

Before the cycle is over: Subcycle time refraction at optical frequencies

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Large and abrupt variations in the electromagnetic properties of materials lead to dramatic effects: Even a single steplike change in the refractive index induces striking phenomena, such as time refraction and time reflection. When the refractive index varies periodically in time, multiple time refractions and time reflections interfere, giving rise to photonic time crystals (PTCs). Importantly, PTCs display momentum bands separated by gaps in which the modes experience exponential amplification, drawing energy from the modulation in a nonresonant fashion. Ordinary nonlinear optics does not operate in this regime: The material response is either very weak or very slow. One of the immediate consequences is that time reflection of light at optical frequencies has never been observed in experiments. Here, we experimentally realize an order-unity change in the refractive index occurring at subcycle rates and explore the phenomena emerging from it. By varying the duration of the index change from extending over many cycles to being significantly below a single cycle, we observe that the frequency shift of the time refraction is enhanced as the index variation occurs faster. Our experiment is the gateway for realizing sharp time interfaces at optical frequencies, which are the key for experimenting with time reflection, PTCs, and exciting phenomena expected from light-matter interactions in time-varying media.

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The physics of light in materials with time-varying refractive index has been gaining considerable attention in recent years. This is mainly because optical waves can strongly couple to matter, and combining time-varying refractive index and strong light-matter interaction holds great promise for exotic applications such as nonresonant amplification of light, coherent light sources not relying on atomic resonances [1], new mechanisms of nonlinear frequency conversion [2], and generation of quantum light controlled by the temporal modulation of the index [3]. These applications and many others [4–13] rely on interference between two fundamental processes: time refraction and time reflection [14,15]. When an electromagnetic wave is traveling in a homogeneous material with time-varying refractive index, its energy (frequency) is altered (as time-translation symmetry is broken by the index variation) but its momentum (wave vector) is conserved. The conservation of wave vector implies that, if the refractive index is decreased, the frequency of the wave is blueshifted (or redshifted if the index is raised). Time refraction is characterized by such spectral translation caused by the temporal variation of the refractive index, and was measured in many

optical systems [16–19]. On the other hand, time reflection is essentially a new wave generated from the index variation, and it is propagating counter to the original wave, at the same frequency as the time-refracted wave but with a conjugate phase. Time reflection requires refractive index variations that are much harder to produce in experiments because the variations need to occur within a time frame shorter than a single cycle of the optical wave and should be of order unity. Because of these extreme conditions, time reflections have not yet been observed in the optical regime, but were thus far observed with low frequency waves such as water waves [20], cold atoms [21], microwaves [22,23], and in synthetic dimensions [24,25]. Once time reflections of sufficient amplitudes are observed, one would be able to advance to the next level and explore the effects of multiple time interfaces and the resulting interference between time refractions and time reflections, for example, photonic time crystals (PTCs), which form from periodic variations (in time) of the refractive index. The periodic modulation causes interference between time-refracted and time-reflected waves, which manifests into a band structure with band gaps in momentum (wave number) instead of energy (frequency) [26–28]. The electromagnetic modes of the momentum gaps are exponentially increasing (or decreasing) in their amplitude, drawing energy from the modulation (or transferring energy to it). PTCs were thus far observed at low rf frequencies [27] and very recently also in microwaves [29,30]. These exponentially increasing modes associated with the momentum band gap of a PTC were suggested as a source of radiation and nonresonant amplification of light [1,9], not relying on any atomic resonance and not

