Quantum electron motion control in dielectric

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Abstract:

Attosecond science capitalizes on the extreme nonlinearity of strong fields, driven by few-cycle pulses, to attain attosecond temporal resolution and give access to the electron motion dynamics of matter in real-time. Here, we measured the electronic delay response of the dielectric system triggered by a strong field of few-cycle pulses to be in the order of 425 ± 98 as. Moreover, we exploited the electronic response following the strong driver field to demonstrate all-optical light field metrology with attosecond resolution. This field sampling methodology provides a direct connection between the driver field and the induced ultrafast dynamics in matter. Also, we demonstrate the quantum electron motion control in dielectric using synthesized light waveforms. This on-demand electron motion control realizes the long-anticipated ultrafast optical switches and quantum electronics. This advancement promises to increase the limiting speed of data processing and information encoding to rates that exceed 1 petabit/s, opening a new realm of information technology.

Main text:

Advancements in attosecond pulse generation and spectroscopic measurements by high harmonic generation in gases and solids opened a new window to study the electronic response driven by strong fields in real-time\textsuperscript{1-11}. The strong-field-induced electron dynamics and the related phase transition to a semimetal-like state of dielectric systems have been studied theoretically\textsuperscript{12-15} and experimentally by XUV attosecond spectroscopy\textsuperscript{16-19}. Accordingly, the strong-field interaction induces a current in the dielectric nanocircuit as
demonstrated elsewhere\textsuperscript{18} and used for the Carrier-Envelope-Phase (CEP) and the waveform detection of the driver’s few-cycle pulses\textsuperscript{18,20-25}. Also, the strong-field interaction with thin films of SiO\textsubscript{2} dielectric has been utilized to generate a wideband coherent EUV radiation extended up to 40 eV\textsuperscript{9}. Based on these studies, the strong-field-induced electron dynamics in the dielectric can be explained by electron motion in the conduction band (illustration in Figure 1a&b). In a strong field (Figure 1a), the electron with initial wave vector (q) is moving in the reciprocal space by acquiring a time-dependent wave vector $K_T(q, t)$ from the driving field, which can be expressed by\textsuperscript{12,16}

$$K_T(q, t) = q + \frac{e}{\hbar} \int_{-\infty}^{t} F_x(z, t_1) dt_1$$

(1)

where $F_x(z, t_1)$ is the optical field strength, and $e$ is the electron charge. Therefore, all electrons are shifted in the reciprocal space by the same wave vector

$$\Delta q(t) = \frac{e}{\hbar} \int_{-\infty}^{t} F_x(z, t_1) dt_1$$

(2)

At a certain critical field strength value (Figure 1b), the shift ($\Delta q$) becomes greater than the Brillouin zone extension $k = 2\pi / a$, causing electron Bragg reflection and Bloch oscillations. Thus, the dielectric constant ($\varepsilon$) and the dielectric material’s optical properties are altered due to the strong polarizability. As a result, the dielectric system undergoes a semimetal-like
phase transition\textsuperscript{12,16}, and the reflectivity changes in real-time following the driver field. Hence, the dielectric time-resolved reflectivity measurement provides direct access to the induced electron motion dynamics in the system. Here, we exploited this field-driven electronic response and the related dielectric reflectivity modulation to directly measure the SiO\textsubscript{2} dielectric system's electronic delay response in a strong few-cycle pulse. Also, we demonstrate all-optical light field sampling metrology with attosecond resolution based on the same principle. Finally, we utilized the light field synthesis to control the electron motion in the dielectric using complex synthesized waveforms.

