

Contribution ID: 4284 Type: Oral Competition (Graduate Student) / Compétition orale (Étudiant(e) du 2e ou 3e cycle)

(G*) A Novel Approach to Tunable, Broadband Stimulated Raman Spectroscopy

Monday 27 May 2024 15:15 (15 minutes)

Stimulated Raman spectroscopy in the femtosecond (1 fs = 1×10^{-15} °s) regime provides a versatile route to measuring the dynamics of molecules on the timescale at which they occur. A tunable and broadband probe pulse allows for detecting molecular signatures across a wide range of energies (frequencies). We develop a novel method for generating the probe pulse that results in the broadest and most tunable probe pulse reported to date.

Four-wave mixing (FWM) occurs when two pump photons (ω_p) amplify a signal photon (ω_s) to create an idler (ω_i) : $\omega_p + \omega_p = \omega_s + \omega_i$. We show that at high intensities, FWM can be extended to include the nonlinear response of the gain medium. We exploit the $\chi^{(3)}$ (Kerr) nonlinearity of materials to amplify broad spectra. We use the resulting amplified spectrum as the probe pulse for stimulated Raman scattering. The benefits of this approach are twofold. First, there is an inherent tunability of the amplified spectrum, defined by the phase-matching condition. Second, we generate Raman frequencies that span the terahertz, fingerprint, and OH-stretching regimes in a single shot.

We prove the usefulness of our method by measuring the methyl stretching mode of 1-decanol, shown in Fig. 1.

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Keyword-1

Raman spectroscopy

Keyword-2

Ultrafast physics

Keyword-3

Nonlinear optics

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Session Classification: (DAMOPC) M2-8 Scattering & Spectroscopy | Diffusion et spectroscopie (DPAMPC)

Track Classification: Technical Sessions / Sessions techniques: Atomic, Molecular and Optical Physics, Canada / Physique atomique, moléculaire et photonique, Canada (DAMOPC-DPAMPC)