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Temperature dependent of IVR investigated by steady-state and time-frequency resolved CARS for liquid nitrobenzene and nitromethane[1]

YANQIANG YANG, GANGBEI ZHU, LIN YAN, XIAOSONG LIU, China Acad of Engr Phys, YANGS ULTRAFAST SPECTROSCOPY GROUP TEAM — Intramolecular vibrational energy redistribution (IVR) is important process in thermal decomposition, shock chemistry and photochemistry. Anti-Stokes Raman scattering is sensitive to the vibrational population in excited states because only vibrational excited states are responsible to the anti-Stokes Raman scattering, does not vibrational ground states. In this report, steady-state anti-Stokes Raman spectroscopy and broad band ultrafast coherent anti-Stokes Raman scattering (CARS) are performed. The steady-state anti-Stokes Raman spectroscopy shows temperature dependent of vibrational energy redistribution in vibrational excited-state molecule, and reveal that, in liquid nitrobenzene, with temperature increasing, vibrational energy is mainly redistributed in NO2 symmetric stretching mode , and phenyl ring stretching mode of ν_{CC} . For liquid nitromethane, it is found that, with temperature increasing, vibrational energy concentrate in CN stretching mode and methyl umbrella vibrational mode . In the broad band ultrafast CARS experiment, multiple vibrational modes are coherently excited to vibrational excited states, and the time-frequency resolved CARS spectra show the coincident IVR processes.

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