Efficient control of electron localization by subcycle 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 timescales and efficient control is usually needed to balance the dynamics of the different timescales. 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 motions. 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 timescale. It has been demonstrated that, using a CEP stabilized few-cycle pulse, the electron can be localized to a specific nucleus and therefore exhibits asymmetric electron localization for hydrogen molecules (or its isotopes) and CO molecules. However, the localization asymmetry of the total dissociation fragments by a few-cycle fundamental pulse is very small (asymmetry parameter 0.03). The reason is that the nuclear motion is too slow to respond directly to the few-cycle pulse. In other wards, the few-cycle pulse is efficient to control the electron motion, but not efficient for the nuclear motion. To effectively control the electron localizatioin, we need to balence both the nuclear motion and electron motion. Nevertheless, precise steering of the electronic motion requires the control of sub-cycle waveforms; thus, a few-cycle pulse is desired. On the contrary, a multicycle pulse is prefored to control the nuclear motion.

To meet this challenge, we propose and numerically demonstrate a novel scheme with the aim of effectively steering both the electronic and molecular motions of hydrogen molecules and its isotopes. Our scheme is based on the idea of sub-cycle waveform shaping (SWS) by synthesizing coherent multicycle IR pulses of different wavelengths. By controlling the amplitudes and phases of two IR pulses, the waveform of the synthesized pulse can be precisely manipulated. We can show that 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, thus enabling precise control of the electronic motion. Moreover, the synthesized pulse contains shoulder peaks maintaining a duration of about 15 fs, which is comparable to the vibration period of hydrogen molecules ion. The nuclear motion can respond to the shoulder pulse. Therefore, we can control more effectively of both the electronic and molecular motions using this SWS field.

We develope a thoery model to study the electron localization in the ultrashot laser field. This model solves the time-dependent Schrodinger equation for the nuclear wavepacket in the Born-Oppenheimer

representation. Our simulation shows that in the case of H_2+, a localization probability of 68% can be realized. It means that 68% electrons can be localized to the left or right nucleus by using our SWS scheme. This localization asymmetry is one order of magnitude higher compared with the conventional few-cycle pulse scheme. In the case of H_2, the localization possibility can be as high as 89% by using our SWS scheme. We also preform the simulation for D_2 and T_2, efficient electron localization still can be realized. Another advantage of our scheme of synthesizing coherent IR pulses outputted from OPA is that the central wavelengths of the IR lights can be adjusted in a broad range. This provides additional freedom to control the sub-cycle waveform and electron localization. Moreover, 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 can anticipate an extension of our scheme to efficiently control the electron localization of more complex molecules or other laser-material interaction processes, e.g., photoemission from a metal nanotip, by multicolor SWS.