

Selective Suppression of Stimulated Raman Scattering with Another Competing Stimulated Raman Scattering

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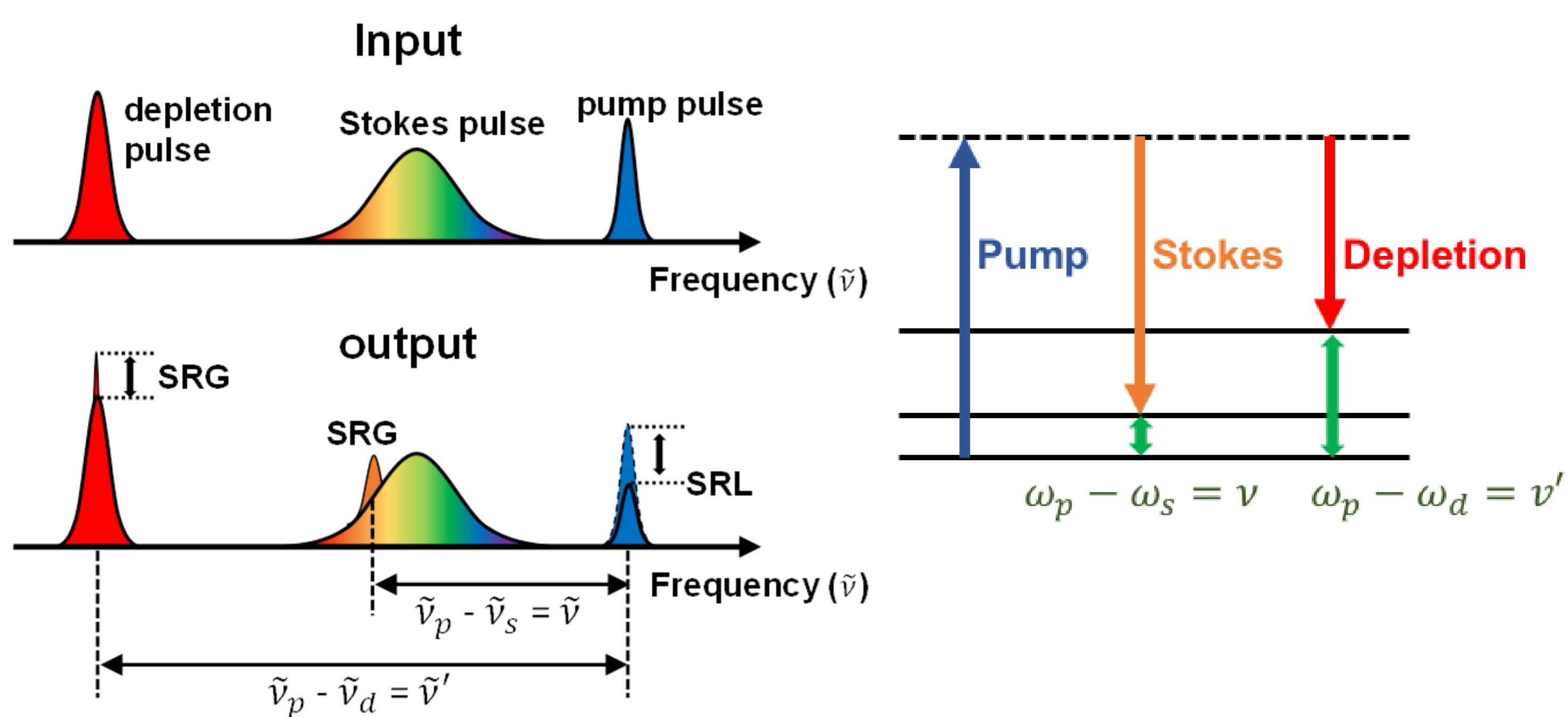
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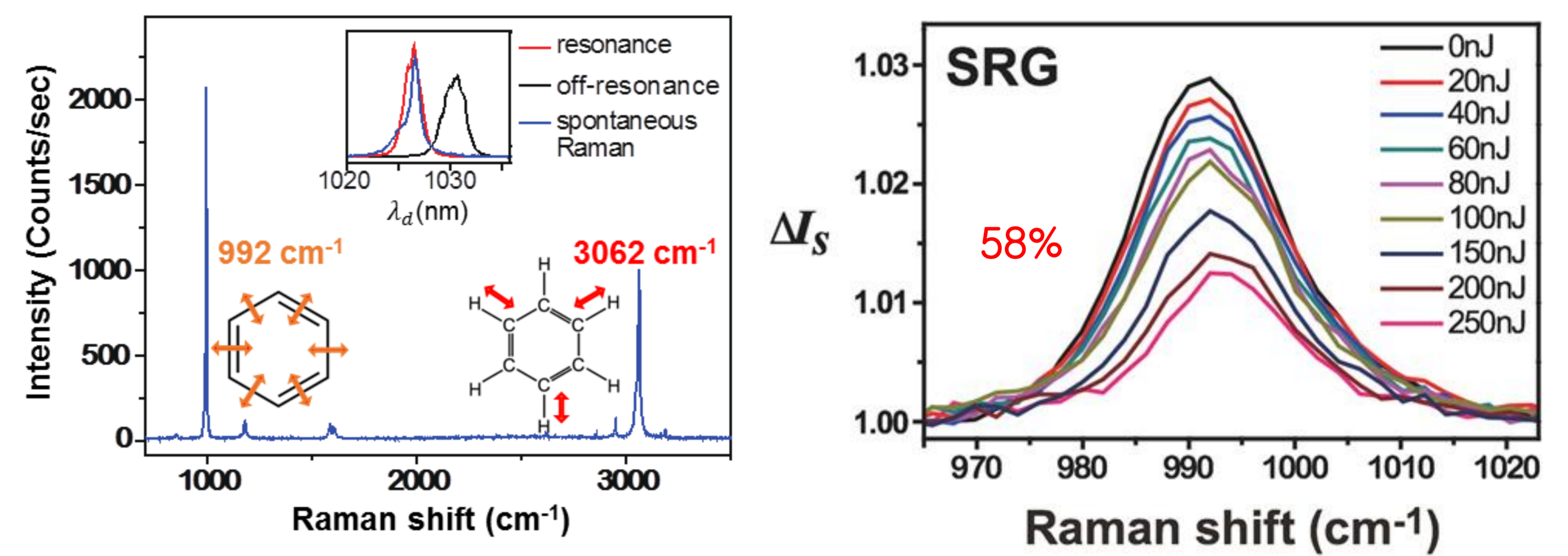
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Three-beam double SRS (Stimulated Raman Scattering)



