Photonic time crystals: A materials perspective

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Abstract: Recent advances in ultrafast, large-modulation photonic materials have opened the door to many new areas of research. One specific example is the exciting prospect of photonic time crystals. In this perspective, we outline the most recent material advances that are promising candidates for photonic time crystals. We discuss their merit in terms of modulation speed and depth. We also investigate the challenges yet to be faced and provide our estimation on possible roads to success.

1. Introduction

The realization of Photonic-Time-Crystals (PTCs) holds great potential to unlock new physics and phenomena applicable to non-resonant light amplification, tunable lasing, and a plethora of yet unexplored quantum light-matter interactions [1–10]. In order to realize photonic time-crystals operating in the optical frequency range, several challenges need to be addressed [11]. Most importantly, to achieve a detectable time-reflected signal and/or wide momentum bandgaps, the refractive index of the optical medium must be modulated by an appreciable amount (in the range 0.1-1) all while occurring in a time scale of a single optical cycle of the wave propagating in the medium. Such sharp transitions of permittivity are challenging to realize experimentally. Conventional nonlinear optics, which employs virtual electronic transitions in solids, albeit instantaneous – is orders of magnitude too weak. Other nonlinear optics mechanisms that can yield large index changes typically require transport of some kind, hence are orders of magnitude too slow (photorefractive materials, liquid crystals, thermal nonlinearities, etc.). In this perspective, we will first explore possible material platforms for both large and ultrafast index changes. We will then discuss the most viable options for overcoming challenges in both aspects.

2. Materials and mechanisms to trigger large refractive index changes

2.1 Large refractive index changes

The demanding requirements of achieving an appreciable change in the refractive index within ultra-fast time scales call for extended search for unconventional photonic materials. For different material platforms, various mechanisms have been studied that enable significantly large refractive index change, which at least promises to meet the first criteria for PTC implementation.

One such material class is phase change materials that can undergo large refractive index changes as they transition from an amorphous to a crystalline state, and vice versa [12,13]. Vanadium oxide has been shown to have large modulations in kilohertz frequencies under thermal stimulation, with cycle times reaching nanoseconds under an optical impulse [14].

GeSeTe, another prototypical phase change material, has been shown to undergo large relative permittivity changes from 30 in the crystalline phase to 11 in the amorphous phase [15]. Because this transition occurs from thermal changes, the minimum time scale is usually on the order of picoseconds. However, when the optical excitation is below what is required for a full phase change, sub-picosecond modulation in the NIR has been observed in both the crystalline [15,16] and amorphous phase [15]. A similar response time of 250 femtoseconds was observed in the optical range by P. Martinez et al [17]. In GeSeTe, even if a full phase change is not triggered, the relaxation time is still on the picosecond time scale. Ge–Sb–Se–Te (GSST), another emerging phase change material, shows large changes (Δn = 0.5 to 3) (Fig. 1b) in the mid-infrared frequency range, but the nature of the transition is still thermal, with transition times of 30-100 nanoseconds [18].

Another promising material class is transparent conducting oxides (TCOs) where the refractive index can be modulated both electrically [19] and optically [20]. Under electrical modulation, an applied voltage accumulates free carriers near the surface of an electrical gate in a TCO, locally changing the refractive index via Drude dispersion [21]. Unity order refractive index changes have been observed in conducting oxides by electrical control [22], with potential speeds reaching the picosecond scale so far [23]. However, despite the large magnitude index changes, the relaxation times with most thermally or electrically activated changes are too slow to implement photonic time crystals. For the same reason, mechanical (MEMS-based) [24,25]and structural modulation [18,19] of the effective permittivity are unsuitable for PTC implementation.

