Stimulated Raman *X* waves in ultrashort optical pulse filamentation

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We demonstrate that ultrashort pulse filamentation in liquids with strong Raman gain leads to the spontaneous formation of nonlinear *X* waves at a Raman-shifted wavelength. We measured as much as 75% energy conversion efficiency into a Raman *X* wave in ethanol starting from 1 ps pulses due to the group velocity matching between the pump and Raman *X* pulses. Large Raman gain of a weak seed signal was observed in water, associated with a strong spatiotemporal transformation of the seed into an *X* wave. © 2006 Optical Society of America

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Stimulated Raman scattering (SRS) is a well-known nonlinear process by which an intense laser field experiences frequency conversion to a redshifted wavelength that depends on the specific material. This process has been widely investigated and led to many applications ranging from the compression of 10–100 ps laser pulses to optical signal regeneration in telecommunication transmission systems.[1] In the past several years, femtosecond laser pulse filamentation has attracted interest due to the many potential applications such as lightning protection and remote sensing of the atmosphere,[2] single optical-cycle pulse generation,[3] waveguide writing,[4] and, more recently, as a tunable-wavelength ultrashort laser pulse source.[5] Ultrashort pulse filamentation has been recently interpreted in terms of the dynamical interaction of spontaneously generated *X* waves,[6,7] i.e., a particular class of stationary, nondiffractive and nondispersive conical waves.[8,9]

Femtosecond pulse durations involved in the filamentation dynamics (pulse splitting and temporal compression, shock front formation) would seem to exclude the possibility of observing efficient SRS processes, due to the negative role played by the group velocity mismatch (GVM) between the pump and Raman pulses. In this Letter, we show that, on the contrary, it is possible to achieve 75% conversion efficiency for the SRS wavelength from an ultrashort-pulse filament in ethanol. The result is obtained owing to the spontaneous formation of a Raman *X* pulse i.e., an *X* wave centered at the Raman shifted wavelength, whose group velocity matches exactly that of the pump. In a second experiment, performed in seeded configuration, we were able to lock the Raman *X* pulse either to the leading or to the trailing split pump pulse, by adjusting the pump-seed delay.

The idea of the following experiments stems from the observation of weak but clearly visible emission at the expected Stokes wavelength associated with filamentation in water.[7] However, Raman gain in water is rather weak so we carried out a first set of experiments in ethanol that has a SRS gain that is roughly 26 times larger.[10] Beam filamentation was induced by launching 1 ps pulses at 527 nm, delivered from a mode-locked, chirped-pulse regeneratively amplified Nd:glass laser with a 10 Hz repetition rate (Twinkle, Light Conversion, Ltd., Vilnius, Lithuania) and focused to a 100 μm diameter FWHM at the input facet of the sample. For input energies greater than $E_{in}=2\ \mu$J, filamentation accompanied by conical emission was observed. The filament far-field spectrum (θ,λ) was measured by collecting the output pulse with an $f=5$ cm achromat lens, placed at a distance f from the input slit of an imaging spectrometer.[11] The spectrum was then recorded either by high 16-bit dynamic range (Andor) or color (Canon) CCD cameras.

Figure 1(a) shows the measured (θ,λ) spectrum for an input energy of $E_{in}=2.5\ \mu$J. Around the pump wavelength, there is a clear *X* shape, indicating the transformation of the input Gaussian pulse into two *X* waves,[6,7,11,12] each of which is featured by a frequency gap separating two half-*X* branches. The half-*X* branch indicated with a solid (dashed) curve in Fig. 1(a) pertains to the leading (trailing) split pulse. The redshifted and blueshifted half-*X* branches in the near-infrared and ultraviolet region (not shown) characterize the separation in time of the split pulses.[7] Around the wavelength of 623 nm, i.e., the Raman-shifted wavelength in ethanol,[10] strong conical emission (CE) is observed. We measured the conversion efficiency from the pump to the Raman wave-

