Experimental confirmation of high-stability of fluorescence in a femtosecond laser filament in air

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Femtosecond laser filamentation is particularly interesting for remote sensing pollutant in the atmosphere. In this work, we investigate the local shot-to-shot stability of the filament induced fluorescence of nitrogen in air. It is found that the root-mean square fluctuation of the fluorescence signal is at least one order of magnitude lower than that of the linear propagation case. In practice, it would contribute to improve the robustness of long distance spectroscopic analysis of the fluorescence of pollutant molecules inside the filament. We further point out that this unique property of filament induced fluorescence spectroscopy is because of the intensity clamping, a profound phenomenon of filamentation.

Femtosecond laser filamentation in air has attracted considerable research interest recently, not only from the point of view of fundamental physics, but also due to its potential applications in various fields [1–4]. Among them, remote sensing of pollutant in the atmosphere is a highly promising one [5–7]. Different types of samples, including gas, dust, aerosol, have been successfully identified by the filamentation based technique at distances as far as 100 m. The physical principle behind this technique is that due to the high laser intensity (about $5 \times 10^{13}$ W/cm$^2$), the molecules inside a filament would be ionized or fragmented leading to fingerprint fluorescence emission. Then by introducing a LIDAR (light detection and ranging) setup, the scattered fluorescence light could be collected, normally in the backward direction, and analyzed spectroscopically. It is well known that due to dynamic counteracting of optical Kerr effect induced self-focusing and the defocusing effect of the self-generated weak plasma, the high laser intensity could be retained over a long distance, even up to kilometers during the filamentation process [1–4]. Recently, Chen et al. has found that the diameter of the filament core is almost constant over a long distance followed by another with weak ionization. Due to self-spatial filtering, the core diverges out linearly with a low divergence when filamentation ends [8]. Therefore, linear diffraction of the laser beam will be overcome and the remote detection of fluorescence can be realized. In a similar way, by using remote filament induced breakdown spectroscopy, noninvasive optical analysis of metallic sample has also been reported [2,6].

In fact, significant extension of the detection distance is not the only advantage that filamentation brings to the above mentioned spectroscopic techniques. Recent results have also demonstrated that the filament induced fluorescence shows unique invariable feature as a function of distance [9,10]. It is due to the profounder phenomenon of intensity clamping when filamentation occurs, which limits the laser intensity that could be achieved inside a filament [1–4,11]. In addition to this established knowledge we explore in the present work, another aspect of the filament induced fluorescence spectroscopy – local high-stability of the fluorescence signal in air, which is also explained by the phenomenon of intensity clamping.

In our experiment, a femtosecond laser pulse was sent in ambient air. In order to produce a long filament, the initial laser beam was squeezed by an inverse telescope consisting of a concave mirror ($f = 40$ cm) and a plan-concave lens ($f = -20$ cm). The displacement between the mirror and the lens was 20 cm. At the output of the lens, the laser beam diameter was about 3 mm at 1/e$^2$ level. The pulse duration was 45 fs (full width at half maximum, FWHM).
The central wavelength of the laser pulse was 800 nm, and the laser energy was 2 mJ/pulse. In this case, a filament was produced starting from a distance 95 cm with respect to the plan-concave lens. In Fig. 1a, we present a typical emission spectrum from the filament region taken by a cryogenically-cooled CCD equipped spectrometer. In consistent with the previous report [1,2], N₂ fluorescence lines can be clearly distinguished in Fig. 1a, while the contribution of the plasma continuum to the spectrum is essentially weak. Afterwards, perpendicular to the propagation axis, the emitted fluorescence light was collected by a 1 in.-diameter fused silica lens, whose focal length was 1 in., onto a photomultiplier tube (PMT). In front of PMT, an interference filter was inserted to isolate one particular nitrogen fluorescence spectral line, namely, the (1–0) vibrionic transition of the second positive band system of N₂ at 357 nm, from the scattered light of the pump beam. Then the PMT signal was sent to an oscilloscope for analysis.

At the first step of the experiment, the longitudinal fluorescence intensity distribution was measured by scanning the PMT along the propagation axis. The obtained results are present in Fig. 1 as the black squares (left label) and each datum was averaged over 20 shots. In Fig. 1b, significant fluorescence signal starts to be recorded at a distance of 90 cm. It corresponds to the starting point of the filament. Then the fluorescence signal reaches a maximum when the propagation distance is 103 cm. Due to the re-focusing phenomenon, a second peak is observed at the distance of 124 cm. The ending distance of the filament is 140 cm.