- Here, we performed SRS experiments using three different laser pulses to selectively suppress one SRS process by the other SRS process for improving the spatial resolution of Raman microscopy.

Suppression of stimulated Raman gain signal in benzene



- We used the fundamental output at 1031 nm of a high-power OPA for a stimulating Stokes beam for the C-H stretch mode of benzene. With the depletion beam energy increasing from 0 to 250 nJ, we measured SR gain spectra of the ring breathing mode of benzene. The SRG intensity of the ring breathing mode is strongly suppressed by the competing SRS process of C-H stretch induced by the pair of pump and depletion beams.

Theoretical derivation

Three-color double SRS processes

$$\frac{d\Delta I_s(z)}{dz} = G_s \left(\frac{\mu_p V}{\hbar \omega_p c} I_p(0) - \frac{\mu_s V}{\hbar \omega_s c} \Delta I_s(z) - \frac{\mu_d V}{\hbar \omega_d c} \Delta I_d(z) \right) (I_s(0) + \Delta I_s(z))$$

$$\frac{d\Delta I_d(z)}{dz} = G_d \left(\frac{\mu_p V}{\hbar \omega_p c} I_p(0) - \frac{\mu_s V}{\hbar \omega_s c} \Delta I_s(z) - \frac{\mu_d V}{\hbar \omega_d c} \Delta I_d(z) \right) (I_d(0) + \Delta I_d(z))$$

