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Spatial and Temporal Control of Populations, Branching ratios and Electronic Coherences in LiH by a Single One Cycle IR Pulse

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ABSTRACT

Dynamical computations demonstrate considerable selectivity over the fragmentation channels of the LiH molecule via the polarization and the carrier envelope phase (CEP) of a single ultrashort one cycle strong IR pulse. For aligned molecules, control of the CEP allows building non stationary coherent electronic wave packets of contrasting ionic character, either $\mathrm{Li}^{\delta+}\mathrm{H}^{\delta-}$ or $\mathrm{Li}^{\delta-}\mathrm{H}^{\delta+}$. The complementary coherences are maintained all the way to dissociation. The direction of the electric field at its maximum points either towards the Li or towards the H atom, which selectively steers the nuclear dynamics to specific dissociation products.

I. INTRODUCTION

Due to their broad bandwidths, ultrashort attopulses [1-5] excite molecules to coherent non equilibrium electronic wavepackets on a time scale shorter than the response of the nuclei. As a result, the non equilibrium electron density is a coherent superposition of several electronic states and can beat between different parts of the molecule. For modular molecules this beating causes migration of charge between the different modules. Tailoring the electronic density in space and time via the parameters of the pulse [6], and in particular by varying the value of the Carrier Envelope Phase (CEP) [7] for tailoring the electric field was suggested early on as means for controlling reactivity by steering the nuclear motion towards desired products. This control of the asymptotic channels is possible if the selectivity of the excitation is maintained all the way to the products. In this paper, we demonstrate by quantum dynamical coupled pulse-electron-nuclei computations that the coherence imprinted by the ultrashort pulse is preserved all the way to the fragmentation for aligned LiH molecules excited by a single ultrashort one cycle strong IR pulse.

Electron localization has been probed in diatomic cations [8-12] and larger systems [13-16] using pump-probe schemes involving atto and fs optical pulses. In these experiments the molecule is locally ionized by the pump which creates a non stationary state in the cation. The cationic dynamics is then probed by monitoring the fragments by photoexcitation or ionization. Several experimental schemes rely on the CEP phase value of a CEP stable few cycle pulse [17] for tailoring the electric field profile of the pump pulse and controlling electron localization and dissociation. [8, 18-25] Photoinduced coherent electronic dynamics can also be probed using high harmonic generation [13, 26-

28], transient absorption spectroscopy [29-31], time-resolved molecular-frame photoelectron-angular-distribution [32] and attoclock measurements [33, 34].

Here, we couple the non equilibrium electronic dynamics induced by the interaction with a single, strong, ultrashort optical pulse [6, 35-40] to nuclear motion including non adiabatic coupling.[8, 41-52] We show that switching the CEP value of the pump pulse from 0 to π controls the fragmentation yields of the different dissociation asymptotes. Nearly one cycle CEP stable IR pulses can be experimentally synthesized for a wide range of IR wavelengths. [18, 20, 53-57] For the polar LiH molecule, changing the value of the CEP of the nearly one cycle pulse allows selecting excited states of opposite polarities, depending on whether the electric field at its maximum points to the Li or to the H atoms.[7, 58] As a result, the non stationary electronic wave packet built at the end of the pulse has a different ionic character, either $\mathrm{Li}^{\delta+}\mathrm{H}^{\delta-}$ or $\mathrm{Li}^{\delta-}\mathrm{H}^{\delta+}$, and this steers the nuclear dynamics to different dissociation asymptotes. We further show that changing the value of the CEP by π prepares at the end of the pulse non stationary states that are not only of complementary polarity but also of opposite coherence. We demonstrate that this coherence can be spectroscopically monitored by transient absorption that probes the phase of electronic coherences and of the charge migration.

II. NON EQUILIBRIUM ELECTRON-NUCLEI DYNAMICS

LiH is a heteronuclear diatomic molecule with a dense manifold of low lying excited electronic states of different polarities in the UV range that fragment into chemically different species. LiH is therefore an ideal prototype for investigating charge migration and dissociation control.

