Lasing Action in Air
Induced by Ultrafast Laser Filamentation

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Researchers are devoting an increasing amount of attention to the propagation of intense, ultrafast laser pulses in air in which nonlinear effects become dominant. The dynamic interplay and balancing between two nonlinear effects—Kerr self-focusing and the subsequent defocusing by the self-generated plasma—result in a series of self-foci that are usually called a filament. After the laser pulse passes, a low density plasma column remains. It has been shown that the peak intensity inside the series of foci is clamped down to about $5 \times 10^{13}$ W/cm$^2$ in air.$^{2,3}$

Self-phase modulation and self-steepening of the pulse through both the neutral gas and the plasma give rise to strong broadening of the spectrum, i.e., the pulse transforms itself into a white-light laser pulse, known in popular terms as a supercontinuum.$^{5}$

Filaments can persist for lengths of several kilometers,$^{5,6}$ which makes them very promising for practical applications such as lightning discharge control and remote sensing.$^{7}$

Thanks to the low density of the plasma, clean fluorescence spectra of nitrogen ($N_2$) molecules and ions from the filaments have been observed. By clean fluorescence, we mean there is no contamination induced by the plasma continuum. The spectra are assigned to the first negative band ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$) of $N_2^+$ and the second positive band ($C^3\Pi_u \rightarrow B^3\Pi_g$) of $N_2$. When the $N_2$ molecules first interact with the intense femtosecond laser field inside the filament, tunnel ionization occurs.$^9$ This means that the molecular systems are initially prepared in the ionization continuum—some of them in the excited ionic state—through the ejection of an inner valence electron.$^{10}$ Subsequent radiative decay of the excited ion gives rise to the first negative band (see Fig. 1).

Recombination between the electrons and the ions would sequentially relax the population to lower excited states. When an upper radiative excited state such as $N_2^* C^3\Pi_u$ is populated from above, the lower state $N_2^* B^3\Pi_g$ remains empty. Because there is no intermediate resonance and the transition between the ground electronic singlet state $X^1\Sigma_g^+$ and the triplet $C^3\Pi_u$ or $B^3\Pi_g$ states is forbidden, in the first approximation, the probability of direct optical excitation from the ground state to the $B^3\Pi_g$ and $C^3\Pi_u$ states is negligible. Our expectation that population inversion and gain would exist has been confirmed by recent observations.$^{11}$

**Figure 1. Schematic potential energy diagram for $N_2$ and $N_2^+$.**

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In our experiment (see Fig. 2), a laser beam with central wavelength of 800 nm and maximum power of 15 mJ is focused by an f = 3 m lens (L). A dielectric mirror (M1) with high reflectivity at around 800 nm and high transmission for ultra-violent (UV) light is placed just after the lens to reflect the beam at a 45 degree incident angle. Along the propagation direction of the laser beam, the filament starts before the geometrical focus; with increasing input laser energy, the length of the filament increases toward the focusing lens. To reduce the strong scattering from the wall, a beam dump, located 7 m away from M1, blocks the laser beam after filamentation. In the inset, a photograph of the filament (in blue) records the near UV fluorescence of the nitrogen molecules and ions it induces. The backscattered fluorescence is collected by a fused silica lens and detected by use of a photomultiplier tube (PMT) with interference filters to detect the (0-0) vibronic transition of the second positive band system of N\(_2\) at 337 nm; the laser energy is measured simultaneously by an energy meter.

Figure 3 shows a typical waveform from the PMT (solid line) with input laser energy at 13 mJ. Each point of the waveform corresponds to the fluorescence intensity at different distances along the propagation path of the beam. The correction for different collection angles at different positions was accomplished by multiplying the signal by \(R^2\), where \(R\) is the distance between the signal position and the detector. The signal is detected between the filament’s starting point and the dump position. We carried out a parallel measurement of filament length to detect the acoustic wave (AW) that arises from the sudden expansion of the hot gas column caused by the recombination of the plasma inside the filament (dotted line). The AW measurement shows that the filament stops just after the geometrical focus (Fig. 3, point A).\(^4\) Two sources contribute to the fluorescence signal (solid line). One is the backward fluorescence (BF) signal from the starting point to the end of the filament (Fig. 3, point A). The other is the Rayleigh scattering of the forward-propagating amplified spontaneous emission (ASE) pulse [forward fluorescence (FF)]. The FF is shown by the low level signal after point A. Because the cross section of Rayleigh scattering is very small, the scattered FF signal is much weaker than the BF signal. Thus, there is no reason to consider any contribution to the BF signal from scattering of the FF signal. The integration of the signal before point A can be considered backward ASE intensity.