**Results and discussion:**

**Electronic delay response in dielectric**

The strong-field-induced current in the dielectric has been exploited to indirectly determine the carrier delay response by measuring the carrier injection delay time in the dielectric nanocircuit\textsuperscript{22}. In this work we directly determined the light-field-induced electronic delay response by measuring the time-resolved reflectivity modulation of the dielectric SiO\textsubscript{2} substrate at different field strengths (see Method). The pump beam is modifying the optical properties and the reflectivity of the SiO\textsubscript{2} which is probed by another weak probe beam. The integration of the reflected probe spectrum is calculated at
each instant of time to obtain the reflectivity modulation trace (normalized and shown in Figure 2a), as explained in Supplementary Information (SI) Section I. This reflectivity modulation carries the signature of the dielectric’s electronic response under the influence of the driver pulse. Hence, we determine the relative electronic delay response by measuring the phase delay shift between the reflectivity modulation traces under the same experimental conditions at different field strengths: $F_1=0.78$ V/Å, $F_2=0.96$ V/Å, and $F_3=1.1$ V/Å. The pump beam intensity was controlled by a neutral density filter introduced in the beam path. Moreover, the reflectivity modulation was recorded (in a time window between -2.6 to 3.2 fs) as a function of the time delay between the pump and probe pulse with high resolution (delay steps of 25 as). The reflectivity modulation traces from the average of four recorded measurements at each field strength are plotted in Figure 2b. The traces show relative phase delays of $425 \pm 98$ as and $575 \pm 45$ as. Note, the driver pulse CEP is passively stabilized (phase jittering during the measurements is on the order of 100 mrad). Thus, the CEP jittering contribution to the measured relative phase delay is reflected in the merged standard deviation (SD). The measured phase delay (Figure 2b) is attributed to the electronic delay response in a strong field\textsuperscript{22}. The delay response increases at higher driver field strengths due to the increase of the system’s polarizability and the excited carrier density. We calculated the number of excitation carriers at different field strengths using the driver pulse’s measured electric field. The electric field presented in Figure 2c is retrieved
from the derivative of the measured reflectivity modulation in Figure 2a, representing the driver field’s vector potential, as explained in the SI Section I. Remarkably, the retrieved temporal intensity profile of the driver field ($\tau_{FWHM} = 10.5$ fs) is similar to the measured temporal profile obtained by the convenient Transient-Gating Frequency-Resolved Optical Gating (TG-FROG) measurement ($\tau_{FWHM} = 10.3$ fs), as shown in Figure S2. The calculated number of excited carriers $n_{ex}(t)$ (the calculation is explained in SI Section II) at different field strengths is shown in Figure 2d in contrast with the instantaneous intensity of the carrier’s triggering field (plotted in black dashed line). The total number of excited carriers at field strength F2=0.96 V/Å and F3=1.1 V/Å is almost double (1.86 times) and triple (2.9 times) the number of excited carriers triggered by the field strength F1=0.78 V/Å, respectively. The number of excited carriers plays a significant role in the electronic delay response due to the electron-electron interaction and the screening effect\textsuperscript{22}. Moreover, in Figure 2d, the excited carrier number $n_{ex}(t)$ is behaving as a function of the instantaneous electric field of the driver pulse (black dashed line in Figure 2d). The excited carrier’s $n_{ex}(t)$ has maximum values at the maxima, and minimum values at the minima of the pulsed electric field. Both the maximum population (at $t \approx 2$ fs) and the residual CB population (for $t > 9$fs) monotonically increase with the excitation field amplitude, indicating high reversibility of the excitation, which happens at the high interband coupling matrix element\textsuperscript{16-19}. 


The reversible electronic dynamic response directly gives access to the triggering field of the pulse with high temporal resolution. The time-resolved measurement of the reflectivity modulation changes at different field strengths opening a direct window to the electronic response in the dielectric.

**All-optical light field sampling**

The direct connection between the dielectric system's reflectivity modulation and the incident driver field shape allows the establishment of direct-simple all-optical light field metrology. First, we conducted a numerical simulation to demonstrate the basic principle of this approach by calculating the reflectivity change and the reflected field of the SiO$_2$ thin substrate in the strong field of a one cycle-pulse (spans over a broadband spectrum and centered at 800 nm) at different field strengths (the calculations is explained in the SI Section IV)\textsuperscript{13}. The reflected and incident fields are normalized, overlapped in time, and plotted in Figure S3 (SI). The reflected field exactly follows the incident field at different intensities with a maximum SD < 1.5%. This calculation proves that the dielectric reflectivity modulation due to the strong-field interaction follows the driver field waveform shape.

