Polarization-dependent supercontinuum generation from light filaments in air

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We investigate polarization-dependent properties of the supercontinuum emission generated from filaments produced by intense femtosecond laser pulses propagating through air over a long distance. The conversion efficiency from the 800-nm fundamental to white light is observed to be higher for circular polarization than for linear polarization when the laser intensity exceeds the threshold of the breakdown of air.

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The propagation of intense femtosecond laser pulses is attracting considerable interest.[1–4] Femtosecond laser pulses can propagate over long distances because of balances between the opposing nonlinear effects. Initially, the intensity-dependent refractive index in air leads to self-focusing. After the intensity of the laser reaches the ionization threshold of air, a low-density plasma is produced. This leads to defocusing of the laser pulse. The self-focusing and the defocusing processes repeat and guide laser pulses to propagate over a distance much longer than the Rayleigh length.[1] Many interesting phenomena have been observed, such as supercontinuum radiation,[5] conical emission,[2] third-harmonic generation,[6] and other plasma-related phenomena such as the laser's backscattering enhancement.[7]

Our Letter concentrates on supercontinuum emission (SCE), which has been observed in both solid and liquid media since 1970.[8] SCE is considered a useful light source with an ultrashort pulse duration and a large spectral extension from the ultraviolet to the infrared.[9] For instance, it can be applied to laser spectroscopy, such as remote detection of atmospheric pollutants[3] and lidar remote-sensing measurements.[9] However, the detailed physical mechanisms involved in supercontinuum generation remain an open question. Self-phase modulation, four-wave mixing, and plasma production are the processes most commonly invoked to explain continuum generation. Many parameters of the laser pulse, such as energy and polarization, affect supercontinuum generation. Some works have addressed supercontinuum generation in condensed materials[10] or in terms of the ion yield rate in air.[11] In this Letter the polarization-dependent properties of supercontinuum generation in air are presented. This information is important for a better understanding of the physical mechanisms involved in filamentation and of optimization of white-light generation for spectroscopic applications.

Our experimental setup is shown in Fig. 1. We used a homemade 10-Hz Ti:sapphire chirped-pulse amplifier laser system. It delivers pulses of 30-fs duration (FWHM) with a central wavelength of $\lambda = 800$ nm and a maximum energy of up to 650 mJ per pulse. In experiments we obtained different laser polarization states by rotating the quarter-wave plate placed at the output of the compressor. Laser pulses were launched in air and focused slightly by a positive lens with a focal length of 2 m. Approximately 70% of the laser energy was in the focusing area, producing an intensity of $\sim 10^{15}$ W/cm^2 in vacuum. A scatter screen was used in the path of the laser to observe the scattered

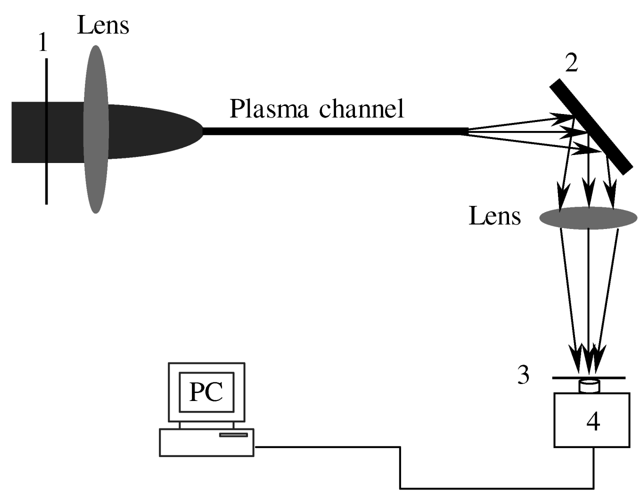

Fig. 1. Experimental setup: 1, quarter-wave plate; 2, scatter screen; 3, bandpass filter; 4, spectrometer.

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radiation, which was collected after a bandpass filter by a silica lens and sent into a CCD coupled spectrometer with a detection range of 200–1100 nm.