In order to study the stability of the fluorescence signal, another scan of the signal was made using the same setup. At this time, a variable neutral density filter was added between the interference filter and the PMT to attenuate the amount of light received by the PMT down to the same level at different distances. It was ensured by obtaining constant waveform amplitude on the oscilloscope. It is necessary to mention that during the whole scan, the supplied high voltage of the PMT was kept constant. At the same time, in order to explicitly exclude the saturation effect of our PMT, which starts to occur when the signal amplitude reaches 200 mV, the amplitude of the experimentally recorded waveform was kept around 120 mV. Then, the RMS fluctuation of the peak value of the signal was calculated for 200 shots at every position. Note also that the RMS fluctuation calculation was done purely on the basis of shot-to-shot measurement, being free from any instrumental pre-averaging. In Fig. 1b, the red circles show the trend of the RMS fluctuation of the signal as a function of the propagation distance (right label). Clearly, the data could be divided into three zones as separated by two dotted lines in Fig. 1b. Zone A corresponds to the region where the propagation distance $d < 96$ cm. The maximum fluctuation is about 13% at 90 cm. As the propagation distance increases, it quickly declines to less than 1% at $d = 97$ cm. Afterwards, the fluctuation of the fluorescence signal displays a very high-stability until $d = 132$ cm. We identify this range as zone B. Throughout zone B, the signal fluctuation is less than 1% and the minimum is even below 0.5%. Note that the energy fluctuation of the input pump beam is about 0.5%. Note that the energy fluctuation of the input pump beam is about 1.5%. We have known that in the case of filamentation the plasma is generated through tunnelling ionization in air [1]. Since the ionization rate of nitrogen is characterized by an effective non-linearity order of 7.5 [12], one would expect that the fluctuation of the fluorescence signal should be at least 1.5% $\times$ 7.5 $\approx$ 11% in the perturbative regime. It is more than one order of magnitude higher than that measured within zone B in our experiment. This phenomenon implies that the laser intensity at the given position inside the filament has been clamped to a constant value with very little shot-to-shot fluctuation. After $d = 132$ cm, i.e. in zone C, the stability of fluorescence signal becomes worse rather quickly. It reaches 14% at the distance of 140 cm. For further distances, the signal received by the PMT was not strong enough to make consistent measurement. Similar stability measurements were also carried out for three other important nitrogen fluorescence lines by using the same setup except that different interference filters were used. The results are shown in Fig. 2: black squares for 337 nm, red circles for 391 nm and blue triangles for 423 nm, respectively (note that in Fig. 1a, due to the coarse resolution used in the experiment, 423 nm line merges with another spectral line located at 427 nm). They all show the same behaviour as that of 357 nm. Ultra-high-stability is the common property of the fluorescence signal emitted by the filament. As comparison, we also tried to focus (focal length $f = 12.5$ cm) the laser pulse at energy as low as 50 μJ. In this case, a plasma spot was produced by linear focusing solely and the measured RMS fluctuation by the same detection setup was about 17%, which has been indicated as the blue dashed line in Figs. 1b and 2.

In order to shed further light on the understanding of the highly stabilized fluorescence signal, the following numerical simulations have been performed. The simulations were based on the nonlinear wave equation under the slowly varying envelope approximation, which can be expressed as:

\[
2ik_0 \frac{\partial A}{\partial z} + \Delta_0 A - k_0^2 \frac{\partial^2 A}{\partial t^2} + 2 \left( 1 + i \frac{\partial}{\partial t} \right) \frac{k_0^2}{\eta_0} (\Delta A_{\text{ker}} + \Delta A_{\text{plasma}}) A - i k_0^2 x A = 0
\]
On axis free electron density \(10^{-16}\) cm\(^{-3}\) shown in Fig. 3. It hints the occurrence of the re-focusing process.

In Fig. 3, the thin lines outline the normalized on axis peak intensity for twenty different initial energies (left label, \(E_0\) denotes the initial laser peak intensity), while the thick blue line indicates the corresponding RMS fluctuation of the peak intensity (right label). Fig. 3 shows that due to self-focusing, the laser peak intensity undergoes quick increase at the beginning. Starting from the propagation distance of 0.5 m, it goes through a plateau region and decreases again after 1 m. The overall length of the plateau region is about 0.6 m. The appearance of the plateau is due to the intensity clamping phenomenon. During the laser propagation within the plateau region, the on axis peak intensity has two maxima as shown in Fig. 3. It hints the occurrence of the re-focusing process. However, since twenty curves are too close to each other to distinguish their difference in Fig. 3, a small part of the figure indicated by the dashed rectangle is enlarged in Fig. 4 for explicit illustration.