We sampled an unknown synthesized waveform generated by four spectral channels (250-1000 nm) using a Light Field Synthesizer (LFS) apparatus mentioned above\textsuperscript{26-28} (LFS is explained in the SI Section V and shown in Figure S4 and) to prove the viability of this methodology experimentally. The output
beam from the LFS with an unknown waveform is divided into two separate beams as explained in the previous section (setup is shown in Figure 1c). The first high-intensity beam (pump) is used to alter the dielectric reflectivity. The pump beam’s field strength is ~1.33V/Å (well below the damage threshold ~2.7 V/Å)\(^{18,22}\). In addition, the second beam (probe) has a lower intensity (~10% of the pump beam). The probe beam spectrum is recorded as a function of the time delay between the pump and probe pulses (with delay step size=100 as). Afterward, the unknown synthesized waveform of the strong driver field (henceforth called WF1) is retrieved from the reflectivity modulation measurement (average of three scans are normalized and shown in Figure 3a) and plotted in Figure 3b. Next, we changed the relative phase delay between one of the spectral channels (Ch\(_{\text{VIS-UV}}\)) with respect to the other channels in the LFS by -2fs to generate a new waveform, WF2. The reflectivity modulation caused by WF2 is measured and shown in Figure 3c. The field of WF2 is obtained and shown in the red line in Figure 3d. Subsequently, we calculated the expected waveform of WF2 from WF1 and plotted it in synchrony with the sampled WF2 field (black dash line and red line in Figure 3d). This expected waveform is computed by analyzing WF1 to obtain the fields and relative delay phases between the individual four spectral channels of the LFS forming the WF1 waveform. Then we introduced the -2fs delay of Ch\(_{\text{VIS-UV}}\) mathematically and summed all four channel fields (as explained in the SI Section V).
expected and measured fields of WF2 (Figure 3d) are in good agreement (SD ~10%) and have similar main field features.

Moreover, we changed the relative phase of the ultraviolet spectral channel (Ch_{DUV}) in the LFS with respect to the other channels by \(-2f_s\) to generate a new waveform (WF3). The measured reflectivity modulation by WF3 is shown in Figure 3e. The retrieved field of WF3 (red line in Figure 3f) is plotted alongside the calculated expected waveform (dashed black line). The estimated SD between the measured and expected waveforms is <10%.

The asymmetric shape of the reflectivity modulation in Figure 3a, c, and e is due to the increase and the residue of the excited carrier numbers as it evolves in time depending on the field strength and the shape of the driver field waveform. Notably, the change in the relative amplitude of the main field cycles in WF1, WF2, and WF3 (marked by small black arrows in Figure 3b, d, and f)—due to the relative phase change of the LFS channels—were captured in our measurements, indicating the high temporal resolution of the demonstrated field sampling approach. Furthermore, the constructed spectrum—calculated from the Fourier transform of the measured WF1 field—is similar to the measured spectrum of the pump pulse (identical to the probe spectrum since they deviate from the same beam; see SI, Figure S5). Thus, the presented field sampling methodology can resolve all the frequency components in the waveform that spans two octaves (250-1000nm).
The demonstrated all-optical field metrology exhibits field sampling capability with attosecond temporal resolution for a single broadband waveform spanning two octaves, which was beyond reach\textsuperscript{17,18,22}. This approach can be used under any experimental conditions, enabling the direct connection between the triggering few femtosecond/attosecond field and the measured dynamics in potential time-resolved measurements, providing more insights into the ultrafast physics dynamics of matter. Also, this simple field sampling metrology promises a profound advancement in light field synthesis technology and the attosecond electron motion control in matter.

**Quantum control of electron motion in dielectric**

The light field-induced electron motion in the dielectric can be controlled on-demand by tailoring the driver field's shape with attosecond resolution. We synthesized a few complex waveforms by changing the relative phase of the four channels and intensities in the LFS to control the electron motion in SiO\textsubscript{2}. Some examples of these complex synthesized waveforms are measured based on tracing the reflectivity modulation altered by the electron dynamics driven by these synthesized fields, as explained in the previous section. These measured complex waveforms are illustrated in the left column of Figure 4. The corresponding instantaneous intensity profiles of these waveforms—presented in blue lines in the right column in Figure 4—reflect the triggered electronic response of the SiO\textsubscript{2} dielectric system in real-time. Accordingly, we calculated
the carrier density \(n_{ex}(t)\) triggered by these measured synthesized waveforms and depicted in contrast with the intensity profiles in black lines in the right column of Figure 4.

At an estimated field strength of 1V/Å, the carrier density \(n_{ex}(t)\) follows the field intensity \(E^2\) profile and the number of triggered electrons is maximized at the highest field crests. Using the optical attosecond pulse\(^{27}\) in Figure 4a (II), the electron triggering is maximized at the highest field crest (shaded in red). In Figure 4b (II), the maximum electron triggering signal (shaded in red) occurs at two-time instants separated by 0.9 fs. The separation interval is controlled to be 2.7 fs using the waveform displayed in Figure 4c (II). Figure 4d (II) & 4e (II) demonstrate fine control of the electron motion through the generated and measured complex synthesized waveforms. The electron's maximum triggering occurs at three-time instants (shown in shaded red in Figure 4d (II)) with equal time intervals of 0.9 fs when utilizing these waveforms. In Figure 4EII, the electron’s highest triggering signal arises at four events (illustrated by shaded red) where the four signals are separated in time. The first and second signals are separated by 0.9 fs, the second and third signals are separated by 3.6 fs, and 0.9 fs separates the third and fourth signals.

