Electronic Supplementary Information:

Cooperative inter- and intra-layer lattice dynamics of photoexcited multi-walled carbon nanotubes studied by ultrafast electron diffraction

Shuaishuai Sun,^{1, †} Zhongwen Li,^{1,2, †} Zi-An Li,¹ RuijuanXiao,¹ Ming Zhang,^{1,2} Huanfang Tian,¹

Huaixin Yang,^{1,2} and Jianqi Li^{1,2,3,} *

¹Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese

Academy of Sciences, Beijing 100190, China

²School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China
³Collaborative Innovation Center of Quantum Matter, Beijing 100084, China



Figure S1. The full cycle of ultrafast structural dynamics of MWCNTs upon 45 mJ cm⁻² fs-laser excitation at microsecond time scale. The data was collected in UTEM with a nanosecond stroboscopic mode. (a) Peak position evolution of (002) plane; (b) Diffraction intensity evolution of (002) plane. The relaxation process was fitted by biexponential function with the indicated time constants, revealing the thermal diffusion of MWCNTs and heat transport between the materials and the supports.



Figure S2. Pump laser fluence dependence of structural dynamics of MWCNTs. (a) and (b) show the temporal evolutions of the (002)- and (100)- plane lattice spacings for the laser fluences indicated. (c) fluence dependence of the magnitudes of thermal lattice changes of the (002)- and (100)-plane estimated at time delay > 30 ps. (d) fluence dependence of the maximal magnitudes (at ~ 3 ps) of nonthermal lattice changes of the (002)-plane.



Figure S3. TEM lattice images of MWNCTs taken after pump laser irradiation of (a) 35 mJ cm⁻² and (b) 100 mJ cm⁻², respectively. In (a) the MWCNTs remain intact after pump laser excitation, while in (b) visible structural damages occur, as evidenced by the disordered layers and the ablated layers.