LiH electronic states have been extensively studied by us [7, 52, 58-60] and others. [61-64] The electronic states were computed at MRCI level using the quantum chemistry code MOLPRO [65], see Supplemental Materials (SM) for details. Since we report on pulses polarized along the molecular axis, we restrict the dynamics to the Σ manifold of states. The computed potential energy curves of all the excited Σ states have shallow minima, as can be seen from Fig. 1a. The lowest excited Σ and Π states are about 3.5 eV-4eV above the ground state, (GS), and can be accessed by strong IR or UV pulses. The ground state and sequence of lowest excited Σ states exhibit alternating polarities. An excess of negative charge is localized on the H atom in the GS and in the second excited Σ state, Σ_2, which have a chemical structure $\mathrm{Li}^{\delta+}\mathrm{H}^{\delta-}$. In contrast, the first excited state, Σ_1, and the third one, Σ_3, are of opposite polarity, $\mathrm{Li}^{\delta-}\mathrm{H}^{\delta+}$. The different Σ excited states dissociate to different asymptotes. The H atom is in its GS, H(1s), and the Li atom in different excited states : Li(2s) for the GS and Li(2p), Li(3s) and Li(3p) for Σ_1, Σ_2 and Σ_3 respectively. The fourth excited state, Σ_4 , dissociates to ionic products, $\mathrm{Li}^{+} + \mathrm{H}^{-}$. The alternating polarity of the sequence of states implies that the Stark shifts of their potentials will be in opposite directions and this is a key to the understanding of the control and the selectivity that we report. Such a control cannot be obtained in the perturbative, one photon limit. Opposite Stark shifts of the potential energy curves of electronic states of different polarities as well as multiphoton processes are essential. This mechanism is different from the one reported in [23, 24] that involves inelastic recollisions of the outgoing electron with the cation.

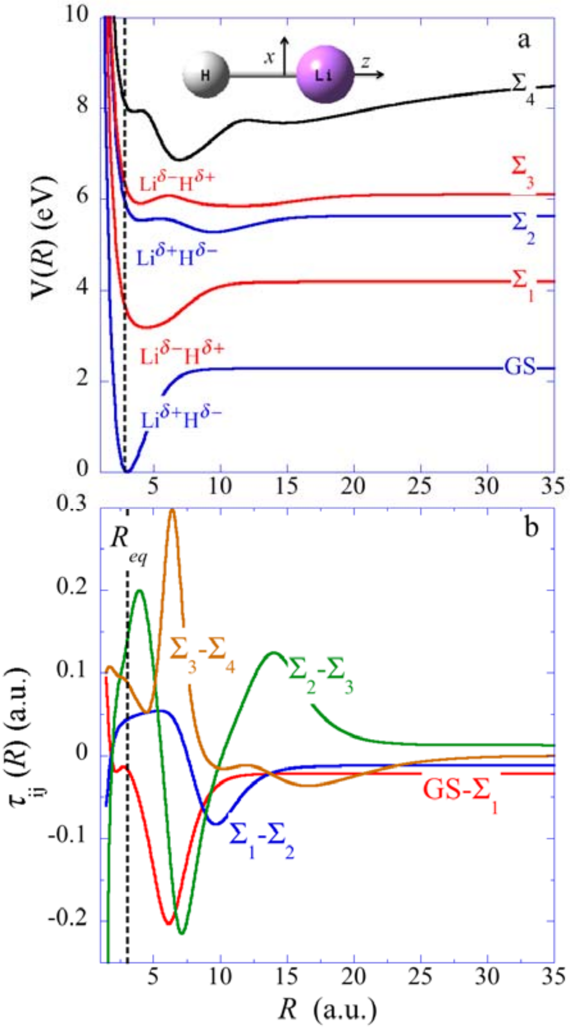

Figure 1. a) Potential energy curves (in eV) of the sequence of five consecutive Σ states of LiH computed at the MRCI level as a function of the internuclear distance R, in a.u. . The alternating polarity of the electronic states in the Franck-Condon region is indicated. b) NAC curves, $\tau_{ij}\left(R\right)$, in the Σ manifold. The computed value of R_{eq} (3.08 a.u., 1.63 Å) is marked by a dashed line. See SM for details on the quantum chemistry computations.

The non adiabatic couplings (NAC), $\tau_{ij}\left(R\right)$, between the different adiabatic states are plotted in Fig. 1b for selected pairs of Σ states, *i* and *j*. Only the pair of $\Sigma_{\square}$ and $\Sigma_{\square}$ states, which are close in energy for the whole range of internuclear distances, is significantly affected by non adiabatic coupling. Other NAC matrix elements are more localized at smaller internuclear distances.

The computed permanent dipole curves and transition dipole moments along the bond direction are plotted in Figs. 2a and 2b respectively. About the equilibrium position (R_{eq}=3.08 a.u.), the transition dipoles reflect the polarity of the different Σ states. The equilibrium dipole moment of the GS is large enough (+ 2.24 a.u., see Fig. 2a) that LiH could be spatially oriented experimentally.[66-68] The molecular frame orientation is shown as an inset in Fig. 1a where the bond is along the *z* axis with the Li atom in the +*z* direction.