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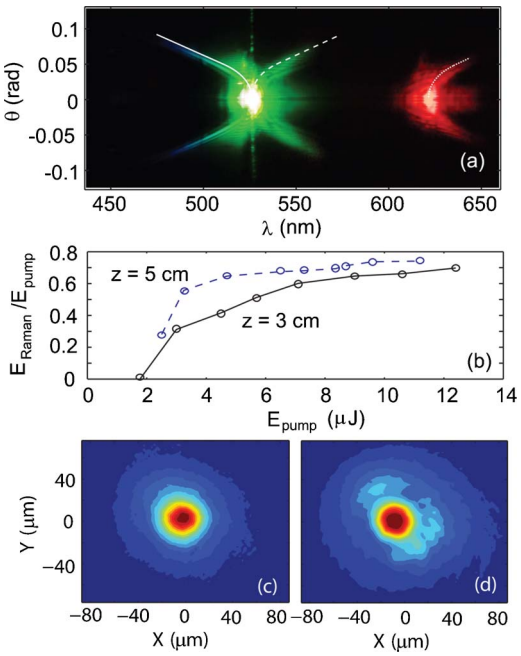

Fig. 1. (Color online) (a) Experimentally measured (θ,λ) spectrum of a filament in 5 cm of ethanol with $E_{III}=2.5\ \mu$J. The solid (dashed) curve indicates the leading (trailing) pump X waves, and the dotted curve indicates the Raman X-pulse spectrum. (b) Measured Raman gain as a function of the pump energy. (c) Measured near-field fluence profile of the Raman pulse and (d) of the filament.

length by spectrally filtering out the pump pulse. Figure 1(b) shows the results for 3 and 5 cm long cells. For the longer cell conversion, efficiencies of 75% are reached. It is worth noticing that a 75% conversion efficiency to the Raman frequency in ethanol has already been reported[13] albeit with pump pulse durations of 30 ps. As derived from the known Sellmeier relations,[14] the GVM between the Raman and the pump pulse in ethanol is 34 ps/m, which would lead to a separation length of 1.5 mm for a typical pulse duration of 50 fs expected in liquids after pulse splitting and temporal compression of the pump (see, e.g., Ref. 6). This indicates that a temporal locking between the pump and the Raman pulse has occurred in our setting. Finally Figs. 1(c) and 1(d) show the near-field fiuence profiles of the Raman pulse and of the filament, respectively, which indicate locking in the space domain as well.

To interpret the results, we first note that X waves may assume any arbitrary value for the group velocity while maintaining the property of stationarity.[7] On the other hand, high SRS conversion efficiency may be expected if GVM is reduced to zero, i.e., the pump and Stokes pulse have the same group velocity, a possibility offered in the case that these are indeed X waves. We therefore computed the group velocity relative to each portion of the spectrum by taking into account the angular dispersion as measured in Fig. 1(a). The longitudinal component of the group velocity v_g^z may be evaluated by finding the longitudinal component of the wave vector from $k_z=\sqrt{k^2-K_\perp}=\sqrt{k^2(1-\theta^2)}$ where $K_\perp$ is the transverse wave vector component and $k=\omega n(\omega)/c$, with $n(\omega)$ evaluated from Eq. (5) in Ref. 14. v_g^z at each frequency $\tilde{\omega}$ of the X wave is then found from $v_g^z=1/(\mathrm{d}k_z/\mathrm{d}\omega)|_{\tilde{\omega}}$. The result leads to two different but well-defined values $v_{g1}^z=2.185\pm0.002\times10^8$ m/s in the 470–527 nm spectral region [solid curve in Fig. 1(a)] and $v_{g2}^z=2.092\pm0.005\times10^8$ m/s in the 527–560 nm spectral region [dashed curve in Fig. 1(a)], thus confirming that the two X branches centered at 527 nm in Fig. 1(a) pertain to two different X waves that separate in time due to pulse splitting. Notably, we find $v_{g3}^z=2.090\pm0.005\times10^8$ m/s in the 620–650 nm spectral region where SRS radiation is observed (dotted curve). The virtually constant velocity in the fairly broad excited bandwidth indicates that a stationary X-type wave is formed in this region as well. Furthermore, v_{g3}^z is equal within experimental error to v_{g2}^z, which shows locking between the Raman X-wave and the trailing X-pump wave.

