TEMPORAL EVOLUTION OF MULTI-ORDER STIMULATED RAMAN SCATTERING IN DROPLET

J. Kohno

Gakushuin University, Tokyo, Japan

jun-ya.kohno@gakushuin.ac.jp

Nonlinear Raman scattering proceeds under an intense light field of the incident and the Raman scattering lights. A liquid droplet provides the intense light field because it acts as a high-quality optical cavity. We previously found that a colliding droplet acts as an optical cavity with higher quality factor than the single droplet by using carbon tetrachloride (CCl_4) as sample liquid, where multi-order stimulated Raman scattered light emerges with significant intensity.[1] In the present study, we invesitigated the mechanism of the multi-order Raman scattering from temporal evlution of the stimulate Raman scattered light in the single/colliding droplet.

Liquid droplets of CCl_4 were produced by a set of piezo-driven nozzles. The single/colliding droplet was irradiated with a second harmonic of a Q-switched Nd:YAG laser for excitation of the stimulated Raman scattering. The Raman scattered light passed through a long-pass filter for removal of the Rayleigh scattering, and divided into two components for a simultaneous measurement of the collision image and the spectrum. For the measurement of the temporal prifile, the Raman scattered light was monochromated and detected by a photomultiplier.

Figure 1 shows the stimuated Raman spectrum obtained from the single and the colliding CCl_4 droplet. For single droplet, peaks in the spectrum are assignable to an integer multiple of 460 cm^{-1}, the wavenumber of the ν_1 mode of CCl_4. For colliding droplet, on the other hand, peaks appear at Raman shift with linear combinations of the vibrational modes of CCl_4, such as ν_1 (460 cm^{-1}), ν_2 (240 cm^{-1}), ν_3 (770 cm^{-1}), and ν_4 (340 cm^{-1}). The Raman bands are denoted as [*a b c d*], where *a* – *d* represent the number of the ν_1 – ν_4 modes, respectively, included in the band.

Figure 2 shows the temporal profile of the Raman bands observed from the single and the colliding droplet. From the single droplet, higher-order Raman scattered lighte emerges with longer delay time from the incident laser. From the colliding droplet, on the other hand, the higher order Raman scattered light originating from the ν_1 mode emerges with almost no delay time, whereas those originating from ν_2 and ν_4 modes have delay of several nanoseconds which is significantly smaller than the delay of the single droplet. These results are explained by assuming the multi-order Raman scattering proceeds after building up light field of the fumdamental and the most intense Raman mode inside the droplet.

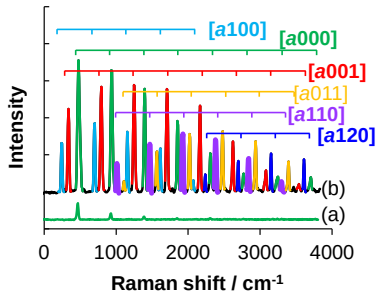

Figure 1 Raman spectra of single (a) and colliding (b) CCl_4 droplet.

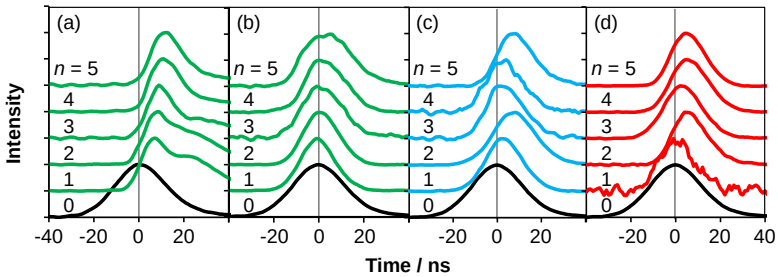

Figure 2 Time profiles of multi-order Raman bands of $n\nu_1$ mode obtained from single (a) and colliding (b) CCl_4 droplet, and those of $\nu_2+n\nu_1$ (c) and $\nu_4+n\nu_1$ (d) modes from colliding CCl_4 droplet.

1. Negishi K., Suzuki S. and Kohno J. 'Multi-Order Stimulated Raman Scattering in Colliding Droplets' *J. Phys. Chem. A*, **2018**, *122*, 6473.