Supporting Information

First-Principles Demonstration of Non-adiabatic Thouless Pumping of Electrons in A Molecular System

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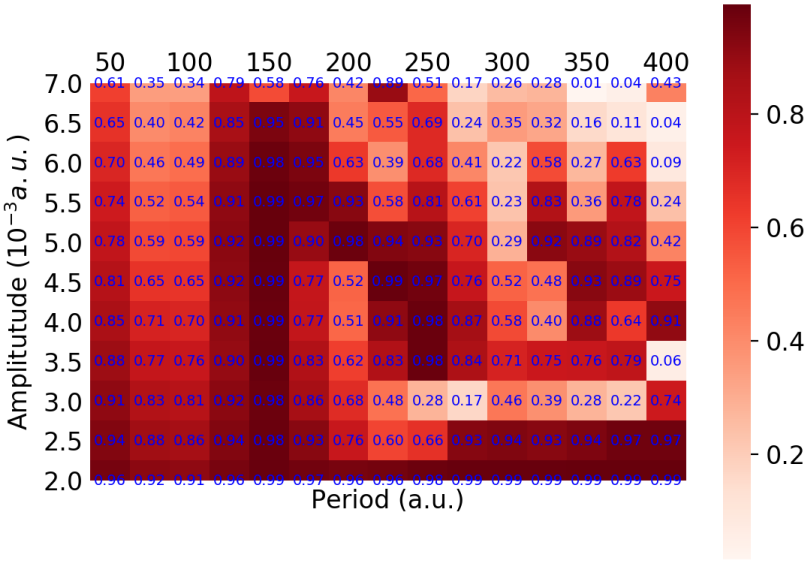


Figure S1. Determinant of the overlap matrix between the initial and final time-dependent states over a single driving cycle as a function of the driving field period and amplitude, sampled at uniform intervals of 25 a.u. and 0.5×10^{-3} a.u., respectively.

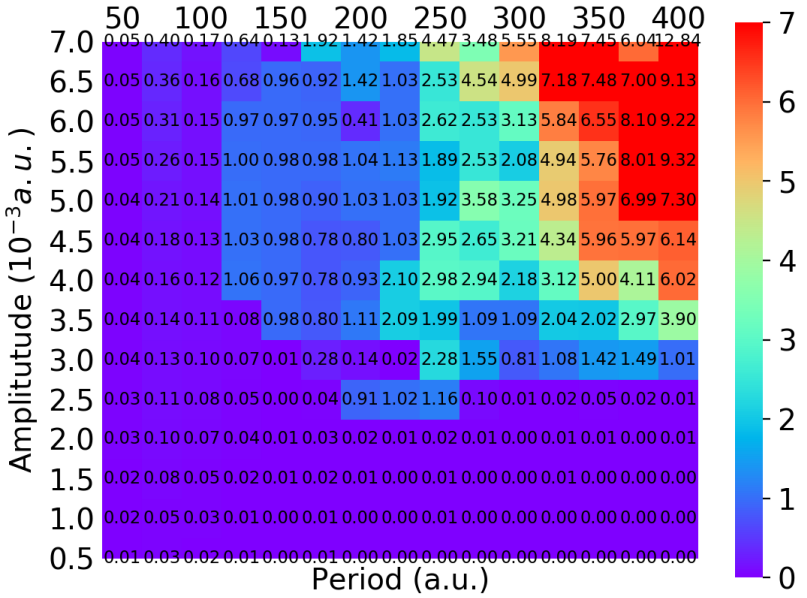


Figure S2. Integrated current over one driving cycle per C-C monomer unit, as a function of the driving field period and amplitude, sampled at uniform intervals of 25 a.u. and 0.5×10^{-3} a.u., respectively.