The presented waveforms can be used to induce and control current signals—lasting ~ 400 as (Fig 4a (II))—in dielectric-based nanocircuits, as presented in other research\(^{18,20,22}\). Setting a certain current signal threshold (the shaded blue
area in Figure 4 right column) in this dielectric-nanocircuit allows for the on and off switching of the light-induced current in the attosecond time scale and establishment of ultrafast optical switches. Remarkably, this demonstrated approach works under ambient conditions, and it is viable in miniaturized ultrafast optical switch devices based on implementing the well-established programmable pulse shaping technology\textsuperscript{29} into dielectric nanocircuits. This development promises an increase in the data processing speed and information encoding to rates that exceed one petabit per second, a million times faster than the current technology. Figure 4 (right column) displays potential binary coding signals generated by the synthesized waveforms in dielectric nanocircuits\textsuperscript{30}.

We exploited the dielectric's strong-field interaction to determine the attosecond electronic delay response in the dielectric. Also, we demonstrate an all-optical direct-simple approach to sample the light field spanning two octaves with attosecond resolution. This field sampling approach can be implemented in different environments and experiment setups to provide a real-time connection between the ultrafast dynamics in matter and its driver field. Consequently, using this realistic sampled field in simulations, calculations, and fitting algorithms related to the measured spectroscopic response of matter provides more accurate interpretation and insight into the underlying physics of these dynamics. Moreover, we utilized synthesized
waveforms to exhibit full control of electron motion in the dielectric. This electron control can be used to develop quantum electronics, paving the way to extend the frontiers of modern electronics and data information processing technologies into the petahertz realm.

**Method**

**Field-induced reflectivity modulation measurement of SiO$_2$ dielectric:** in this experiment (setup is illustrated in Figure 1c), conducted in an ambient environment, the beam of few-cycle visible (500-700 nm, centered at $\lambda = 600 \text{ nm}$, and p-polarized) laser pulses is split into two beams by passing the laser through a two-hole mask. The mask is designed to have two different hole diameters (3 mm and 1 mm). Therefore, the two beams that emerge through the mask have different intensities. The first beam has a high-intensity (pump beam) to induce the phase transition and alter the reflectivity of the SiO$_2$ substrate. The estimated pump beam's field strength is 0.78 V/Å (at a lower field strength $\leq 0.67 \text{ V/Å}$; no significant reflectivity modulation signal was observed) which is lower than the damage threshold$^{18,22}$. Note, the reflectivity signal disappears when the SiO$_2$ substrate is damaged, so all the presented measurements were collected at field intensity lower than the damage threshold. The second beam (probe beam) has a lower intensity ($\leq 0.1 \text{ V/Å}$) than the threshold field strength required to induce any degree of phase
transition in SiO$_2$. The two beams deviate from the mask, are incident on two
D-shape focusing mirrors (f=100 mm), and focused onto the 100 μm thick SiO$_2$
substrate (incident angle < 5º). An imaging system has been used to ensure
perfect spatial overlapping between the two beams. One of these mirrors is
attached to a piezo-stage device to control each beam’s relative delay with
attosecond resolution. The reflected probe beam (off the substrate’s front
surface) is tightly focused into an optical spectrometer entrance after
propagating enough distance to be spatially isolated from the pump beam; a
polarizer and a one-hole mask were introduced to filter out the pump beam.
The measured probe beam spectrum (in the presence of the reflected pump
beam, as shown in Figure S1) is recorded as a function of the time delay
between the pump and probe pulses with delay step size=100 as. Since the
spectrum does not show any interference fringes, the reflected pump beam has
no contribution to the reflectivity modulation measurements, providing a high
signal/noise ratio.
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Author Contributions:

H.A. and D.H. conducted the experiments and analyzed the data. S.Y. and K.Y. carried out the simulations and calculations. V.P. designed and measured the optics of the LFS. M.H. conceived, supervised, and directed the study. All authors discussed the results and their interpretation and wrote the manuscript.

