Study of non-resonant effects of stimulated Raman scattering with intensive Raman pump pulse

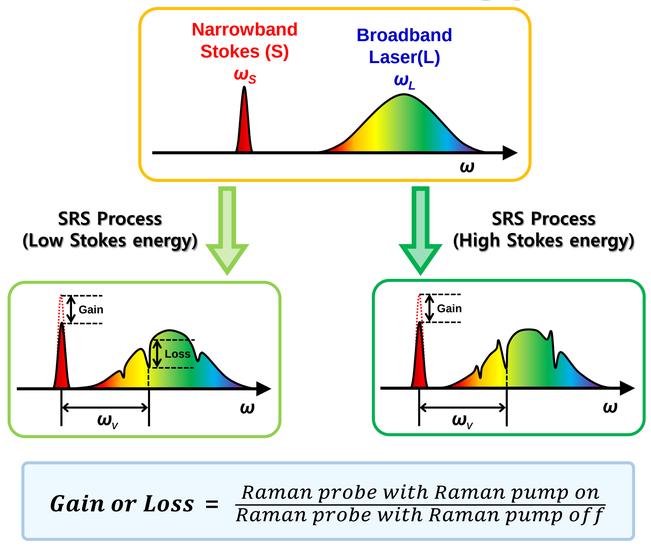
Sohee Lim[1,2], Bonghwan Chon[1,2], Hanju Rhee*[,3] and Minhaeng Cho*[,1,2]

[1] Center for Molecular Spectroscopy and Dynamics, Institute for Basic Science (IBS)
[2] Department of Chemistry, Korea University
[3] Korea Basic Science Institute (KBSI)
*hjrhee@kbsi.re.kr, *mcho@korea.ac.kr

Introduction

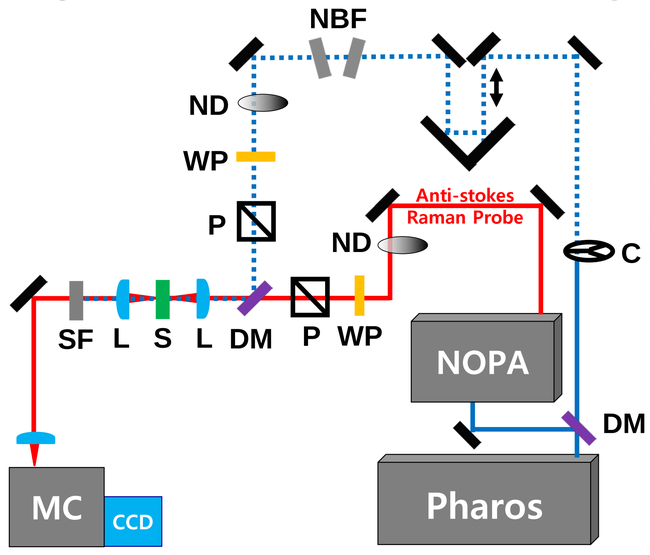
Unlike spontaneous Raman effect, stimulated Raman scattering (SRS) generates fields allowing Raman signals from individual scatterers to add up into a highly directional, high-brightness coherent beam. In our experiment, however, at intensive Raman pump energy, the SRS spectra is distorted dramatically. The experimental results are compared with theoretical simulation by numerically solving the third order susceptibility. These results show that nonlinear effects such as cross-phase modulation (XPM), self-phase modulation (SPM) can be included in SRS signals with intense Raman pump pulse.