Weak Stokes and strong depletion beams

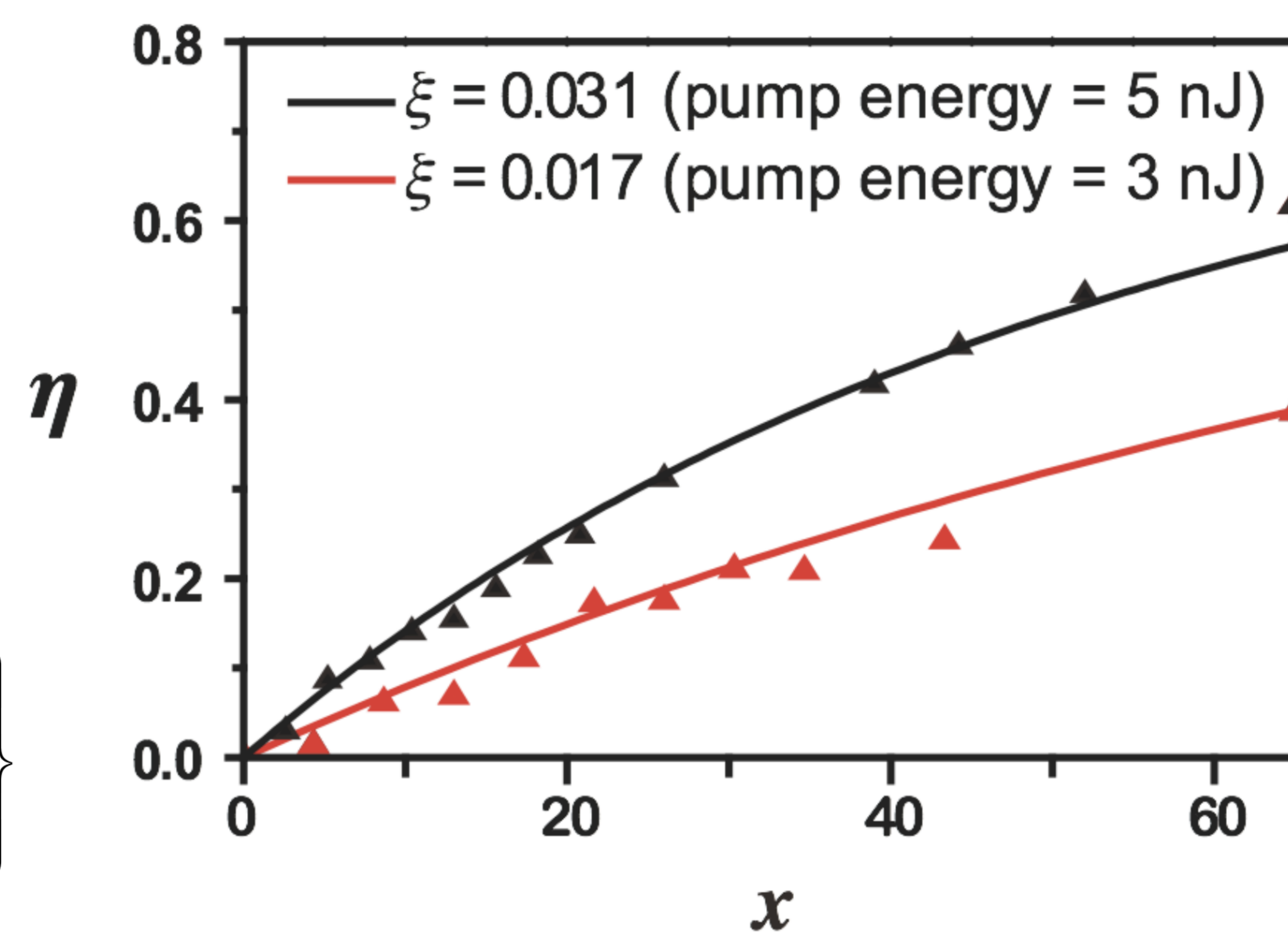
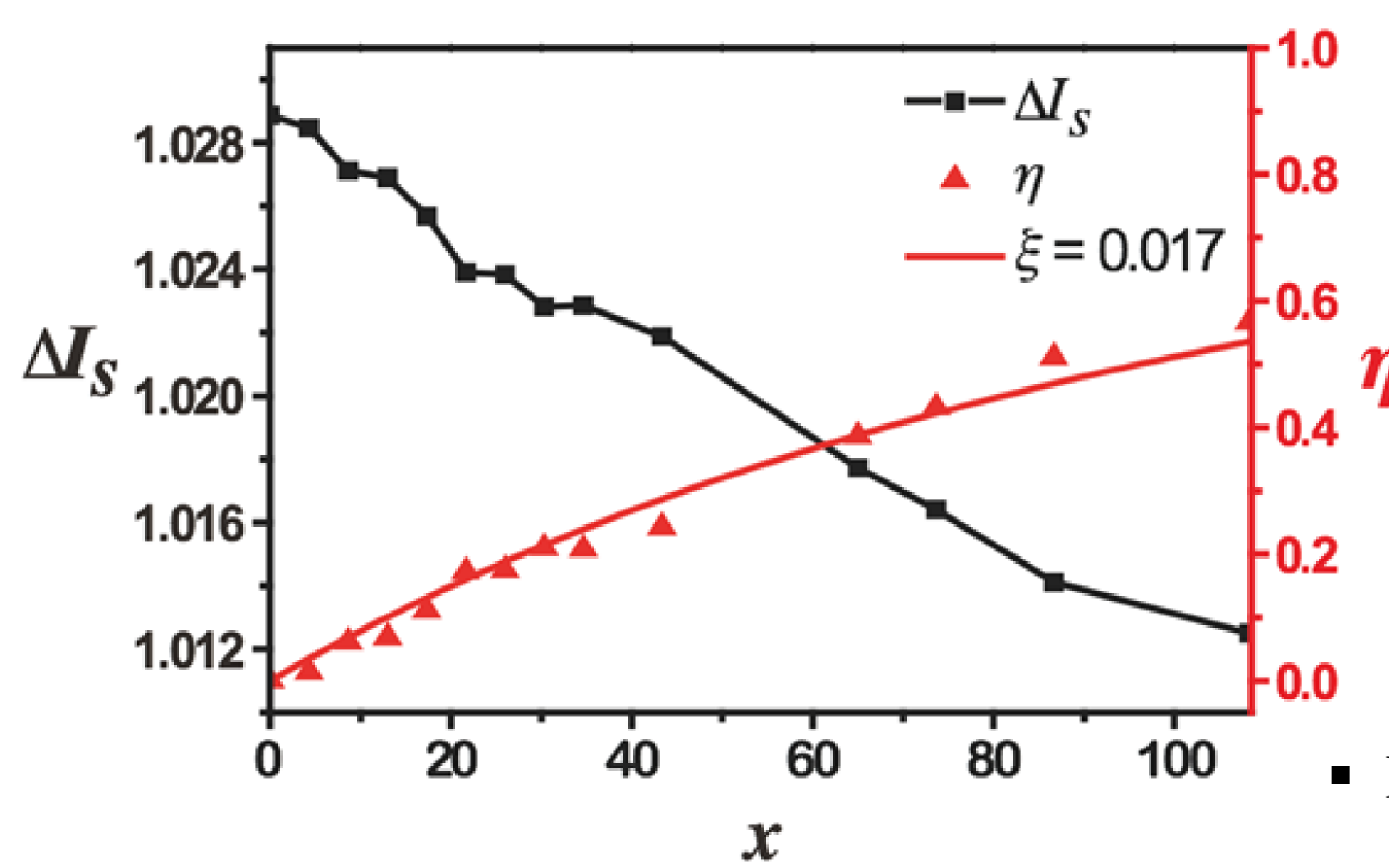
$$I_d(0) > I_p(0) > I_s(0)$$

