## SUPPORTING INFORMATION

## Enhanced Double Resonance Raman Scattering in Multilayer Graphene with Broadband Coherent Anti-Stokes Raman Spectroscopy

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AFM experiments are conducted both in the tapping mode and contact mode to avoid the influence of surface adsorbents. The height difference between the substrate and test region is about 10nm which equals to 30 layers graphene.



Figure S2: Spectrum of the supercontinuum pulse

Supercontinuum pulses are measured with a fiber optic spectrometer (Ocean, USB2000+) for the 780nm-1000nm part as figure S2(a) shown after blocking the narrow band pulse and removing the short pass filter. The longwave part is relative weak compared with the short part which result in the same trend in the emission spectra as figure 2d shown because of the four-wave mixing relation. Figure S2 (b) are measured with a near infrared spectrometer (Ocean, flameNIR) which shows the supercontinuum are broadened to 1050nm.



Figure S3: Time-resolved B-CARS spectrums of CNT powders

The time-resolved B-CARS spectrums of CNT powders are measured under the same experimental conditions with MLG. CNT powders are deposited on the cover glass directly. Clear G peak is detected around 696.976nm in 0-time delay, as the time delay increase to 1.2 ps, the center of the peak gets a 0.651nm blue shift because of the influence of the dispersive part in Eq. (1) when the NVRB is suppressed gradually. The peak position settles down after 1.6ps in the position of 696.976nm which means the phononic response is dominant.



Figure S4: Autocorrelation curve of the supercontinuum pulse

The time duration of the supercontinuum is about 650 fs which is short enough to excite phonons inside the pulse.