Enhanced remote filament-induced breakdown spectroscopy with spatio-temporally chirped pulses

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A spatio-temporal focusing technique can be used to enhance the peak intensity in filamentation [Phys. Rev. A 84, 063819 (2011)]. We experimentally applied this technique to remote filament-induced breakdown spectroscopy. Significant enhancement of the fluorescence signals was achieved on iron and aluminum samples from 22 m away using spatio-temporally chirped pulses when compared with using normally compressed pulses. © 2012 Optical Society of America

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Femtosecond laser filamentation [1–4] propagating tens of meters to several kilometers with high intensity has found many applications in remote sensing spectroscopy [5–8]. One example is the strong white light continuum generated during propagation. It provides an ideal light source for absorption in light detection and ranging techniques [7]. Another example is to use the high intensity inside the filament to induce ionization and fragmentation of gaseous targets or breakdown on solid targets, resulting in the emission of characteristic fluorescence spectra [5,9–11]. The latter is called remote filament-induced breakdown spectroscopy [9] and is a powerful tool for real-time surface analysis of materials from a distance. Filament-induced breakdown of solid target is a highly nonlinear process that crucially depends on the laser intensity. However, the peak intensity in a remote filament in air is always clamped even if we continue to increase the input laser energy [12–15]. Although the peak intensity in the filament core could be increased via tight focusing [16,17], the filament could not be launched to a long distance for remote applications. Recently, we have demonstrated the use of spatio-temporally chirped pulses to increase the peak intensity in air filaments and proposed the applicability of this technique in remote sensing spectroscopy [18]. In this work, we demonstrate the applicability of this technique to remote sensing of solid targets, namely, iron (Fe) and aluminum (Al) from up to a distance of 22 m. We shall first show that the above proposed technique using a 100 cm focusing lens could indeed be extended to using telescopic long distance focusing for the increase of intensity as compared to using normally compressed pulses. This was followed by remote filament-induced breakdown on Fe and Al targets showing that a significant improvement of the fluorescence signal at a remote distance of ~22 m on Fe and Al samples could be obtained.

A 10 Hz chirped pulse amplified Ti:sapphire laser beam at a central wavelength of 800 nm was delivered directly from the amplifier without compression. The beam pattern was slightly elliptical with 4.1 mm × 3.2 mm in full width at half-maximum (FWHM) due to the usual performance of such lasers. A cylindrical convex lens (f = 300 mm) and a cylindrical concave lens (f = −50 mm) were employed before the grating pair to reduce the laser beam size in the horizontal direction [18]. After passing a pair of gratings, the temporally chirped pulses were converted into spatio-temporally chirped pulses, and then focused by a 100 cm focal length lens [Fig. 1(a)] to generate a filament. The distance between the gratings was optimized to create the shortest pulse duration only at around the geometrical focus. It is implemented through changing the grating distance and observing the strongest white light generation under single filamentation (lower pulse energy) condition. Spatio-temporally chirped pulses can also be generated by simply removing the cylindrical lenses. In this case, the uncompressed pulses were directly sent to the gratings. The difference between the cases with and without cylindrical lenses will be discussed later. As shown in Fig. 1(a), a 6.7 cm focal length lens, perpendicular to the propagation axis, was used to image the nitrogen fluorescence from a small part of the filament to a fiber bundle (2f-to-2f imaging); the latter was connected to a SpectraPro-500i spectrometer equipped with a PIMAX:512 intensified charge-coupled device (ICCD) camera. The detection allowed us to measure the longitudinal distribution of the fluorescence signal from nitrogen by moving the 100 cm lens, i.e., moving the filament longitudinally (along the propagation direction). The laser pulse energy was adjusted via a half-wave plate and a Glan–Taylor prism inside the amplifier. For comparison, the fluorescence from normally compressed pulses without using the spatio-temporal
focusing technique was also measured. The pulse duration of the Fourier-transform-limited pulses was 50 fs.

Typical longitudinal distributions of the spectral intensities of 337 and 391 nm fluorescence are shown in Fig. 2(a) using the spatio-temporally chirped pulses at 8 mJ per pulse through the cylindrical lenses. The zero position was artificially defined at the peak position of the 337 nm signal. When the pump energy was increased, the peak position shifted slightly away from the geometrical focus [circles in Fig. 2(b)]. The shift of the peak using spatio-temporally focusing without cylindrical lenses [triangles in Fig. 2(b)] is a little more. But the shift of the compressed pulses [diamonds in Fig. 2(b)] was much larger than that due to the spatio-temporally chirped pulses. These shifts indicate that filaments were formed in all the cases.