Competing Interests statement

Authors declare no competing interests.
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Figure 1: Light field-induced electron motion in a dielectric. a & b, Illustration of the electron motion dynamics in the reciprocal space at the strong and critical strong-field strength, respectively. See text for explanation. c, the electronic delay response and all-optical field sampling experiment setup. A mask splits the main beam into two strong (pump) and weak (probe) beams. The two beams are focused on a hundred microns dielectric (SiO₂ substrate). One of these D-shape mirrors is connected to a piezo-stage to control the relative delay between the pump and probe pulses with attosecond resolution. An optical spectrometer measures the reflectivity modulation of the reflected probe beam spectrum from the substrate. A polarizer and a one-hole mask are introduced in the probe beam path before the spectrometer to enhance the signal-to-noise ratio of the reflectivity modulation measurements.
Figure 2: Electronic delay response in SiO$_2$ dielectric system. a, The normalized measured reflectivity modulation of the SiO$_2$ substrate under the influence of the strong-field of a few-cycle pulse. Each point represents the integration of the probe beam spectrum measured at each time delay between the pump and probe pulses. b, The measured reflectivity modulation traces in a time window between -2.6 to 3.2 fs at different driver field strengths; 0.78 V/Å, 0.96 V/Å, and 1.1 V/Å, are shown in blue, red, and orange lines, respectively. The relative delay responses between these measured traces are 425 ± 98 as and 575 ± 45 as. c, The retrieved driver pulse-field from the derivative of the reflectivity modulation measurement in a. d, The calculated number of excited carriers $n_{ex}(t)$ triggered by different field strengths are shown in contrast to the driver's instantaneous intensity ($E^2$) plotted in a black dashed line.
Figure 3: All-optical field sampling of synthesized waveforms. a, The normalized measured reflectivity modulation and b, the retrieved field of the synthesized waveform WF1. c, The normalized measured reflectivity modulation and d, the obtained field of the synthesized waveform WF2 after moving ChVIS-UV with respect to the other channels in LFS by -2fs, from the four channels phase setting of WF1. The sampled field of WF2 is plotted in the red line, and the expected field of WF2 is plotted in the black dashed line. The expected and measured fields have similar main features. By comparing the WF1 and WF2 in b & d, the introduced phase delay of the ChVIS-UV modifies the relative amplitude of the main four field half-cycles (pointed by small black arrows). The gray dashed line shows the maximum amplitude of the field for eye guidance. e, The measured reflectivity modulation altered by a waveform (WF3) synthesized from WF2 by shifting the ChDUV with respect to the other channels in LFS by -2 fs. f, The synthesized waveform WF3 is plotted in a red line, and the corresponding expected calculated field is shown in the black dashed line. The relative amplitude of the half-cycles on both sides of the main half-cycle changed from WF2 to WF3.
Figure 4: Attosecond quantum electron motion control and optical switch in dielectric. Synthesized waveforms (in red line) and the corresponding intensity profile (blue line) for controlling the electron motion in the dielectric are shown in the left (a (I), b (I), c (I), d (I), &e (I)) and right columns (a (II), b (II), c (II), d (II), &e (II)), respectively. The calculated number of carrier density $n(x,t)$ triggered by these waveforms is plotted in contrast with the intensity profiles (right column) in black lines. Using the waveform in a (I) & a (II), the number of triggered electrons is maximized in one instant of time at the highest field crest (shown in a shaded red). The temporal profile FWHM of this crest is ~400 as. The synthesized waveform in b (I) & b (II) triggers the maximum number of electrons at two different time instants (as indicated in shaded red) separated by 0.9 fs. This separation time interval becomes 2.7 fs by using the waveform in c (I) & c (II). The highest triggering signals of electrons induced by the synthesized field plotted in d (I) & d (II) occur at three events separated equally in time by 0.9 fs. e (I) & e (II), shows a
complex measured synthesized waveform that triggers the maximum electron number signal four
times. The first two signals (separated from each other by 0.9 fs) lead the third signal by 3.6 fs.
The fourth signal appears after 0.9 fs from the third signal. The blue shaded area in the right column
presents a virtual threshold that would be potentially introduced in dielectric-nanocircuit to switch
on/off the light-induced current with attosecond resolution. Potential binary coding signals
generated inside this nanocircuit using the presented waveforms are shown in 0 & 1 numbers in
the right column.
Supplementary Files

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