In Fig. 4, to the left of 0.5 m, the laser intensity has evident fluctuation (%)

\[
\Delta n_{\text{num}}(t) = n_2 \left\{ |A(t)|^2 + \int_{-\infty}^t H(t-t')|A(t')|^2dt' \right\}
\]

(2)

where \(n_2\) is the nonlinear index of refraction. The delayed Raman response function \(H(t)\) was approximated based on the damped model by the following equation:

\[
H(t) = \frac{1}{T_k} \theta(t) \exp(-t/T_k)
\]

(3)

where \(\theta(t)\) is the Heaviside function and the fitting parameter \(T_k = 70\) fs [13]. The initial laser parameters applied in this work read: central wavelength \(\lambda_0 = 800\) nm, pulse duration \(\Delta t = 50\) fs (FWHM) and beam radius \(r_0 = 500\) \(\mu\)m (1/e\(^2\)), respectively. However, the laser energy was a variable parameter. 20 randomly chosen values have been adapted for 20 simulations. All the energies were limited within the range of \(800 \pm 12\) \(\mu\)J, i.e. within \(\pm 1.5\%\) error. The radial resolution used in our simulation is \(2\) \(\mu\)m and the propagation step is \(500\) \(\mu\)m. Care has been taken to ensure that our results converge by using these simulation grid sizes. The simulated results are summarized in Figs. 3–5.

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In Fig. 4, to the left of 0.5 m, the laser intensity has evident fluctuation (%).
ation. For example, when the propagation distance is 0.42 m as indicated by the dashed lines, the maximum and minimum laser intensity of these twenty cases are 11 l0 and 9 l0, respectively. The difference is roughly 20%. We could explain it by the change of the filament onset position when unstable laser energy is used. According to the well known Marburger equation, the starting position of the filament produced by a Gaussian beam is given by [14]:

$$z_{sf} = \frac{0.367k\alpha^2}{\left( \left( \frac{l_0}{\alpha} \right)^{1/2} - 0.852 \right)^2 - 0.0219}$$

where $ka^2$ indicate the diffraction length, $k$ being the wave number and $\alpha$, the radius at 1/e level of the beam profile. $P_{cr}$ denotes the critical power for self-focusing. Eq. (4) indicates that the starting position of the filament is proportional to the square of the initial beam radius. Previous study also implies that filamentation started with smaller beam could give rise to longer extension of the filament zone [8]. These explain the discrepancy of the starting position and the filament extension observed in the experiments (Figs. 1b and 2) and the simulations (Figs. 3 and 5) since smaller beam diameter (1 mm) has been used in the simulations. After substituting all the necessary parameters into Eq. (4), we have found that $z_{sf}$ changes at best from 0.65 to 0.68 m for all the energy values used in our simulations. Taking into account that the intensity evolution could be described by catastrophic collapse when the propagation distance approaches the onset position of the filament [15], the variation of the laser intensity would be significant even though the change of the filament position is less than 5%. This leads to the large intensity fluctuation in Fig. 3 when the propagation distance is less than 0.5 m as indicated by the thick blue line.

