Ultrafast All-Optical Metasurfaces: Challenges and New Frontiers

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Abstract

Dynamic metasurfaces have emerged as a disruptive change in the way the response of optical systems can be tailored, combining the flexibility of flat optics in spatially engineering materials at the nanoscale with the opportunity to reconfigure the metasurfaces' properties reversibly upon external stimuli over time. In this context, the far-reaching interest in pushing the tuning speed has driven the development of 'ultrafast all-optical metasurfaces', in which transient nonlinearities photoinduced by femtosecond laser pulses empower to achieve >GHz modulation rates. While holding great promises to unlock forefront applications, the future frontiers of this class of spatiotemporal all-optical metasurfaces are accompanied by formidable challenges. In this Perspective, alongside a brief panorama of the state-of-the-art, we spotlight some among the emerging frontiers for ultrafast light-driven metasurfaces, with special emphasis on the all-optical control of light, the enhancement of light-matter interactions, and the time-variant frequency conversion, hoping our vision will prompt new ideas and horizons to explore.

Keywords

Dynamic metasurfaces, Ultrafast reconfiguration, Photoinduced nonlinearities, All-optical control, Light-matter interactions, Time-variant metamaterials

1. Introduction

Optical metasurfaces have realized a paradigm shift in the way light can interact with matter. These are ultrathin photonic components consisting of planar sub-wavelength arrangements of (usually periodic) resonant nanoantennas, referred to as 'meta-atoms'.[1,2] Thanks to their quasi two-dimensional (2D) ordered configuration in space, these nanopatterned layers exhibit extraordinary optical properties with no counterpart among thin layers of naturally

available materials or other nanophotonic structures.[3,4] Meta-atoms can feature virtually any shape, and the materials most often employed span from plasmonic media (e.g. Au, Ag, Cu, Al, Ni, or TiN, HfN, etc.) and epsilon-near-zero (ENZ) materials (transparent conductive oxides as indium-tin-oxide, ITO), to semiconductors (e.g. Si, Ge, GaAs, AlGaAs, SiO_2, TiO_2, etc.) and 2D materials, such as graphene, or transition metal dichalcogenides (TMDs), as sketched in Fig. 1. In parallel, tweaking the periodic ordering of nanoresonators allows for broadening the range of optical resonances supported by metasurfaces, which encompass: (i) localized resonances governed by the single meta-atoms, as localized surface plasmons or Mie-like resonances, which occur when either metallic or all-dielectric media are utilized, respectively; and (ii) extended modes with a collective, nonlocal character, as (quasi-)bound states in the continuum (BICs), surface lattice resonances, or guided mode resonances, featuring much higher quality (Q)-factors (depicted in Fig. 1). Owing to such flexibility, metasurfaces have come into the spotlight as the ideal platform for a wealth of diverse photonic applications and functionalities, implemented within thicknesses of hundreds of nanometers beyond the rules of diffractive optics.[5–14] With the progress of nanofabrication, even large-scale devices for real-world applications are nowadays available and commercialized.[15]

While outstanding advances have been demonstrated and the number of configurations explored so far is notably ample, most of the current metasurfaces implement static operations and exhibit optical properties which are irreversibly fixed at the fabrication stage. The interest in benefitting instead from tunable platforms has recently prompted the increasing momentum gained by 'active' metasurfaces. The underlying concept is simple: an external stimulus is used to alter dynamically the optical properties of the metamaterial, whose response is thus driven and reversibly modulated in time. This creates a new class of (2+1)D 'spatiotemporal' nanostructures in which time, akin to space, acts as a new degree of freedom to control the metasurface behaviour.

Driven by the great promises of dynamic metasurfaces, various strategies and reconfiguration regimes have been explored, which are characterized by modulation speeds spanning several orders of magnitude.[16,17] Indeed, the timescale to achieve the sought active tuning is concurrently dictated by (i) the nature of the external stimulus, and (ii) the operational physical mechanism regulating the modulation of the optical properties. Successful implementations[18] have for instance employed electrical gating, voltage-controlled carrier injection, light-driven free carrier density variation, mechanical actuation (e.g. with MEMS), phase transitions (e.g. in liquid crystals or phase-change materials), magnetic or thermal gradients. The achieved modulation times range from ms to μs (rates of kHz to MHz), with phase-change materials or mechanical modulations, to ns and sub-ns, e.g. for thermo- and magneto-optic effects, down to the tens of ps regime, when the free carrier density is modified by electrical or optical means, e.g. in ENZ or excitonic 2D materials.