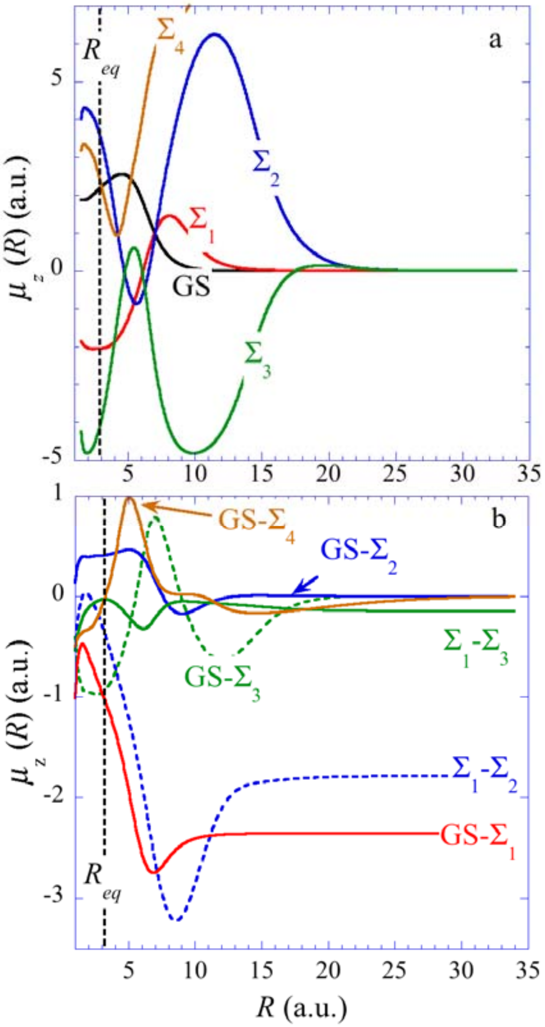

Figure 2: a) permanent dipole curves of the five states of the manifold, b) transition dipole curves for selected pairs of states. The value of the equilibrium distance in the ground state, R_{eq} is indicated by a dashed line.

Using pump pulses of different polarizations with respect to the molecular axis one can create non stationary electronic densities that rotate perpendicular or parallel to the LiH bond. [69] In this study we report on the dynamics induced by pump pulses polarized along the molecular axis. As a hydride LiH has a rather long rotational period so we take the molecule to be non rotating. The quantum nuclear dynamics computations were carried out by solving the time-dependent Schroedinger equation (TDSE) on a grid for the nuclear motion. [70-73] The states coupled are the manifold of the 5 lowest bound Σ states □□ plotted in Fig.1a and do not include the high Rydberg states that converge to the continuum (IP is 7.8 eV). The nuclear Hamiltonian includes the coupling of the molecule to the time-dependent electric field of the pump and the probe pulses in the dipole approximation and the non adiabatic coupling (NAC) between the Σ states. Since the pulse is an essentially one-cycle IR pulse that photoexcites the low lying Σ states, the probability that the molecule photoionizes through a multiphoton process during the pulse is low and not included in the dynamical simulations. The matrix form of the Hamiltonian on the grid in the basis of the adiabatic electronic states is given by:

$$\begin{aligned} H_{ig,jg'}(t) = {} & \left(-\frac{\hbar^2}{2\mu}\nabla_R^2\right)_{g,g'} \delta_{ij} + V_{ig,jg'}\delta_{ij}\delta_{gg'} - \mathbf{E}(t)\left(\boldsymbol{\mu}^{elec}_{ig,jg'} + \boldsymbol{\mu}^{nucl}_{gg'}\delta_{ij}\right)\delta_{gg'} \\ & -\frac{\hbar^2}{2\mu}\left(2\tau_{ig,jg}\left(\nabla_R\right)_{gg'} + \sum_l \tau_{ig,lg'}\tau_{lg',jg}\delta_{gg'} + g_{ig,jg'}\delta_{gg'}\right) \end{aligned} \tag{1}$$

In Eq. (1), *i* and *j* are the index of the adiabatic electronic states and *g* and *g'* are indices of grid points. μ is the reduced mass of LiH, $V_i\left(R\right)$'s are the potential energy curves plotted in Fig. 1a, $\boldsymbol{\mu}\left(R\right) = \boldsymbol{\mu}^{elec}\left(R\right) + \boldsymbol{\mu}^{nucl}\left(R\right)$ is the total molecular dipole moment vector, see Eq. (2) below. **E**(*t*) is the electric field vector of the pump and the probe pulses and the last terms in Eq. (1) are the non adiabatic coupling matrix elements with $\tau_{ij} = \left\langle \psi_i\left(r;R\right)\middle|\hat{\nabla}_R\middle|\Psi_j\left(r;R\right)\right\rangle$ (see Fig. 1b) and $g_{ij}\left(R\right) = \nabla_R\left(\tau_{ij}\left(R\right)\right)$.