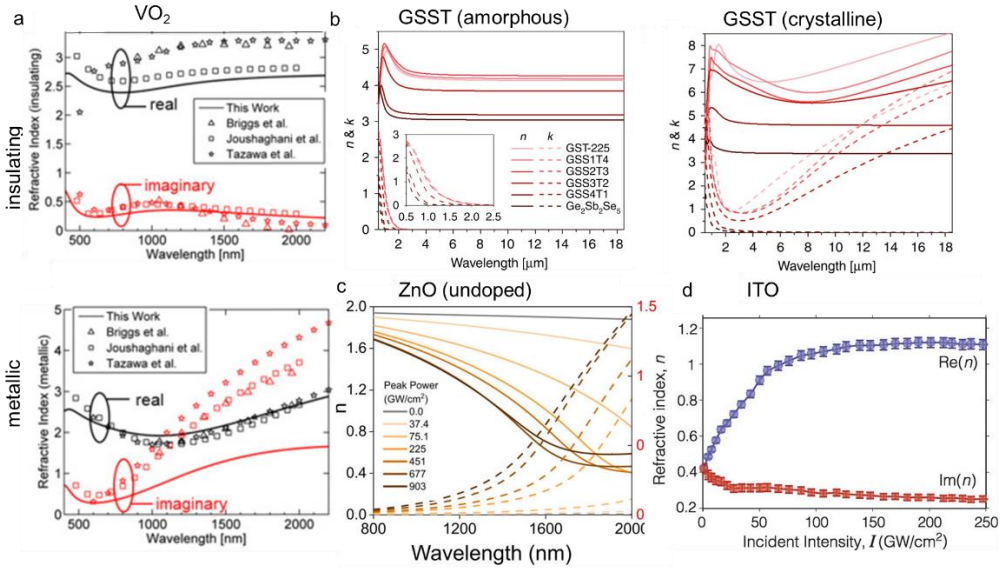

Figure 1 (a) Vanadium dioxide shows large refractive index changes as it transitions from an insulating to a metallic phase under thermal stimulation [14] (b) GSST can transition from an amorphous to a crystalline stage under an optical pulse, showing unity order changes in the mid infrared wavelength regime. (c) Transparent conducting oxides like zinc oxide have large laser damage threshold, and can tolerate large interband optical pump intensities, enabling large index changes [26](d) Operating near the epsilon-near-zero regime of transparent conducting oxides such as ITO enables large index changes at low pump intensities [20].

2.2 Ultrafast index change

The second criteria that needs to be met for PTC implementation is the periodic modulation of refractive index, which is more difficult to attain at the required timescales. Since the refractive index change must occur in a time duration comparable to the optical pulse,

modulations based on thermal phase-change, which are generally in the nanosecond regime, and electrical changes which are in the picosecond range, are not viable for PTC implementation in the optical frequency domain. Longer wavelengths relax the constraints of the ultrashort cycle time. It is, therefore, no surprise that the first observation of the momentum gap in a time-modulated medium was done at radio frequencies [27] and the first PTC-dominated effects such as the exponential growth of EM waves in a temporally varying metasurface, have been demonstrated in the microwave regime [28]. These experiments are also promising signs that for proof-of-concept devices, plasmas or RF circuits can be viable media [29]. Faster nonlinearities in materials such as lithium niobate and barium titanate are another option, but the resistive-capacitive delays of circuits for electrooptic modulation need to be circumvented. Emerging materials such as Gallium phosphide have also been shown to have sub-30 fs optical response times, with large reflectance modulation observed for patterned substrates [30,31], and warrant further exploration in the time-varying field.

So far, all-optical excitation seems to be the most promising route to realize PTCs. One potential route for the implementation of PTCs is the optical excitation of free carriers in TCOs. In TCOs, significant changes in the dielectric permittivity can occur much faster than any thermal, thermoelastic, or electrooptic modulation. There are two major pathways to achieve all-optical modulation utilizing the free carriers in TCOs. Firstly, under illumination by an interband pump pulse, electrons jump from the valence band to the conduction band of TCOs, increasing the free carrier concentration and decreasing the permittivity. The material, in this case, can exhibit a large decrease in the refractive index (0.1 in aluminum doped zinc oxide, AZO [32], 1.6 in undoped ZnO [26]). Since the excitation is optical, through absorption of photons, the transition is instantaneous (follows the pump pulse), which can be as fast as a few femtoseconds, and the density of electrons accumulated at the conduction band follows the energy deposited in the medium. The relaxation of electrons back to the valence band, on the other hand, is slower: as the material relaxes through trap-assisted recombination over sub-picosecond to few picosecond timescales. However, since the relaxation happens through defect assisted Shockley-Read Hall mechanisms, it can, in principle, be sped up by increasing the defect density in the crystals [33]. In fact, sub-picosecond optical relaxation has been observed in aluminum doped-zinc oxide films operating near epsilon-near-zero (ENZ) (Fig 2a). Altogether, the excitation speed of free carriers in these materials is instantaneous (photoabsorption time), which is limited only by the temporal pulse width of the excitation laser. This allows for ultra-sharp changes in the refractive index upon light absorption (Fig. 2b), which can result in large frequency shifts [34,35], with the potential to achieve time-reflection [36] under a fast-enough change. In such interband processes the bottleneck is the relaxation speed. However, femtosecond-speed relaxations with intraband pumping are possible, where a sub-bandgap pump energizes conduction electrons, and changing the free carrier concentration which leads through an index change. As the carriers relax through thermalization to their original positions in the conduction band, the material returns to the original refractive index. This change can occur in a short time spanning a few hundred femtoseconds [20,37,38]. The use of TCOs has been demonstrated in reflectance and transmittance modulation, as well as in optical polarization switches [13,20,21], making them a promising platform for PTC realization. Furthermore, the large optical nonlinearities near their epsilon-near-zero regimes result in lower power requirements for the same refractive index changes, relaxing the power constraints [20,37].