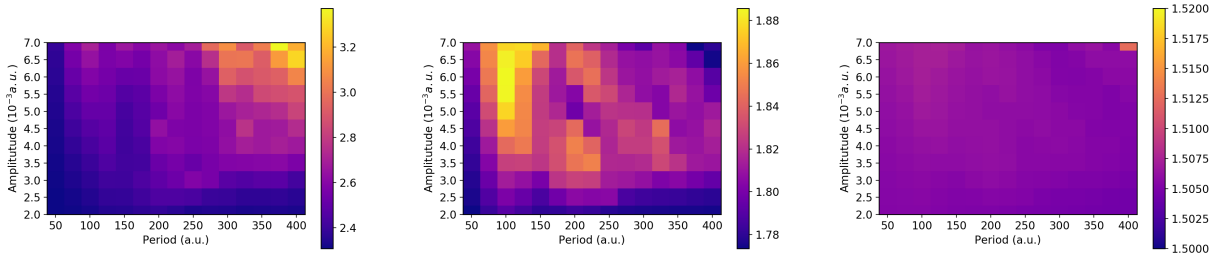


Figure S3. Averaged spreads (a.u.^{2}) of time-dependent MLWFs for C-C double bond (Left), C-C single bond (Center), and C-H bond (Right) as a function of the driving field amplitude and period.

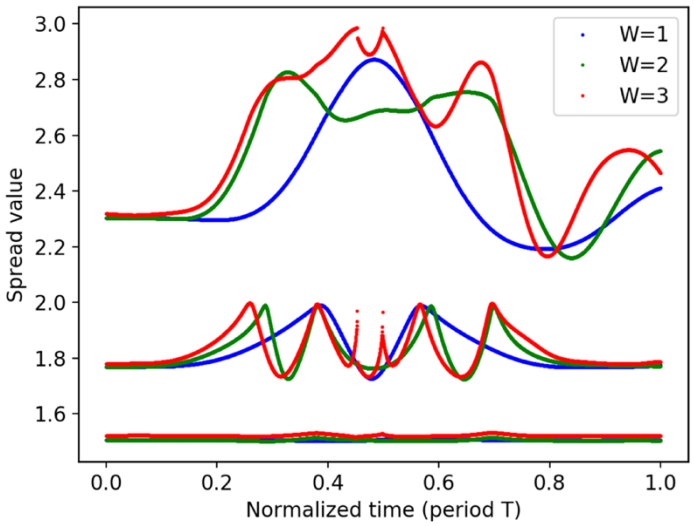


Figure S4: Averaged spreads of the time-dependent MLWFs for W=1 (period=150 a.u./amplitude=4.0×10^{-3} a.u.), W=2 (period=250a.u./amplitude=3.5×10^{-3} a.u.), and W=3 (period=250 a.u./amplitude=4.0×10^{-3} a.u.), during a single driving cycle. The values are averaged for the C-C double bond MLWFs, C-C single bond MLWFs, and C-H bond MLWFs separately from the top to the bottom.

