Supercontinuum emission from water using fs pulses in the external tight focusing limit

S. Sreeja,[1,2] S. Venugopal Rao,[1] P. Radhakrishnan,[2] Surya P. Tewari,[1] P. Prem Kiran[1,*]
[1]Advanced Centre of Research in High Energy Materials (ACRHEM) University of Hyderabad, Hyderabad 500046, India.
[2]International School of Photonics, Cochin University of Science and Technology, Cochin 682022, India.

ABSTRACT

From the initial observation of self-channeling of high-peak power femtosecond (fs) laser pulses in air, propagation of intense ultrashort laser pulses in different media has become one of the most investigated research areas. The supercontinuum emission (SCE), a spectral manifestation of the spatio-temporal modifications experienced by a propagating ultrashort laser pulse in a nonlinear medium, has many practical applications. However, the extent of blue shift of SCE is reported to be constant due to the phenomenon of "intensity clamping". To further explore the recently observed regime of filamentation without intensity clamping, we measured the evolution of spectral blue shift of SCE resulting from the propagation of fs pulses (800 nm, 40 fs, 1 kHz) in distilled water under different focusing geometries. The efficiency of SCE from tight focusing (f/6) geometry was always higher than the loose focusing (f/12) geometry for both linear and circular polarized pulses. The blue edge of the SCE spectrum (λ_{min}) was found to be blue shifted for f/6 focusing conditions compared to f/12 focusing geometry. The lower bound of the intensity deposited in the medium measured from the self-emission from the filament demonstrated the existence of intensities ~ 6 ×10^{13} Wcm^{-2}, far beyond the clamping intensities achieved erstwhile.

Keywords: intense fs laser pulses, Supercontinuum emission, tight focusing, propagation in water

1. INTRODUCTION

The propagation of intense femtosecond pulses in transparent condensed media or gases results in strong modification of its spatial profile due to self focusing/ defocusing and temporal profile due to self phase modulation [1-3]. The broad frequency sweep, so called Supercontinuum emission (SCE) of the pulse extending from ultraviolet to the infrared range is spectral manifestation of spatiotemporal modifications of the focused laser beam in a nonlinear medium. SCE associated with filamentation of fs laser pulses occurs when the laser power (P) exceeds the critical power for self-focusing (P_{cr}) in the medium [3,4]. Understanding the dynamics of the filamentation and the associated phenomenon benefits both fundamental science from the recently proposed analogy between Hawking radiation and the optical filament propagating inside glass [5] and the applications of SCE as a tunable source for pump–probe measurements [2], optical pulse compression, [6] fs-LIDAR, and remote sensing to name a few [7]. Despite wide variety of applications, generation of SCE phenomenon associated with filamentation has been associated with long range propagation of intense fs laser beams in diverse media mainly confined to unfocused or loosely focused geometries [8,9]. While many applications using SCE require larger blue shifted spectrum, the extent of the blue shift of SCE from condensed media is reported to be constant due to the phenomenon of intensity clamping [9]. The phenomenon of intensity clamping proposed by Braun et al. [10] has been a limiting factor to the blue shift of the SCE spectra. However, many recent works have predicted and demonstrated the presence of high peak intensities (~10^{14} Wcm^{-2}) in gaseous media under the external tight focusing conditions [11-13]. In view of this we ask the following question: Can the external tight focusing lead to higher intensity inside a filament in water? To understand this, we present our results from studies on the effect of external focusing on SCE evolution (more specifically spectral blue shift) resulting from the propagation of focused ~40 fs pulses in distilled water achieved with different focusing geometries. Our experimental results suggest that under tight focusing geometry the blue shift of SCE spectrum is enhanced. We investigate effects of linearly polarized (LP) and circularly polarized (CP) pulses on the spectra obtained for two different focal geometries.

*premsp@uohyd.ernet.in; phone: +91 40 2313-8842; fax +91 40 2301-2800

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2. EXPERIMENT

The experimental setup for generating SCE from water is illustrated in figure 1. Transform limited pulses (confirmed from the measurements using Silhouette, Coherent) with duration of ~40 fs, 800 nm, p-polarized with a repetition rate of 1 kHz (Coherent; Legend-USP) are focused into an 8 cm long glass cuvette containing doubly distilled water. The amplifier was seeded with ~15 fs pulses from an oscillator (MICRA, Coherent, ~1W average power, 80 MHz repetition rate, and 800 nm). BK-7 lenses of focal length 60 mm and 120 mm were used to attain the focal geometries of f/6 and f/12 respectively. The input diameter ($1/e^2$) before the focusing element was 10±0.1mm.

