

サニャック干渉計を用いた高速低波数振動分光法

High-speed low-frequency vibrational spectroscopy using a Sagnac interferometer

東京大学¹, JST², ○(D)ピーターソン ヨン ウォーカー¹, リンドリー マット¹, ガラデパブロー
ジュリア¹, 平松 光太郎¹, 合田 圭介^{1,2}

The University of Tokyo¹, JST.², ○Jorgen Walker Peterson¹, Matt Lindley¹, Julia Gala de Pablo¹,
Kotaro Hiramatsu¹, Keisuke Goda^{1,2}

E-mail: peterson@g.ecc.u-tokyo.ac.jp

High-speed vibrational spectroscopy, by virtue of its chemical specificity and label-free nature, as well as its suitability to imaging or capturing short-lived phenomena, has many applications in biological, medicinal, and material sciences¹. One powerful technique in this domain is Fourier-transform coherent anti-Stokes Raman scattering (FT-CARS) spectroscopy, based on a third-order nonlinear process where Raman-active vibrational modes are coherently excited and read out by pump and probe pulses, respectively². It is a promising technique due to its broadband Raman spectral range, rapid spectral acquisition rate, and its relatively simple optical setup. However, rapid-scan FT-CARS is limited by inherent poor sensitivity in the information-rich low-frequency Raman region (typically below $\sim 200\text{ cm}^{-1}$)³. This weakness is a result of 2 factors: (1) FT-CARS detection typically occurs at probe frequencies far from the probe center, where low-frequency Raman signals are strongest; and (2) optical shortpass filtering schemata for FT-CARS detection disproportionately attenuate low-frequency Raman modes.

In order to push the capability of high-speed vibrational spectroscopy beyond these limitations, we demonstrate rapid-scan Sagnac-enhanced impulsive stimulated Raman scattering (SE-ISRS) spectroscopy, a technique for high-speed vibrational spectroscopy based on a common-path Sagnac interferometer (Fig. 1a). The Sagnac interferometer provides two benefits: (1) probe background reduction by destructive interference and (2) low-frequency vibration sensitivity enhancement by detection of molecular vibrations at the probe center frequency^{4,5}. Comparing Raman power spectra of bromoform acquired at 24 kHz using both SE-ISRS and FT-CARS spectroscopies, we demonstrate improvements of the signal-to-noise ratios of the 157 cm^{-1} , 222 cm^{-1} , and 539 cm^{-1} vibrational modes by factors of 7.6, 2.5, and 4.3, respectively (Fig. 1b)⁶. This technique is a step towards more sensitive and versatile low-frequency vibrational spectroscopy.

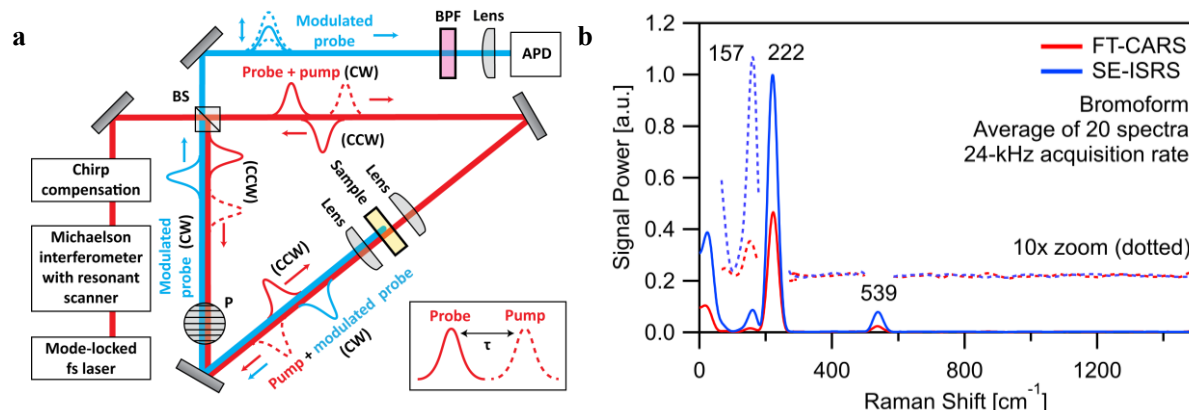


Figure 1: Rapid-scan SE-ISRS. **a)** Schematic of rapid-scan SE-ISRS. SE-ISRS signal (blue) generated in Sagnac interferometer in clockwise (CW) direction. CCW, counter-clockwise; BS, beamsplitter; P, polarizer; BPF, bandpass filter; APD, avalanche photodiode. **b)** Raman spectra of tribromomethane acquired at 24 kHz with FT-CARS (red) and SE-ISRS (blue), average of 20 spectra. 10x zoomed and +0.2 offset regions (dotted line) showing background and lowest-frequency bromoform mode.

¹C. Chung et al., *Annu. Rev. Phys. Chem.* **64**, 77 (2013) ²Jennifer P. Ogilvie et al., *Opt. Lett.* **31**, 480 (2006) ³K. Hashimoto et al., *Sci. Rep.* **6** (2016) ⁴J. K. Wahlstrand et al., *Opt. Lett.* **30**, 926 (2005) ⁵W. Peterson et al., *Opt. Lett.* **44**, 5282 (2019) ⁶Shimanouchi, *Tables of Mol. Vib. Freq. Consolidated Vol. I*, National Bureau of Standards, 1 (1972)