Supplementary Information

Non-adiabatic excited-state time-dependent GW (TDGW) molecular dynamics simulation of nickel-atom aided photolysis of methane to produce a hydrogen molecule

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We demonstrated that the H ejection away from the Ni side facilitates the formation of a hydrogen molecule, i.e., $CH_4 + Ni \xrightarrow{\hbar\omega} CH_2-Ni + H_2$, with the quasiparticle level corresponding to it having the energy close to the negative ionization potential of an isolated H_2 molecule. We additionally perform a contour analysis of the charge densities shown in Figs. 3(a32.6) and (d32.6) in the main text to obtain quantitative estimates of charge populations around the H_2 fragment [Fig. S1(a)]. We compare this with the charge contour of an isolated H_2 molecule obtained from a single-point LDA calculation [Fig. S1(b)]. We find an extremely good agreement between the TDGW-MD and the LDA results indicating that a H_2 molecule is produced for the H away from Ni trajectory within a short timescale.
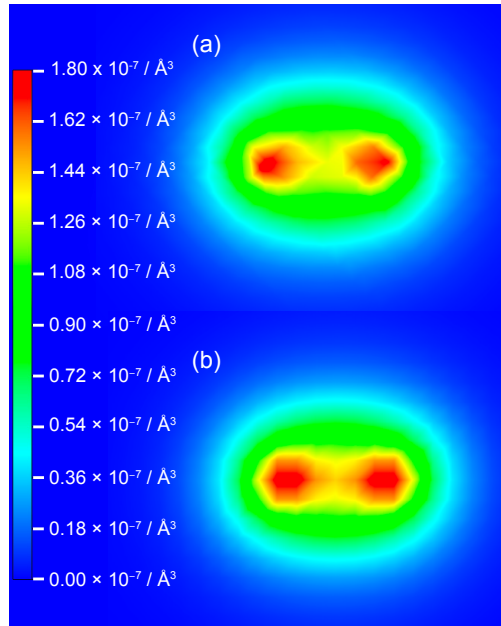

Figure S1: Contour plots of the H_2 charge density obtained from (a) the on-the-fly TDGW-MD result for the CH_4-Ni system at $t = 32.6$ fs [Fig. 3(d32.6) in the main text] and (b) the self-consistent LDA result for the purely isolated H_2 system.