In the grid representation for the nuclear coordinate, *R*, all the operators in Eq. (1) are local except the nuclear kinetic energy and the nuclear momentum dependent term of the non adiabatic coupling terms, $\tau_{ij}\left(R\right)$. We define the Gaussian electric field time profile, $\mathbf{E}_p$(t), of each pulse from the vector potential, $\mathbf{A}_p$(t), $d\mathbf{E}_p\left(t\right)/dt = \left(-1/c\right)\partial\mathbf{A}_p\left(t\right)/\partial t$, with $\mathbf{A}_p = \frac{-\mathbf{E}_p c}{\omega_p} f\left(t\right)\sin\left(\omega_p t+\phi_p\right)$ where ϕ_p is the CEP, c the speed of light and ω_p the carrier frequency . This ensures that there is no DC or low frequency component in the Fourier transform of $\mathbf{E}$(t). The vector potential, $\mathbf{A}_p\left(t\right)$, for each pulse, p, has a Gaussian envelope, $f\left(t\right)=\exp\left(\left(t-t_p\right)^2/\sigma_p{}^2\right)$ centered at the time t_p. The polarization of the pulse is along the bond axis, $\mathbf{E}_p = \left|E_z^p\right|\mathbf{e}_z$. The amplitudes $c_{ig}\left(t\right)$ of the wave function in the electronic state i at the grid point g, $\Psi\left(t\right)=\Sigma_{i,g}c_{ig}(t)\left|i\right\rangle$, are computed by numerically integrating the TDSE, $i\hbar\, dc_{ig}\left(t\right)/dt=\sum_{jg'} H_{ig,jg'}\left(t\right)c_{jg'}\left(t\right)$, for the Hamiltonian of Eq. (1).

III. SELECTIVE FRAGMENTATION BY A SINGLE CEP CONTROLLED ONE CYCLE IR PULSE

The pump is a CEP controlled essentially one cycle pulse polarized along the molecular axis with a carrier frequency, ω_p, of 1.55 eV (800 nm). The pulse we use is strong ($5.61 \cdot 10^{13}$ W/cm^2, $\left|E_z^p\right|$ = 0.04 a.u.) and its envelope is short (σ = 0.68 fs, FWHM= 1.13 fs) so as to span a single large maximum, which is necessary for the selectivity mechanism discussed here. There are also inevitably two secondary small maxima because we impose that the area of $\mathbf{E}(t)$ is equal to zero, so as not to have a component at zero frequency. There are 1.5 oscillations of the electric field within the short pulse envelope, oscillations that are faster than the period of the carrier wave (800 nm corresponds to a period of 2.66 fs). This is reflected in the Fourier transform of the pulse given in Figure S1, which is an asymmetric broad peak with a maximum at 2.27 eV. The essential feature of the pulse for the control mechanism by the value of the CEP is that it is strong, so that multiphoton excitation is allowed and that the envelope is such that the electric field exhibits a main maximum significantly higher than the secondary ones. A CEP ϕ_p equal to 0 corresponds to a maximum of the electric field in the +*z* direction, pointing to the Li atom, while $\phi_p = \pi$ corresponds to a maximum of the electric field pointing to the H atom, in the –*z* direction. We start the dynamical simulation from the ground vibrational state of the ground electronic state. The populations, $P_i(t) = \sum_g \left| c_{ig}(t) \right|^2$, in the different excited Σ states are plotted in Fig. 3a for the pulse with a CEP ϕ_p=0 and Fig. 3b for that with ϕ_p=π. The two panels of Fig. 3 show that changing only the value of the CEP of an essentially one cycle pulse leads to very different non equilibrium electronic densities, as reflected in the different values of the populations in the Σ manifold at the end of the pulse. For the strong pulses used in Fig.3, different values of the phase, ϕ_p, lead to different populations in the excited states. Pumping with a CEP = 0 one cycle pulse, Fig.3a, the highest population is in Σ_2 while for the CEP=π pump pulse, the highest population is in Σ_1 and Σ_3. The selectivity of the excitation can be qualitatively understood on the basis of the sign of Stark shift in the energies of the excited electronic states which will be opposite for states of different polarities. Excited states with a permanent dipole antiparallel to that of the GS, a dipole pointing to the H atom in the Franck Condon region, like Σ_1 and Σ_3, are preferentially accessed by the CEP=π pulse, while Σ_2, which is of opposite polarity, is preferentially populated by the CEP = 0 pulse. Changing the value of the CEP controls the fragmentation yields of the two states that dissociate along Σ_2 and Σ_3. Changing the CEP by π inverts the yield ratio between the two states. A value of the CEP=0 leads to a branching ratio of $Y_{\Sigma_2}/Y_{\Sigma_3} = 2.45$ while the pulse with a CEP = π leads to a branching ratio $Y_{\Sigma_2}/Y_{\Sigma_3} = 0.29$. The rates for these direct dissociations are in the picosecond range, 0.01 fs^{-1} for Σ_2 and 0.006 fs^{-1} for Σ_3.

