Shaping of broadband Near-Infrared and Mid-Infrared pulses for Mid-Infrared-, CARS-, and Sum-Frequency spectroscopy in a nonlinear microscope

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Nonlinear microscopy have seen a tremendous development during the last decades. In this work, we demonstrate two applications of a versatile, broadly tunable mid-infrared light source in a shaper based single beam setup for multimodal microscopy: (i) the combination of coherent anti-Stokes Raman scattering (CARS) with mid-infrared (MIR) microscopy [1] and (ii) the application of sum frequency (SF) spectroscopy. Spectral Focusing [2] with flexible pulse shaping allows high spectral resolution in MIR and CARS spectroscopy by generating narrowband excitation in both difference frequency based processes. The imprinted phases generate broadly tunable (>2000 cm^{-1}) and narrowband (<20 cm^{-1}) MIR light in the setup (Fig.1(a)) consisting of a Ti:Sa oscillator, providing sub 10 fs pulses, a 4f liquid crystal pulse shaper setup and a thin $LiIO_3$ crystal.

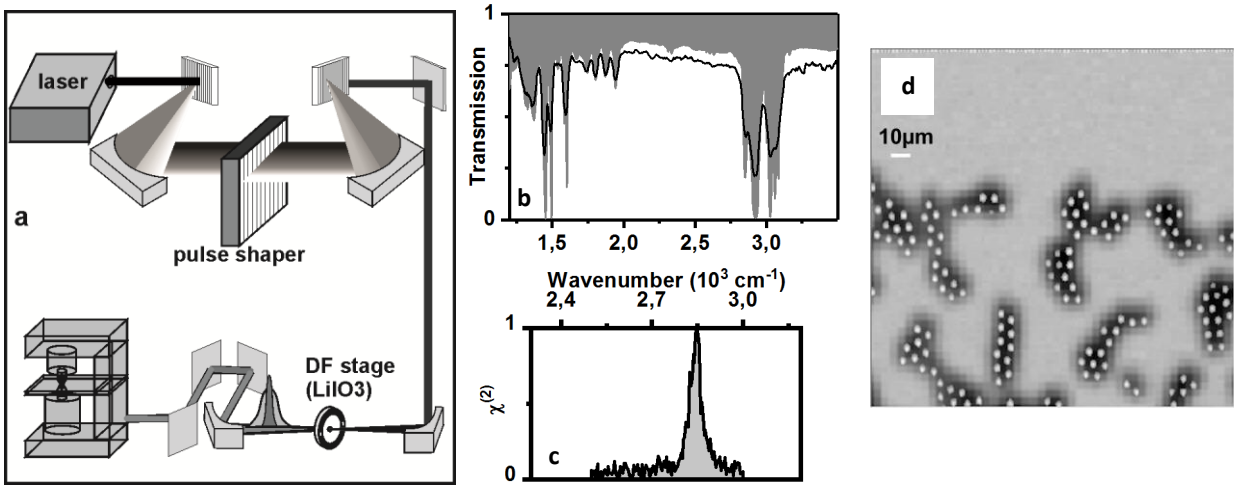

Fig. 1: (a) Single beam setup consisting of laser, 4f-shaper setup, DFG stage and microscope for MIR and CARS spectroscopy. (b) MIR transmission of polystyrene (black) and its reference (grey background). (c) Sum frequency signal of cholesterol. (d) Image of polystyrene beads by MIR transmission at 3060 cm^{-1} (grey scales) and CARS light (bright spots).

The successful implementation of combined CARS and MIR microscopy in the same setup is demonstrated on two applications: firstly, on human skin tissue (not shown) and, secondly, on polymers (Fig. 1) by measuring the MIR transmission of a polystyrene film (Fig.1(b)) and by imaging polystyrene beads with CARS and MIR microscopy (Fig.1(d)) [1]. Moreover, the use of narrowband MIR and broadband NIR pulses yield applications in SF spectroscopy, which were verified by recording the susceptibility ($\chi^{(2)}$) of cholesterol (Fig. 1(c)).

[1] N.Müller, L.Brückner, and M.Motzkus, "Invited Article: Coherent Raman and mid-IR microscopy using shaped pulses in a single-beam setup", *APL Photonics,* **3**, 092406 (2018)
[2] G. Veitas and R. Danielius. "Generation of narrow-bandwidth tunable ps pulses by DFM of stretched pulses". *JOSA B*, **16**, 1661 (1999).