Supplemental Material for "*Ab Initio* Simulation of Laser-Induced Water Decomposition close to Carbon Nanotubes"

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S.1. ESTIMATION OF POSSIBLE OPTICAL E-FIELD STRENGTH

The power-conversion relationship between the optical E-field and the power density is given as 1 V/Å , which corresponds to 1.327×10^{13} W/cm^2. (Note that the square of the E-field is proportional to the power per section.)

A recently commercialized femtosecond laser has a power of 1 W with a repetition rate of 1 GH$_Z$, according to the manufacturer's specifications. This corresponds to an energy output of 1 J per second and 1 nJ per single pulse. Although laser powers exceeding 10 W have been achieved in some laboratories, we restrict our consideration to commercially available lasers. By using an additional amplifying device, a power gain of a factor 10^5 with a reduced frequency of 10 kH$_Z$ is possible, resulting in an energy of 1×10^{-4} J per pulse. For a pulse width with a full width at half-maximum (FWHM) of 10 fs (10^{-14} second), the corresponding power is 1×10^{10} W per pulse.

The laser beam can be focused using a commercially available device based on silver mirrors. For practical purposes, the current feasible focusing size is approximately 30 μm $\times$ 30 μm area for a laser pulse with an FWHM of 10 fs. Since 1 μm is 10^{-4} cm, this corresponds to an area of 9×10^{-6} cm^2.

Therefore, if a laser beam with a power of 1×10^{10} W is focused on the above area, the power per section is $1/9 \times 10^{16} = 1.1 \times 10^{15}$ W/cm^2, which corresponds to $\sqrt{(1.1 \times 10^{15})/(1.327 \times 10^{13})} = \sqrt{(110/1.327)} = 9.1$ V/Å of optical E-field strength according to the aforementioned relationship between the optical E-field and the power per area. Therefore, the range of E-fields examined in this study is accessible using currently commercially available lasers.

S.2. DETAILS OF MD CALCULATIONS

We performed RT-TDDFT-MD simulations to examine the H_2O decomposition for the case with 15 H_2O molecules around an (8,0) CNT. The stable atomic coordinates were determined by geometry optimization from 15 randomly placed/oriented H_2O molecules around the (8,0) CNT. Several geometries should be obtained as metastable structures, and testing all of them may give a statistically correct answer. However, we observed no quantitative difference in the threshold intensity upon testing several initial geometries as reported in Ref. [8] of the main text. In the present relaxed geometry, as shown in Fig. 1 of the main text, the distances between the H_2O molecules and the CNT wall were kept within a typical range for van der Waals interactions (3.0∼3.4 Å). At the same time, the locations of the H_2O molecules along the tube axis and their molecular orientations were dispersed. Therefore, a single MD trajectory with randomly distributed H_2O molecules provides a suitable approximation for understanding the trend of H_2O decomposition unless temperature effects are being considered.