Of all the possible operational mechanisms to tailor metasurfaces in time, special attention has been drawn to optical nonlinearities as the elective approach to maximize the speed of reconfigurability. Indeed, optical media exhibiting Kerr-like effects (i.e., changes of the complex refractive index proportional to light intensity) respond to stimuli down to the sub-ps timescale.[16] The challenge of driving dynamically such nonlinearities in this temporal regime can be addressed by pulsed mode-locked lasers, which provide ultrafast (femto- to picoseconds), highly frequency-tunable light pulses.[19] By capitalising on the high peak intensities and short temporal durations of the fs-light pulses inducing the Kerr nonlinearities, ultrafast modulation schemes known as 'all-optical' (i.e., where light both produces and experiences changes in the material optical properties) can be realised, to reach tuning rates up to tens of GHz or even higher, beyond those achievable with any other approach.[20] Metasurfaces, thanks to their structural resonances and distinctive light confinement effects, contribute to enhancing the (often weak otherwise) interaction of photons and nonlinear

media, a relevant advantage compared to conventional architectures. The combination of optical nonlinearities, femtosecond lasers, and the flat optics advantages creates what is referred to as an 'ultrafast all-optical metasurface', which paves the way to conceive unique reconfiguration approaches operating on demand upon photoexcitation in the ultrafast regime.

The arsenal of mechanisms empowering the ultrafast modulation of the meta-atoms' optical properties is rather diverse. Although different light-induced processes preside over the onset of the nonlinearities at play depending on the material (a brief overview is presented in the Supporting Information Section S1), the dominant effects can all be interpreted as a delayed third-order nonlinearity governed by 'hot' carriers, namely photoinduced electron-hole pairs featuring high energies and ultrafast relaxation times.[21–28] The nonequilibrium occupancy distribution of these short-lived carriers introduces changes in the allowed optical transitions within the material, varying its permittivity in the spectral domain.[29–34] Aside from the specific platform and modulation mechanisms involved, the metasurface response is modified on the timescale from tens of femtoseconds to a few picoseconds, positioning the laser-pulse-driven control via hot-carrier-mediated Kerr nonlinearities among the fastest reconfiguration approaches.

With this Perspective, we aim to discuss some of the intriguing emergent prospects for ultrafast all-optical metasurfaces. By firstly presenting a brief overview of some advanced, experimental and theoretical methods, we outline the potential of these tools to investigate the dynamical response of reconfigurable metasurfaces on femtosecond timescales. We then focus on possible applications and challenges, by spotlighting a few recent (i.e. within the last five years) results suggesting new horizons for these optical active nanostructures. A special emphasis is given to the ultrafast control over the properties of light, the manipulation of strong light-matter interactions, and the frequency conversion in time-variant systems. It is not our intent to provide a comprehensive review on dynamically tunable metasurfaces, for

which we address the readers to more focused publications (see e.g. refs.[18,35–41] to mention a few). Here we rather aim to highlight forefront techniques and emerging scenarios to exploit optically tunable metasurfaces in the ultrafast regime, which might be useful to conceive future experiments and design novel enhanced metamaterials.

The interest in pushing the current speed bounds of spatio-temporal metasurfaces and unseal full control over their behaviour in the ultrafast regime stems from the intriguing prospects of demonstrating active, ultracompact and energy-effective photonic architectures with >GHz modulation rates. Several exciting applications can be envisaged.

- Similar optical components could serve to dynamically switch between two states following an optical command signal, towards the development of logic circuits and optical units for processing and transmitting data in the form of optical signals at unparalleled speeds. This could transform the fields of signal processing, optical computing and communication, with integrated nanophotonics synergistically complementing electronics. Besides, the ultrafast active control over the optical properties of a highly flexible system as metasurfaces is highly attractive for applications such as dynamic imaging, augmented reality, light detection and ranging (LIDAR), holographic displays, waveform active shaping and light structuring, or thermal emission engineering.

- Light-driven active metasurfaces could also grant access to the dynamic manipulation of ultrafast light-matter interactions in molecular or solid-state systems coupled to the meta-atoms array. This would bring additional knobs to the exploration of hybrid effects resulting from the coupling, such as resonance interferences or polaritonic states, of potential relevance beyond photonics across physics and chemistry.

- The temporal control of dynamic metasurfaces at ultrafast speed has been shown to lead to entirely new physical phenomena which are out of reach in the steady state or upon slower modulations. Exotic effects have been examined,[42–48] posing completely

new fundamental questions and technological challenges. In this vein, ultrafast all-optical metasurfaces become the pivot to enter into this peculiar regime otherwise inaccessible, where modulations are as fast as the optical signal. In principle, by further engineering space- and time-varying meta-atoms, periodic modulations with rates approaching the optical carrier frequency could lead to photonic Floquet phenomena (i.e., peculiar of systems characterised by a periodic dependence on time[49,50]).

This Perspective is organised as follows. After the brief panorama provided above of the state-of-the-art in reconfigurable metasurfaces, in Section 2 we present both experimental and modelling techniques to characterise and exploit the unique properties of all-optical metamaterials in the ultrafast regime. Section 3 discusses some of the emerging applications at the forefront of research on ultrafast active metasurfaces. Finally, Section 4 concludes the manuscript and draws prospect blueprints that, we hope, will motivate future works and inspire new intriguing directions to explore.

