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Abstract: Short lived plasma channels generated through filamentation of femtosecond laser pulses in air can be revived after several milliseconds by a delayed nanosecond pulse. Electrons initially ionized from oxygen molecules and subsequently captured by neutral oxygen molecules provide the long-lived reservoir of low affinity allowing this process. A Bessel-like nanosecond-duration laser beam can easily detach these weakly bound electrons and multiply them in an avalanche process. We have experimentally demonstrated such revivals over a channel length of 50 cm by focusing the nanosecond laser with an axicon.

OCIS codes: (190.7110) Ultrafast nonlinear optics; (350.5400) Plasmas

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1. Introduction

Much attention has been given in the last few years to the propagation of ultrashort laser pulses in air. With short pulses (pulse duration < 10^{-12} s), high peak intensities are achieved with a modest energy per pulse. As a consequence, a dielectric medium such as air is readily driven into a highly nonlinear regime. This can lead to spectacular effects. For instance, several groups have reported the occurrence of femtosecond (fs) filamentation (for recent reviews see [1-3]). In this process, an intense ultrashort laser pulse self-organizes into a contracted beam, 10-100 µm in diameter, fed by a surrounding laser energy reservoir. The contracted beam, often called filament, carries a peak intensity of I ~ 5×10^{13} W/cm² sufficient to ionize air molecules in a multiphoton process. It maintains this high intensity over distances which can reach several hundreds of meters in air because of the dynamic balance between self focusing (due to the optical Kerr effect) and beam defocusing (due to multiphoton ionization of air molecules) [1-3].

The plasma channel formed in the wake of the propagating pulse has captivated the interest of many researchers especially because of its unusual characteristics and potential applications [4]. It is at the same time weakly ionized but highly collisional cold plasma since only a ratio of 10^{-3} of the air molecules are typically ionized at atmospheric pressure. Its special geometry, in the form of a channel with a very high aspect ratio (>10^4) is responsible for new effects. For instance, the plasma generates an intense terahertz (THz) radiation in the form of an ultrashort burst emitted in a narrow forward directed cone [5, 6]. Several meters long straight discharges could be triggered and guided in air by such fs filaments [7]. It was shown that such discharges could carry a current of several hundreds amperes with very little power dissipation [8]. This raises the prospect to use filaments in order to capture high currents, to preventively discharge electrodes and clouds or even to act as a lightning rod for the safeguard of sensitive sites [9, 10]. It was also shown recently that plasma channels in air can guide microwaves [11, 12].

However, the short plasma lifetime remains a major obstacle to many applications, because it prevents a fast discharge to build up over a long distance. The origin of the short plasma lifetime is well understood. For a filament laser intensity of 5×10^{13} W/cm², the initial free electron densities obtained in air reaches 10^{16}-10^{17} cm^{-3}, mostly from multiphoton ionization of oxygen molecules of lower ionization potential (12 eV against 16 eV for N_2). At such densities electron recombination is dominated by the capture by parent ions (see Eq. (1) below). This process leads to a decrease of the electron density by two orders of magnitude within a few nanoseconds (ns) [13, 14]. Below 10^{14} cm^{-3}, capture by neutral oxygen molecules becomes the main recombination process. It gives rise to an exponential decay with a time constant τ ~ 150 ns [14].

2. Experimental methods

In this paper, we present what we believe is a significant progress in resolving the problem of obtaining a long-lived plasma channel. We demonstrate experimentally that it is possible to reestablish a conducting air column even milliseconds (ms) after the passage of the fs pulse and to prolong its lifetime with the help of a second laser. The principle is to exploit the on-site storage of weakly bound electrons onto oxygen molecules following the formation of the original plasma channel [15]. The attached electrons provide a reservoir with a memory of the special geometry of the filament over a time corresponding to the ion-ion recombination and diffusion of the negative oxygen molecules out of the initial filament core. This corresponds to times of the order of several ms. By using a Bessel-like ns duration laser beam to detach these electrons we show that it is possible to restore a plasma over a distance much longer than could be achieved with a conventional focusing system. In the present case, the restored plasma length is 50 cm, but it could be considerably extended with more powerful lasers. More importantly, the focused Bessel like beam has on-axis intensity sufficient not only to liberate the bound electrons, but also to multiply them through inverse bremstrahlung and avalanche processes. We further show that by using a two-color scheme with sufficient pulse energies, the electron density in the revived plasma channel can even exceed the initial electron density from the fs laser. The restored plasma channel lifetime lasts beyond one microsecond.