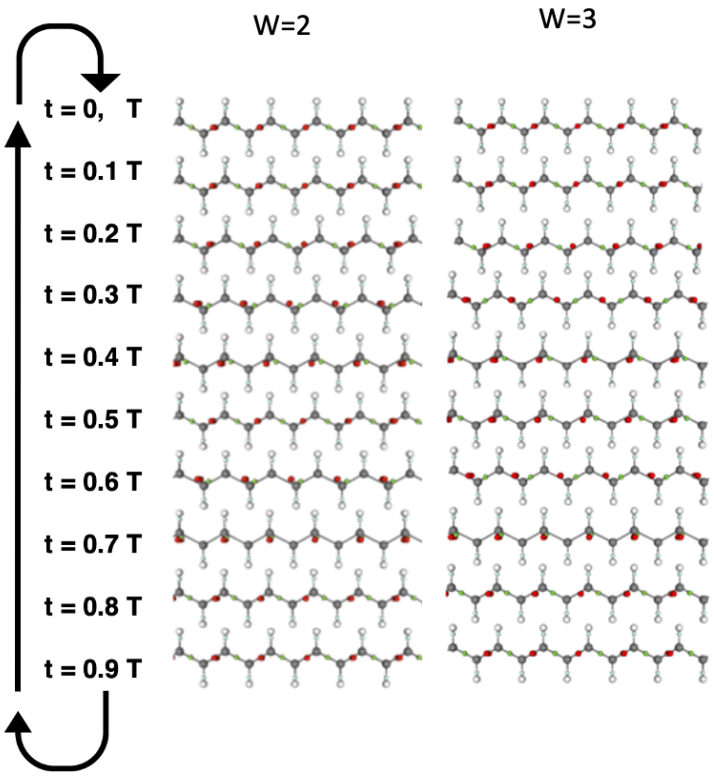
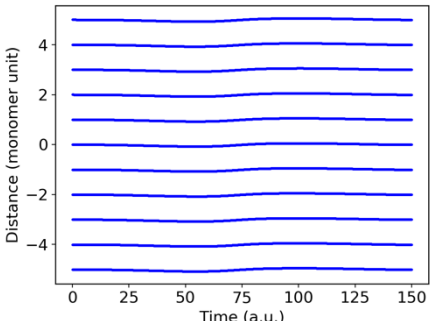
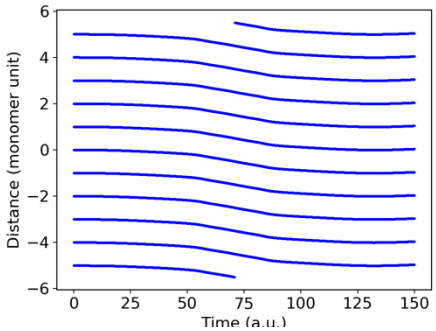


Figure S5: Snapshots of the geometric centers of the time-dependent MLWFs (Wannier centers) at different instances of time in a single driving cycle for W=2 (period=250 a.u./ amplitude=3.5×10^{-3} a.u.), and W=3 (period=250 a.u./amplitude=4.0×10^{-3} a.u.).

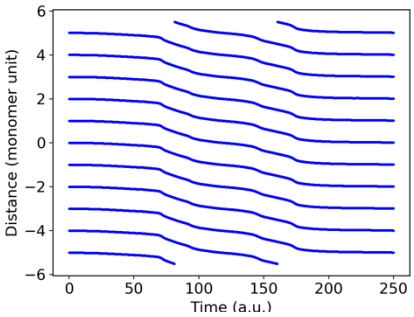
(a) W=0 : 150 a.u. / 0.0020 a.u.



(b) W=1 : 150 a.u. / 0.0040 a.u.



(c) W=2 : 250 a.u. / 0.0035 a.u.



(d) W=3 : 250 a.u. / 0.0040 a.u.

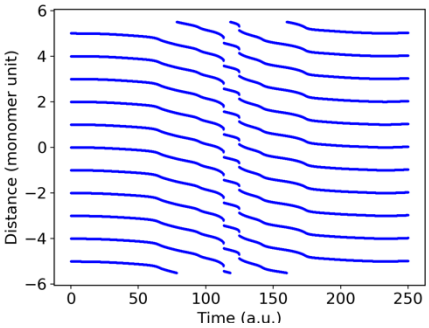


Figure S6: The movement of the Wannier centers in a single driving cycle for "C=C double bond" MLWFs for (a) W=0 (period=150 a.u./amplitude=2.0×10^{-3} a.u.), (b) W=1 (period=150 a.u./amplitude=4.0×10^{-3} a.u.), (c) W=2 (period=250 a.u./amplitude=3.5×10^{-3} a.u.), and (d) W=3 (period=250 a.u./amplitude=4.0×10^{-3} a.u.).