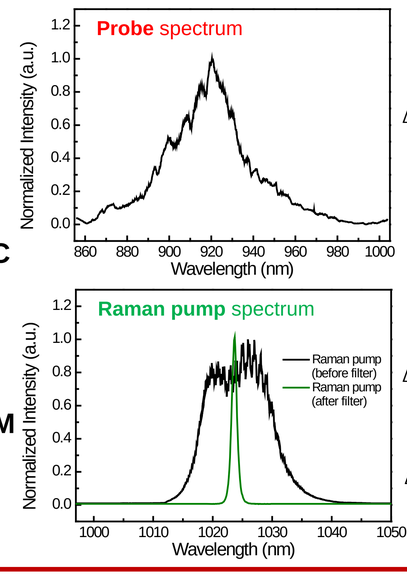
Stimulated Raman scattering process



$$Gain\ or\ Loss = \frac{Raman\ probe\ with\ Raman\ pump\ on}{Raman\ probe\ with\ Raman\ pump\ off}$$

Layout of the stimulated Raman spectrometer





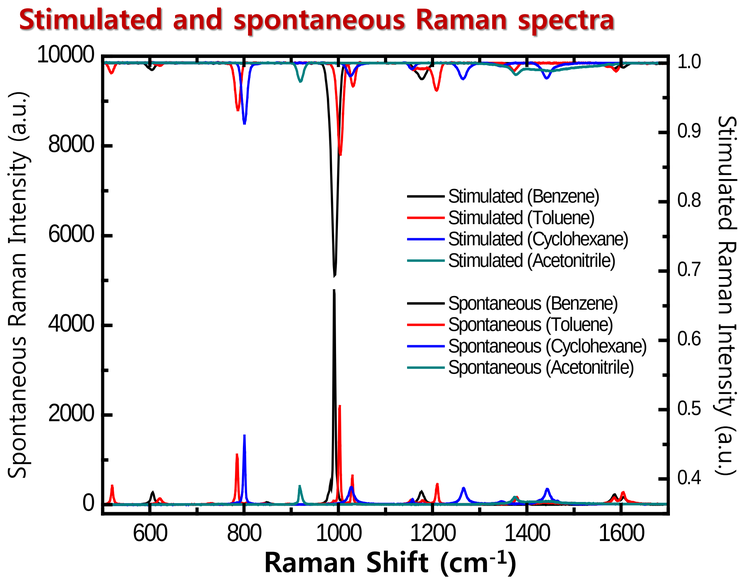
Raman probe
$\Delta\nu$ (FWHM) = ~ 507 cm^{-1}
= ~ 39 nm

Fundamental Raman pump
$\Delta\nu$ (FWHM) = ~ 156 cm^{-1}
= ~ 12 nm

Raman pump after NBP filter
$\Delta\nu$ (FWHM) = ~ 12 cm^{-1}
= ~ **1 nm**

Experimental results

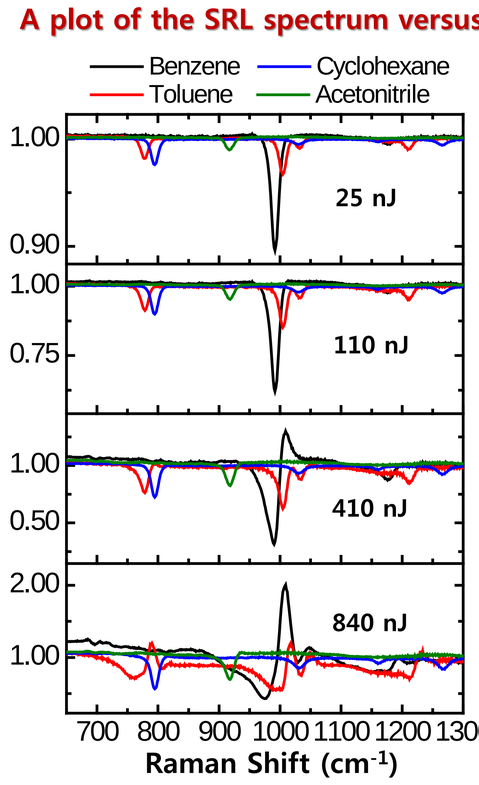
Stimulated and spontaneous Raman spectra

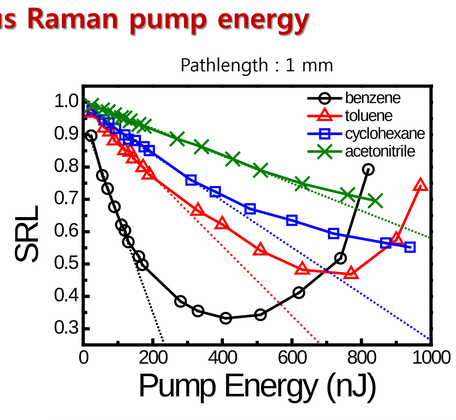


✓ The stimulated Raman spectra (upper) are well correlated with the spontaneous Raman spectra (lower).

