Optical excitation and control of coherent lattice motions in carbon nanotubes

Ki-Ju Yee, Ji-Hee Kim, Yong-Sik Lim*, and Junichiro Kono**
Depearment of Physics, Chungnam National University, *Department of Applied Physics, Konkuk University, **Department of Electrical and Computer Engineering, Rice University, USA
kyee@cnu.ac.kr

Having a cylindrical shape with a diameter of around 1 nm, , single-walled carbon nanotubes (SWNTs) constitute a prototype of one-dimensional nano-structures, where the quantum size effects become important and the lattice vibrational frequency depends on its diameter.(1)

When femtosecond pulses with the duration being shorter than the phonon period, are indident on a material, coherent oscillation of the lattice modes can be invoked impulsively.(2) In this talk, we demonstrate on excitation and control of coherent radial breathing mode (RBM) and G-mode oscillations, by applying femtosecond pump-probe and pulse shaping technique for carbon nanotubes. Using ultrashort pulses with about 10 fs FWHM and the spectrum covering from 710 nm to 900 nm from a Ti:sapphire laser, we have measured coherent lattice motions in micelle-suspended SWCNTs. Fig. 1(a) shows the transmitted probe intensity modulations at the detection wavelength of 750 nm, in which both coherent G-mode and RBM lattice vibrations are observed as denoted in the Fourier-transformed spectra of Fig. 1(b).

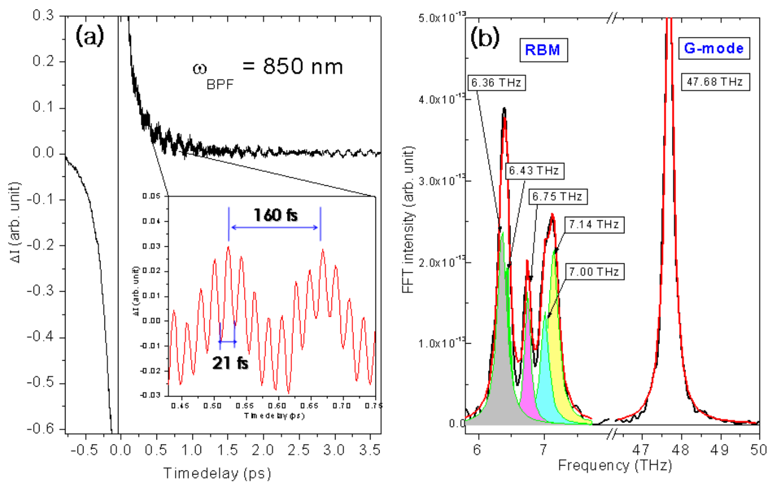

Fig. 1 (a) Probe intensity changes as a function of the time delay between pump and probe pulses showing coherent RBM (~160 fs peroids) and G-mode (~21 fs period) oscillations. (b) Fourier-transformed spectra of time-domain oscillations.

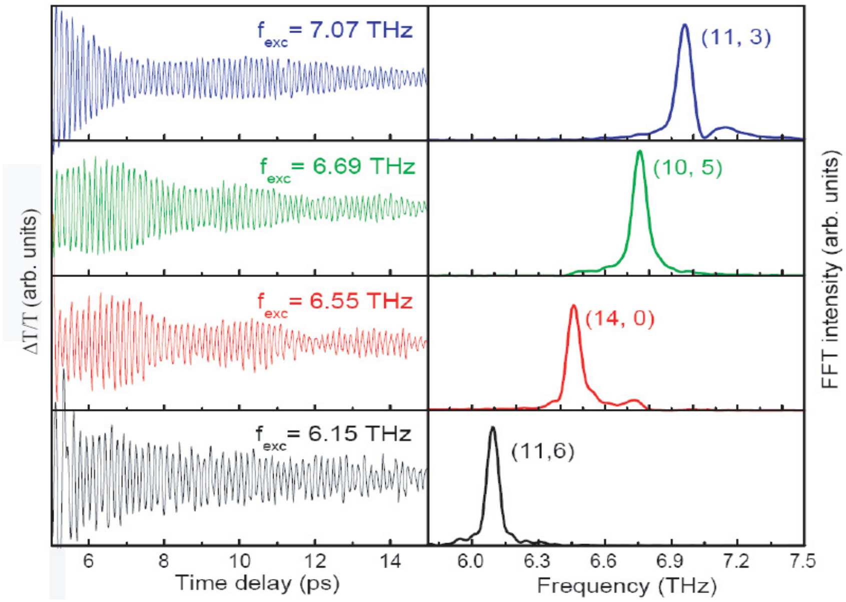

Fig. 2 Coherent RBM oscillations excited by multiple pulse trains and the corresponding Fourier-transformed spectra.

Among diverse chiralities with different diameters as is excited by single femtosecond pulses shown in Fig. 1, a specific chiral excitation for coherent RBM motions could be successfully fulfilled with the help of the repetitive pulse trains prepared by the femtosecond pulse shaping apparatus as shown in Fig. 2. With the appropriate repetition rate of the pulse trains, a single, specific chirality dominantly contributes to the signal, while other nanotubes are suppressed. For example, by choosing a pump repetition rate of 7.07 THz, we can selectively excite only the (11,3) nanotubes. Similarly, with a pump repetition rate of 6.69 THz, the (10,5) nanotubes are selectively excited. The accuracy of selectivity depends on the number of pulses in the tailored pulse train as well as on the distribution of chiralities in the nanotube ensemble.

We also address the detection mechanism of the band gap modulations for the RBM mode and the impulsive stimulated Stokes/anti-Stokes Raman scattering for the G-mode oscillations.

1. M. S. Dresselhaus, G. Dresselhaus, and P. Avouris, "Carbon Nanotubes: Synthesis, Properties, and Applications", Springer-Verlag, Berlin (2001).
2. Y. S. Lim, K. J. Yee, J. H. Kim, E. H. Hároz, J. Shaver, J. Kono, S. K. Doorn, R. H. Hauge, and R. E. Smalley, "Coherent lattice vibrations of single-walled carbon nanotubes", Nano Lett. **6**, 2696-2700 (2006).