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

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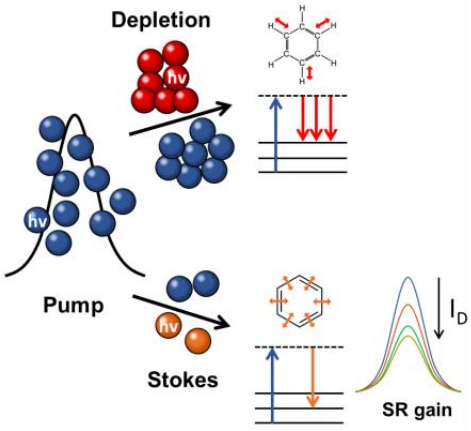

Raman scattering microscopy offering a label-free, chemical-specific contrast mechanism based on the intrinsic vibrations of chemical bonds or groups is limited in spatial resolution by the optical diffraction limit. We aim to develop a new method for super-resolution Raman imaging by implementing the depletion strategy of STED (Stimulated Emission Depletion) microscopy to a stimulated Raman scattering (SRS) technique. In order to reduce the effective focal spot of a SRS signal, we overlap the focal spot with a donut shaped beam that eliminates SRS signal at the periphery of the focal spot. In order to achieve resolution <100 nm, it is required to device a way to suppress SRS signal with efficiency >40% [1]. Our depletion principle is based on the competition of two different stimulated Raman processes on the same molecule. When two SR gains shares the same pump pulse, the two processes become coupled and complete: As one of the Stokes beam intensity increases, the other SR gain is selectively suppressed. We obtained up to 70% efficiency for the depletion of the ring breathing mode of benzene by using the CH stretching mode with 2.1 TW/cm2 of 1026.5 nm [2]. At the biological damage threshold of 1 TW/cm2, the depletion efficiency is 40 % and the expected resolution is 6-fold enhanced from the diffraction limit. With the theoretical foundation and experimental demonstration, we expect a practical biological imaging based on Raman scattering with sub-100-nm resolution.

[1] Cho, M., "Three-beam double stimulated Raman scatterings" *The Journal of Chemical Physics*, **148**, 014201 (2018).

[2] Kim, D.; Choi, D. S.; Kwon, J.; Shim, S.-H.; Rhee, H.; Cho, M., "Selective Suppression of Stimulated Raman Scattering with Another Competing Stimulated Raman Scattering" *The Journal of Physical Chemistry Letters*, **8**, 6118-6123 (2017).