✓ All peak intensities observed two different Raman spectroscopic methods are the same tendency, indicating the SRS method is a qualitative analytic method like to the spontaneous Raman spectroscopy.

A plot of the SRL spectrum versus Raman pump energy

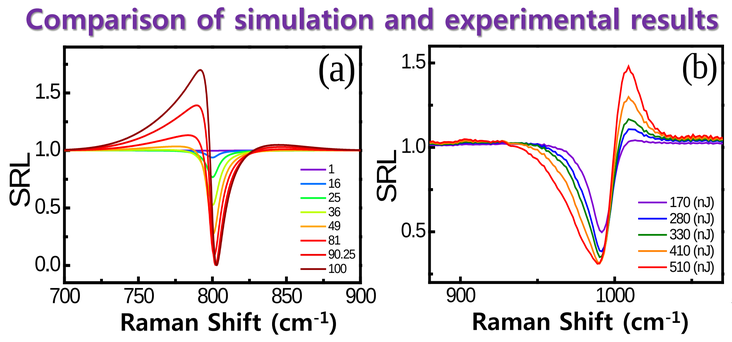




✓ At Raman pump energy above 400 nJ, the SRL spectra changes dramatically around strong Raman resonant mode. The results were inconclusive and difficult to interpret because the SRL spectra includes unpredictable non-resonant contributions such as pump-induced Optical Kerr Effect (OKE), cross-phase modulation (XPM), self-phase modulation (SPM) and so on.

✓ A plot of the SRL intensity at maximum Raman vibrational mode versus Raman pump energy. At low pump energy, linear fitting (dotted line) is well-matched with SRL intensity at maximum.

Comparison of simulation and experimental results



Raman gain (or Loss), G

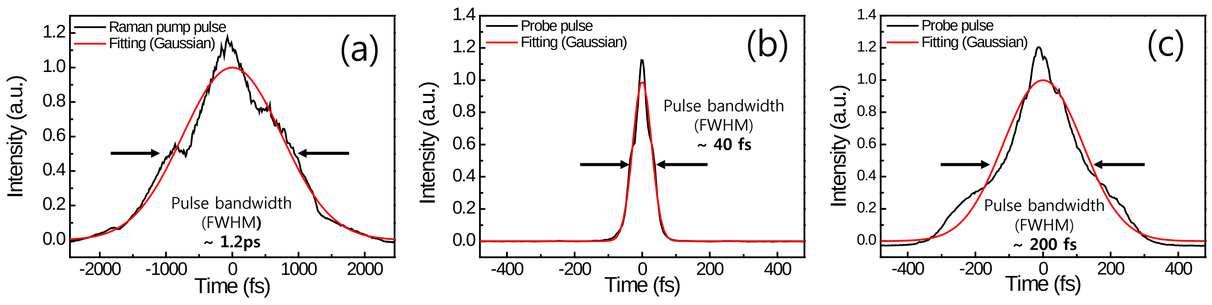
$$G = \frac{I_{on}}{I_{off}} = \frac{|E_{sig} + E_p|^2}{|E_p|^2}$$

