Fiber delivered two-color picosecond source through nonlinear spectral transformation for coherent Raman scattering imaging

Ke Wang^{a)} and Chris Xu

School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853, USA

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We demonstrate a two-color, fiber-delivered picosecond source for coherent Raman scattering (CRS) imaging through nonlinear spectral transformation. The wavelength tunable picosecond pump is generated by nonlinear spectral compression of a prechirped femtosecond pulse in a fiber wavelength division multiplexer (WDM). The 1064-nm synchronized picosecond Stokes pulse is generated through pulse carving of a continuous wave laser, nonlinear spectral broadening in 100-m standard single-mode fiber, and subsequent dispersive compression with a fiber compressor. The pump and Stokes beams are combined and delivered by the fiber WDM. CRS imaging of mouse skin is performed to demonstrate the practicality of this source. © 2012 American Institute of Physics. [doi:10.1063/1.3686149]

Coherent Raman scattering (CRS) microscopy, with contrast from coherent anti-Stokes Raman scattering (CARS)¹ or stimulated Raman scattering (SRS),² allows label-free imaging of biological samples with endogenous image contrast based on vibrational spectroscopy. An important technical challenge in CRS microscopy is the requirement of two synchronized picosecond excitation sources. Current excitation sources for CRS (Refs. 2–11) suffer from limitations¹² such as bulky layout, high cost, limited spectral resolution, or low power. Recently, we have demonstrated a scheme for two-color, synchronized picosecond light sources for CRS imaging based on the time-lens concept.^{12,13} This technique benefits from robust all-fiber configuration and radio-frequency (RF) electronic tuning of pulse delay for the temporal overlap between the two pulse trains. Through nonlinear spectral compression of a prechirped femtosecond pulse, we further demonstrated a fiber delivered two-color picosecond source derived from a femtosecond pump instead of a picosecond pump,¹² thus expanding the options of sources for CRS imaging since femtosecond titanium:sapphire (Ti:S) lasers are much more common in research labs. However, the time-lens source still necessitates phase modulation with phase modulators as well as proper RF filter and amplifiers for spectral broadening,^{12,13} which adds complexity and cost to the whole system.

In this paper, we demonstrate a fiber delivered picosecond source through nonlinear spectral transformation. Specifically, the 2.7-ps pump pulse is generated through nonlinear spectral compression of a prechirped femtosecond pulse in a fiber wavelength division multiplexer (WDM) as mentioned in Ref. 12. The 1064-nm Stokes pulse is first generated through pulse carving from a continuous wave (CW) laser with a Mach-Zehnder (MZ) intensity modulator, which yields a 55-ps pulse train synchronized to the pump. Then after self-phase modulation (SPM) induced spectral broadening in 100-m standard single mode fiber (SSMF), and subsequent temporal compression with an all-fiber compressor, the Stokes

pulse is compressed to 1.9 ps. This nonlinear scheme of spectral broadening replaces the phase modulators in the time-lens source, potentially reducing the complexity and cost of the source. The pump and the Stokes pulses are combined and delivered in a fiber WDM, which also serves as the nonlinear medium for spectral compression of the femtosecond pump pulse. The resultant fiber delivered two-color source retains the advantage of automatic synchronization of the time-lens source^{12,13} but requires no phase modulators. We further demonstrate its application to CARS imaging of *ex vivo* mouse ear.

In our experiment (Fig. 1), the 80-MHz, 90-fs pump pulses centered at 817-nm are generated from a mode-locked Ti:S laser (Tsunami, Spectra-Physics). The prechirp of the femtosecond pulse is provided by a rotating cylindrical lens system in a 4f configuration,¹⁴ which provides tunable anomalous dispersion up to $-1.4 \times 10^5 \text{ fs}^2$ (described in details in Ref. 14). The prechirped 817-nm pulse is coupled into a WDM for nonlinear spectral compression, fiber-optic beam combining, and delivery. To generate the 1064-nm Stokes pulse, a fast GaAs photodetector (ET-4000, 10 GHz, EOT)

FIG. 1. The experimental setup of the fiber-delivered two-color picosecond source for CARS imaging. M: mirror, G: 1800 line/mm grating, L1: $f = 30 \text{ cm}$ collimating lens, CL: $f = 10 \text{ m}$ concave cylindrical lens, BS: beam splitter, HWP: half-wave plate, QWP: quarter-wave plate, L2: aspheric lens, CFBG: chirped fiber Bragg grating, L3: collimating lens, LPF: 800 nm long pass filter, PC: polarization controller, BB: broadband, MZ: Mach-Zehnder intensity modulator. Electrical paths are labeled with dashed lines, and optical paths are labeled with solid lines.

