mechanical delay line. The probe beam diffracts on the pump-generated filament, and the resulting diffraction pattern is photographed by a linear CCD camera placed at a distance of 75 cm from the center of the filament. The 800 nm pump light incident on the camera is blocked by a color-glass filter. In this setup, the probe pulse nondestructively samples index changes that are experienced by the intense pump pulse as it undergoes filamentation.

To ensure that the results presented are entirely due to the argon gas, we need to rule out plasma defocusing in the glass windows of the cell as a factor [20]. In our setup we use $L = 150$-μm-thick cell windows made of borosilicate glass with a band gap of 4 eV. The pump beam at the entrance window has peak intensity $\simeq 4 \times 10^{11}\ \mathrm{W/cm^2}$ and radius $a = 1.25$ mm. The power density is safely below the threshold for optical damage. Ionization in the window by the pump pulse at 800 nm is a three-photon process. Using the ionization data reported in Ref. [21] and a pulse duration of 35 fs, we estimate the peak plasma density generated in the window to be $2 \times 10^{15}\ \mathrm{cm^{-3}}$. This plasma density in turn leads to an on-axis phase shift of $\phi \simeq -3.5 \times 10^{-4}$ rad over the window thickness L at the probe wavelength $\lambda_p = 400$ nm. Approximating the radius of the plasma channel in the window as $a = 1.25$ mm, we estimate the effective focal length due to plasma defocusing as $f \approx \pi a^2/\lambda_p \phi = -35$ km. This estimate shows conclusively that plasma defocusing in the entrance window plays a negligible role in our experiment.

In the filamentation zone, the incident probe beam is much broader than the transverse refractive-index distribution due to the pump-induced Kerr effect and plasma. Thus we may treat diffraction of the probe as scattering of an incident plane wave by a line source representing the refractive-index perturbation, each longitudinal position along the filament giving rise to an outgoing spherical wave. Under our experimental conditions, the longitudinal extent of this line scatterer is much larger than the Rayleigh range corresponding to its transverse dimensions. Furthermore, the scatterer has a complex and time-dependent transverse profile: In both cases of the old and new filamentation theories, the periphery of the filament is focusing, while the net effect of the filament center is defocusing. The interference between the collimated incident probe beam and the spherical waves coming from various points along the filament gives rise to a complex far-field beam pattern. The fringes in the pattern are not expected to be uniform near the beam axis but should become more regular, though dumped, away from the axis.

We conducted extensive numerical simulations of our experiment by using the old and new filamentation theories. Both models correctly predict the total filament length of about 20 cm. For peak values of the generated plasma density, the old and the new models yield 3×10^{16} and $1 \times 10^{15}\ \mathrm{cm^{-3}}$, respectively. The former value is consistent with several previously reported experimental results, including the most recent report [22].

We found that fine details of the generated diffraction patterns near the beam axis, in particular, for the cases of small pump-probe delays, depend on experimental conditions and model parameters. The resulting far-field patterns of the probe beam, in general, have aperiodic distributions of interference fringes. Away from the beam axis, however, the simulated patterns are more regular and much less dependent on the material parameters.

In Fig. 2, we show the results of the simulation using the old theory, which is based on plasma defocusing as the main regularization mechanism in laser filaments. The curves show differences between radial intensity distributions of the probe pulse with and without the pump pulse present. At zero delay between pump and probe pulses, the probe experiences the focusing action of the strong pump beam. The corresponding diffraction pattern has a maximum on the beam axis. At large pump-probe delays (8 ps in this case), the pump pulse is gone, and the probe is defocused by free electrons left in the wake of the filament. The resulting diffraction pattern has a minimum on the beam axis. Near the origin, the oscillation

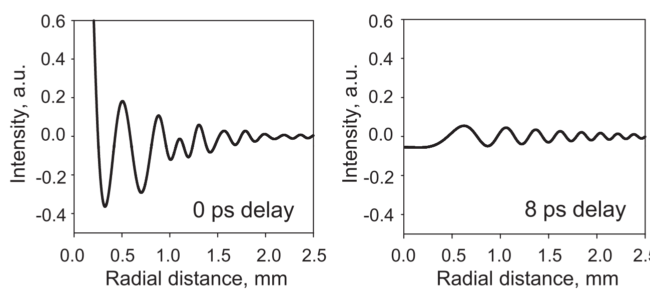

FIG. 2. Radial intensity variations of the probe pulse in argon that have been numerically simulated by using the old theory (plasma defocusing), for two different values of the pump-probe delay. Both patterns are plotted on the same scale. The oscillations of the peripheral parts of the two patterns have comparable amplitudes.