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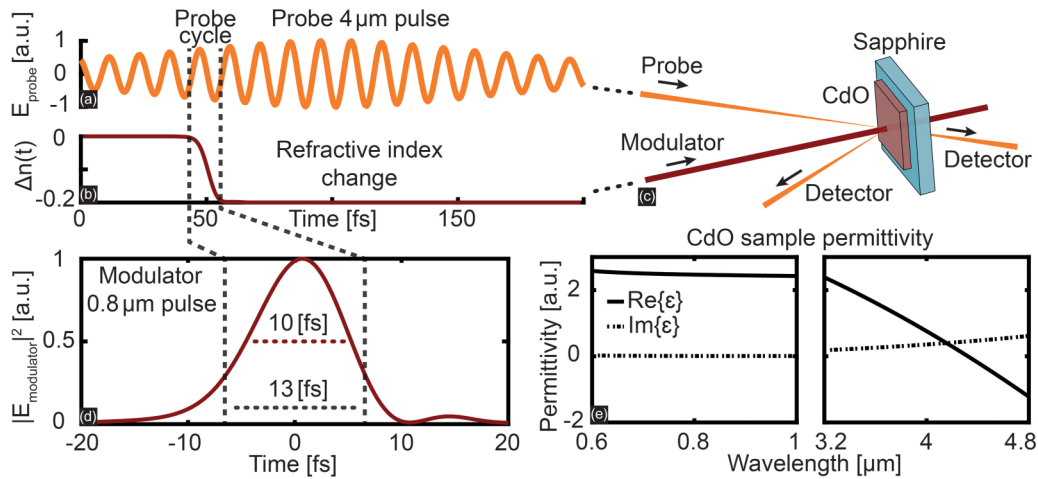


FIG. 1. Experimental scheme of the subcycle refractive index variation. (a) Electric field of the probe pulse (4 μm mean wavelength), reconstructed using cross-correlation frequency-resolved optical gating (XFROG). (b) Illustration of the temporal dependence of the sub-probe-cycle refractive index variation, calculated by integrating the squared intensity profile of the modulator pulse ($\Delta n(t) \propto \int_{-\infty}^t I_{\text{modulator}}^2(\tau) d\tau$). (c) Schematic of the experimental modulator-probe setup. The Fresnel-reflected probe power and transmitted probe power and spectrum are measured for each modulator-probe delay. (d) Intensity profile of the intense 10 fs modulator pulse (0.8 μm mean wavelength), reconstructed using frequency-resolved optical gating (FROG). (e) Permittivity of the CdO sample in the two relevant wavelength windows, with its ENZ point at 4.4 μm.

requiring population inversion. These band gaps in momentum appear at a region of wave numbers centered around half the modulation frequency. Modulating a material periodically at twice the period (half the frequency) of an optical wave implies that, for one optical period, the refractive index is falling, and for one optical period it is rising, or that either the fall or the rise of the refractive index is subcycle. Hence, to achieve interference between time refractions and time reflections, to create PTCs and make other exotic applications possible, large (order unity) index changes occurring at subcycle time frames are the key.

Until recently, all the experimental observations of order-unity index changes were in the multicycle regime [16,17]. Two years ago, the first order-unity index changes occurring within a single optical cycle were reported [18,19]. In these experiments, a probe pulse is launched into a transparent conductive oxide (TCO) sample that is modulated by an intense optical pulse (so-called “modulator”) [16,17,31–35]. The TCO sample has an epsilon-near-zero (ENZ) point at the mean wavelength of the probe pulse, and due to the modulator pulse this ENZ point moves, creating a large change (~ 0.5) in the refractive index for the probe pulse. To directly induce single-cycle index variation, Ref. [18] employed a short intense modulator pulse at a shorter wavelength, such that the duration of the modulator pulse is roughly one cycle of the probe period. The TCO samples with their ENZ points near the mean wavelength of the probe were not chosen by coincidence. Rather, these materials display the only mechanism known today that can achieve order-unity variations in the refractive index within a few femtoseconds while remaining relatively transparent. However, thus far, the time it took to change the refractive index in all experiments was longer than one cycle of the probe wave, which was too long to enable significant time reflections, not to mention the formation of PTCs. For this reason, it is essential to have

order-unity index changes occurring with deep subcycle time frames.