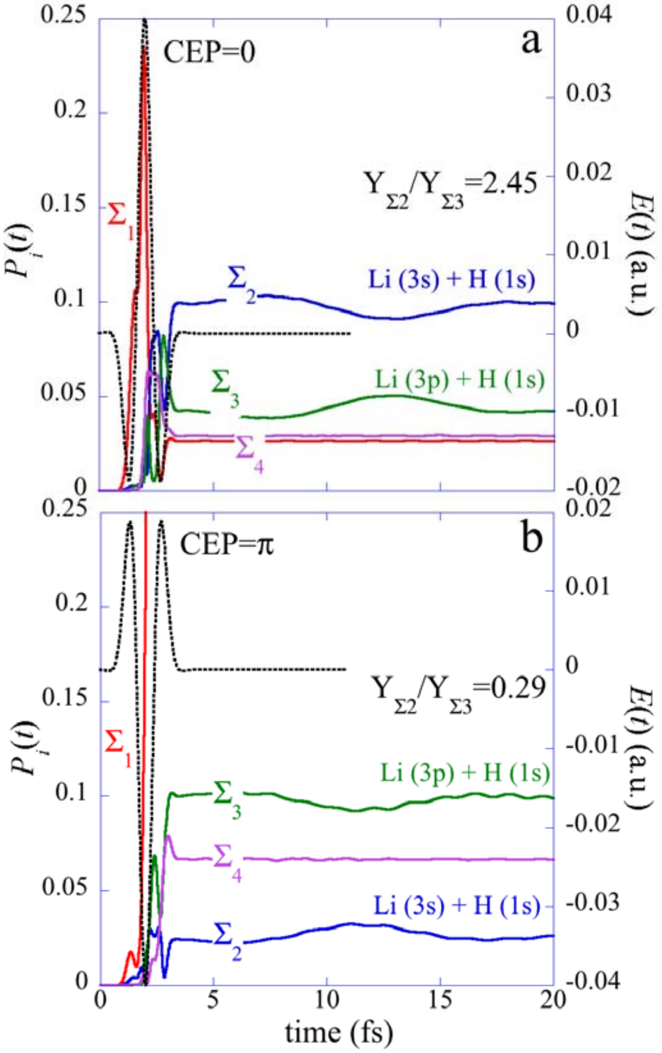

Fig. 3 Effect of the CEP ϕ_p on the populations of the different electronic states, $P_i(t)$, in the Σ manifold. a) CEP = 0. The highest population is in the Σ_2 state, asymptotically 9.72 % with 4.40 % in Σ_3. b) CEP = π, the maximum of population is in Σ_1 (48.7%, not shown) and the branching ratio of the populations in Σ_2 and Σ_3 is opposite (Asymptotically 2.65% in Σ_2 and 9.82% in Σ_3). The Σ_1 state does not dissociate for these excitation pulses. The small population in Σ_4 is about equal for either value of the CEP. The profile of the electric field of the pump pulse is shown in dotted line (scale on the right y axis). The small exchange of populations between the Σ_2 and Σ_3 states is due to their non adiabatic coupling. By about 20 fs the molecule is in the asymptotic range so that the dissociation is very direct.