We also measured the variation of fluorescence intensity that occurs at different laser energies. The plot in Fig. 4 shows the typical results obtained when an f = 3 m lens is used. The filament length in Fig. 4 is calculated as follows. The starting point is determined by the input laser energy based on the known relations between the self-focusing position and the peak laser power.\(^9\) The end of the filament is set at the point at which the filament stops, as determined by the

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**Figure 2.** Experimental setup. L: focusing lens with f = 1 m, 3 m or 5 m; M1: dielectric mirror for 800 nm laser beam at 45 degree incident angle; M2: dielectric mirror for 800 nm laser beam at 0 degree incident angle; F: interference filter. The distance between M1 and the beam dump is around 7 m. (Inset) Digital camera picture of filament obtained by use of an f = 1 m lens to focus the laser beam.

**Figure 3.** Typical waveform of fluorescence signal detected by PMT (solid line) and acoustic wave (AW) signal intensity measured along the filament (dotted line).
parallel AW measurement for the highest input laser energy. For an $f = 3$ m lens, the end of the filament is set at position $A$, which is 4 m away from the detector.

In an initial approximation, the filament can be considered a uniform column of plasma generated by a constant clamped intensity. In the presence of ASE, the total fluorescence intensity detected by the PMT would increase exponentially with filament length. If the gain coefficient were $g = 0$, the detected signal intensity would be proportional to the length of the filament. Our experimental results (Fig. 4) show an exponential dependence on filament length, which is a direct indication of the existence of gain.

In the backward direction, depletion of the inverted population is to be expected because of the lifetime of the upper state. The population on the upper state decays to the lower state because of stimulated emissions, collisions and other relaxation processes, and the population difference between the $C^1\Pi_u$ and $B^3\Pi_g$ states is decreased. The backward ASE thus becomes increasingly less amplified as it propagates back along the filament. When the population difference reaches equilibrium, there is no further amplification of the backward fluorescence, despite the fact that the filament can be rather long. The amplification processes are thus limited by the lifetime of the upper state. As a result, the gain coefficient, which is proportional to the population difference, will saturate with the increase in the intensity of the ASE. In qualitative terms, the effect of gain saturation can be described by introducing the saturation intensity $I_s$ into the gain coefficient calculation. The saturation intensity is directly related to the lifetime of the upper state.

The lifetime of the detected $N_2$ fluorescence is around 1-2 ns, a level which corresponds to 30-60 cm in propagation length. As shown in Fig. 3, when an $f = 3$ m lens is used, the filament can be as long as 1 m. It is clear as a result that gain saturation plays an important role in backward ASE in this case.

The fluorescence intensity curve shown in Fig. 4 reproduces the measured data well. The curve is calculated by use of an exponential function with $g_0$ (the small signal gain) and $I_s$ (the saturation intensity) as fitting parameters. The experimental results obtained by use of an $f = 5$ m and 1 m lens are similar to those obtained with an $f = 3$ m lens. In the case of an $f = 1$ m lens, even when the highest energy (13 mJ) is used, the filament is several centimeters long. This is short compared to the lifetime of the $C^1\Pi_u$ state. In this case, the gain saturation effect is therefore not obvious.

In the forward direction, the fluorescence follows the pump laser pulse. The pump pulse continues to ionize the nitrogen molecules as long as the energy remaining in the pulse is higher than the critical power needed for self-focusing. The upper state is always populated through the recombinations and relaxation processes, while the lower state is empty. Forward fluorescence, which arrives just after the inverse population is created, is always amplified. The amplification process stops only when there are no more filaments. For this reason, more efficient amplification is expected in the forward direction. We are currently investigating gain in forward ASE.

In conclusion, we have observed an exponential increase of the backscattered fluorescence from $N_2$ molecules with an increase in filament length. This indicates that the fluorescence has been amplified through ASE as it propagates along the filament.

We expect that other molecules in the strong field inside a filament would undergo similar tunnel ionization, dissociation and fluorescence processes. In this case as well, the fluorescence would undergo amplification in the forward and backward directions. The population inversion which results from direct ionization followed by recombination from the ionization continuum seems to be a universal phenomenon in all gases that show fluorescence. This finding could be very important in the future in applications such as remote sensing.

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