^{a)} Author to whom correspondence should be addressed. Electronic mail: kw376@cornell.edu.

FIG. 2. (a) Measured 817-nm pump spectra from the WDM at linear propagation (dashed line) and maximum spectral compression (solid line). (b) Cross correlation trace between the 90-fs and the spectrally compressed 817 nm pulses.

converts the 80-MHz pump pulse train into a synchronized RF pulse train, which is amplified by broadband RF amplifiers and used to drive the MZ intensity modulator. The intensity modulator carves a synchronized 80-MHz, 55-ps optical pulse train from a 1064-nm CW laser (QFBGLD-1060-30PM, QPhotonics). The preamp and the power amplifier boost the energy of the optical pulse for nonlinear spectral broadening in 100-m SSMF, which replaces the phase modulators in the time-lens setup.^{12,13} For dispersion compensation, the spectrally broadened 1064-nm pulse is compressed with an all-fiber dispersion compressor consisting of a circulator and a chirped fiber Bragg grating. The output port of the compressor is spliced to the fiber WDM. In our setup, the fiber WDM serves three purposes: combining the pump and the Stokes beams spatially, nonlinear spectral compression of the pump, and fiber delivery of both beams. An in-line fiber polarization controller before the WDM adjusts the polarization of the 1064-nm output, ensuring that the Stokes beam is linearly polarized and parallel to that of the pump to maximize the CARS signal. An electronic discrete RF delay (PDL-10A, Colby Instruments) and a continuously tunable RF delay provide a large tuning range (0.795 ns) to adjust the relative time delay between the pump and the Stokes pulses without any mechanical optical delay line. After collimation and an 800-nm long-pass filter (LPF), which removes the four-wave mixing background generated in the fiber coupler,¹⁵ the spatially and temporally overlapping pump and Stokes pulses are sent into a microscope (described in detail in Ref. 16) for CARS imaging. CARS signal is excited and collected in the epi-direction with a water immersion objective lens (XLUMPlanFI, Olympus, NA = 0.95) and detected by a GaAsP photomultiplier tube (PMT).

We first characterize the 817-nm pump pulse. Spectral compression of a negatively prechirped femtosecond pulse

due to SPM in optical fibers is explained in detail in Ref. 12. At maximum group delay dispersion ($-1.4 \times 10^5 \text{ fs}^2$) and an output power of 145 mW from the WDM, the full-width-at-half-maximum (FWHM) bandwidth of the pump spectrum is compressed to its minimum, from 7.4 nm to 0.72 nm [Fig. 2(a)] with a spectral compression ratio of 10.3. To obtain the temporal profile of the spectrally compressed pump pulse, we measured cross correlation trace between the 90-fs 817-nm pulse and the spectrally compressed pump pulse, through background-free noncollinear sum-frequency (SF) generation in a barium borate (BBO) crystal, shown in Fig. 2(b). The measured FWHM pulse width is 2.7 ps.

We then characterize the 1064-nm Stokes pulse and the synchronization performance. As power increases, the 1064-nm spectrum continuously broadens due to SPM [Fig. 3(a)]. At the maximum output power of 186 mW from the WDM (820 mW output from the 100-m SSMF), the measured bandwidth [defined by the two outmost peaks indicated by the arrows Fig. 3(a)] is 1.14 nm. To determine the temporal profile, we measured the cross correlation between the 1064-nm Stokes pulse and the 90-fs 817-nm pulse directly from the Ti:S laser [Fig. 3(b)]. To characterize the timing jitter, cross correlation of the Stokes pulse and the 2.7-ps spectrally compressed 817-nm pump pulse is performed [Fig. 3(c)]. All cross correlation measurements use SF generation in the BBO crystal. A bandpass filter placed before the photodiode removes the residual second harmonic of the 817-nm pulse and the 1064-nm Stokes pulse, which guarantees background-free SF measurement in the collinear configuration. The relative delay between the pulses is readily scanned by tuning the electronic RF delay line, instead of a mechanical delay line used for Fig. 2(b). Figure 3(b) shows that the FWHM pulse width of the compressed 1064-nm pulse is 1.9 ps. The FWHM of the cross correlation between the 1064-nm pulse