As can be seen from Figure S2, which reports the *R* dependence of the coefficients, $c_{\Sigma_2 g}(t)$ and $c_{\Sigma_3 g}(t)$, computed at the end of the pulse (*t* = 4 fs) for the two values of the CEP, the value of the CEP is imprinted in the coefficients of the wave packet. The CEP = 0 pulse prepares a state that we approximate as $\left|\Psi_{\mathrm{CEP}=0}\left(R_g,t=4\mathrm{fs}\right)\right\rangle = a_{Rg}\left(t=4\mathrm{fs}\right)\left|\Sigma_2\left(R_g\right)\right\rangle + b_{Rg}\left(t=4\mathrm{fs}\right)\left|\Sigma_3\left(R_g\right)\right\rangle$ at the end of the pulse. In the state prepared by the CEP = π pulse, the absolute values of the amplitudes on the two electronic states are almost switched because of the polarity control and there is a phase shift of about π, $\left|\Psi_{\mathrm{CEP}=\pi}\left(R_g,t=4\mathrm{fs}\right)\right\rangle \approx b_{Rg}\left(t=4\mathrm{fs}\right)\left|\Sigma_2\left(R_g\right)\right\rangle - a_{Rg}\left(t=4\mathrm{fs}\right)\left|\Sigma_3\left(R_g\right)\right\rangle$. The electronic coherence between the states Σ_2 and Σ_3 at a given value R_g on the grid is given by

$\rho_{\Sigma_2-\Sigma_3}\left(R_g,t\right)=c^*_{\Sigma_2 g}\left(t\right)c_{\Sigma_3 g}\left(t\right)$. Using the simplified forms above for the wave function, we get $\rho^{\text{CEP=0}}_{\Sigma_2-\Sigma_3}\left(R_g,t\right)=a^*_g\left(t\right)b_g\left(t\right)$ for the CEP = 0 pulse, while for the CEP=π pulse, the sign is opposite $\rho^{\text{CEP=}\pi}_{\Sigma_2-\Sigma_3}\left(R_g,t\right)\approx -a_g\left(t\right)b^*_g\left(t\right)$. The two coherences beat with a frequency given by the energy difference, $\Delta E_{\Sigma_2-\Sigma_3}(R)=V_{\Sigma_3}(R)-V_{\Sigma_2}(R)\approx 0.5$ eV (period = 8.2 fs) between the field free state electronic energies and they have opposite sign for their real parts. We show in Figure 4 heatmaps of the real part of the $\rho_{\Sigma_2-\Sigma_3}\left(R_g,t\right)$ as a function of time (ordinate) and of its localization in *R* (abscissa), computed for electron-nuclei dynamics induced by the two pulses of opposite CEP shown in Figure 3. Figure 4 shows that the difference in the phase of the electronic coherence prepared by the two pulses is preserved in the presence of nuclear motion both in the part of the wave packet that remains bound and in the part that dissociates all the way to the dissociation asymptote. The heatmaps of the imaginary parts are shown in Figure S3.

The difference in the wave functions prepared by the two pulses explains why in Figure 3, the small oscillations in the populations of Σ_2 and Σ_3 that are due to the non adiabatic coupling have opposite phases.

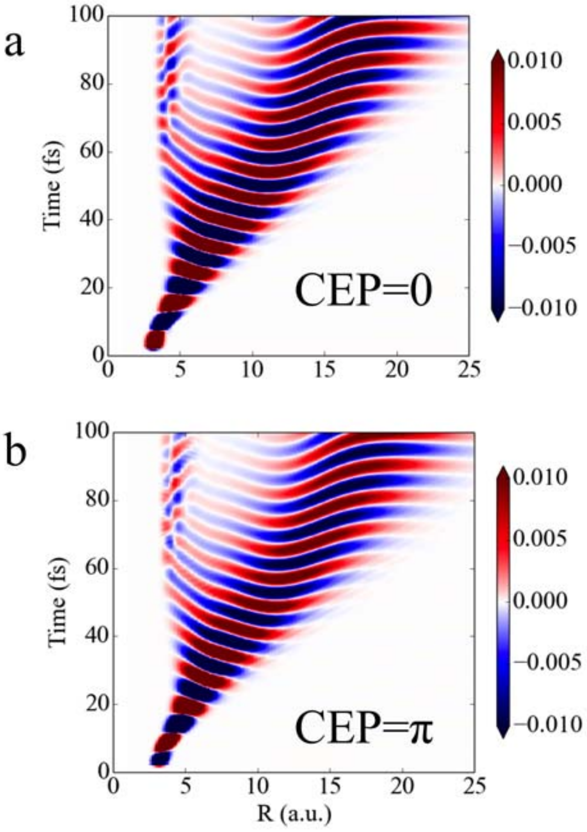

Fig. 4: Heatmaps of the real part of the electronic coherence, $\mathrm{Re}\left[\rho_{\Sigma_2-\Sigma_3}\left(R_g,t\right)\right]$, between the states Σ_2 and Σ_3. Plotted as a function of the Li-H distance R (x axis) and of time (y axis), for a pulse with a CEP = 0 (upper panel) and π (lower panel), with the same parameters as in figure 3.