$$\frac{d\Delta I_s(z)}{dz} = G_s I_s(0) \left(\frac{\mu_p V}{\hbar \omega_p c} I_p(0) - \frac{\mu_d V}{\hbar \omega_d c} \Delta I_d(z) \right)$$

$$\frac{d\Delta I_d(z)}{dz} = G_d \left(\frac{\mu_p V}{\hbar \omega_p c} I_p(0) - \frac{\mu_d V}{\hbar \omega_d c} \Delta I_d(z) \right) (I_d(0) + \Delta I_d(z))$$

$$\Delta I_s(z) = G_s I_s(0) \left(\frac{\mu_p V}{\hbar \omega_p c} I_p(0) + \frac{\mu_d V}{\hbar \omega_d c} I_d(0) \right) z$$

$$+ \frac{G_s}{G_d} I_s(0) \left\{ \ln \left(\frac{\mu_p \omega_d I_p(0)}{\mu_d \omega_p I_d(0)} + 1 \right) - \ln \left(\frac{\mu_p \omega_d I_p(0)}{\mu_d \omega_p I_d(0)} + e^{G_d \left[\frac{\mu_p V}{\hbar \omega_p c} I_p(0) + \frac{\mu_d V}{\hbar \omega_d c} I_d(0) \right] z} \right) \right\}$$



