Intramolecular Vibrational Energy Redistributions in Nanocarbons Controlled by Tailored Intense Near-Infrared Pulses

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Interactions between nanocarbons and intense near-infrared (NIR) pulses with femtosecond pulse length $T_p$ and intensity $I \approx 10^{15}$ W/cm$^2$ induce structural rearrangements and fragmentations. Laarmann et al. experimentally showed that tailored NIR pulse trains can optimize the specific fragment yields of fullerene C$_{60}$ [1]. We theoretically showed that selective impulsive Raman excitation of specific vibrational modes with vibrational period $T_{vib}$ in C$_{60}$ can be achieved by setting $T_p \approx T_{vib}/2$ and the fragmentation process depends on the vibrational modes initially excited [2]. These results suggest that intramolecular vibrational energy redistributions (IVR) leading to fragmentations depend on the pulse shape.

We performed the on-the-fly trajectory calculations on NIR-pulse-induced vibrational dynamics in C$_{60}$ with various $T_p$ to clarify the effects of the pulse shape on the IVR dynamics. We combined the time-dependent adiabatic state approach [3] and density-functional based tight-binding theory [4] for simulations. We found that exciting different vibrational modes drastically changes the IVR dynamics as in Fig. 1 even if the total vibrational energy $E_{in}$ acquired remains the same. We will also discuss IVR dynamics in a semiconducting single-wall carbon nanotube.

**Fig. 1:** Temporal change of the vibrational energies in low frequency modes of C$_{60}$ induced by single pulse excitation ($I = 7.0 \times 10^{14}$ W/cm$^2$, $\lambda = 1800$ nm). (a) $T_p = 70$ fs: the $h_g(1)$ mode ($T_{vib} = 140$ fs) is mainly excited and the IVR is slow. (b) $T_p = 30$ fs: the $a_g(1)$ mode ($T_{vib} = 60$ fs) is relatively enhanced and fast IVR between the $a_g(1)$ and $h_g(2)$ modes is observed.