

Efficient control of electron localization by sub-cycle waveform synthesis

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The control of quantum dynamics through the coherent feature of laser pulses has very important applications in various fields. Many of these applications involve processes that occur at different time scales and efficient control is usually needed to balance the dynamics of the different time scales. For instance, control of the quantum dynamics of chemical reactions has been an exciting goal of chemistry and ultrafast physics. Such a process naturally involves both electron and nuclear motion. On the one hand, femtosecond chemistry has made remarkable advances in controlling the pathway of molecular fragmentation using an optimally-shaped multicycle pulse. On the other hand, the advent of carrier-envelope phase (CEP) stabilized few-cycle (~ 5 fs) pulses and attosecond pulses has provided a robust tool for steering electronic dynamics on the attosecond time scale. An interesting question therefore is how to efficiently steer both electronic and molecular motion by combining these two developments.

It has been demonstrated that, using a CEP stabilized few-cycle pulse, the electron wavepacket can be localized to a specific nucleus and therefore exhibits asymmetric electron localization for hydrogen molecules (or its isotopes) [1]. Other approaches of electron localization have also been reported, that includes synthesizing a fundamental laser with another extreme ultraviolet or near infrared probe pulse. However, the localization asymmetry of the total dissociation fragments by a few-cycle fundamental pulse is very small (asymmetry parameter ~ 0.03) even though the kinetic energy release (KER) spectra exhibit large asymmetry. If the molecular wavepacket could respond to the electronic motion, it would be possible to efficiently control the electron location in a very direct way. Nevertheless, precise steering of the electronic motion requires the control of sub-cycle waveforms; thus, a short few-cycle pulse, e.g., 3.5-fs, is desired. On the contrary, the vibration period is longer than 14 fs even for the simplest molecular ion, H_2^+ . The nuclear motion is too slow to respond directly to electronic motion and the few-cycle pulse. To meet this challenge, in our work, we propose and numerically demonstrate a novel scheme with the aim of effectively steering both the electronic and molecular motion of hydrogen molecules and its isotopes. Our scheme is based on the idea of sub-cycle waveform shaping (SWS) by synthesizing coherent multicycle infrared (IR) pulses of different wavelengths.

With the development of laser techniques, it has become possible to synthesize and control two (or three) coherent light sources outputted from different laser systems, such as, fiber lasers and optical parameter amplifier (OPA) or optical parameter chirp pulse amplifier (OPCPA) systems. Our basic idea of SWS stems from the temporal profile modulation by the beat wave of two IR waves. By controlling the amplitudes and phases of two IR pulses, the waveform of the synthesized pulse can be precisely manipulated. As shown in Fig. 1(a), except for the weak shoulder peaks, the field synthesized with a 15-fs fundamental (800 nm) pulse and a 25-fs IR (1200 nm) pulse is almost equivalent to a 4-fs fundamental one-color (OC) field, which thus enables precise control of the electronic motion. Moreover, the synthesized pulse contains shoulder peaks maintaining duration of about 15 fs, which is comparable to the vibration period of H_2^+ . The nuclear motion can respond to the shoulder pulse. Therefore, we can expect more effective control of both the electronic and molecular motion using this SWS field. We need to emphasize that to precisely control the electronic dynamics, it is important to keep the waveform of the SWS pulse similar to that of an OC few-cycle (~ 5 fs) pulse. This condition requires the pulse duration of the synthesized IR pulse to be less than 30 fs.

Figure 1(b) shows the electron localization asymmetry as a function of the CEP. The results are obtained by a numerical simulation based on the time-dependent Schrodinger equation [2]. The solid magenta and blue lines correspond to two typical cases: the interaction of the SWS pulse with the 4th and 8th vibrational initial states of H_2^+ , respectively. For comparison, a similar simulation was performed for a 5-fs fundamental field and the results are presented by the dashed red and green lines in Fig. 1(b). In both situations, we can observe sensitive CEP dependence of the electron localization. Nevertheless, in comparison with the 5-fs OC pulse, the asymmetry amplitude in the SWS pulse is two and three fold higher for the 8th and 4th vibrational states, respectively. Moreover, note that the preparation mechanism of H_2^+ generally indicates an incoherent Franck-Condon (FC) distribution of vibrational states in experiments. Thus it is necessary to average the observables over the initial vibrational states weighted by FC factors, which is called FC averaging. Figure 1

(c) shows the FC-averaged localization asymmetry. Very distinct result can be observed. In the OC field, the asymmetry has been significantly smeared out (dashed red line). In contrast, very large asymmetry with an amplitude of 0.35 can still be obtained in the SWS pulse (solid blue line), which corresponds to an electron localization probability of about 68% of all fragmentation events. We have verified this result by changing the intensity of the OC fundamental pulse from 2×10^{13} to 2×10^{14} W/cm² and the pulse duration from 3 to 10 fs. But the FC-averaged asymmetry in the OC few-cycle pulse does not change significantly and is below 0.06, which is only one-sixth of that in the SWS pulse.

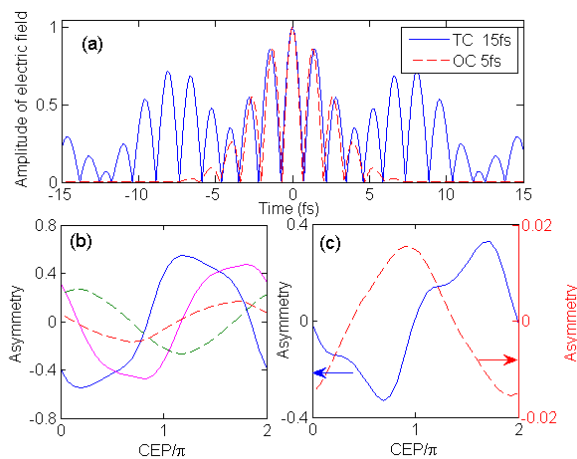


Fig. 1. (a) Temporal profiles of laser amplitude of the 5-fs fundamental (800 nm) OC pulse (red dashed line) and SWS pulse (blue line, 15 fs/800 nm + 25 fs/1200 nm). (b) CEP dependence of the electron localization asymmetry. The solid magenta and blue lines correspond to the interaction of the SWS pulse with the 4th and 8th vibrational states of H_2^+ , respectively. The dashed red and green lines correspond to the interaction of the 5-fs OC pulse with the 4th and 8th vibrational states of H_2^+ , respectively. (c) CEP dependence of the FC-averaged localization asymmetry for the 5-fs OC field (red dashed line) and SWS pulse (solid blue line).

Even though the above results are obtained with hydrogen molecules, state-of-the-art laser technology has made it possible to synthesize and stabilize the phases of multicolor coherent pulses, such as the CEP-stabilized pump, signal and idle lights of optical parametric synthesizers and supercontinuum optical field synthesizers. We anticipate an extension of our concept to efficiently control the electron localization of more complex molecules or other laser-material interaction processes, e.g., the electron localization of CO [3] and photoemission from a metal nanotip [4], by multicolor SWS.

References

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