After all, what interests us most in Fig. 4 is the merging of twenty curves with increasing propagation distance. The superposition begins to take place at 0.5 m. This distance coincides with the detuning point of the laser intensity to the plateau region. It implies that intensity clamping should be responsible for both cases. It was confirmed by the on axis free electron density plots of all the 20 random cases in Fig. 5 (thin lines, left label). In Fig. 5, the electron density reaches its first maximum at 0.5 m and keeps at relatively high level for almost 60 cm before it fades away. The phenomenon of the intensity clamping tells us that owing to the crucial role of plasma generation, the laser intensity will be constrained giving rise to the plateau in Fig. 3. Furthermore, for the same intensity plateau, the thick blue line in Fig. 3 demonstrates extraordinary stability of the laser intensity despite the initial energy fluctuation of the input pulses. The intensity RMS fluctuation within this range is always less than 0.5% with the minimum approaching 0.05%. Such extremely stabilized laser intensity naturally leads to constant ionization rate and results in invariable free electron density. In fact, it is depicted by the thick blue line in Fig. 5. The simulated fluctuation of the free electron density within the range from 0.5 to 1.1 m is as low as 0.5%. Since the fluorescence signal detected in the experiment is proportional to the free electron density, Fig. 3 qualitatively reproduces the experimental results obtained in Figs. 1 and 2. Therefore, our interpretation in principle applies to the experimental results too. Here it is necessary to emphasis that each longitudinal fluorescence measurement shown in Fig. 1b is the result of 20 shots average. Therefore, the longitudinal fluctuation of the fluorescence single indicated in Fig. 1b could not truly reflect the corresponding shot-to-shot stability. For example, in Fig. 6 we demonstrate the averaged on axis peak intensity (red line) and the averaged on axis free electron density (black line). The averaged free electron density varies more than 3% per centimetre between 0.5 and 0.6 m. However, the RMS fluctuation falls below 1% (see Fig. 5). Hence, based on the evidence obtained in our work, we believe that the phenomenon of intensity clamping not only sets an upper limit for the achieved laser intensity during the filamentation process, but also constrains the laser intensity inside filament to a highly stabilized level.

In order to shed further light into the underlying physics mechanism of the high laser intensity stability, we have studied the longitudinal evolution of the laser beam diameters in the simulations. Twenty sets of data are shown in Fig. 7 simultaneously. Similar to the laser peak intensities plots (Fig. 3) and the free electron density plots (Fig. 5), the fluctuation of the input energy induces pronounced shift of the position where the first diameter minimum is reached. Soon after this characteristic position, twenty plots overlap well between 0.5 and 1.1 m. It could be understood as the nature consequence of the balance between the Kerr self-focusing and the plasma defocusing. Since the balance is essentially determined by two intrinsic properties of the optical media, namely, the coefficient of the nonlinear refractive index and the effective photo-ionization rate [16], the stabilized laser beam diameter has weak dependence on the initial laser energy. On the other hand, it has been stated that when filamentation occurs, the power coupled into a filament is constant [17,18]. Hence, the finally obtained laser peak intensity would be independent to the input energy fluctuation (see Fig. 3). In Fig. 7, the noticeable small variation of the diameter inside the filament region could be associated with the systemic pulse length change during the filamentation.

Fig. 7. Simulated laser beam diameter at full width at half maximum.
tion process. We have known that due to the presence of the Raman effect in air, the coefficient of the nonlinear refractive index would be faintly influenced by the longitudinal precision of the measurements. The assumption has been confirmed by binning the simulated on axis free electron density over various spatial steps ($D_b$) and recalculating the corresponding RMS fluctuation of each data. Our results are represented by Fig. 8. The binning steps are 1, 2, 5 mm, 1 and 2 cm, respectively. In Fig. 8, no significant change of signal RMS fluctuation is found in the filament zone. The dominant effect of increasing spatial binning step is the smoothening of the curve, which is apparently due to the averaging effect.

In summary, we have studied the shot-to-shot stability of nitrogen fluorescence signal emitted from the filament as a function of propagation distance. Our results demonstrate that there exists a region where the fluorescence signal is highly stabilized. The measured root-mean-square (RMS) fluctuation of the signal within this range is at least one order of magnitude lower than that of the linear propagation case. With further numerical simulations, we have pointed out that this highly stabilized range is consistent with the intensity clamping zone, where the laser peak intensity is roughly constant. However, when a filament dies the balance between the self-focusing and the plasma defocusing will be broken. In this case, intensity clamping would not play a dominant role any more. The ultra-high-stability of the laser intensity will then be lost, inducing unstable signal.

As a consequence, any outcome determined by the laser intensity, including fluorescence emission, will be characterized by an ultra-high-stability. For example, it has been demonstrated that the energies of visible short laser pulses generated through four-wave mixing inside a femtosecond laser filament were highly stabilized [19]. In this case, the observed RMS energy fluctuation of the visible pulse is about 1.8%, which is only one third of the values expected in the perturbative limit. Our results have established another example of the highly self-stabilized outcome for nonlinear optical interactions taking place inside filament. We believe that its application in spectroscopy can benefit the robustness of the filamentation based techniques. It has also been confirmed that as long as the longitudinal detection resolution is significantly shorter than the filament length, the stability measurement is weakly affected by the longitudinal precision of the measurement.

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