IV. PROBING OF THE ELETRONIC COHERENCE

The non-stationary character of the coherent state prepared by the pulse means that the LiH molecule will have a time-dependent dipole, $\mu(t)$, [51, 52] which includes both nuclear and electronic contributions:

$$\begin{aligned}
\boldsymbol{\mu}_{tot}(t) &= \sum_{g} \rho\left(R_g,t\right)\boldsymbol{\mu}_{tot}\left(R_g\right) \\
&= \sum_{ig,jg} c_{ig}^{*}(t)\, c_{jg}(t)\, \mu_{i,j}^{ele}\left(R_g\right) + \sum_{ig} \left|c_{ig}(t)\right|^2 \boldsymbol{\mu}^{nucl}\left(R_g\right) \qquad (2) \\
&= \sum_{ig} \left|c_{ig}(t)\right|^2 \left[\boldsymbol{\mu}_{ii}^{elec}\left(R_g\right) + \boldsymbol{\mu}^{nucl}\left(R_g\right)\right] + \sum_{g} 2\,\mathrm{Re}\left(\sum_{i>j} c_{ig}^{*}(t)\, c_{jg}(t)\, \boldsymbol{\mu}_{i,j}^{ele}\left(R_g\right)\right)
\end{aligned}$$

and which has both a diagonal and a coherent, transition dipole, contribution. Figure 5 shows the time-dependent transition dipole component, that is, the part of the dipole that is non diagonal in the electronic states, $\mu_{trans}(t) = \sum_{g} 2\,\mathrm{Re}\left(\sum_{i>j} c_{ig}^{*}(t)\, c_{jg}(t)\, \boldsymbol{\mu}_{i,j}^{ele}\left(R_g\right)\right)$, computed for the two pulses of opposite CEP used in Figure 3. $\mu_{trans}(t)$ clearly exhibits oscillations with a period of ≈ 8 fs, which corresponds to the period of the $\Sigma_2 - \Sigma_3$ coherence. Moreover these oscillations have opposite phase for the two pump pulses, which reflects the π difference in the values of their CEP. In addition, the rapid oscillations of $\mu_{trans}(t)$, of the order of 1fs, reflect the transition frequencies between the excited states and the GS. The slower modulation is due to nuclear motion in the bound electronic states. The vibrational period of the GS is 20fs and that of the first excited state Σ_1 equal to 80 fs.[52]

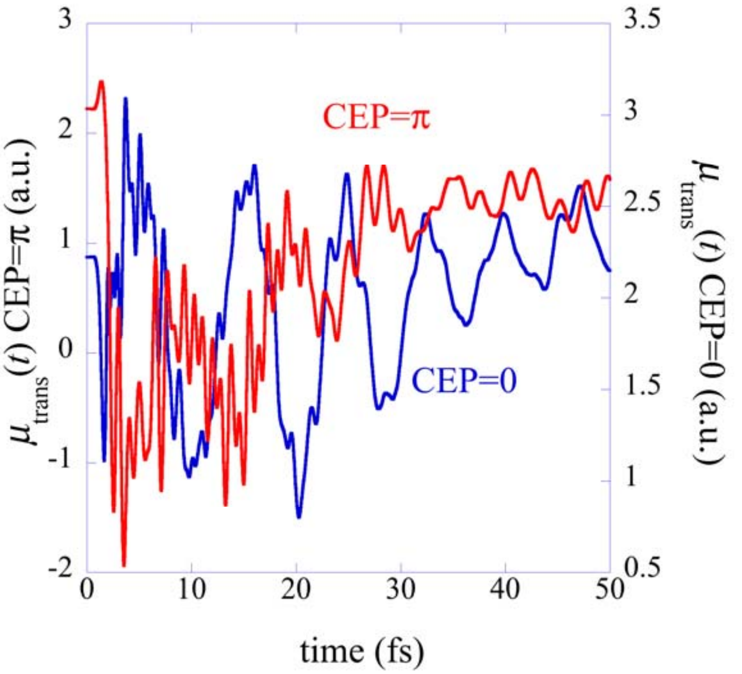

Figure 5: Computed transition dipole, $\mu_{trans}(t)$, for the two pulses. Note that the oscillations with a period $\approx$ 8 fs, which correspond to the $\Sigma_2 - \Sigma_3$ coherence, have opposite phase for the two pump pulses.