There have been several attempts to increase the filament plasma lifetime by using a second laser [15-20]. However, in all of these previous works, the plasma lifetime is increased at early times while free electrons are still present and the longitudinal extent of the effect is limited to the Rayleigh length of the focused laser beam.

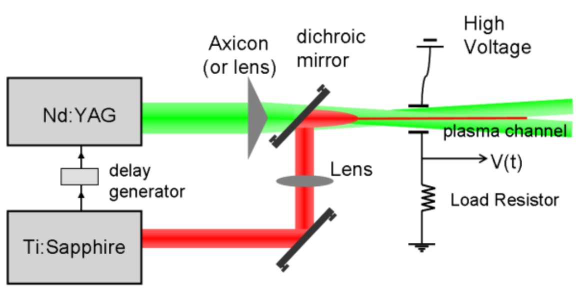

Fig. 1. Experimental setup. The fs laser generates the plasma channel and the delayed ns laser revives it. The plasma channel is detected using electrodes with high voltage and/or a PMT.

The experimental setup is shown in Fig. 1. The initial plasma channel was generated through the filamentation of fs laser pulses. We have used a Ti:Sapphire chirped pulse amplifier chain, which can provide up to 15 millijoules (mJ) pulse energy at a pulse duration of 50 fs (at 800 nm center wavelength) and the teramobile laser [21] which can provide up to 250 mJ pulse energy at a pulse duration of 150 fs. To revive the plasma channels, we used a Nd:YAG laser at 1064 nm and/or its second harmonic at 532 nm, which can provide up to 250 mJ pulse energy at both wavelengths with a pulse duration of around 10 ns. The different laser systems were synchronized at 10 Hz repetition rate, with controllable delay between their pulses. The Nd:YAG laser was focused with an axicon (0.5 degree base angle, 5 cm diameter), which generated a Bessel-like transverse profile with a long focal region [22, 23]. The longitudinal focal range of the Bessel-like beam was about 50 cm with a central peak of 100 µm, which provides good longitudinal and transverse overlap with the filament. The fs laser beam was focused with a 75 cm focal length lens and aligned such that the filament overlapped with the axicon line focus. In order to detect the presence of plasma, we passed both beams between two square metal plates with ~ 5 kV voltage and 5 mm separation. To avoid the laser beams directly hitting the metal and generating photocurrent, the plates were covered with an insulator. In the presence of an external electric field, the charges in the plasma redistribute, screen out the external field [24], modify the potential and generate a current through the grounded arm. The corresponding potential drop across the resistor was measured using an oscilloscope. We also used a photomultiplier tube (PMT) with a narrow interference filter centered at 338 nm (10 nm bandwidth) to detect the emission from N_2^+ ions fluorescence [25]. The former method proved more sensitive in detecting low plasma densities, while the latter gives a faster temporal response.

3. Results and discussion

Figure 2(a) shows measurements of the revival of plasma channel generated by 90 mJ of the teramobile laser and reestablished by the Nd:YAG laser (at 532 nm) at 1 ms delay. The electrical method described above was used with a 1 MΩ load resistance in order to enhance detection sensitivity. Due to the circuit response the decays exhibited by the signals were slower than the actual plasma lifetime. The true plasma lifetime, obtained with a 50 Ω load resistance, is shown in the inset of Fig. 2(a). The first peak around $t=0$ in Fig. 2(a) is caused by the filament plasma of the fs pulse. When the Nd:YAG beam focal axis was carefully aligned with the filament, we observed a second peak at the arrival time of this laser. The second peak disappeared when the detection system was enclosed in a box purged with nitrogen or when a strong air current was flowing between the detecting electrodes. The on axis intensity of the Nd:YAG laser around the focal region of the axicon was slightly above 10^{10} W/cm^2, which is well below what is required to photoionize air. As a result, we attribute the appearance of the second peak to the detachment of the electrons from O_2^- ions and subsequent multiplication through an avalanche process. We observed this ns laser *revived* plasma at delays up to several ms in quiet atmosphere.

