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CONICAL EMISSION OF TI:SAPPHIRE FEMTOSECOND LASER PULSES PROPAGATING IN WATER

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Conical emission is investigated for Ti:sapphire femtosecond laser pulses propagating in water. The colored rings can be observed in the forward direction due to the constructive and destructive interference of transverse wavevector, which are induced by the spatio-temporal gradient of the free-electron density. With increasing input laser energy, due to filamentation and pulse splitting induced by the plasma created by multiphoton excitation of electrons from the valence band to the conduction band, the on-axis spectrum of the conical emission is widely broadened and strongly modulated with respect to input laser spectrum, and finally remains fairly constant at higher laser energy due to intensity clamping in the filaments.

Keywords: Conical emission; self-focusing; self-phase modulation; plasma.

1. Introduction

An intense ultrashort laser pulse propagating in nonlinear media can generate an output with a nearly white light, called the supercontinuum (SC).[1–3] The nonlinear phenomenon has attracted considerable attention for several decades because of its extensive applications, such as tunable ultrashort pulse generation,[4] optical parametrical amplification,[5] lighting control,[6] and the detection and identification of pollutants in the atmosphere.[7] Various models have been used to describe the phenomenon, self-phase modulation following self-focusing and optical breakdown is considered as the dominant mechanism for the supercontinuum spectrum generation.[3] In addition, self-steepening,[8] material dispersion,[9] and stimulated Raman scattering[10] have contributions to the supercontinuum spectrum as well. The experimental studies are also faced with many challenges, and the main experimental difficulty is focused on the measurement of filaments since the peak intensity far exceeds the damage threshold of any measurement instrument. Some indirect ways, such as the measurement of output spectra,[3] fluorescence spectra[11] and CCD camera image,[12] have been used to investigate the propagating properties of the intense laser pulse in nonlinear media.

The broadening of the spectrum is usually accompanied by angular emission. The conical emission (CE) appears in the forward direction at a small angle, which has been extensively investigated in experiments and numerical simulations.[13–17] Golub[13] believed that the CE arose from the Čerenkov effect, and Xing *et al.*[14] attributed it to the four-photon parameter process, however there the ionization is not considered. It is known that (quasi-) free electrons via multiphoton excitation can easily generate the intense laser pulse propagating in a medium. Therefore, Kosareva *et al.*[17] attributed the CE to the spatio-temporal self-phase modulation of the pulse in the plasma generated by the moving focus. In this paper, we report the conical emission of intense Ti:sapphire femtosecond laser pulse propagating in water. Many colored rings are observed, and the on-axis spectrum of the conical emission is widely broadened and strongly modulated with respect to the laser spectrum, and their mechanisms will be explicitly elucidated in the present context.

2. Experiments

In our experiment, a mode-locked Ti:sapphire laser (Spectra-Physics TSA regenerative amplifier) is used as the excitation source with a pulse duration of about 40 fs, a repetition rate of 10 Hz and a center wavelength of 800 nm. The laser output peak power far exceeds the self-focusing threshold of water, therefore the filament in water can be observed without lens focusing. As shown in Fig. 1, the output beam is sent into different long cells (10 cm, 50 cm, and 120 cm) by a reflector. The photograph of conical emission is taken on a white screen placed at a distance of 20 cm after the cell with a CCD camera. The OOIBase32 spectrometer is used to measure the on-axis spectrum of the conical emission. At higher input laser energies, a blue filter BG18 is placed before the spectrometer to filter the strong central part of the laser spectrum around 800 nm.

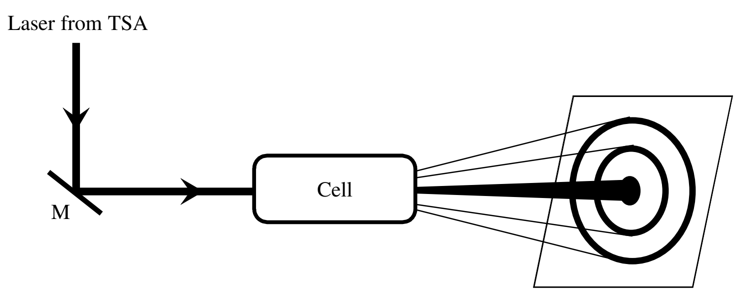
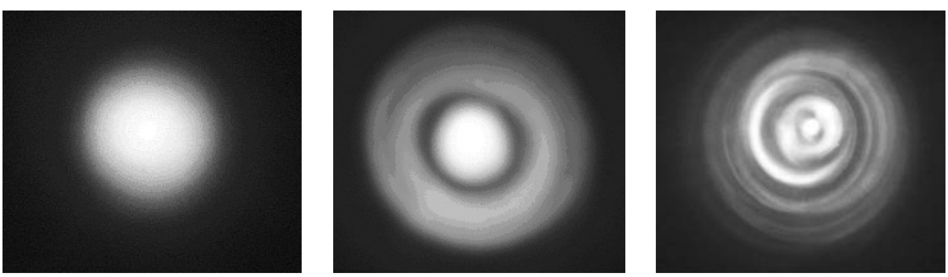


Fig. 1. Experimental setup for measuring conical emission.