The non stationary electronic polarization, a signature of the excitation of a coherent state, can be probed by a second pulse.[31] We computed the linear response function, $S(\omega)$, of the time dependent dipole prepared by the pump pulse as in refs. [31, 74, 75], $S(\omega) = -2\,\mathrm{Im}\left[\boldsymbol{\mu}(\omega)\mathbf{E}^*(\omega)\right]$. $\boldsymbol{\mu}(\omega)$ and $\mathbf{E}(\omega)$ are the Fourier transform (FT) of the dipole, $\boldsymbol{\mu}(t)$, Eq. (2), and of the complete time course of the electric field, $\mathbf{E}(t)$, including the pump and the probe pulses. The probe is a one cycle IR pulse with the same parameters as the CEP = 0 pulse used for the pump, but a far lower strength, $\left|E_z^p\right|$ = 0.001 a.u. . Figure 6 shows heatmaps of the response, $S(\omega)$, computed for the two pump pulses, top with a CEP = 0 and bottom with a CEP = π. The heatmaps are plotted as a function of the delay time between the pump and the probe (abscissa) and as a function of the frequency ω (ordinate). The frequency axis is centered on the transition frequency, $\Delta E_{\Sigma_2-\Sigma_3}$ ≃0.5 eV. Absorption corresponds to a positive value of $S(\omega)$ and is plotted in blue, emission which corresponds to a negative value is plotted in red. The delay time range starts just after the end of the pump pulse at 4 fs, up to 25 fs. Comparing the two heatmaps shows that the response function $S(\omega)$ provides a suitable probe for the electronic coherence and its phase. Figure 6 shows that the response function oscillates as a function of the delay time with the period of ≈ 8fs that corresponds to the period of the electronic coherence between Σ_2 and Σ_3. Moreover the response oscillations are of opposite phase depending of the value of the CEP of the pump pulse and reflect the π phase difference in the real part of the electronic coherence created at the end of the pulse. The value of the CEP phase is also reflected in the heatmaps of the response centered at Σ_1 - Σ_2 and Σ_1 - Σ_3 transition frequencies, at ≈ 2.5 eV, plotted in figure S4. In figure S4, in addition to the fast beating of ≈ 1.6 fs, which corresponds to the Σ_1 - Σ_2 and Σ_1 - Σ_3 transitions, one also distinguishes a clear modulation due to the Σ_2 - Σ_3 coherence, with a period of ≈ 8 fs.

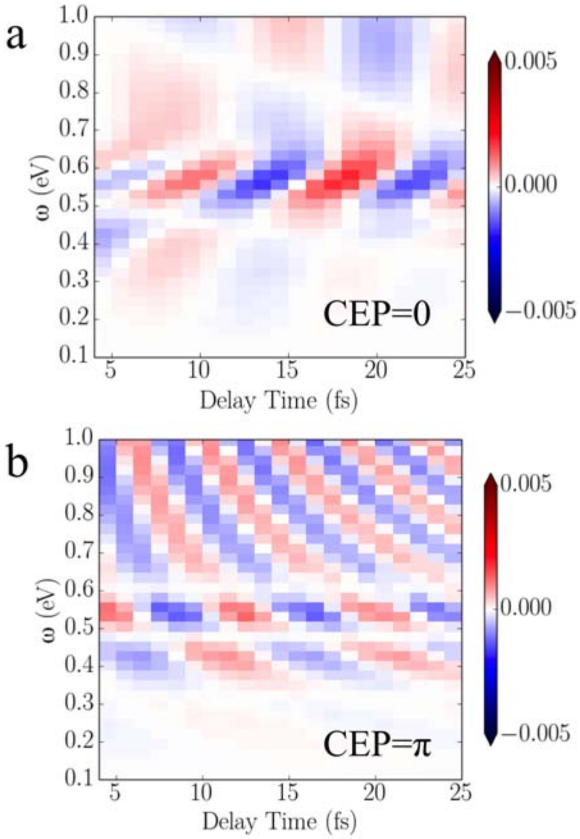

Figure 6: Heatmaps of the response $S(\omega)$ computed for the pump pulse with a CEP = 0 (top) and with a CEP = π (bottom) for a range of delay times between the pump and the probe of 20 fs (x axis). The y axis is centered on the frequency range of the Σ_2 - Σ_3 electronic coherence.

V. CONCLUDING REMARKS

We demonstrated a switching the polarity of the non equilibrium charge density in a modular molecule by altering the CEP of a one cycle laser pump pulse. Controlling the polarity allows choosing alternative dissociation channels by selectively accessing different electronic excited states. Switching the polarity with the carrier phase also induces a switch of the coherence of the excited electronic wavepacket. Controlling the coherence selects contrasting directions for the non stationary charge migration, along the bond or in an opposite direction as can be probed by a second laser pulse. Controlling the coherence is also essential in quantum memory and quantum computing applications.

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