Suppression efficiency

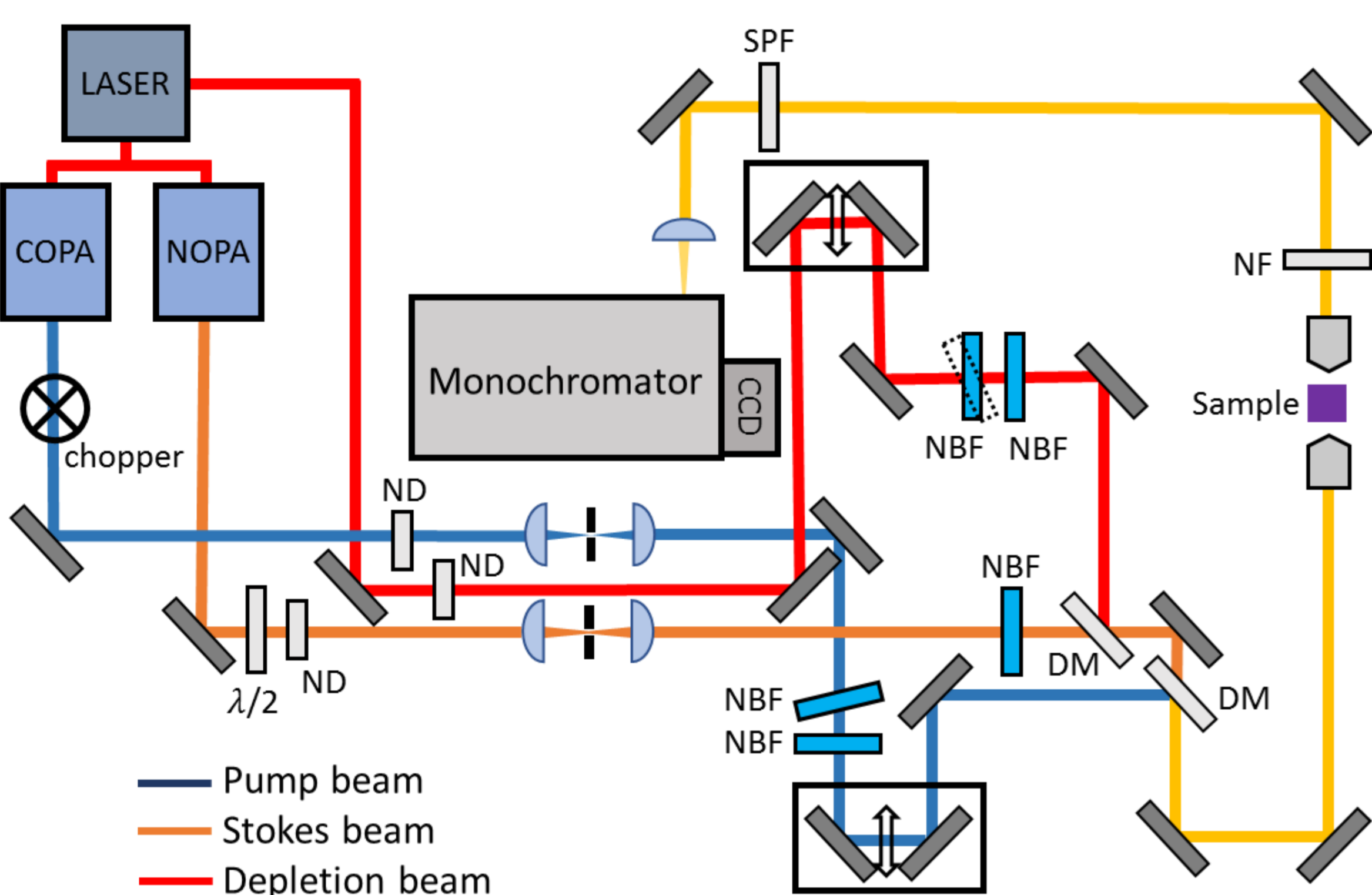
$$\eta \equiv \frac{\Delta I_s(z, I_d(0) = 0) - \Delta I_s(z, I_d(0))}{\Delta I_s(z, I_d(0) = 0)}$$

$$= 1 - \left\{ \left(1 + x \right) + \frac{1}{\xi} \ln \left(\frac{x + 1}{x e^{\xi(1+x)} + 1} \right) \right\}$$

