

## サニャック干渉計を用いた時間領域ラマン分光法

### Time-Domain Raman Spectroscopy using a Sagnac Interferometer

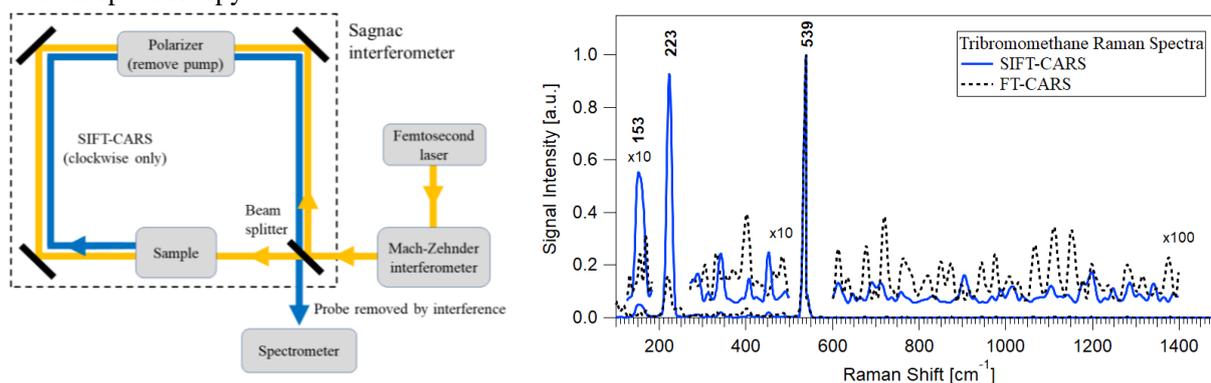
東京大学<sup>1</sup>, JST<sup>2</sup>, ○ (M2) ピーターソン ヨン ウォーカー<sup>1</sup>, 平松 光太郎<sup>1</sup>, 合田 圭介<sup>1,2</sup>

The University of Tokyo<sup>1</sup>, JST.<sup>2</sup>, ○Jorgen Walker Peterson<sup>1</sup>, Kotaro Hiramatsu<sup>1</sup>, Keisuke Goda<sup>1,2</sup>

E-mail: hiramatsu@chem.s.u-tokyo.ac.jp

Time-domain Raman spectroscopy, by virtue of its chemical specificity and label-free nature, has many applications in biological, medicinal, and material sciences<sup>1</sup>. One phenomenon in this domain is Fourier-transform coherent anti-Stokes Raman scattering (FT-CARS), a third-order nonlinear process where Raman-active vibrational modes are coherently excited with a pump pulse and read out by a subsequent probe pulse<sup>2</sup>. It is a powerful technique due to its wide Raman spectral range, high-speed Raman signal acquisition, and high signal intensity. However, FT-CARS is limited by its reliance on optical filters for isolating the frequency-shifted CARS signal from the strong background of the unscattered probe light. Molecular vibrational signatures at wavelengths outside the filter cutoff are lost. As a result, typical Raman spectra acquired with FT-CARS are attenuated or altogether vacant in the low-frequency Raman region (typically below  $\sim 200$   $\text{cm}^{-1}$ )<sup>3</sup>.

In order to push the capability of FT-CARS beyond these limitations, we demonstrate Sagnac interferometry FT-CARS (SIFT-CARS), a technique for time-domain Raman spectroscopy that does not rely on optical filtering (Figure 1a). The technique makes use of a common-path Sagnac interferometer as a means of background reduction. Counter-propagating probe beams in the Sagnac interferometer recombine at its output and destructively interfere, while the CARS signal generated propagates only in the clockwise direction and escapes the interferometer, isolated from the probe background. Comparing Raman spectra of tribromomethane acquired using both SIFT-CARS and FT-CARS, we demonstrated improvements of the signal-to-noise ratios of the 223  $\text{cm}^{-1}$  and 539  $\text{cm}^{-1}$  vibrational modes by factors of 21.8 and 3.7, respectively (Figure 1b). Furthermore, the 153  $\text{cm}^{-1}$  mode peak is observable with SIFT-CARS, but not with FT-CARS<sup>4</sup>. This technique is a step towards more sensitive and broadly applicable time-domain Raman spectroscopy.



**Figure 1: SIFT-CARS.** **a)** Schematic of SIFT-CARS. Pump-probe interaction generates SIFT-CARS signal (blue) in Sagnac interferometer only in clockwise direction. **b)** Normalized Raman spectra of tribromomethane acquired with FT-CARS (dotted black) and SIFT-CARS (blue). Peaks at 153, 223, and 539  $\text{cm}^{-1}$  apparent with SIFT-CARS. 10x and 100x zoomed-in regions show higher noise level of FT-CARS compared to SIFT-CARS.

<sup>1</sup>C. Chung and E.O. Potma, *Annu. Rev. Phys. Chem.* **64**, 77 (2013) <sup>2</sup>Jennifer P. Ogilvie et al., *Opt. Lett.* **31**, 480 (2006) <sup>3</sup>C. H. Camp Jr et al., *Nat. Photonics* **8**, 627 (2014) <sup>4</sup>Shimanouchi, *Tables of Mol. Vib. Freq. Consolidated Vol. I*, National Bureau of Standards, 1 (1972)