$$P^{(3)} = \chi^{(3)} E_s E_S^* E_p$$

third order susceptibility

$$E_{sig}^{(3)} \propto i\,P^{(3)} = A\,i\,\chi^{(3)} E_s E_S^* E_p$$

Autocorrelation of Raman pump and (chirped) probe pulses



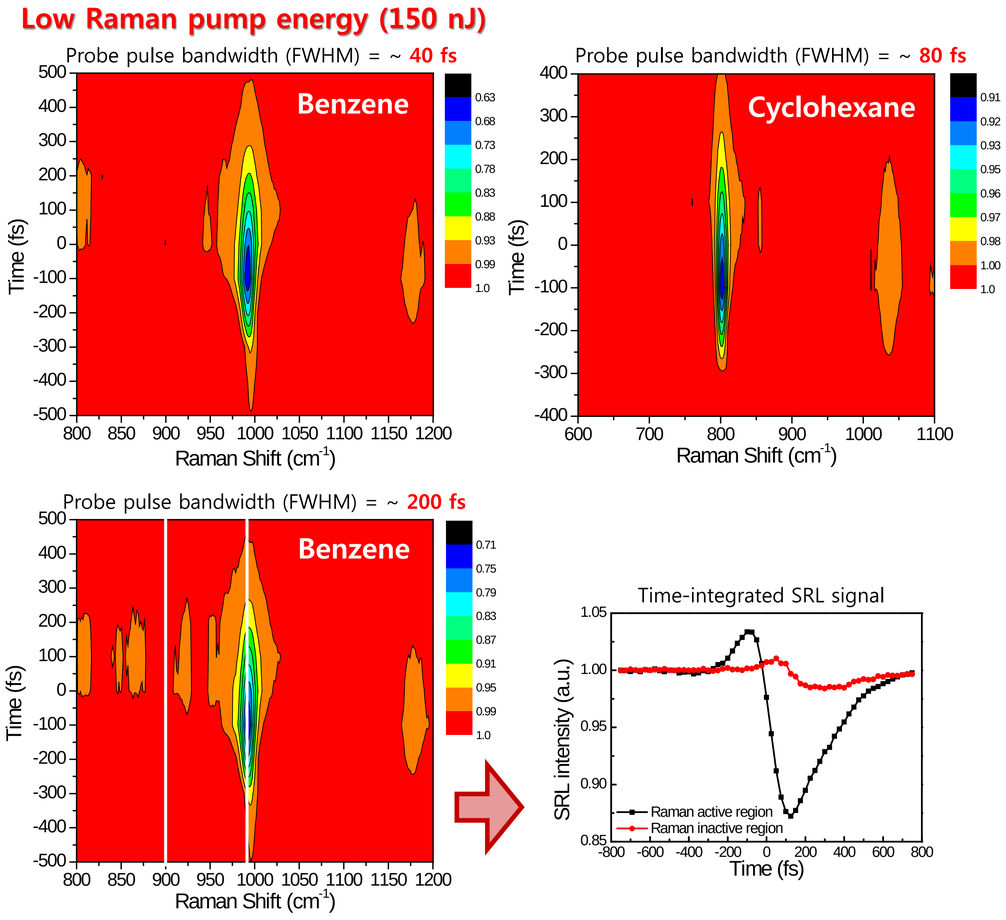
(b) : **transform limited** Gaussian

✓ We produce the chirped probe adjusting by prism compressor in Non-Collinear Optical Parametric Amplifier (NOPA) for measuring chirp effect.