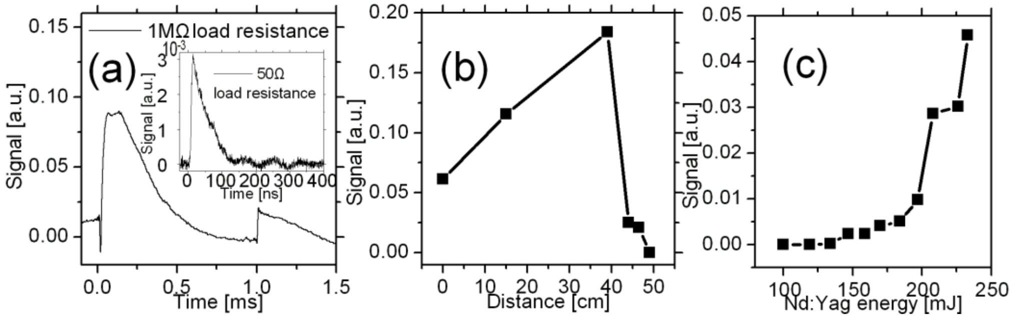

Fig. 2. (a) Signal detected using the electrical method, showing plasma channel initially created by the fs laser and then revived by the ns laser (532 nm) after 1ms. (b) Magnitude of the signal from the revived plasma at various distances from the beginning of the initial plasma filament. (c) Magnitude of the signal from the revived plasma as a function of the Nd:YAG laser pulse energy. In (b) and (c) each point corresponds to the average of 100 measurements. The mean fluctuation around the average is 5%.

Figure 2(b) shows the measured magnitude of the second peak induced by the revived plasma, as a function of distance of the detection system from the beginning of the filament. It is seen that the plasma channel is revived over about half meter propagation distance. Note that a longer distance would be obtained with different axicon and higher ns laser energy. We also measured the magnitude of the signal as a function of the Nd:YAG laser energy (Fig. 2(c)). The sharp rise of the signal with laser energy above ~ 190 mJ reveals that an avalanche process is involved.

The principal interactions involved in the revival process have different frequency dependences. The photo detachment rate of O_2^- ions increases with increasing photon energy [16, 26], whereas the avalanche process is more efficient at lower frequencies [27]. This brings up the opportunity of using together both the fundamental and the second harmonic frequencies of the YAG laser. In this case, the second harmonic can detach the electrons and the fundamental can accelerate and multiply them. We performed such experiments using two separate YAG lasers emitting at 532 and 1064 nm respectively. By focusing both beams with conventional lenses and using the appropriate chronological order for the pulses, sufficient combined laser intensity was available to induce a full evolution of the plasma towards dielectric breakdown. The initial plasma filament in this case was generated by the 15 mJ fs laser focused by a 75 cm focal length lens. The fundamental beam made 10 degrees angle with the other two laser beams. The fundamental and second harmonic of the ns laser (focused with 40 cm and 75 cm focal length lenses, respectively) arrived after 53 microseconds (μs) delay, the second harmonic pulse was followed after 10 ns by the fundamental pulse. The characteristic spark due to optical breakdown was easily observed (see Fig. 3(b)). We verified that no breakdown occurred when the fs laser was blocked. Figure 3(a) compares the revived plasma optical signal observed through the UV interference filter to that of the initial filament plasma. We note that the revived plasma lasts at least 1 μs, and that the corresponding optical signal exceeds that of the filament more than 200 ns. The probability to obtain a fully developed breakdown was measured as a function of the relative delay between the two ns pulses, for a fixed delay of 53 μs with respect to the fs laser pulse. In consistence with the expectations mentioned above, we observed a maximum probability of 100% when the 532 nm pulse arrived first and the 1064 nm pulse comes after a delay of 10 ns (see Fig. 3(c)).

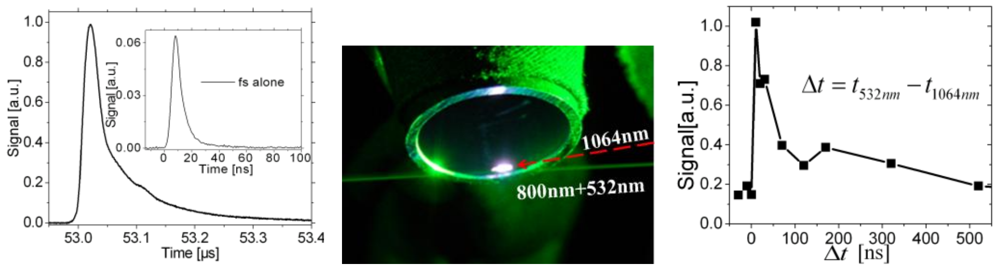

Fig. 3. Three beam configuration when both YAG laser pulses are delayed by 53 μs from the fs laser pulse: (a) Signals detected by the PMT with a 338 nm filter in the presence of breakdown. The laser pulse at 1064 nm arrives 10 ns after the pulse at 532 nm. The optical signal recorded under the same conditions with the fs laser alone is shown in the inset (the magnitudes can be compared). (b) Photograph of the breakdown spark in a three laser beams configuration. The beam paths are shown with dotted lines. (c) Revival plasma triggered breakdown probability as a function of delay between the 1064 nm and 532 nm. A positive delay corresponds to the 532 nm pulse arriving first. Each point is an average over 1000 shots.

