Backward supercontinuum emission from a filament generated by ultrashort laser pulses in air

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Backward emission of the supercontinuum from a light filament induced by high-intensity femtosecond laser pulses propagating in air has been observed to be enhanced compared with linear Rayleigh–Mie scattering. This enhancement is interpreted as a nonlinear scattering process onto longitudinal refractive-index changes induced by the laser pulse itself. The spectral dependence of the supercontinuum angular distribution is also investigated. © 2001 Optical Society of America

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High-intensity ultrashort laser pulses propagating in air have been observed to self-collimate into long filaments over a distance that substantially exceeds the Rayleigh length.[1] Such light filaments provide long interaction paths, leading to an ultrabroadband continuum from the UV to the IR.[2–4] In the forward direction, this supercontinuum exhibits conical emission with a spectacular pattern of concentric colored rings.[5–7] The self-channeling model interprets filamentation as the result of a balance between self-focusing owing to the Kerr effect and the combined effects of natural diffraction and refraction from a low-density plasma.[1,5] Use of the moving focus model to explain filamentation as well as conical emission in the femtosecond regime has also been proposed.[6,7] More recently, use of the spatial replenishment model[8] to describe the filamentation that results from dynamic guiding has been suggested. Longitudinal effects on a pulse wave packet, such as pulse self-shortening[9,10] and pulse splitting,[11] have been reported. Notice that numeric simulations have been carried out.[12]

Recently, supercontinuum emission and propagation of terawatt laser pulses were observed over several kilometers in air by use of a lidar arrangement.[13,14] For the new applications in atmospheric remote sensing, key issues are the origin and the underlying physical processes of white light detected at large distances. The question is whether this white light is due to *in situ* backward emission from remotely located filaments or to subsequent Rayleigh–Mie backscattering of white light generated in the forward direction by filaments at shorter distances.

In this Letter we show, for the first time to our knowledge, that the supercontinuum emission from a filament is enhanced in the backward direction compared with linear backscattering. This enhancement is interpreted as being due to a backscattering process in laser-induced longitudinal refractive-index changes caused by Kerr and plasma effects.

The experimental setup that we used to measure the angular dependence of the supercontinuum emitted by a filament in air was based on a chirped-pulse amplification femtosecond laser system providing 6 mJ of energy in 120 fs at 810 nm. The 12 mm-diameter output beam was slightly focused by a spherical mirror of 10-m radius of curvature. The beam propagated in free space over 80 m after the region investigated (1.5 m downstream from the geometrical focus). The direction of propagation of the incident beam was considered the forward-scattering direction and defined as $\theta = 0°$ (see Fig. 1). The supercontinuum emitted by the filament was collected by a 6-mm-diameter liquid optical fiber with a restricted field of view of 0.86° by a nontransparent guiding tube. The overlap between the field of view of the detection system and the filament (and hence of the signal) is proportional to $1/\sin(\theta)$. We mounted the fiber onto a stepper-motor-driven goniometer to measure the angular dependence from $\theta = 5°$ to $\theta = 176.5°$. In most of the experiments the fiber transmitted the collected light to a photomultiplier tube (PMT) through a blue-green color filter, which rejected the fundamental and transmitted the 350–600-nm region. The atmosphere in the region investigated was controlled with a hood that was able to generate a dust-free laminar air flow.

The experimental setup that we used to quantify the contribution of elastic Rayleigh-Mie scattering to the measured signal was identical, except for the input laser beam. Inasmuch as the linear processes had to be investigated in the same blue–green spectral region as the detected part of the supercontinuum, the output beam of the laser was frequency doubled in a KDP crystal. The second harmonic at 405 nm was separated from the fundamental by a dichroic mirror (~200 μJ after the dichroic mirror), which provided a low-intensity and linearly propagating blue beam. The divergence of this beam was 0.065° (half-angle).