As Xu et al. have shown recently [19], the strength ratio \( R = \frac{\text{Signal}_{391 \text{nm}}}{\text{Signal}_{337 \text{nm}}} \) of two nitrogen fluorescence lines, namely 391 and 337 nm, can be used to estimate the peak laser intensity in filamentation, where the 391 and the 337 nm lines are assigned to the first negative band system of \( N_2^+ (B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+) \) and the second positive band of \( N_2 (C^3\Pi_u \rightarrow B^3\Pi_g) \), respectively [20]. The peak intensity can then be calculated by the empirical formula [19]

\[
I_0 = 79 \times (2.6/R - 1)^{-0.34} \times 10^{12} \text{ W/cm}^2. \tag{1}
\]

The peak intensity inside the filament as a function of input laser energy is shown in Fig. 2(c) according to the above formula. The flattening of the two curves in Fig. 2(c), one using transform limited pulses and the other using spatio-temporally chirped pulses without the cylindrical lenses, shows that intensity clamping still prevails [21–23]. Multiple filamentation might have occurred at higher pulse energy. However, the peak intensity inside each filament at higher energy is constant; thus, the increase of the number of filaments does not change the ratio \( R \) in Eq. (1) if the peak intensity inside each filament is not changed.

According to Fig. 2(c), the peak clamped intensity in the filament can obviously be enhanced by \(-30\% \) by using spatio-temporal pulses (without cylindrical lenses) as compared to compressed pulses when using the 100 cm focal length lens. We thus extended this technique to the longest available distance in the laboratory for the remote detection of solid targets. The experimental setup is shown in Fig. 1(b). A variable focal length telescope was used to focus the spatio-temporally chirped pulses with cylindrical lenses at \(-22 \text{ m} \) away from the telescope. The telescope consisted of a 5 cm diameter convex mirror, whose focal length was \(-50 \text{ cm} \), and a focusing lens with focal length of 100 cm (diameter of 8 cm). The focusing lens was mounted on a computer controlled motorized stage, allowing us to change the effective focal length of the telescope.

The filament-induced fluorescence signal was collected by a photomultiplier tube (PMT) with a 4\( ^\circ \) imaging lens scanning along the propagation axis. An 800 nm high-reflection mirror and a bandpass interference filter center at 337 nm with a bandwidth of 10 nm were placed in front of the PMT to block the scattered laser beam and other stray lights. However, filamentation could not be formed when the focal length was \(-22 \text{ m} \). The reason can be explained as follows. As one knows, the beam size at the focal plane can be estimated through \( \omega = f_{\text{eff}}/\pi \omega_0 \) when a Gaussian beam with a beam size of \( \omega_0 \) enters a focusing system with effective focal length of \( f_{\text{eff}} \). Only linear propagation is considered in the equation and \( \lambda \) is the laser wavelength. After the cylindrical lenses, the beam size is horizontally reduced. This makes the beam size at the effective focal plane of the focusing telescope

\[
\omega = f_{\text{eff}}/\pi \omega_0
\]
very large, i.e., diffraction dominates. Thus the laser beam defocused rather than focused after the telescope. In order to solve this problem, we removed the cylindrical lenses in the setup. Filamentation was observed. The reason is that the natural beam size resulted in a smaller beam diameter at the effective focus of the telescope. Note that there was still spatio-temporal focusing in this case. With an input energy of 8 mJ, the signal of the 337 nm fluorescence is shown in Fig. 3 as a function of the propagation distance. We used the same telescope configuration to project the spatio-temporally chirped pulses without the cylindrical lenses as well as the compressed 50 fs pulses. In this propagation experiment, there was no temporal chirp in the two kinds of pulses before sending to the telescope. We can see that the filament was shorter and occurred at a farther distance with spatio-temporally chirped pulses (solid curve) than that with compressed pulses (dashed curve). This observation confirms that the spatio-temporal shaping technique can significantly reduce the $B$-integral $B = \frac{2\pi}{\lambda} \int n_2 I(z) dz$, $\lambda$ is laser wavelength, $I(z)$ and $z$ are the optical intensity along the beam axis and the position in beam direction, and $n_2$ is the Kerr nonlinear index after the focusing optics, which can postpone the occurrence of self-focusing and filamentation. In addition, the peak value of the fluorescence signal is about five times stronger.