3. Results and Discussion

Self-focusing is a dynamical optical Kerr effect, which causes beam shrinkage and the peak intensity to increase. Self-phase modulation is a consequence of self-focusing in the temporal domain and the essential origin of spectral broadening.[8] However, self-phase modulation in space on the transverse profile of a beam is also possible.[1] It appears as a distortion on the wavefront and results in self-focusing. For a beam with a Gaussian-like transverse profile, the phase increment $\Delta\phi(r)$ across the beam profile has a bell shape centered at $r = 0$. If $[\Delta\phi(r)]_{\max}$ is much larger than 2π, then the output beam in the transverse wavevector space shows peaks and valleys due to constructive and destructive interference. The interference rings will appear on a projection screen in the forward direction, and the number of bright rings is approximately given by the integer of $[\Delta\phi(r)]_{\max}/2\pi$. Figure 2 shows a photograph of conical emission on the white screen through three different length cells (10 cm, 50 cm, and 120 cm) at a laser energy of 23 mJ. No ring is observed in a 10 cm-long cell, one ring is observed in a 50 cm-long cell, and four rings can be observed in the 120 cm-long cell. The spectral characteristics of the conical emissions are observed by transversely scanning the output laser beam with the OOIBase32 spectrometer. The central part exhibits a continuous distribution of angular anti-Stokes radiation, and its wavelength appears blue-shift with the emission angle increasing. The outer rings consist of the visible spectrum, and their emission wavelengths also appear blue-shift with the emission angle increasing.

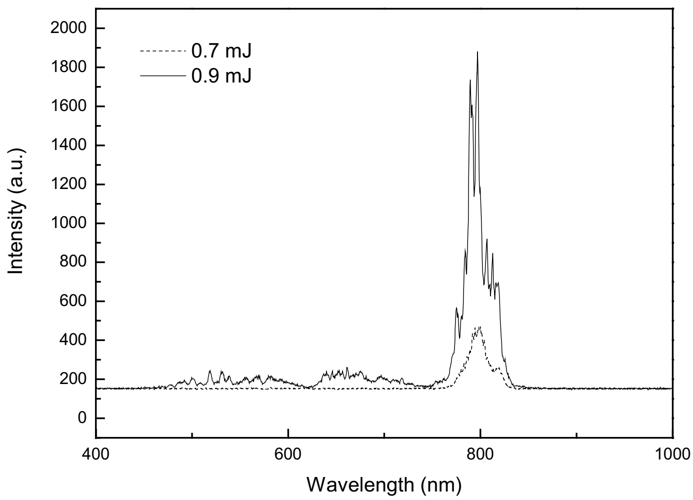
We assume that the input laser pulse is Gaussian profile in space and time, such that $\mathrm{E}(r, z = 0, \tau = t - z/\upsilon_g) = \mathrm{E}_0(-r^2/2a^2 - \tau^2/2\tau_p^2)$. When the laser pulse propagates in a nonlinear media, the pulse experiences time-dependent self-focusing with little ionization up to $z = z_f$ (self-focus distance), where a self-focus is formed at the peak slice ($\tau = 0$). At this point, the self-focusing action is balanced with the plasma defocusing created by the multiphoton excitation from the valence band to the conduction band.[18–20] With further propagation, the other slices further self-focus along the z-axis, and the self-focus moves towards the front of the pulse. Meanwhile, the back of the pulse is defocused by the free electrons produced by the



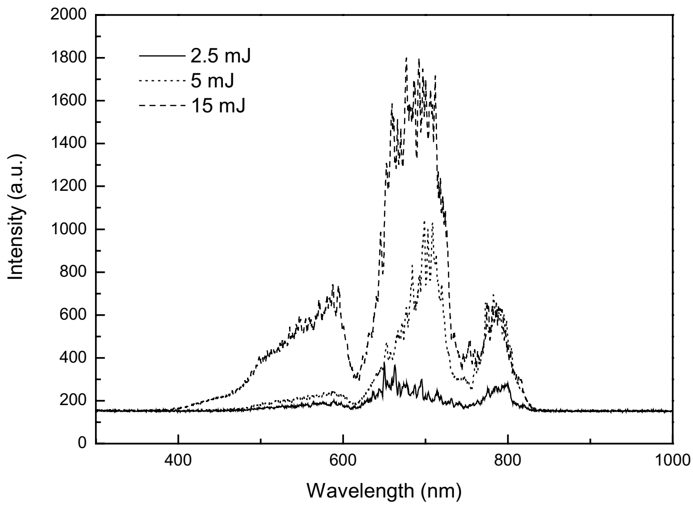
(a) (b) (c)