Here, we demonstrate exactly that: subcycle order-unity variation of the refractive index. We use a sample of doped cadmium oxide (CdO), which has its ENZ point around 4.4 μm free-space wavelength, and launch a probe pulse with 4 μm mean wavelength (~ 13 fs period) through the sample, while it is modulated by an intense 10 fs modulator pulse (800 nm mean wavelength). From measurements of the transmission and Fresnel reflection of the probe through the modulated sample, we infer that the refractive index is decreased by ~ 0.3 during the ~ 0.77 cycle index variation. We gradually vary the duration of our modulator pulse from 40 fs (~ 3 cycles) to 10 fs (subcycle) and observe enhanced blueshift of the time refraction. This experimental result opens the door for realizing sharp time interfaces, time reflection, and ultimately photonic time crystals for light waves at optical frequencies.

As explained above, generating subcycle refractive index variations requires several components. The first is an optical probe pulse (incident wave), shown in Fig. 1(a). The second is a sample that, while the probe is traveling through it, undergoes an order-unity refractive index variation within a timescale shorter than a single cycle of the probe (few femtoseconds), as illustrated in Fig. 1(b). The third is that the ENZ point of the TCO sample is at the close vicinity of the (mean) wavelength of the probe pulse. In our experiments, the change in the refractive index in the sample is induced by an intense modulator pulse of duration shorter than a single cycle of the probe [Fig. 1(d)]. Clearly, the second requirement is extremely hard to achieve at optical frequencies. In fact, the only known mechanism that yields subcycle index change of order unity is the one associated with TCO materials, when the frequency of the probe pulse is in the vicinity of the ENZ point, where the index change is highly enhanced. In this article, we rely on a family of TCO

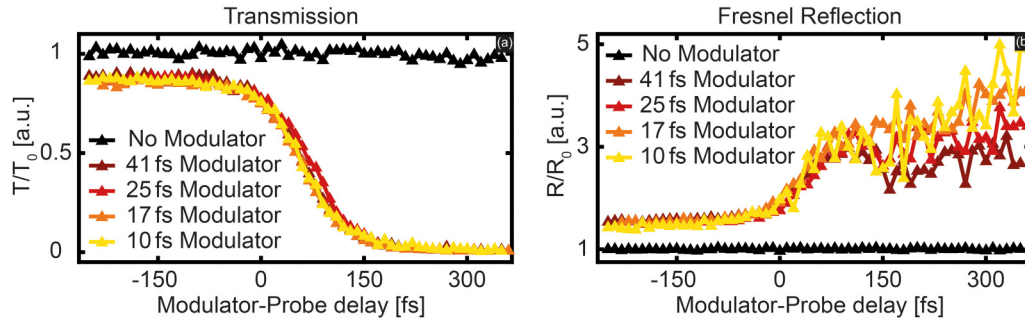


FIG. 2. Transmissivity and reflectivity of the probe beam incident on the CdO sample vs modulator-probe delay and for different durations of the modulator pulse. (a) Transmitted power of the probe vs modulator-probe delay. Negative times correspond to the probe pulse arriving at the sample before the modulator pulse. The modulator pulse causes a decrease in the transmission of the probe pulse. The transmission does not depend on the duration of the modulator pulse. As a reference, black triangles correspond to transmission without any modulator pulse. (b) Fresnel reflection of the probe pulse vs modulator-probe delay [same zero delay as in panel (a)]. The modulator pulse causes an increase in the reflection from the sample. Here, shorter modulator pulses enhance the increase in reflectivity. For comparison, black triangles are without any modulator pulse. At negative delays, there is a small difference between the unmodulated and the modulated measurements due a weak ghost replica of the probe pulse.

materials that have their ENZ point around $4\mu\text{m}$ free-space wavelength.

