Coherent anti-Stokes Raman scattering microscopy with near-infrared ps pulses synchronized within 50 fs

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Abstract: By employing a new method of stabilizing the relative timing jitter between two, tunable ps lasers to <50 fs we significantly improve the vibrational imaging capability of coherent anti-Stokes Raman scattering microscopy.

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Confocal microscopy

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Coherent anti-Stokes Raman Scattering (CARS) microscopy [1] has enjoyed a resurgence following a series of recent improvements such as co-linear excitation geometry [2], and epi-direction detection [3]. An additional refinement has been the use of synchronized ps Ti:Sapphire lasers [4, 5]. Due to their wide tunability, ability of access the fingerprint region (1000 to 2000 cm^{-1}) and near-infrared center wavelengths, these lasers are almost the ideal light source for CARS imaging. However, the relative timing jitter between the two lasers has remained a fundamental limitation. As CARS is inherently a multi-photon (four-wave mixing) microscopy technique, the presence of timing jitter is directly mapped to fluctuations in the CARS signal itself. In this work, we utilize a new locking method to reduce the timing jitter to below 50 fs. At this level of jitter, stability of the CARS signal is nearly shot-noise limited.

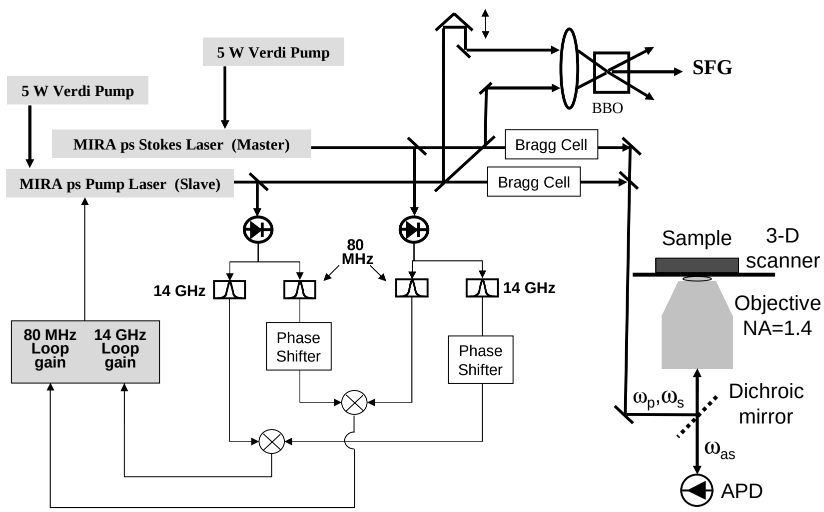

Fig. 1. Schematic of CARS experimental setup. In our case the pump laser is slaved to the Stokes using high harmonic locking technique. After emerging from individual pulse pickers running at 250 kHz, the beam are combined and co-linearly launched into the CARS microscope. Detection of the anti-Stokes signal is in the backward (epi-) direction via an avalanche photo-diode (APD).

The experimental setup is shown in Fig. 1. Two picosecond lasers tunable from 700-1000 nm (Coherent Miras individually pumped by two 5 W Coherent Verdis) are employed to generate the pump and Stokes signals. To tightly synchronize the two pulse trains we slave the pump laser to the Stokes laser using a recently reported high harmonic locking technique [6]. Briefly, the fundamental repetition rate (80 MHz) and the 175th harmonic (14 GHz) are simultaneously detected from each laser. The pump laser is first synchronized to the Stokes laser using only the error signal generated from the 80-MHz loop. Next, after temporally overlapping the two pulses trains by adjusting the 80-MHz phase shifter, the feedback loop is gradually switched over to the 14-GHz loop (via an analog switch). By switching to the 175th harmonic, the sensitivity of the feedback loop to any relative timing jitter is greatly enhanced, thereby enabling an extremely tight lock between the two pulse trains. Using this stabilization scheme, the amount of timing jitter can be accurately controlled by changing the amount of 100 MHz and 14 GHz loop gains. A portion of each laser is tapped off to monitor the jitter via sum frequency generation (SFG). To measure the jitter, the pulses are temporally offset in the cross-correlator by one-half of a pulsewidth. Then, amplitude fluctuations recorded in the SFG signal can be converted to timing fluctuations via the width and shape of the cross-correlation pulse. With the pump (Stokes) laser tuned to 753 (857) nm, an autocorrelation measurement yielded 3.4 (4.1) ps, while a cross-correlation width of 3.8 ps was measured.

To evaluate the effect of jitter on the CARS signal and imaging capability, the 1600 cm^{-1} transition in polystyrene beads was investigated. Figure 2(a) displays both the SFG signal (when the pulses are offset as described above) and the CARS signal at two different jitter levels. From the SFG fluctuations (the lower two traces), we measure the RMS jitter in a 160 Hz bandwidth to be 32 fs and 770 fs over a 10 second record length. The upper two traces show the CARS signal, recorded with a 1 ms bin-width, at these two jitter levels, clearly showing the enormous improvement with reduced jitter. In fact, at the lower jitter level, fluctuations of the CARS signal become shot-noise limited as shown in Fig. 2(b) where a linear relationship exists between the signal-to-noise ratio of the CARS signal and the square-root of the CARS intensity.

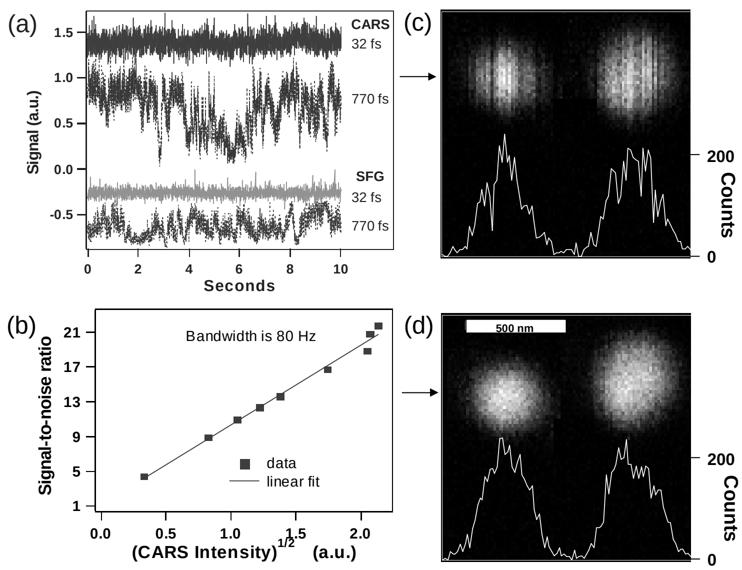

Fig. 2. (a), CARS and SFG signals recorded at two different jitter levels. (b), Plot showing nearly shot-noise limited stability (signal-to-noise ratio $\propto \sqrt{I}$) of the CARS signal. (c) and (d), Images of two polystyrene beads taken with RMS jitter levels of 770 and 32 fs. A horizontal slide taken at the arrow for each image is also shown

At low jitter levels, the enhanced stability of CARS signal significantly improves the imaging capability of CARS. Figure 2(c) and (d) shows images of polystyrene beads at jitter levels of 770 and 32 fs, respectively. Also shown is a horizontal slice of the image, taken at the coordinate indicated by the arrow. Clearly, there is a significant improvement in the image quality when the jitter level is reduced below 50 fs, which is sufficiently stable to reveal fundamental shot noise.

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