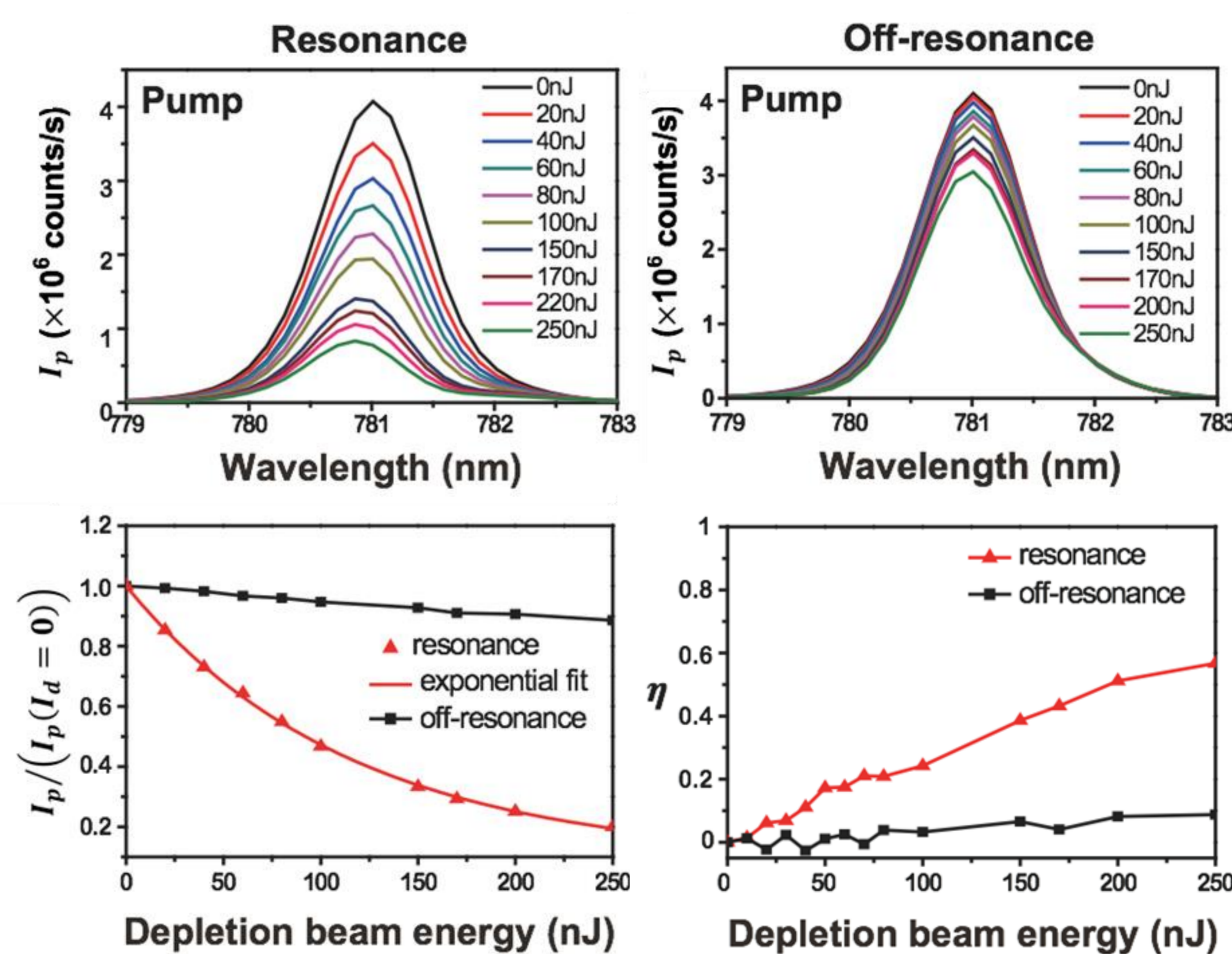
$$(\xi \equiv G_d n_p(0) z, x = n_d(0) / n_p(0))$$

- If the intensity of depletion beam is increased, the Stokes Raman gain becomes coupled with Raman gain of the depletion beam. Using approximate rate equations, we found the analytical solution for the Stokes gain signal ΔI_s .
- To estimate how strongly the SRG of ring breathing mode is suppressed, we calculated the suppression efficiency η using our experimental result. We numerically calculated the value by using known coefficients and compare that with the value obtained by fitting to experimental results with equations. With 3 nJ pump energy, the calculated value is 0.07, whereas our fitted value is 0.017. Although a few approximations were used to derive the equation, our calculated value appears to be in good agreement with the experimental value.