FIG. 3. (a) The measured 1064-nm pulse spectra at various output power from the WDM. Cross correlation trace between the 186-mW 1064-nm pulse with the 90-fs 817-nm pulse from the Ti:S laser (b) and the spectrally compressed 2.7-ps 817-nm pump pulse (c). Inset in (c) shows the measured sum-frequency signal at the half maximum of the cross-correlation trace over 160 s.

FIG. 4. CARS image of sebaceous glands at the CH_2 stretching frequency in mouse ear. 512×512 pixels, 4 s/frame, no average.

and the 2.7-ps spectrally compressed 817-nm pulse is 3.3 ps. To measure the relative timing jitter between the pump and Stokes pulses, we record the SF intensity fluctuation at half maximum of the cross correlation trace [inset in Fig. 3(c)].^{3,13} The RMS timing jitter over a measurement time of 160 s is 180 fs, only a small fraction of the pulse widths.

With this fiber-delivered two-color source, we perform CARS imaging of sebaceous glands at the CH_2 stretching frequency (2845 cm^{-1}) in *ex vivo* mouse ear. Figure 4 highlights the practicality of the two-color, fiber-delivered picosecond source for CARS imaging. While CRS imaging of the CH_2 bond is demonstrated, the wavelength tunability of the femtosecond Ti:S laser, together with proper adjustment of the grating angle and choice of fast photodetectors responsive to the pump wavelength, ensures that a wide range of stretching bands can be covered by the two-color source. For example, tuning the pump wavelength to 909 nm, we have previously demonstrated CRS imaging of drug delivery in mouse skin at the polyene stretching frequency (1600 cm^{-1}).¹³

Compared with the time-lens source we demonstrated before,^{12,13} Stokes pulse generation through nonlinear spectral broadening is simpler and more cost effective to implement. High speed phase modulators and relevant RF filter and amplifiers are no longer needed, eliminating the major cost of the time-lens system. On the other hand, the timing jitter measured in this paper is larger than that of the time-lens source, and there is some polarization instability over a long imaging session of several hours. We attribute these to the long SSMF (100 m) we used. A long fiber is more susceptible to environmental variations such as temperature fluctuations and mechanical vibrations. For future improvement, a higher speed photodetector and RF amplifiers for pulse carving can be used to shorten the optical pulses before nonlinear spectral broadening. A shorter input pulse would

require shorter length of SSMF and thus reduce timing jitter. For example, our numerical simulations show that if the initial pulse width after pulse carving is 27.5 ps (instead of the 55-ps pulse width used in this paper), only 24-m SSMF is needed to obtain the same spectral bandwidth at the same input average power. We also note that polarization maintaining fiber can be used to eliminate the polarization instability.

In conclusion, we have demonstrated a fiber-delivered two-color, picosecond source for CARS imaging through nonlinear spectral transformation. The pump pulse is generated through nonlinear spectral compression of a prechirped femtosecond pulse, while the Stokes pulse is generated through SPM-induced spectral broadening of a 55-ps pulse and subsequent dispersive compression. Spectral compression of the femtosecond pump pulse, beam combining, and fiber delivery are readily accomplished in a fiber WDM. The temporal overlap of the two pulses is electronically adjusted without any mechanical optical delay line, greatly facilitating the temporal alignment of the excitation beams for CRS imaging. Mouse skin imaging at CH_2 stretching frequency is performed to demonstrate the practicality of this source. The nonlinear spectral broadening scheme of the Stokes pulse replaces phase modulation with phase modulators in the time-lens source, which potentially reduces the complexity and cost of the Stokes source for CRS imaging.

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