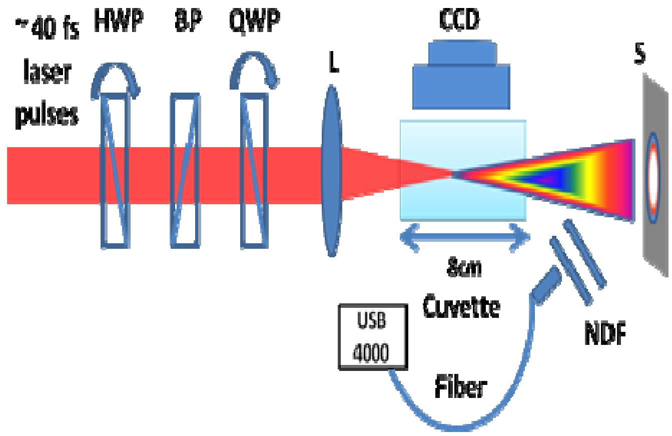

Figure 1 Experimental setup for investigating SCE from water using focused fs pulses. The various components in the figure are identified as follows HWP-Half Wave Plate, BP- Brewster Polarizer, QWP-Quarter Wave Plate, L- Lens, C-Cuvette, S-Screen, NDF-Neutral Density Filter and CCD - Camera.

An attenuator, combination of a half wave plate (HWP) and a Brewster polarizer (BP) was used to vary the input pulse energy into the medium. A quarter wave plate (QWP) was employed after the attenuator to change the polarization of the pulse. The SCE spot was scattered using a Teflon sheet kept at a constant distance from end face of the cuvette for both the focusing geometries. The scattered light was collected using a fiber optic coupled spectrometer (USB 4000, Ocean Optics Spectrometer). A CCD camera was used to image self-emission from the propagating filament inside the water cuvette and to estimate the diameter of the energy reservoir surrounding the filament inside the medium. A set of calibrated neutral density filters were placed in front of the spectrometer to avoid saturation.

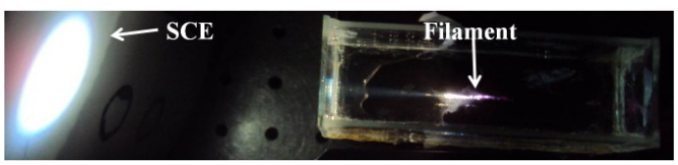

Figure 2 Top view of the filament within the water cuvette and the SCE generated with f/6 focusing geometry at a power of 6700 PCr for linear polarized (LP) pulses.

SCE spectra were collected for 350 shots to reduce the noise from pulse to pulse fluctuations. The experiment was performed for both linearly and circularly polarized pulses. The pulse energy was monitored using a power meter (Coherent, PM30). The critical power for self focusing in water (P_{Cr}) is ~4.4 MW according to the equation $P_{Cr} = 3.77\lambda^2/8\pi n_0 n_2$ where λ is the central wavelength, n_0 and n_2 are the linear and nonlinear refractive indices, respectively. The corresponding input powers are in the range of 20 – 7000 P_{Cr} for both f/6 and f/12 geometries.

3. RESULTS AND DISCUSSION

Figure 2 shows the image of filament propagating inside water and the associated SCE at an input power of 9100 P_{Cr} for LP pulses in the f/6 focusing geometry. The femtosecond pulses propagating in water produced SCE with conical colored rings with predominantly white color in the central portion. Figure 3 depicts the SCE spectrum recorded at different input peak powers. The spectra demonstrate broadening towards both sides of the central wavelength. The symmetrical spectral broadening about the incident laser wavelength (800 nm) is ascribed to the self-phase modulation (SPM) that arises from Kerr nonlinearity. The asymmetric component in blue spectral region occurs due to processes such as space-time focusing, self-steepening, [14] and plasma formation that arise from free electrons generated by multi-photon ionization (MPI) in gases and multi-photon excitation (MPE) in condensed media [1]. In addition to the asymmetric broadening, there is a marked dip near 611 nm that is superimposed on the white light continuum. This dip corresponds to inverse Raman Effect for the OH stretching bond (3650 cm^{-1}) [15, 16]. The Raman dip increases with increasing excitation energy which seems natural since more of the excitation energy is converted into the Raman mode.