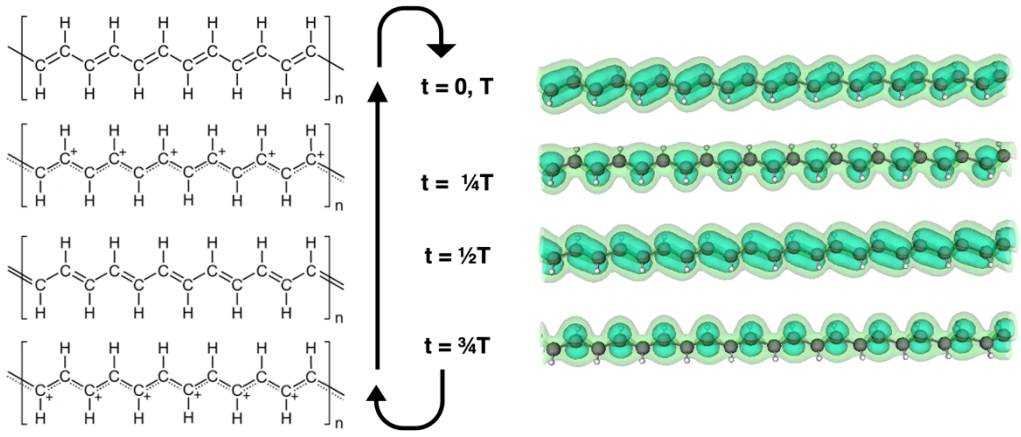


Figure S7: Schematics of corresponding resonance structures for the dominant dynamical transition orbital in a single driving cycle for W=1 (period=150 a.u./amplitude=4.0×10^{-3} a.u.).

Molecular geometry of the *trans*-polyacetylene

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Supercell : 51.3232x15x15 Bohr.
atom C1 carbon -24.4331 -0.617224 0
atom C2 carbon -19.7674 -0.617224 0
atom C3 carbon -15.1016 -0.617224 0
atom C4 carbon -10.4359 -0.617224 0
atom C5 carbon -5.77014 -0.617224 0
atom C6 carbon -1.1044 -0.617224 0
atom C7 carbon 3.56135 -0.617224 0
atom C8 carbon 8.22709 -0.617224 0
atom C9 carbon 12.8928 -0.617224 0
atom C10 carbon 17.5586 -0.617224 0
atom C11 carbon 22.2243 -0.617224 0
atom C12 carbon -22.2243 0.617186 0
atom C13 carbon -17.5586 0.617186 0
atom C14 carbon -12.8928 0.617186 0
atom C15 carbon -8.22709 0.617186 0
atom C16 carbon -3.56135 0.617186 0
atom C17 carbon 1.1044 0.617186 0
atom C18 carbon 5.77014 0.617186 0
atom C19 carbon 10.4359 0.617186 0
atom C20 carbon 15.1016 0.617186 0
atom C21 carbon 19.7674 0.617186 0
atom C22 carbon 24.4331 0.617186 0
atom H1 hydrogen -24.4331 -2.65246 0
atom H2 hydrogen -19.7674 -2.65246 0
atom H3 hydrogen -15.1016 -2.65246 0
atom H4 hydrogen -10.4359 -2.65246 0
atom H5 hydrogen -5.77014 -2.65246 0
atom H6 hydrogen -1.1044 -2.65246 0
atom H7 hydrogen 3.56135 -2.65246 0
atom H8 hydrogen 8.22709 -2.65246 0
atom H9 hydrogen 12.8928 -2.65246 0
atom H10 hydrogen 17.5586 -2.65246 0
atom H11 hydrogen 22.2243 -2.65246 0
atom H12 hydrogen -22.2243 2.65243 0
atom H13 hydrogen -17.5586 2.65243 0
atom H14 hydrogen -12.8928 2.65243 0
atom H15 hydrogen -8.22709 2.65243 0
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atom H16 hydrogen -3.56135 2.65243 0
atom H17 hydrogen 1.1044 2.65243 0
atom H18 hydrogen 5.77014 2.65243 0
atom H19 hydrogen 10.4359 2.65243 0
atom H20 hydrogen 15.1016 2.65243 0
atom H21 hydrogen 19.7674 2.65243 0
atom H22 hydrogen 24.4331 2.65243 0
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