SRS Spectroscopy Set up

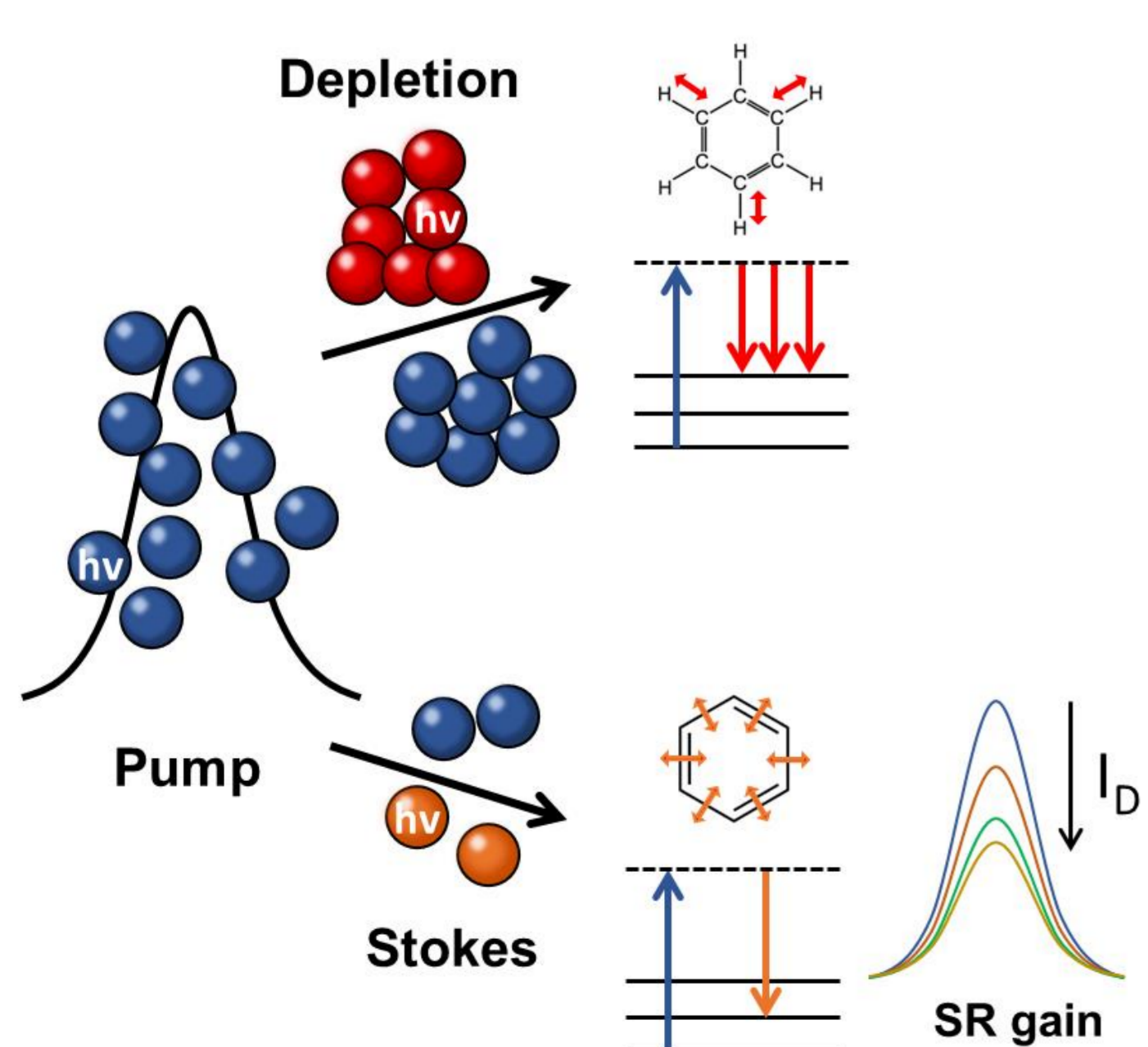


Resonant vs. off-resonant conditions: Pump intensity loss



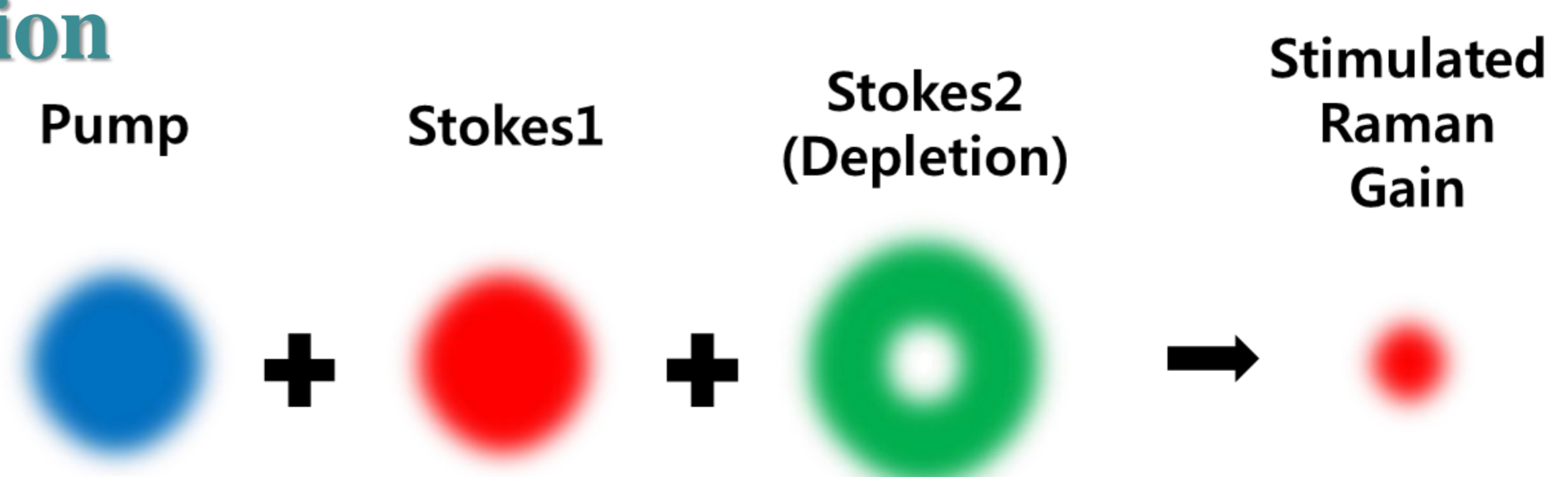
- To check if the depletion is indeed caused by another SRS process, we changed the center wavelength of the depletion beam about 4 nm to make the depletion beam off-resonant, and carried out the same measurements. When the depletion beam is in resonance with the C-H stretching vibration, the pump signal was exponentially decreased and the depletion efficiency was 80% at depletion pulse energy of 250 nJ. However, as the depletion beam becomes off-resonant, the suppression efficiency at the same condition dropped to 11%. This clearly shows that the SRS process induced by the pump and depletion beam pair with coherently excited C-H stretch mode strongly competes with and effectively suppresses the target SRS process of the ring breathing mode.

Conclusion



- Using three-color double SRS measurements, we successfully suppress the SR gain signal with chemical specificity.
- Suppression of SRG signal is associated with the competition between two different but coupled SRS processes by the same pool of pump photons.
- The maximum suppression efficiency of target SR gain signal in our experimental condition using benzene is $\sim 58\%$.

Future direction



- Our scheme can be used to super-resolution SRS microscopy by using doughnut shaped depletion beam for selective suppression of the target SRS signal in the periphery of the focal spot of the pump and Stokes beams. The theoretical resolution is as below.

$$\Delta r_{FWHM} = \frac{\lambda}{2\mu \sin \theta} \frac{4/\pi}{\sqrt{3 + \frac{G_d n_d^0}{G_s n_s^0}}} \approx \frac{\lambda}{2NA} \frac{1.273}{\sqrt{3 + \frac{G_d n_d^0}{G_s n_s^0}}}$$

- We are planning to implement our scheme to microscopy using various samples containing benzene ring such as polystyrene beads. The same vibrational modes used in the spectroscopic study of benzene can also be used as detection (992cm^{-1}) and depletion (3056cm^{-1}) modes.