# Quantum-enhanced stimulated Raman scattering spectroscopy

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# 1. Introduction

Stimulated Raman scattering (SRS) spectroscopy is a powerful tool of vibrational spectroscopy to analyze the chemical structure of molecules. SRS has also enabled vibrational imaging of cells and tissues [1]. However, the sensitivity of traditional SRS spectroscopy is restricted by the laser shot noise [2,3]. Quantum-enhanced SRS (QE-SRS) has been reported to break this physical limit, but previous reports suffered from low sensitivity due to continuous-wave excitation [4] or low average power [4,5]. Here, we demonstrate QE-SRS spectroscopy with pulsed excitation and QE-balanced detection [6], which is advantageous for quantum enhancement in the moderate power regime (>10 mW).

## 2. Experimental setup

Fig. 1 shows the experimental setup. A Ti:sapphire laser provides picosecond SRS pump pulses. Squeezed vacuum is generated via single-pass optical parametric amplification in a periodically poled stoichiometric LiTaO<sub>3</sub> (PPSLT) waveguide. A spatial light modulator is installed for beam shaping of local oscillator light. A Yb fiber laser system provides picosecond SRS Stokes pulses with ~30-nm wavelength tuning range for hyperspectral SRS measurement. A homemade balanced photodetector is used for QE-SRS signal detection.



Fig. 1. Schematic of QE-SRS experimental setup. TiS: Ti:sapphire, AOM: acousto-optic modulator, SLM: spatial light modulator, SHG: second-harmonic generation, OPA: optical parametric amplifier, DM: dichroic mirror, RFSA: RF spectrum analyzer.

### 3. Results and discussion

Fig. 2(a) shows the output of the balanced photodetector without placing a sample. A squeezing level of >3 dB was achieved. Fig. 2(b) shows the SRS spectra of dimethyl sulfoxide-d6 (d-DMSO). With the assistance of squeezed vacuum, >2-dB noise reduction was realized. The squeezing level is presumably limited by the scattering loss when passing through the sample contained in a cuvette.



Fig. 2. (a) Squeezing result without placing SRS sample. (b) SRS spectra with or without squeezed vacuum input. SNL: shot-noise-limited, ASQ: anti-squeezed, SQ: squeezed.

## 4. Conclusions

In this report, we have demonstrated QE-SRS spectroscopy with picosecond laser sources and QE-balanced detection. The SRS sensitivity is improved by 2.06-dB below the shot-noise-limit. Currently, we are preparing the imaging system to achieve ultrasensitive QE-SRS microscopy. We believe our approach will enable new possibilities in SRS imaging.

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