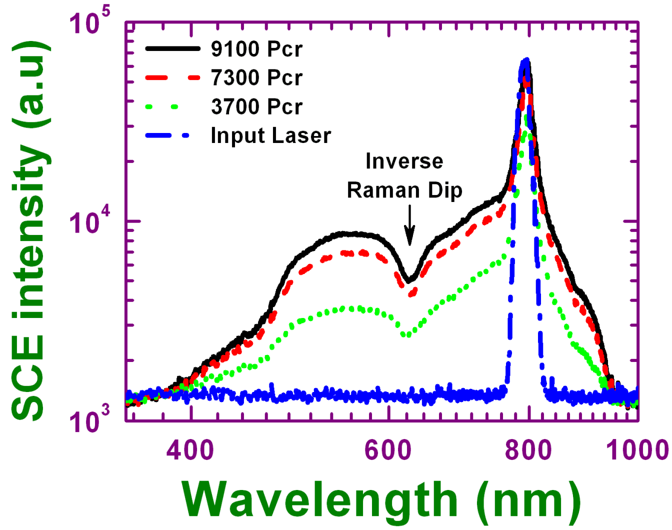

Figure 3 SCE spectra recorded for different input powers in the f/6 focal geometry. Input pulse spectrum is also overlaid for reference. The plots are in log10-log10 scale for convenient comparison.

The SCE spectra observed for different focal geometries of f/6 and f/12 at an input power of 9100 P_{Cr} for CP pulses are shown in figure 4. The spectra show noticeable increase in both the symmetric and asymmetric branch of SCE for tight focusing geometry. Tighter focusing in case of f/6 geometry has resulted in increased conversion of incident laser pulse into SCE, indicating the role of tighter focusing on the evolution of SCE spectrum in terms of the increased peak intensity at the focal plane. As the input polarization of the pulse was changed from LP to CP (simply by rotating the quarter wave plate) a reduction in the SCE intensity for both f/6 and f/12 geometries was observed, which is in agreement with earlier reports [15, 16]. The ratio of 1.5 of the critical powers for self focusing with LP and CP pulses explains the reason for MPE being less efficient for CP pulses [4].

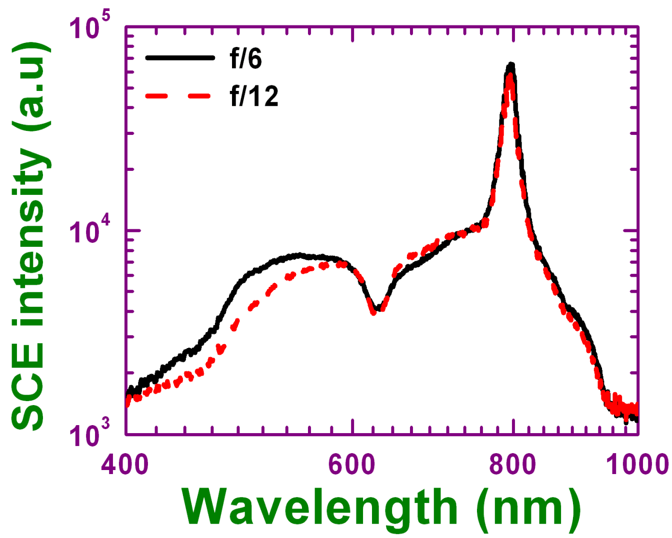

Figure 4 SCE spectra from water with f/6 (black) and f/12 (red) focusing geometries at an input power of 6700 PCr for circularly polarized pulses.

The blue edge of the SCE spectrum i.e., the minimum cutoff wavelength (λ_{min}) or maximum positive frequency shift (ω_{max}) as a function of the input laser power from the collected SCE spectra is found to decrease continuously with increasing input power for both focusing geometries. Figure 5 shows the variation of λ_{min} in the f/12 focusing geometry for LP and CP pulses. At lower input powers (up to 100 P_{Cr}) λ_{min} is coinciding with the blue edge of input laser pulse. Beyond 100P_{Cr} we start observing the shift in λ_{min} going much beyond the blue edge of input laser pulse. Above 300 P_{Cr} the λ_{min} reached a value of 465 nm. Further increase of input power (beyond 1000 P_{Cr} and up to 5000 P_{Cr}) λ_{min} reached a value of 380 nm. Further increase in input power resulted in no appreciable change of λ_{min} observed. The difference between λ_{min} with LP and CP pulses observed is very small (~ 3 nm). The λ_{min} is found to be blue shifted for f/6 focusing conditions compared with f/12 focusing geometry. This clearly indicates presence of higher intensities in the vicinity of interaction region (focal plane) for f/6 focusing geometry revealing the blue shift of the SCE spectra due to the external focusing of the fs pulses inside transparent media. A similar result is observed in the SCE from air and argon under tight focusing conditions [11,13]. However, the variation in λ_{min} is observed to be small compared to that observed with air. This may be attributed to the increased plasma density around the focal plane (within the interaction region) of laser pulse inside water and the nonlinear refractive index of water that governs the extent of self-focusing of fs pulses inside the medium.