The linearly polarized laser pulse produced a plasma channel that was approximately 1 m long. White light was generated, as well as a pattern of concentric color rings from conical emission at the end of the channel. SCE and conical emission were studied previously[2,5] with ultrashort laser pulses, ranging in power from gigawatts to terawatts. The white light was observed at the center of the channel and was surrounded by color rings with increasing diameter for decreasing wavelength. In our experiments, when we gradually changed the polarization of the laser pulse from linear to circular by rotating the wave plate, the brightness of the plasma channel gradually increased correspondingly. It is obvious that white light is produced much more efficiently with circular polarization than with linear polarization.

Detailed spectra of the white light generated in the channel is shown in Fig. 2(a) for the two polarizations with the same laser energy. The spectra are normalized with respect to the maximum with linear polarization. Figure 2(b) shows the ratio of the spectral intensity produced with circular polarization to that with linear polarization. We found that the spectra are dominated by the blueshifted part rather than the redshifted part, which is the same trend others have found.[2,5,10] $\chi^{(3)}$ processes contribute partially to SCE generation. In our case, however, phase modulation from ionization processes dominates SCE generation.[5] When the laser pulse propagates through air, multiphoton ionization (MPI) produces the plasma channel within which the MPI will continue to increase the plasma electron density. Another process that can increase the electron density in the plasma channel is electron impact ionization (EII). Previous investigations of the characteristics of the plasma channel show that the electron density is between 10^{16} and 10^{18} cm^{-3}, which yields a collision ionization time scale between 1 ns and 10 ps if the EII cross section is 10^{-16} cm^2 and the electron energy is approximately 300 eV.[12] This time scale is much longer than the laser pulse duration, meaning that EII plays a small role in the electron density increase where the laser enters the plasma channel. It is the MPI processes that dominate SCE generation. The frequency blueshift caused by the above processes is[13,14] $\Delta\omega = -(\omega_0/c)\int_0^z(\partial n/\partial t)(l)\mathrm{d}l$, where ω_0 is the angular frequency of light, l is the plasma length over which MPI takes place, and $n = (1 - N_e/N_c)^{1/2}$ is the index of refraction of the plasma through which the laser propagates. The continuous increase in electron density from MPI processes decreases n and produces a broad spectrum of emission. Since the laser pulse lasts only several tens of femtoseconds and the plasma expands little while the laser propagates through it, the laser always experiences a net decrease in n and $\Delta\omega$ has positive values, meaning that the spectrum should be dominated by a blueshift wing.

We also found that the polarization dependence of the spectral profile is different from that of Sandhu *et al.*[10] who observed the suppression of supercontinuum generation with circular polarization when the same laser intensity was applied as in the linear polarization case. In contrast, our experiments show that the spectra of the circular polarization case have higher intensities in the visible range; i.e., circular polarization has a higher conversion efficiency from the fundamental into the supercontinuum. Correspondingly, around the fundamental wavelength, the linearly polarized laser pulse reserves more energy, as shown in Fig. 2(b), which is consistent with our qualitative observation made by eye. We found that the ratio is generally between 1 and 3 in the blue wing of the spectra, whereas on the side of the longer wavelength, the signal levels with the two polarizations are similar. Sandhu *et al.*[10] attributed such suppression of SCE to the weaker $\chi^{(3)}$ processes of the circularly polarized laser in the medium, combined with other nonlinear optical processes that are also sensitive to laser polarization. But which processes play such important roles is still not clear. We note that Sandhu *et al.*[10] managed to keep the laser intensity under the damage threshold of the medium. In our case, however, the laser intensity after self-focusing due to nonlinearity is high enough to produce a plasma channel in air by MPI. The plasma scatters and defocuses the trailing part of the laser pulse, which counteracts the $\chi^{(3)}$ processes. Using the generation mechanism of the blue wing of the spectra discussed above, we can explain the polarization dependence of the spectral profile as follows.