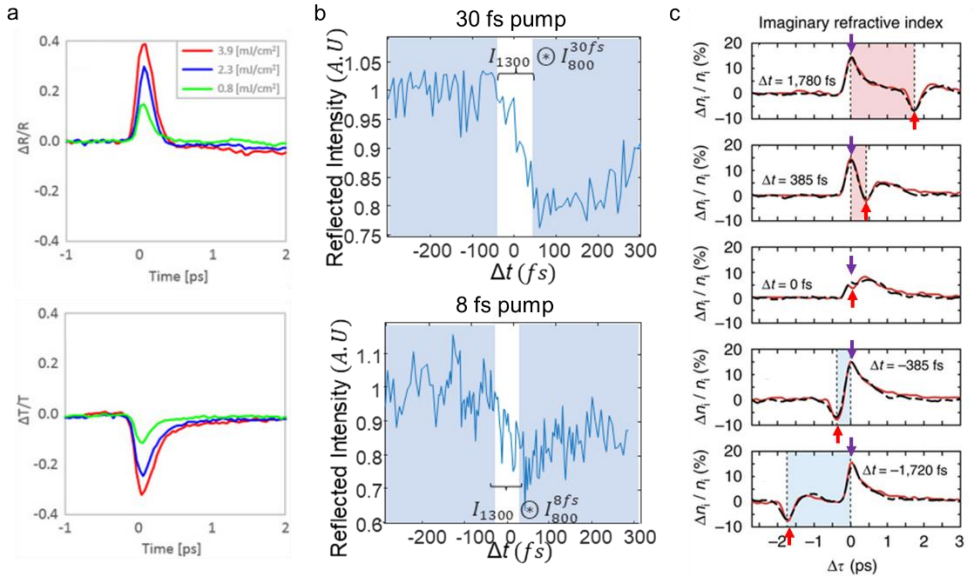

Figure 2 (a) Sub picosecond relaxation has been observed in aluminum-doped zinc oxide (AZO), with interband excitation [18] (b) The index change in free-carrier induced modulation is determined by the photoabsorption time (which is instantaneous), and is generally limited by the temporal width of the optical pump. As seen here, using an 8 fs pump instead of a 30 fs pump results in a much sharper optical transition [36] (c) The interband and the intraband nonlinearities can be added. An interband pump (blue arrow) increases the absorption of AZO, and an intraband pump (red arrow) decreases it. It can be seen that if both pumps hit the sample at the same time, the nonlinearities can cancel each other. [39]

3. Overcoming the limits of fixed relaxation time

While optical excitation of materials can be controlled by controlling the pump temporal width, the relaxation mostly depends on electron recombination and lattice cooling, which are intrinsic properties of the materials. Speeding up the relaxation time of materials requires continued exploration of materials with fast enough relaxation time. One viable method of achieving ultrafast changes (positive and negative) with sub-relaxation cycle times, circumventing the intrinsic relaxation time, could be to use two-color excitation. Addition of nonlinearities via concurrent, time-shifted pumping with both intraband and interband pumps has been showed in AZO [39]. This is achieved by using an interband pump to generate photocarriers, decreasing the refractive index, and instantly energizing the carriers to modify the index. This might provide a method to obtain a sharper temporal contrast in the permittivity, without relying on the material relaxation time. Another route to achieve ultrafast relaxation time, also with two-color excitations, could be to create population inversion within the conduction band for the lower frequency pump. In this situation, the lower frequency pump could force stimulated emission, which would reduce the density of energetic electrons instantaneously. It is not at all clear that this idea is physical, but it is worth trying.

4. Overcoming the power constraints

Finally, large and sharp optically induced changes in the refractive index call for millijoules of energy pumped into optical systems in femtosecond cycles, resulting terawatts of peak power [9]. This calls for an in-depth study of materials with large damage thresholds that can simultaneously undergo large index changes. Unity order changes have been observed in doped and undoped oxides with powers 10x lower than the damage threshold of the materials [26,32]. Refractory metals like TiN have been demonstrated to withstand powers over 1 TW [40], for high harmonic generation experiments, and show large reflectance modulation near their

epsilon-near-zero regimes [41]. Hybrid systems comprising refractory metals and nitrides may serve as another method of reducing the power constraints. Plasmonic antennae strongly coupled with epsilon-near-zero cavities have been shown to greatly boost nonlinearities due to the enhanced light-matter interaction and slow light effects. Such antennae have been shown to increase the effective optical nonlinearity [42], and even lower the power requirements for time refraction [8,32] and photon acceleration [43] in ENZ-nanoantenna systems, making them a promising route to alleviate the high-power requirements. In such system, robust epsilon-near-zero substrates can be used to increase the nonlinear interaction between the incident wave and antennae made of refractory metals, boosting the time-reflected signal.

5. Conclusion

While the ultrafast and large modulation of the material's optical response remains a challenge for the photonic time-crystal implementation, the range of materials that allow for a fast and substantial change of their refractive index is rapidly growing. At present times, the most promising class of materials that offers large optical response changes, fast modulation, and large damage threshold is transparent conducting oxides operating near their epsilon near zero regime. While TCOs promise to be viable candidates to implement PTCs, emerging materials such as monolayer $MoSe_2$ [44], and transdimensional films [45], also show promise in terms of speed and modulation depth. Techniques to reduce the modulation power requirements while sustaining PTC effects call for both rigorous theoretical study of different schemes of refractive index modulation, with a continued quest for finding better, faster, and more laser tolerant materials.

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The authors declare no conflicts of interest.

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