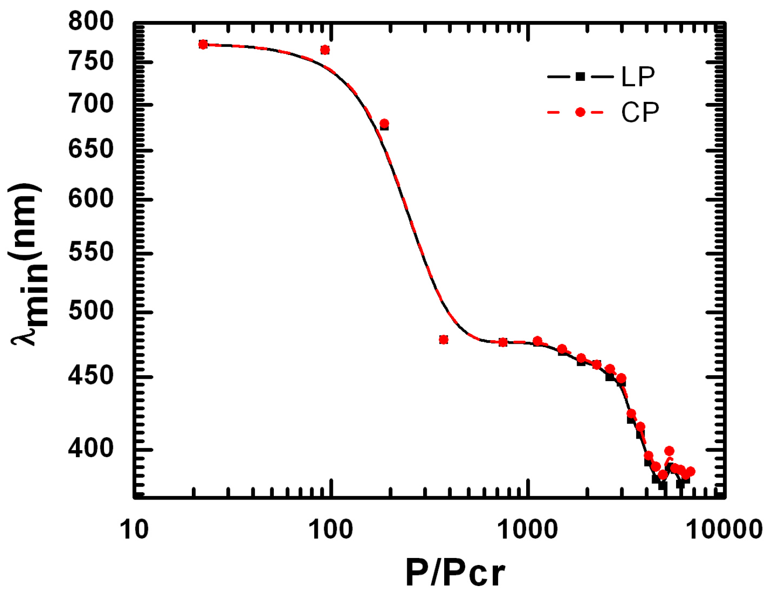

Figure 5 Variation of minimum wavelength (λmin) with P/Pcr for f/12 geometry for linearly and circularly polarized fs pulses.

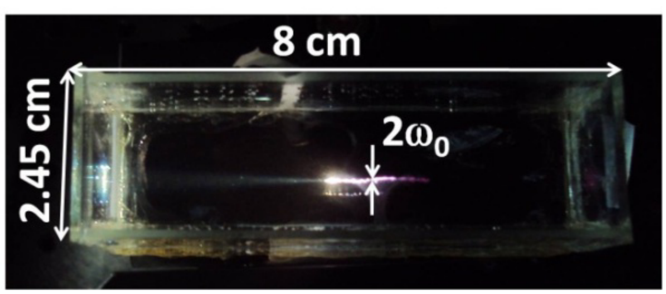

Figure 6 Image of the self-emission due to filamentation inside water with linear polarized pulses for f/6 focusing geometry at an input power of 6700 PCr.

The lower bound of the maximum peak intensity around the focal plane in the f/6 focusing geometry is estimated from the diameter of the energy reservoir **~360 µm** surrounding the filament observed in the water (shown in figure 6). The intensity corresponding to 6700 P_{Cr} is **6.6×10^{13} W/cm^2**. The estimated intensity of ~6×10^{13} W/cm^2 around the focal region inside the medium reveals that under tight focusing conditions, it is possible to have the intensities higher than the values predicted erstwhile. In gaseous media, it is well understood that in the tight focusing geometry initial high beam curvature, due to external focusing, leads to complete ionization of the medium, in turn preventing the plasma defocusing to play any significant role in the intensity clamping [12]. A similar phenomenon paving way for plasma densities corresponding to fully ionized medium [17,18] and intensities larger than the clamping values in air [11] due to the external tight focusing conditions are reported recently. However, in condensed media like water or in solids the phenomenon may be totally different due to low ionization thresholds and higher densities. Many other phenomenon like (i) the propagation of tightly focused fs pulses inside water, (ii) the effect of pulse splitting (iii) ionization and breakdown phenomenon of water at intensities greater than 10^{13} Wcm^{-2} within the filament [19], (iv) interaction of the cavitation bubbles in water [20] needs to be understood completely in order to devise practical high intense SCE source.

4. CONCLUSION

In conclusion, effect of external focusing on the evolution of SCE resulting from the propagation of ~40 fs laser pulses in water with different external tight focusing geometries is studied. The SCE from tight focusing geometry is always higher than the loose focusing geometry for both LP and CP pulses. For tight focusing geometries, ultrashort laser pulse filamentation in water does not lead to a constant minimal cutoff wavelength (λ_{min}) for the SCE. Deposition of laser intensities of approximately 10^{13} W/cm^2 near the focal region with tighter focusing conditions enhancing the blue shift of SCE paves course for novel intense sources of radiation and several possible practical applications.

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