We deepened our investigation of this phenomenon by repeating the experiment with water that is well characterized from the point of view of filament and X-wave formation.[6,7,15] Since the Raman gain of water is much lower than that of ethanol,[10] large conversion efficiencies or spontaneous formation of Raman X waves were not observed. Therefore we seeded the filament-mediated SRS process by a weak signal at the Raman-shifted wavelength (637 nm in water), that is delivered by an optical parametric amplifier (Twinkle, Light Conversion Ltd., Vilnius, Lithuania) pumped by redundant laser energy. The seed had the 1 ps pulse duration of the pump pulse and was a collimated 1 mm FWHM beam with a fluence of 8 μJ/cm^2 that is much smaller than the pump fluence, 11 μJ[2]. A variable delay line allowed us to change the delay between the weak seed and the pump. Figures 2(a)–2(c) show the (θ,λ) spectra around the Raman wavelength for increasing delay, Δt. When the seed is in advance [Fig. 2(a), $\Delta t=-200$ fs] the Raman X-wave that emerges from the seed amplification is featured by CE with tails in opposite direction with respect to Fig. 1(a). On the other hand, for positive delays [Fig. 2(b), $\Delta t=+200$ fs], angular dispersion similar to that in Fig. 1(a) is observed. By adopting the same procedure as above for

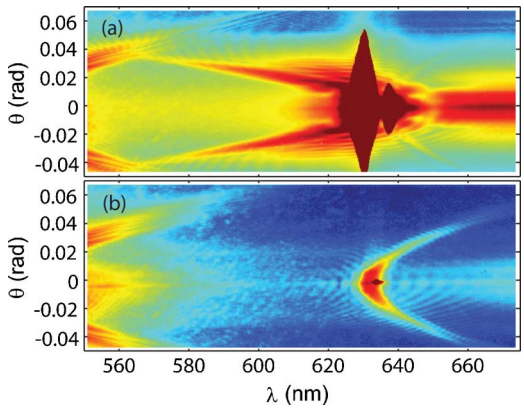

Fig. 2. (Color online) Experimentally measured (θ,λ) spectrum of a seeded filament in 3 cm of water with relative delays of (a) $\Delta t=-200$ fs and (b) $\Delta t=+200$ fs.

computing v_g^z around the pump (data not shown) and the Raman wavelength [Sellmeier relation Eq. (12) in Ref. 16] we obtained: $v_{gp1}^z = 2.2459 \pm 0.006 \times 10^8$ m s for the pump leading X wave, $v_{gp2}^z = 2.186 \pm 0.005 \times 10^8$ m s for the pump trailing X wave, $v_{gr3}^z = 2.246 \pm 0.003 \times 10^8$ m s for the Raman X wave observed with $\Delta t = -200$ fs, and, finally, $v_{gr4}^z = 2.187 \pm 0.005 \times 10^8$ m s for the Raman X wave observed with $\Delta t = +200$ fs. These results prove that the Gaussian seed transforms into an X-wave during the SRS amplification, whose group velocity exactly matches that of the leading or the trailing split pump pulse, depending on the relative delay.

Finally, we measured a SRS gain (defined as the ratio of the peak fluence of the output beam in the 620–660 nm spectral region in the presence and in the absence of the pump pulse) as large as 1000, which corresponds to a 10% conversion efficiency from the pump to the Raman wavelength. The bandwidth of the amplification process was probed by tuning the seed wavelength and was found to be $\sim 200\ \mathrm{cm}^{-1}$, which corresponds to the Raman line width in water.[10]

In conclusion, we have observed the formation of Raman X waves associated with optical pulse filamentation. Both seeded and spontaneous (starting from quantum noise) SRS induce the formation of X waves with group velocity that is matched to the pump pulse within the filament. This result may be compared with the formation of Raman solitons in one-dimensional systems such as optical fibers.[17] In analogy to this case and thanks to the group-velocity-matched interaction, we expect the Raman X wave to have a peak duration of the order of the ~ 50 fs pump X wave with the advantage that the interacting wavelengths may both remain in the normal dispersion regime. We note that the spectral characterization technique adopted here does not reveal if the pulse is Fourier limited, so further measurements in this sense are under way.

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