Fig. 2. Conical emission of femtosecond pulse propagating in water with different long cells: (a) 10 cm; (b) 50 cm; (c) 120 cm.

front of the pulse, resulting in a spatio-temporal self-phase modulation.[20,21] The temporal phase gradient (the frequency deviation $\delta\omega(\tau)$) is proportional to the time derivative of the electron density: $\delta\omega(\tau) = \partial\phi/\partial\tau \sim \partial N_e(\tau)/\partial\tau$. The spatial phase gradient (the transverse wavevector δk_r) is proportional to the radial derivative of the electron density: $\delta k_r = \partial\phi/\partial r \sim \partial N_e(r)/\partial r$. During the propagation of the intense laser pulse in nonlinear medium, the spatio-temporal gradients of the electron density ($\partial N_e(\tau)/\partial\tau$ and $\partial N_e(r)/\partial r$) simultaneously increase due to the



(a)



(b)

Fig. 3. Power spectra obtained for the pulse propagating in water at various average input powers: (a) at low average input power, (b) at high average input power (using BG18 filter).

self-focusing. As a result, the outer rings also exhibit the wavelength-dependent angle, and the shortest wavelength corresponds to largest divergence.

In order to reveal the conical emission involved in different frequency components, we measure the on-axis spectrum of conical emission at various input laser energies in the cell of 120 cm, as shown in Fig. 3. At low laser energy, the laser spectrum is slightly broadened and its peak wavelength is slightly blue-shifted. As the laser energy is increased to 0.9 mJ, the laser spectrum is widely broadened and strongly modulated by a large oscillatory feature, and two weaker peaks appear around the wavelengths of 570 and 670 nm (as shown in Fig. 3(a)). As the input laser energy increases, the spectrum is continuingly broadened and the two peaks gradually intensify (as shown in Fig. 3(b)). Finally, the spectral structure remains unchanged, and the shortest wavelength of about 394 nm can be observed. Moreover, similar spectral evolution is observed in the on-axis spectrum of the conical emission in the cell of 10 and 50 cm.

The supercontinuum spectrum generation correlates with two nonlinear effects: nonlinear polarization and plasma formation.[3,22] The two processes have a different intensity dependence on the laser peak intensity. The spectral broadening is dominated by the nonlinear refractive index at low laser energy and by plasma contribution at high laser energy. When the laser energy is too low for the filament to be formed, the slight broadening of the spectrum is due to the influence of the nonlinear refractive index. As the laser energy is up to 0.9 mJ, the corresponding peak intensity is $P \approx 22.5\,\text{GW}$, several thousand times higher than the self-focusing threshold $P_{\text{crit}} = 3.77\lambda^2/(8\pi n n_2) \approx 2.65\,\text{MW}$ (where $n = 1.34$ and $n_2 = 2.7 \times 10^{-16}\,\text{cm}^2/\text{W}$). The filaments are formed and the laser pulse generates splitting,[23–26] which result in the laser spectrum being broadened and modulated. At high input laser energy, the plasma contribution is considered as the main mechanism for the spectral broadening.[3,8] Since the plasma density $(N_e(r, z, \tau))$ crucially depends on the peak intensity and the intensity of the pulse is clamped by the ionization threshold,[3,11] the maximum intensity will limit the spectral broadening range, and so finally the spectral structure will remain unchanged even if the input laser energy is continually increased.

4. Conclusions

In summary, the conical emission of Ti:sapphire femtosecond laser pulse propagating in water has been observed. The observed colored rings in the forward direction can be attributed to the constructive and destructive interference of transverse wavevector induced by the spatio-temporal gradient of the free-electron density. At low laser energy, the on-axis spectrum is slightly broadened, which can be attributed to the nonlinear refractive index. With increasing laser energy, the on-axis spectrum is widely broadened and strongly modulated due to the filamentation and pulse splitting induced by the plasma created by multiphoton excitation of electrons from the valence band to the conduction band. Finally, its spectrum remains fairly constant at higher laser energy due to intensity clamping in the filaments.

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