## Electronic Supplementary Information:

## Ultrafast Structural Dynamics of Boron Nitride Nanotubes

## **Studied Using Transmitted Electrons**

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Figure S1. Fitting of exponential functions to the experimental (002) peak shifts. (a) mono-exponential (1-exp Fitting, red curve) and bi-exponential functions (2-exp Fitting, blue curve) are fitted to the measurements. Inset of (a) shows the fittings to the measurement at the early time range from 0 to 100 ps. (b) The fit residuals for both mono- and bi-exponential functions. Inset of (b) shows the fit residuals at the early time range from 0 to 100 ps.

The experimental data of (002) peak shifts in Figure. S1(a) cover a large time span from -10 to 700 ps, displaying a steep rising of  $\Delta d/d$  at the early times from 0 to 10 ps and then a slowly increasing of  $\Delta d/d$  up to the time frames of 400 ps. This observed lattice change behavior suggests a bi-exponential function, rather than a mono-exponential function, is needed to better describe the temporal evolution of lattice triggered by pulsed laser. Indeed, when fitting with a mono-exponential function  $\Delta d(t)/d = 0.0081-0.0061 \cdot \exp(-t/112)$  (red curve, Figure. S1(a)), the fitting residual (red curve + symbol, Figure. S1(b)) clearly shows a large deviation to the measured, especially at the early time 0 to 10 ps. By contrast, fitting with a biexponential function (see fitting parameters in main text, blue curve in Figure.S1(a)) yields an overall satisfactory match to the measurement. Hence, the (002) lattice dynamics involves a fast and a slow component, as revealed by the fitting with a biexponential function.



Figure S2. Temporal evolution of interlayer space along the radial direction with pump fluence of 25 and 50 mJ/cm<sup>2</sup>. The relatively slow lattice dynamics is discussed in the data of 50mJ/cm<sup>2</sup> in the main text. The time constants specific to the slow process depend on the laser-pulse fluence, which reflect the fact that the Auger recombination rate depends on the carrier density in semiconducting materials.



Figure S3. Electron energy loss spectroscopy (EELS) measurement of BNNTs. The red curve shows the raw data, and the inset shows the deconvoluted and smoothed results. The band gap of BNNTs is measured to be  $\sim 5.8 \pm 0.3$  eV, as taken by the inflection point of the first derivative of the EEL spectrum.