4. Simulation and Results

In order to compare our experimental results with theoretical models we have also numerically estimated the electron density evolution under conditions similar to the experiments. The chain of events leading to the revival can be cast into the following coupled nonlinear rate equations:

$$\begin{aligned}
\frac{dn_e}{dt} &= -\beta_{ep}\, n_e\, n_p - \eta\, n_e + \alpha(I_2)\, n_e + \sigma\left(\frac{I_2(t)}{h\upsilon}\right) n_n \\
\frac{dn_p}{dt} &= -\beta_{ep}\, n_e\, n_p - \beta_{np}\, n_n\, n_p + \alpha(I_2)\, n_e \\
\frac{dn_n}{dt} &= -\beta_{np}\, n_n\, n_p + \eta\, n_e - \sigma\left(\frac{I_2(t)}{h\upsilon}\right) n_n
\end{aligned} \tag{1}$$

where n denotes density and the subscripts e, n and p denotes electrons, negative ions and positive ions, respectively. β_{ep} , β_{np} and η are the rates for electron-ion recombination, ion-ion recombination and attachment to neutral molecules. The attachment coefficient η is a function of the amplitude of the laser electric field, and is found from the addition of the two-body and three-body interactions [15]. α is the impact ionization rate, which depends on the pulse intensity, ionization potential of the neutral molecules and laser wavelength; it is calculated according to Drude model [27]. I_2 is the intensity of the ns pulse, σ is the cross section for absorption of a photon and consequent detachment of the electron (calculated from the experimental results of Burch *et al*. [26]), υ is the ns laser frequency and h is Planck's constant.

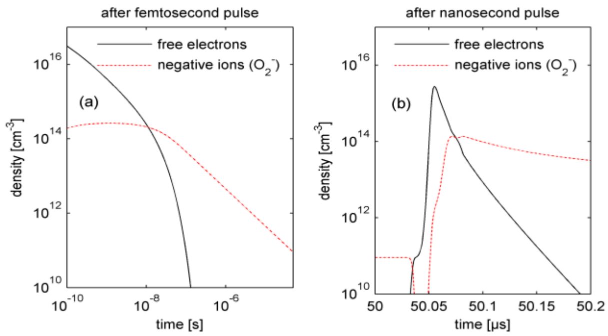

Fig. 4. Evolution of free electron and O_2^- ion densities: (a) After the fs laser excitation. (b) During and after the delayed ns pulse (at 532 nm) excitation. The end of the time axis in (a) coincides with the beginning of time axis in (b). The fs pulse arrives at $t = 0$ and creates the initial plasma channel of density 10^{17}cm^{-3}. The ns pulse is a Gaussian with 10 ns (FWHM) duration, has its peak 50 ns after the beginning of time axis of (b) (at 50.05 µs) and it has a peak intensity of 10^{11} W/cm^2.

At $t=0$, n_e and n_p are taken to be 10^{17} cm^{-3} and n_n to be zero consistent with measurements performed in filaments [1]. We numerically solved Eq. (1) in the absence of the last terms to obtain the densities at the arrival time of the ns laser, and reinserted these values as initial conditions for the same rate equations, last term included, which describe the change in the densities during and after this long pulse excitation. The results of the calculations Fig. 4 agreed well with the experimental observation. The above model is insufficient, however, to describe the full evolution towards dielectric breakdown for higher intensities used during experiments shown in Fig. 3. since it does not take into account the increase of absorption cross section with electron temperature.

4. Conclusion

In conclusion, we have demonstrated experimentally and verified theoretically that a fs laser generated plasma channel can be reconstructed in air long after the passage of the fs laser pulse. The attachment of free electrons to oxygen molecules, which is usually perceived undesirable, can be turned to advantage as it provides storage of low affinity electrons. With a delayed ns laser, these electrons can be detached and multiplied through impact ionization. We have observed revivals at delays as large as several ms. With sufficient ns laser intensity, the avalanche process could be pushed up to dielectric breakdown. The plasma lifetime in this case reaches µs. These results may prove important for applications of long distance filamentation in air, where the short lifetime of the plasma poses strong limitations.

Acknowledgements

We thank C.L. Arnold and M. Pellet for stimulating discussions. This work has been partially funded by DGA. Teramobile is a joint project funded by CNRS, DFG and ANR.