In our experiments, the probe pulse has an ~ 100 fs duration with energy ~ 0.01 mJ, centered around $4\mu\text{m}$ mean wavelength (a single cycle amounts to 13 fs) [Fig. 1(a)]. This probe pulse is generated by down-converting a pulse from an amplified Ti:sapphire laser pulse using optical parametric amplifier (OPA) followed by difference frequency generation (DFG). The sample is a $2.7\mu\text{m}$ thick doped CdO and the modulator pulse carries energy of ~ 1 mJ centered around $0.8\mu\text{m}$ mean wavelength. The modulator pulse is also generated from our amplified Ti:sapphire laser, but it is compressed to < 10 fs pulses [Fig. 1(d)] using a hollow core fiber filled with Ar gas [36]. The doping of the CdO sample is chosen such that the ENZ point will be at $4.4\mu\text{m}$ [Fig. 1(e)]: just above the mean wavelength of the probe pulse. This choice of ENZ point allows for order-unity refractive index variation in the vicinity of the probe's mean wavelength, induced by the modulator pulse. Figure 1(c) illustrates the experimental setup. To maximize the spatial homogeneity of the refractive index variation experienced by the probe beam, we expand the spot size of the modulator beam on the sample to ~ 2 mm, which is an order of magnitude larger than the diameter of the probe beam on the sample (~ 0.1 mm). We measure the Fresnel-reflected probe power and the transmitted probe power and spectrum, as a function of the delay between the probe and the modulator pulses, for different pulse durations of the modulator.

The permittivity of the CdO sample [Fig. 1(e)] shows negligible linear absorption at $0.8\mu\text{m}$ (modulator mean wavelength). However, the band gap of CdO is relatively small (~ 2.4 eV [37–40]) and therefore, at sufficiently high intensities, the $0.8\mu\text{m}$ (~ 1.5 eV modulator) causes significant two photon interband transitions (see Sec. 1 in Supplemental Material [41] for more details). The two-photon interband transitions increase the density of conduction electrons and lead to a shift of the ENZ point to shorter wavelengths. This mechanism decreases the refractive index for the probe pulse.

To unravel the dynamics of the sample permittivity, we scan the delay between modulator pulse and probe pulse, and measure the power of the transmitted and Fresnel-reflected

probe (Fig. 2). Figure 2(a) shows the transmitted probe power as a function of modulator-probe delay, for different durations of the modulator pulse (represented by different colors). To enable comparison between the response at different delays, we set the zero-delay point (for each duration of the modulator pulse) to 10% change of the transmitted probe with respect to transmission measured for the earliest delay. We find that, for all durations of the modulator pulse, the transmission of the probe decreases in the presence of the modulator pulse, in accordance with the shift of the ENZ to shorter wavelengths. Simultaneously, the power of the Fresnel-reflected probe [Fig. 2(b)] increases due to the index change in the sample, implying that the sample becomes more reflective, which also supports the understanding that the ENZ point shifts to a shorter wavelength. The relaxation time of both the transmission and the Fresnel reflection back to their original values occurs on picosecond timescales (see Sec. 1 in Supplemental Material [41]), further verifying that the origin of the observed index change is an interband transition, which is always associated with long relaxation times (in contrast to interband transitions where the relaxation can be as fast as tens of femtoseconds [18]).

Next, we examine what happens when we shorten the duration of the modulator pulse while keeping its energy constant. As shown in the rightmost part of Fig. 2(b), the Fresnel reflection rises more for shorter modulator pulses, although we make sure to maintain the same energy for the modulator pulses for all temporal widths. From these experiments, we conclude that, in this specific experiment, the observed change in the electromagnetic properties of the sample does not depend on the modulator pulse energy, but depends on the temporal width. Finally, we note that here the change in the refractive index is negative, unlike our previous experiments [18] where the index change was positive. From all of these insights, we conclude that the index change here is caused by a multiphoton process (most likely two-photon absorption) caused by the modulator pulse, which increases the density of conduction electrons and leads to a blueshift of the ENZ point, and consequently to an order-unity negative index change occurring within a subcycle time frame. Using the data pre-