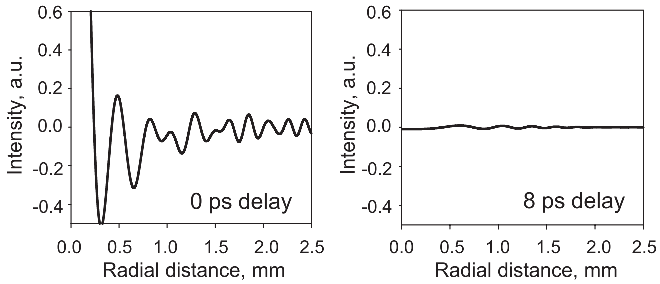

FIG. 3. Radial intensity variations of the probe pulse simulated by using the new theory (sign reversal of Kerr effect). Both patterns are plotted on the same scale. The oscillations in the pattern at 8 ps delay are essentially invisible.

amplitude in the top pattern is much larger than in the bottom one. This difference is not representative of the relative magnitudes of the Kerr focusing and plasma defocusing effects. (They dynamically balance each other in the old theory, on which this simulation is based.) The oscillations of the peripheral parts of the two patterns have similar amplitudes.

Results of the simulation based on the new theory are shown in Fig. 3. The pattern corresponding to zero time delay between the pump and probe looks more irregular than the one computed by using the old theory. For large pump-probe delays, the diffraction pattern is essentially absent. This is to be expected, since, in this case, the sign reversal of the Kerr effect stabilizes the filament before any appreciable plasma is produced.

We emphasize that the purpose of these simulations is not to quantitatively reproduce fine details of the diffraction experiments. Instead, the simulations single out a qualitative feature that can be used for the discrimination between the two filamentation theories. This feature is the oscillation amplitude of the peripheral parts of the diffraction patterns for large pump-probe delays. The amplitude of these oscillations is similar to that for the case of zero delay in the old filamentation theory, and it is vanishingly small in the new theory.

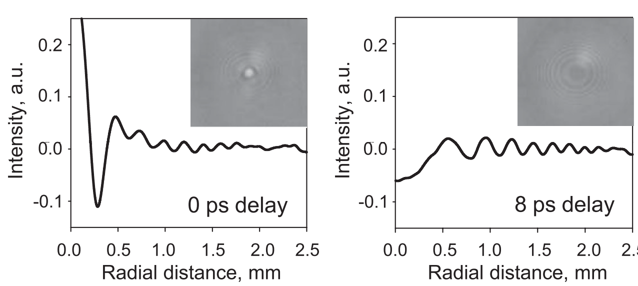

FIG. 4. Experimental data for filamentation in argon under the same conditions as those used in the simulations shown in Figs. 2 and 3. The radial profiles of the probe pulse diffracted on the filament are plotted on the same scale for 0 ps delay (left) and 8 ps delay (right), between the pump and probe pulses. The insets show actual photographs of the probe beam in the two cases.

The experimental results for the case of filamentation in argon under the conditions used in the above simulations are shown in Fig. 4. The curves shown are obtained by digitizing the photographed diffraction patterns along the line passing through the pattern's center and subtracting the profile obtained with the pump blocked. It is evident that the experimental results agree with the simulation based on the established theory and completely disagree with the new theory.

In conclusion, we have conducted two independent experiments testing the new filamentation paradigm, according to which a sign reversal of the Kerr effect is the dominant physical process that stabilizes laser filamentation in gases. Both experiments were designed to test qualitative, not quantitative, differences between the new paradigm and the established theory based on plasma defocusing. Both of our tests consistently disqualified the new paradigm and confirmed the established theory.

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