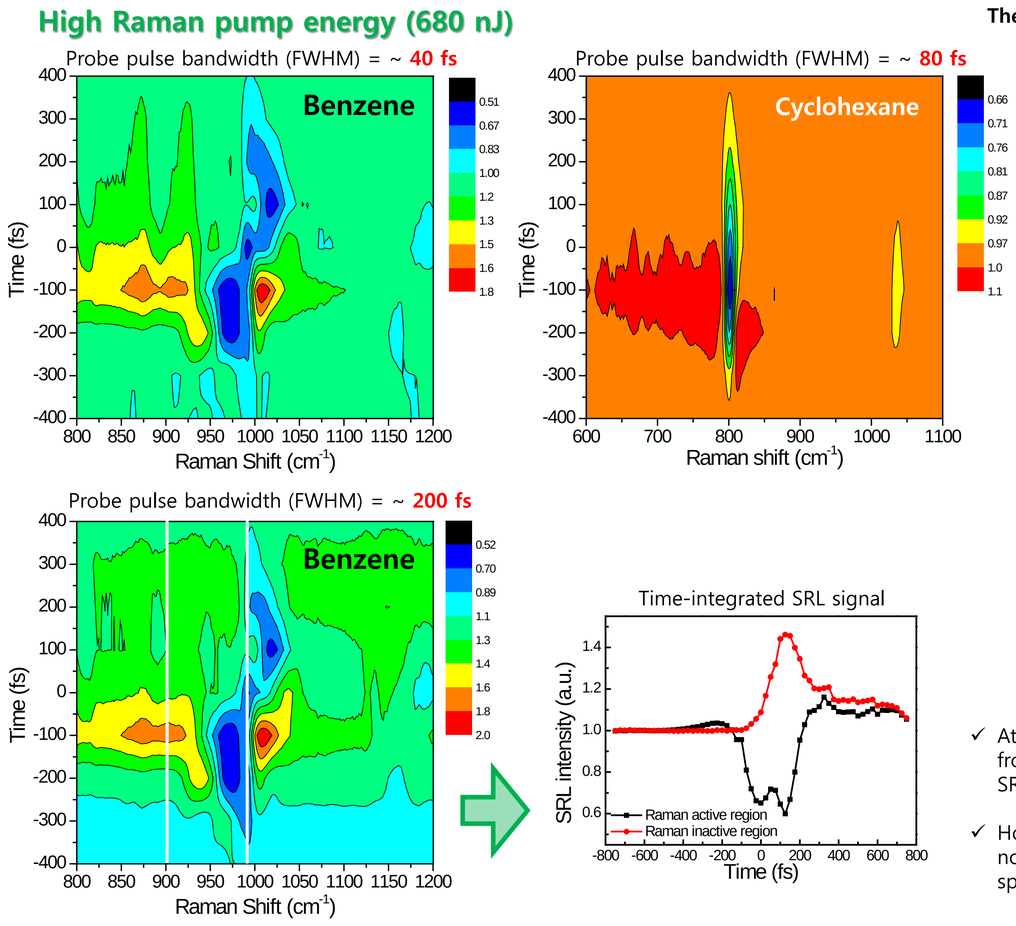
✓ The SRS signal intensity increase linearly with changes in Raman pump pulse energy. However, with intensive Raman pump pulse, SRL spectrum can be distorted. To understand properly dispersion shaped in SRL spectrum, we perform time-resolved SRL measurement with chirped probe pulse.

Time-resolved stimulated Raman scattering spectra

Low Raman pump energy (150 nJ)



High Raman pump energy (680 nJ)



The stimulated Raman Loss (SRL) can be defined as

$$I_c(z) = I_c(0)\exp(Gz)$$

$$G \propto Im\left[\chi_{SRL}^{(3)}(\omega_c)\right] I_{RP}$$

$$\propto \left(\frac{\partial^2\sigma}{\partial\omega\partial\Omega}\right) I_{RP}$$

G	: SRG or SRL
z	: The pathlength of sample
I_{RP}	: **The intensity of the Raman pump pulse**
$\frac{\partial^2\sigma}{\partial\omega\partial\Omega}$	: The spontaneous Raman cross section

The transient gain/loss $D(t_d)$ can be define as

$$D(t_d) \equiv \ln\left\{\frac{[(I_s/I_r)(t = t_d)]}{[(I_s/I_r)(t \ll 0)]_{blank}}\right\}$$

In order to eliminate the coherent artifact, $D(t_d)$ was integrated

$$G = \int D(t_d)dt_d$$

Seung Min Jin et al., Bull. Korean Chem. Soc., Vol.25, 2004

✓ At low Raman pump energy, nonlinear effects from XPM can be eliminated by time-integrated SRL signal on Raman inactive region.

✓ However, at high Raman pump energy, high order nonlinear process is included in time-resolved SRL spectra and it causes misunderstanding.

Center for Molecular Spectroscopy and Dynamics, IBS-Korea University