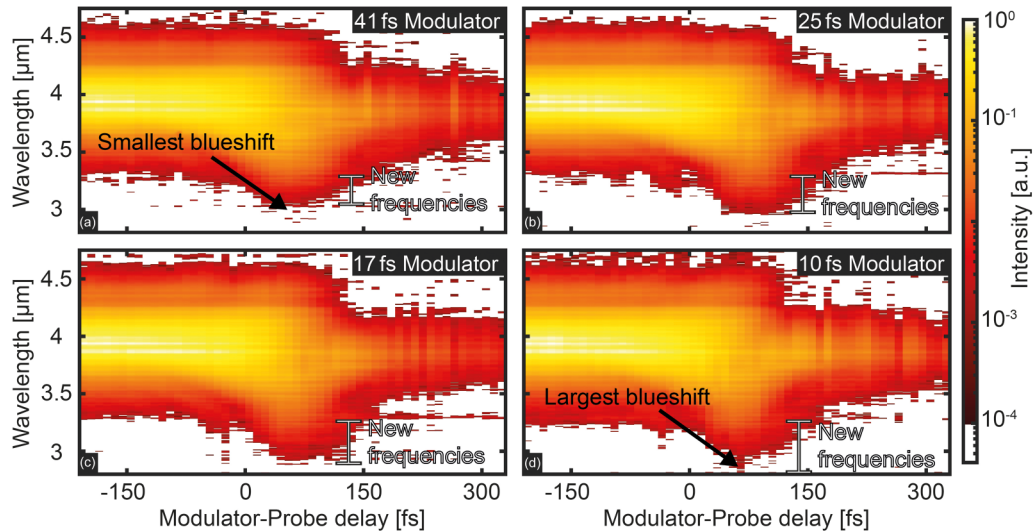


FIG. 3. Spectrograms of the transmitted probe pulse vs modulator-probe delay, for different durations of the modulator pulse. Zero delay is set to be the same as in Fig. 2(a). In all panels, new frequencies appear between 0 and 150 fs delays. Namely, the spectrum is blueshifted to short wavelengths. At larger delays (beyond 150 fs), the spectral shift of the probe becomes unnoticeable, because the refractive index is changing only very slowly. In this range of delays (beyond 150 fs), the intensity is reduced due to the decreased transmission through the sample [as also visible in Fig. 2(a) for the same delays]. The bars in all panels show the range of new frequencies emerging from the modulation. This range is growing as the modulator pulse becomes shorter. Note that the spectrum is not normalized for each delay individually. (a) 41 fs modulator. (b) 25 fs modulator. (c) 17 fs modulator. (d) 10 fs modulator, which is subcycle for the probe (where a single cycle is 13 fs).

sented in Fig. 2, and assuming that the change to the refractive index of the CdO sample can be described by a simple shift of the ENZ wavelength, we estimate that the refractive index variation in our experiments is $\Delta n \approx -0.3$ (see Sec. 2 in Supplemental Material [41] for calculation details).

A strong signature of the temporal dynamics of the variation in the refractive index is imprinted on the time refraction—the frequency shift of the transmitted probe pulse. For each duration of the modulator pulse, we measure a spectrogram of the transmitted probe spectrum versus the modulator-probe delay (Fig. 3). As the modulator and probe start to overlap [between 0 and 150 fs delays in Figs. 3(a)–3(d)], the spectrum of the probe exhibits a blueshift. This blueshift of the spectrum is another hint of the interband transition occurring in the CdO, contrary to intraband transitions, which exhibits mostly a redshift of

the spectrum followed by a blueshift [18]. This is because in the current experiments, the modulator pulse increases the density of conduction electrons, which causes a decrease in the index, whereas for intraband transitions [16–19] the modulator pulse increases the energies of the conduction electrons while keeping their density unchanged, which is manifested in an increase in the refractive index. Finally, we find that compressing the temporal width of the modulator pulse to be subcycle of the probe [Fig. 3(d)] shows enhanced blueshift, extending to below 3 μm wavelength (13% shorter than without the modulator).

