# Supercontinuum generation in an optical fiber capillary filled with Toluene

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Liquid-core optical fibers (LCOFs) have recently been identified as extremely attractive waveguides for applications to frequency conversion and supercontinuum (SC) generation [1-4]. Their advantages over silica optical fibers include broad visible to mid-infrared transparency and enhanced material nonlinearities by physical processes specific to anisotropic liquid molecules [3]. As a result, there has been extensive interest in the use of nonlinear liquids, such as CS_2 or ethanol, for applications in visible and mid-infrared SC generation [2-4].

Here we demonstrate, for the first time to our knowledge, efficient visible supercontinuum generation in an optical fiber capillary filled with Toluene pumped by a picosecond pulsed laser at 532 nm. Although operating in the strong normal dispersion regime, we show in this study that SC is generated through the combined effects of multiple cascaded Raman scattering and stimulated Raman-Kerr scattering (SRKS). The latter enables a large spectral broadening of the Raman lines owing to the strongly anisotropic orientational relaxation of liquid molecules [5]. This is shown experimentally in Fig 1(a) that shows a set of output spectra recorded by increasing the input pump pulse energy from 3 nJ to 200 nJ. As can be seen, flat and broad SC generation from 550 nm to 870 nm is achieved for the highest pump energy (top spectrum). In this experiment, we used a 2-m long 9µm-core-diameter Toluene-filled LCOF pumped by a Nd:YAG 18-ps pulsed laser at 532 nm. The SC dynamics can be examined from the spectra at lower energies. We first observe two cascaded Raman vibrational lines shifted by 30 THz and 60 THz, respectively (blue vertical arrows in Fig. 1(a)), coming from the aromatic ring molecular breathing mode. Increasing further the energy gives rise to an additional far-detuned Raman peak shifted by 92 THz from the pump, (horizontal red arrow in Fig. 1(a)), from the C-H symmetric stretching vibrational mode of Toluene. Once generated, this narrow-band Raman peak plays the role of a secondary pump that gives rise to further red-shifted Raman peaks indicated by blue and red arrows in Fig. 1(a). The strong spectral broadening of all Raman lines arises from SRKS [5]. To confirm our physical interpretation, we numerically solved the nonlinear Schrödinger equation including the full Raman spectrum of Toluene and the group-velocity dispersion (GVD). Figure 1 shows a numerical simulation of the cascaded Raman scattering in the LCOF both in the spectral (b) and time (c) domains. The agreement with the experimental spectrum at 17 nJ is excellent. We observe that the picosecond pump pulse at 532 nm generates simultaneously the two Raman vibrational modes shifted by 30 THz and 92 THz from the pump, respectively. In the time domain, pulse splitting is observed due to the large normal GVD between the pump at 532 nm and the Raman line around 640 nm (120 ps.m^{-1}). The combined effects of all these processes together with SRKS lead to SC generation from the pump up to the absorption band of Toluene near 870 nm, as shown for the highest input energy spectrum in Fig. 1(a).
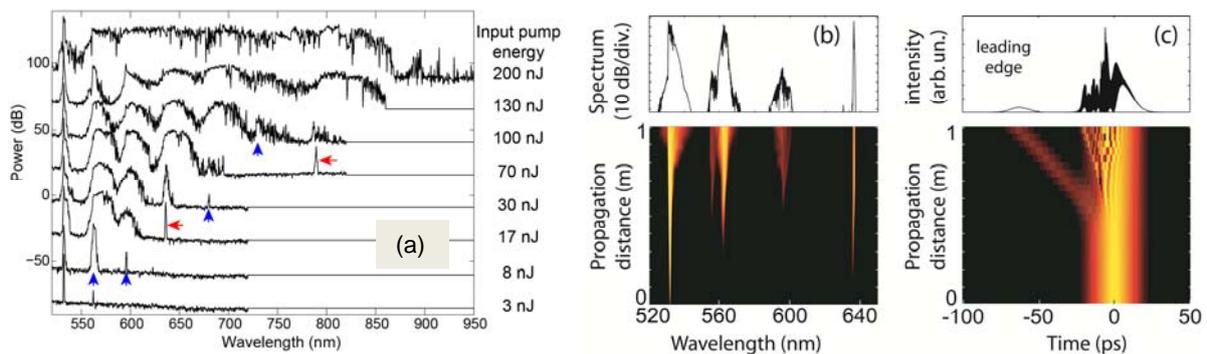
Figure 1 (a) Output experimental spectra with increasing input pump energy. Spectra are separated by 25 dB for better clarity. Resolution of OSA is 0.5 nm, except the highest energy with 2 nm. See text for explanation of blue and red arrows. (b) and (c) Numerical simulations of the generalized nonlinear Schrödinger equation including the Raman spectrum in Toluene and the dispersion in both the spectral and time domains.

References

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