The results obtained for linear Rayleigh–Mie scattering are shown in Fig. 1. The raw data are divided by the geometric overlap function $1/\sin(\theta)$. The use of the laminar flow provided dust-free air, giving access to pure Rayleigh scattering. The experimental data

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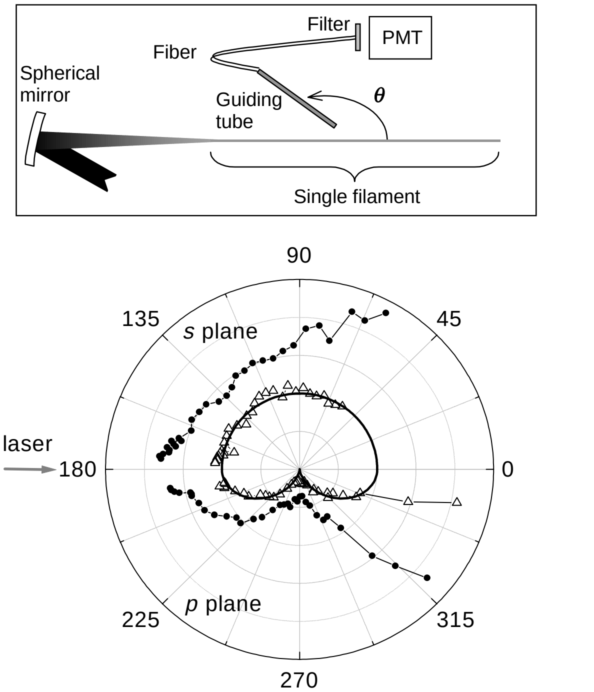

Fig. 1. Experimental setup and angular distributions of linear scattering from a low-energy blue beam. Filled circles, dusty air; open triangles, clean air. The clean-air data are fitted with a Rayleigh distribution (solid curves).

(open triangles) reproduce well the expected Rayleigh scattering angular pattern for both s-plane (displayed from 0° to 180°) and p-plane (from 180° to 360°) polarizations. The filled circles in Fig. 1 display the results for an atmosphere containing aerosol particles (laminar flow off). The angular distributions are thus representative of the combination of Rayleigh and Mie scattering.

The angular distributions of the supercontinuum emitted by a filament were then measured and compared with the linear data. With pulses of 50-GW peak power at 810 nm, a single filament started slightly before the geometrical focal point with conical emission, which had a divergence of 0.12° (half-angle) for its outer ring, in agreement with former observations.[5–7] The angular distributions obtained are shown in Fig. 2 for both clean and aerosol-containing atmospheres. In view of nonlinear lidar applications,[13,14] for which the ratio (α/β) is the relevant parameter[15] (here α is the overall extinction coefficient and β is the backscattering coefficient) we need to compare the shapes of the angular patterns, rather than the absolute intensities, of linear and nonlinear signals. Therefore we arbitrarily normalized the linear and the nonlinear signals to the same intensity at 90° in each polarization plane. A remarkable result is that the supercontinuum emission from the filament is greater (factor of 2) in the near-backward direction (176.5°) than Rayleigh–Mie scattering. The intensity rises steeply in the near-backward direction, which should lead us to extrapolate even stronger enhancements at 180°. These measurements demonstrate that self-generated enhancement of backward emission occurs within the filament. This discovery implies that the supercontinuum detected in the backward direction in a white-light lidar experiment not only is the result of linear Rayleigh–Mie backscattering but is also due to the nonlinear backscattering. The fact that a similar enhancement is observed in clean and in dust-containing air implies that aerosols do not contribute significantly to the nonlinear backscattering enhancement.

Compared with elastic scattering, the additional nonlinear scattering is thus of the same order of magnitude at 176.5°. We interpret this nonlinear backscattering enhancement as a consequence of laser-induced longitudinal refractive-index changes. These changes in the index can occur in a dynamic guided structure[8,16] in which the filament diameter, and hence the intensity, undergoes oscillations. The leading edge of a high-peak-power pulse produces long-period gratinglike index changes in the longitudinal direction (as a result of Kerr and plasma effects), which then backscatter white light generated from the trailing part of the pulse. The order of magnitude of the index changes in our experimental conditions is estimated to be 10^{-5} when the nonlinear refractive index of air is used[17] and for the measured plasma density.[18–20] A rough estimation from the Fresnel formula, and assuming a step-shaped refractive-index change, yields an upper limit for the self-reflection factor of 10^{-5}. This value is much larger than the Rayleigh backscattering efficiency of 2.5×10^{-7} in our experimental conditions in the blue–green band

