Enhancement of laser-induced water decomposition by 2D sheets studied by first-principles simulations

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Photo-induced water decomposition with aid of photo-catalysis has become a hot topic since 1979 [1]. Photo-carrier generation and subsequent hole-transfer to water oxidation level and electron-transfer to reduction level protons are believed to play a main role [2]. Recently graphitic carbon nitride (gC_3N_4) sheets are found to assist water decomposition with UV and visible light [3,4] and usage of graphitic materials as photo-catalysis for water decomposition seems to be promising.

We here propose application of femtosecond laser, which has higher photon-flux than ordinary light thus can decompose water with higher yield. In this study, we have performed first-principles molecular dynamics for studying water decomposition under strong laser field with full-width of half maximum 10 fs and basic wavelength of 800 nm. In performing the simulation, we applied real-time propagation time-dependent density functional theory combined with Ehrenfest type molecular dynamics under presence of the laser field by using a code FPSEID [5] including optical field [6].

The threshold of laser-field intensity for decomposing an isolated water molecule was computed as 9 V/Å by using local density approximation and by using the generalized gradient approximation. On the other hand, this value is reduced less than 7 V/Å when water molecule is located above graphitic sheets, *i. e.* graphene, hexagonal boron nitride, and gC_3N_4. Fig. 1 shows snapshots of the results of water decomposition above a graphene sheet. We will discuss possible mechanisms of reducing threshold intensity and further applications.

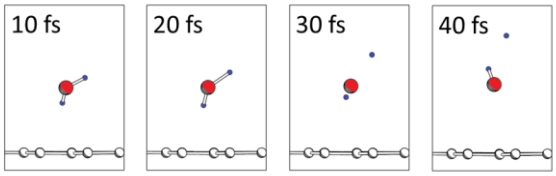

Fig.1 Snapshots of first-principles molecular dynamics of water decomposition above a graphene sheet

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