It is instructive to quantify the blueshifts presented in Fig. 3 by plotting the shortest detected wavelength for each modulator-probe delay and for the duration of the modulator pulse [Fig. 4(a)]. This analysis shows that compressing the modulator pulse dramatically enhances the blueshift of the

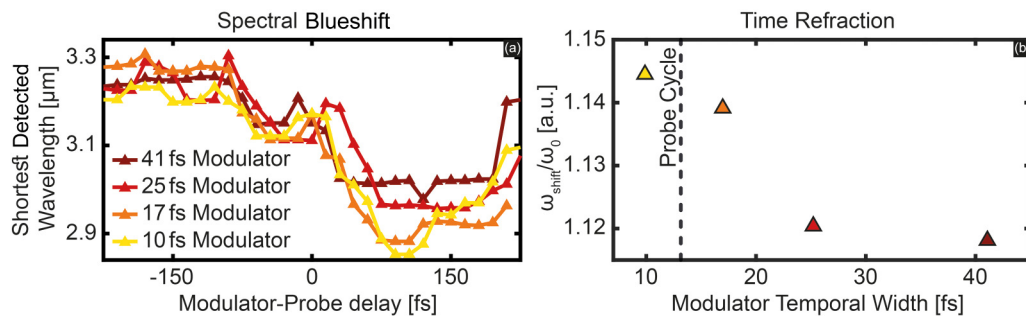


FIG. 4. Spectral blueshift and time-refraction analysis. (a) Shortest detected wavelength at each delay point of the spectrograms of Fig. 3. Different colors correspond to different durations of the modulator pulse presented in the panels of Fig. 3. Shorter modulator pulses create greater blueshift. (b) Estimated spectral shift of the time refraction calculated from the shortest wavelength of each modulator width in panel (a) divided by the shortest transmitted wavelength measured without the modulator. The subcycle index variation generates the largest spectral shift of the time refraction.

probe pulse. Comparing the shortest measured wavelength over all delays, with and without the index variation, gives a quantitative estimation of spectral shift of the time refraction for each duration of the modulator pulse [Fig. 4(b)]. The sub-cycle modulator pulse generates the largest frequency shift.

Our results demonstrate subcycle refractive index variation of order unity in an ENZ CdO thin film, driven by intense 10 fs modulator pulses. The induced $\Delta n \approx -0.3$ over <1 optical cycle yields a substantial enhancement of the frequency shifts, validating that the index variation is both large and temporally sharp. This level of ultrafast control is a key prerequisite for observing optical time reflection, generating sharp time interfaces, and ultimately realizing PTCs at infrared or visible frequencies. Generating a PTC requires periodic modulation of the refractive index where the time-reflections and the time-refractions both play a major role and interfere, giving rise to a band structure displaying a momentum gap. For this to happen, the modulation should be of the order of a single cycle of the probe. As our experiments show, when the modulator pulse excites electrons from valence to conduction - the index indeed varies faster than a single cycle, but the relaxation is via recombination across the bandgap, and therefore very slow. This means that our current samples are not suitable for periodic modulation with single cycle periodicity, due to the slow relaxation. Thus, to generate photonic time-crystals (PTCs), we need to move to modulator pulses at longer wavelengths or to materials with a larger bandgap. Nevertheless, there are numerous related phenomena relying on interference

between just 2-3 time-interfaces [5,7,8], which have now become within reach, following our experiments. For example, using two or more consecutive modulator pulses with various relative delays will allow to study the interference between time refractions and time reflections.

The demonstrated platform opens a promising pathway toward experiments with light at optical frequencies in time-varying media. Perhaps most promising future direction is nonlinear frequency conversion in time-varying media, where recent experiments with individual time-interfaces reported observations that cannot be explained by perturbative nonlinear optics theory [42,43], even though the index changes occurred within several cycles, not yet within an optical sub-cycle. Bringing these effects into the subcycle regime will generate a new frontier in nonlinear optics.

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Data availability. The data that supports the findings of this letter are not publicly available. The data are available from authors upon reasonable request.

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