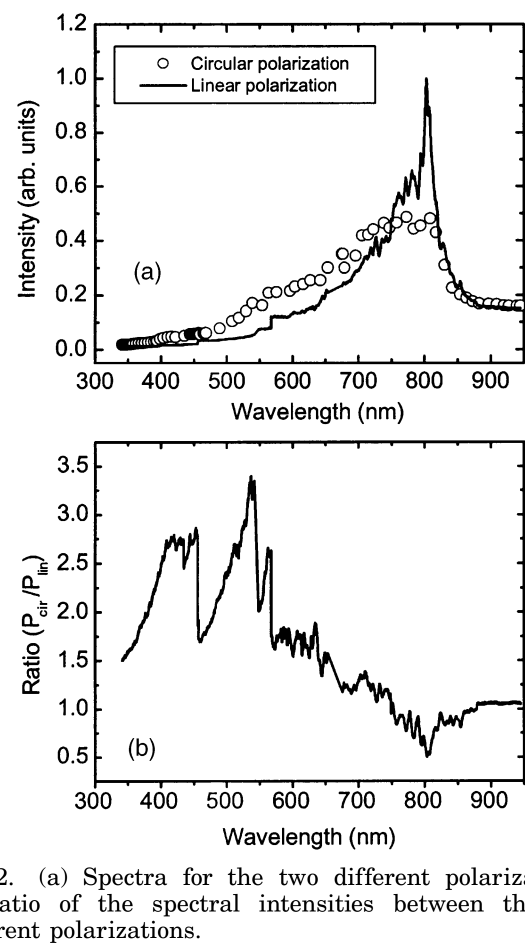

Fig. 2. (a) Spectra for the two different polarizations. (b) Ratio of the spectral intensities between the two different polarizations.

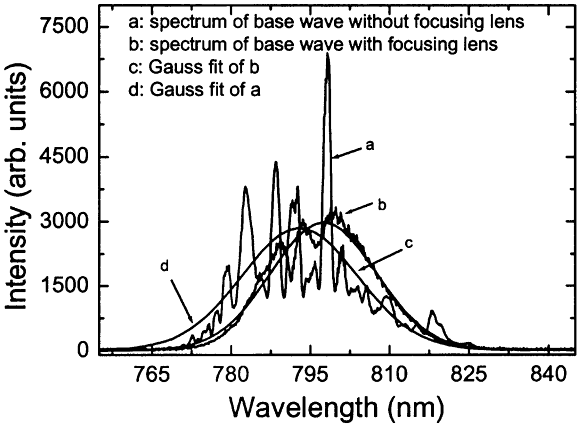

Fig. 3. Blueshifted spectra of the fundamental when intense laser pulses propagate in air.

Self-focusing of the circularly polarized laser pulse occurs later than that for the linearly polarized pulse since $\chi_{\mathrm{cir}}^{(3)}$ is smaller than $\chi_{\mathrm{lin}}^{(3)}$.[15] Because of this effect and the less-efficient MPI for the circularly polarized laser,[16–19] the part of the plasma channel before and after the focus is longer but more tenuous with the circularly polarized laser pulse than with the linearly polarized pulse, allowing the laser pulse to propagate a longer distance along the plasma channel and focus to a higher final intensity. Since the frequency shift is proportional to the temporal differential of the integrated refractive index with respect to distance, stronger SCE appears to be produced in the longer propagation distance within the plasma channel, with the electron density (the refractive index) capable of being continuously increased (decreased) by the MPI and EII processes. Moreover, the higher intensity in the focus of the circularly polarized laser pulse enhances the temporally and spatially abrupt change in the refractive index. The MPI in the refocusing process for the circularly polarized laser pulses is also considered a contributor to the higher conversion efficiency of the fundamental into SCE.[11] The ionization-induced self-phase modulation also plays an important role in the spectral profile difference between linear and circular polarization. It causes only anti-Stokes broadening and blueshifting of the spectrum,[20,21] as shown in Fig. 3. The center of the spectrum is shifted approximately 5 nm with linear polarization when we focus the beam with a lens with a shorter focal length of $f = 50$ cm.

In summary, we have reported polarization-dependent supercontinuum generation when femtosecond pulses propagate in air. Our results show that the spectral intensity in the visible range of the supercontinuum is larger for circular polarization than for linear polarization. It is more efficient to use the circularly polarized laser to transfer the fundamental energy to the supercontinuum. The polarization dependence of the supercontinuum generation can have applications in remote sensing with femtosecond white light or laser lightning control.

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