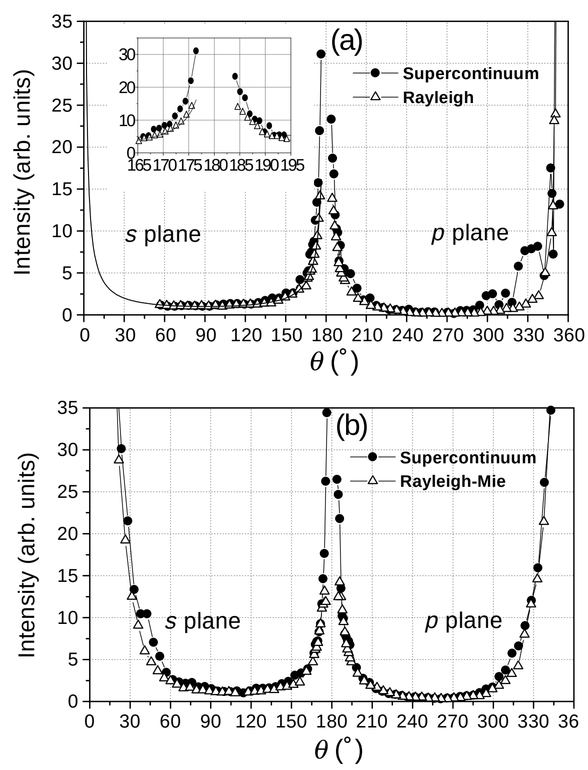

Fig. 2. Angular distributions of the supercontinuum emitted from a filament detected in the blue–green band compared with linear scattering: (a) clean air, (b) dusty air. In clean air and the s plane, the linear data are fitted and extrapolated by a Rayleigh distribution [$1/\sin(\theta)$]. Inset, zoomed-in view of the details from 165° to 195°.

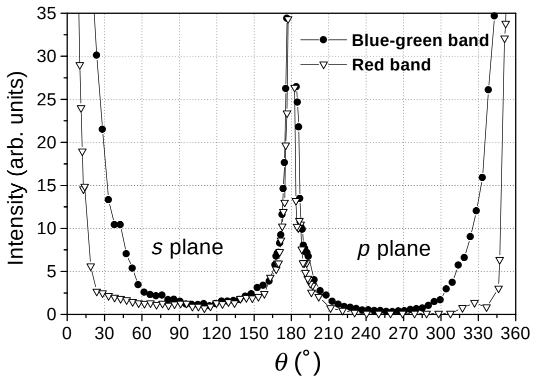

Fig. 3. Angular distributions of the supercontinuum scattered from a filament detected in the red band compared with those in the blue–green band.

[calculated with a 50-cm-long scattering volume, a 0.1-sr detection solid angle, and a Rayleigh backscattering coefficient $\beta = 4.9 \times 10^{-8}\ \mathrm{cm}^{-1}\ \mathrm{sr}^{-1}$ at 405 nm (Ref. 15)]. This estimated value is also 2 orders of magnitude higher than the experimentally observed value at 176.5°. This result should suggest a much larger nonlinear self-reflection coefficient at 180°. We remark, however, that the step-shaped index change is an assumption that is not verified in reality. The actual smooth change of index leads to a smaller reflection coefficient than for a step change.

The spectral dependence of the supercontinuum angular distribution was also investigated (in aerosol-rich air) with a bandpass filter from 600 to 650 nm. In Fig. 3 the results obtained are compared with the blue–green data. We can notice first that in the red band the forward peak is much narrower than in the blue–green region. This can be explained by the angular dispersion in the conical emission: The radius of color rings decreases with increasing wavelength.[5] In these plots, data were normalized in each polarization plane to yield identical signals in the near-backward direction for both spectral bands. This representation shows that, for the same near-backward signal, scattering about 90° decreases in the red band. This behavior is easily explained as being due to the decrease of the efficiency of both Rayleigh and Mie scattering when the wavelength increases. This observation implies that the importance of nonlinear self-reflection relative to linear Rayleigh–Mie scattering increases for longer wavelengths. This statement has important implications for lidar measurements in the mid and far infrared, where Rayleigh–Mie scattering decreases dramatically. We may expect that nonlinear self-reflection will then greatly dominate elastic backscattering and permit infrared lidar measurements at larger distances.

In conclusion, we have demonstrated that the supercontinuum emitted by a filament is enhanced in the backward direction because of longitudinal refractive-index changes induced by the laser pulse itself. These results have strong implications for lidar applications based on high-intensity femtosecond laser pulses.

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