To further understand the role of the cylindrical lens in the spatiotemporal focusing technique, we redid the experiments with the 100 cm focal lens, while removing the cylindrical lenses. From Fig. 2(b) we can see that, with the cylindrical lenses, the filament was nearer the geometrical focus and the enhancement of the peak intensity was more significant [Fig. 2(c), circles]. This can be easily understood by considering that, after the laser beam passes through the cylindrical lenses, the beam size becomes smaller in the horizontal direction. Thus after being dispersed by the gratings, different spectral components of the laser beam have less spatial overlap, hence resulting in a narrower spectral bandwidth for a specific spatial position [Fig. 4(b)] as compared to the one without cylindrical lenses [Fig. 4(a)]. The dispersion induced by the gratings was calculated by Eq. 1 in [18]. The parameters adopted in the calculation were based on the experimental conditions. The FWHM pulse duration near the focus was shown in Fig. 4(c). It can be seen that the pulse durations were the shortest only at the focus both with and without the cylindrical lenses, and then increased quickly while away from the geometric focus. However, when the cylindrical lenses were induced, the pulse duration increases more rapidly as compared to that without cylindrical lenses. As a consequence, self-focusing would occur closer to the geometric focus and a shorter filament would be formed. The same

![Image](image_url1)

**Fig. 3.** (Color online) Measured nitrogen fluorescence intensity at 337 nm as a function of propagation distance with spatio-temporally chirped pulses without cylindrical lenses (solid line) and normal 50 fs pulses (dashed line) with the same telescope configuration and fixed pulse energy of 8 mJ. The dashed-dotted line indicates the effectively geometric focus of the telescope.

![Image](image_url2)

**Fig. 4.** (Color online) Spectral distribution after the grating pair without (a) and with (b) the cylindrical lenses. (c) Pulse duration in the vicinity of geometric focus without (solid line) and with (dash line) the cylindrical lenses.
energy would be confined in a smaller volume to feed the filament resulting in a higher intensity. This effect was dominant in 1 m lens experiments with high pulse energy. But the cylindrical lenses could also enlarge the beam size at the focus of the telescope as discussed above, making it hard to focus the laser pulses at a long distance.

The significant nonlinear increase of the peak fluorescence signal (Fig. 3) suggests that this technique can also be applied to remotely sensing breakdown spectra from solid targets. The experimental setup is shown in Fig. 1(b). Solid targets were placed at a distance of 22 m to the controlling telescope of the laser beam. The backward fluorescence from the targets was collected near the sending telescope with a 1.5 m focal length Al mirror of 25 cm in diameter and focused onto a fiber bundle connected to the spectrometer for spectral analysis. Unlike that in Fig. 3 which used the same telescope configuration for spatiotemporal focused pulse and normally compressed pulses, in this experiment, the fluorescence signal was optimized separately for each input laser pulse condition. By adjusting the effective focal length of the telescope and optimizing the chirp of the incident pulses, the strongest part of the filament was projected on the surface of the solid target, resulting in the strongest fluorescence signal. To minimize the filamentation-induced white light noise, which was partially from the instantaneous white light in air filament and mainly from the plasma from the solid target, the gate of the ICCD detector was optimized to open 10 ns after the fluorescence signal arrived at the ICCD detector and the gate width was set to 4 μs. Figure 5(a) presents the emission spectrum accumulated by 500 shots from an Fe sample located at 22 m away from the sending telescope with 12.6 mJ input pulse energy. The typical lines in the spectrum, for example, 516.75, 522.72, 526.95, 532.80, 537.15 nm, are assigned to neutral Fe I. Around four times enhancement of the fluorescence was observed by using spatio-temporally focusing pulses without cylindrical lenses as compared to using compressed pulses. The reason is that filament-induced breakdown on solid targets is a highly nonlinear process. Thus, even a slight improvement of the peak intensity would significantly increase the emission of the fluorescence spectrum. The typical spectral lines of neutral Al I at 394.4 and 396.1 nm are shown in Fig. 5(b). The spectra were accumulated by 50 shots. A similar enhancement of the fluorescence intensity by ~3 times was also observed from the Al target.

In conclusion, we have demonstrated the applicability of spatio-temporally focusing technique in remote filamentation spectroscopy. Using spatio-temporal focusing technique makes it possible to concentrate more energy at the focus due to the weaker nonlinear Kerr self-focusing effect suffered by the spatio-temporally chirped pulses before arriving at the geometrical focus. That is to say, this spatio-temporal shaping postpones the occurrence of self-focusing and filamentation by reducing the B-integral. A filament forms only near the geometrical focus; more energy is guided into the filament core, thus increasing the intensity. This could not increase too much because of intensity clamping. Nevertheless, significant enhancement of the fluorescence intensity on Fe and Al samples at 22 m away has been observed. So was the nitrogen fluorescence signal. This, in terms of remote sensing, is an advantage because it allows one to send the beam farther and use the beam more efficiently. We believe the technique will find more applications on remote sensing of other targets by filamentation spectroscopy.

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