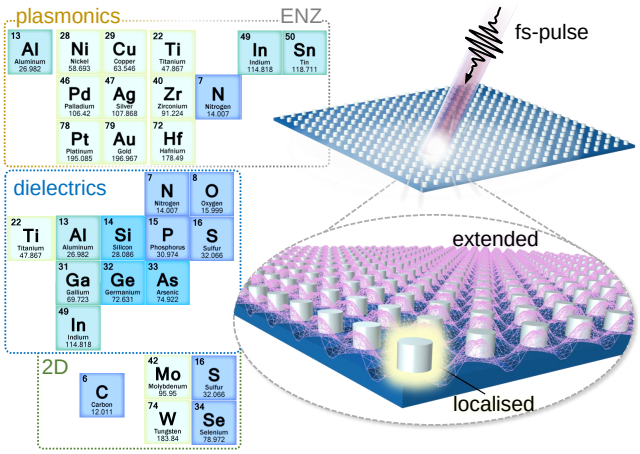


Figure 1: **Ultrafast all-optical metasurfaces.** Sketch of an optical metasurface activated by an ultrafast light pulse. The typical materials employed (left, divided by category) and a pictorial visualisation of the possible resonances (either local or nonlocal) supported by the metasurface are also depicted.

2. Advanced Methods for the Study of Ultrafast Active Metasurfaces

2.1 Experimental Techniques

To characterise all-optical metasurfaces and exploit their functionalities in the ultrafast regime, advanced experimental tools enabled by femtosecond pulses are required. Ultrashort pulses generate nonlinear effects at the nanoscale that can be exploited in optical spectroscopy experimental schemes. While several techniques with different degrees of sophistication have been developed, transient absorption (TA) spectroscopy remains the simplest, yet most powerful approach. TA spectroscopy typically works in a stroboscopic fashion, using a sequence of two pulses: a first intense 'pump' pulse triggers a dynamical photophysical process of interest, while a second delayed broadband pulse, the 'probe', monitors in real time the evolution of the excited system by measuring changes of its absorption spectrum (Fig. 2, top). For details on the technical implementation of TA experiments, we address the readers to more exhaustive reviews.[51] Although conceptually simple, ultrafast TA has provided an enormous amount of information on photoinduced dynamical processes in (bio)-molecules, nanostructures and solids.[19] In the context of all-optical metasurfaces, TA is the natural implementation of an all-optical scheme for multiple reasons: (i) the pump allows pulsed photoexcitation to induce nonequilibrium processes; (ii) the probe resolves the dynamics of the photoinduced optical response of the system; (iii) the rational interplay over time between the two pulses allows to achieve dynamic control of the properties of either the probe light or the metasurface.

Considering the notable variety of possible interactions between light and metasurfaces, it is essential to access a large set of information on the transient state of the detected probe light, by retaining its broadband spectrum (possibly collecting the nonlinear harmonic signals generated by the probe), or by following its properties beyond the mere amplitude

(e.g. polarisation, phase, momentum). Figure 2 (top) schematically reviews some of the pump-probe techniques that, by resolving the probe light over a specific extra-dimension, are suited to perform advanced, comprehensive analyses of this kind.

- *Polarization-resolved*: Determining the polarisation state of light upon interaction with the metasurface requires tailored control of the pulses both at the excitation (before the sample) and detection (after the sample) stage. In the easiest configuration, the probe light is decomposed into orthogonal polarization components (TE/TM for linear polarization or left/right helicity for circular polarisation) at the excitation stage and their intensities are separately recorded at the detection stage, allowing to retrieve the transient dichroism, either linear (difference between TE and TM components) or circular (difference between left or right circular polarizations). More complex schemes introduce optical phase-retarder elements at the detection stage, allowing access to the phase of the probe light via polarimetry measurements,[52] to reconstruct the transient complex birefringence. Furthermore, controlling the polarisation state of the pump pulse at the excitation stage could modify the optical symmetry of the metasurface. Then, by tuning the pump and probe polarisations with respect to each other in various schemes, depending on the measured signal, the photoinduced transient anisotropy can be revealed at the detection stage.

- *Space-resolved*: Ultrafast TA microscopes[53] can be implemented in several configurations: (i) real space ultrafast imaging is typically performed to achieve diffraction-limited spatial resolutions (on the order of a few hundreds of nanometers in the visible spectral range) in a point-scanning confocal geometry or in wide-field geometry. Analyses down to the single meta-atom level, or studies on the beam profiles at the output of the metasurface become accessible. (ii) Fourier space imaging[54] resolves the metasurface response over the reciprocal space. This allows for retrieving the dispersion relations of the metasurface resonances or the dependence of light-matter interactions on the light momentum (or, equivalently, the angle of incidence, in the most common

implementations).[55]

- *Excitation-frequency-resolved*: Complex systems supporting multiple absorption resonances require spectral selectivity over the excitation frequency, while still retaining the ultrafast temporal resolution. Two-dimensional electronic spectroscopy (2DES), which correlates over time excitation and detection frequencies, is the elective tool to dissect correlations, coherences and intricate phenomena.[56] 2DES is typically employed in molecular-based systems, thin semiconductors or hybrid complexes.[19,53,57,58] However, we foresee it might be extended to study excitation-frequency-resolved light-matter interactions in ultrafast metasurfaces.

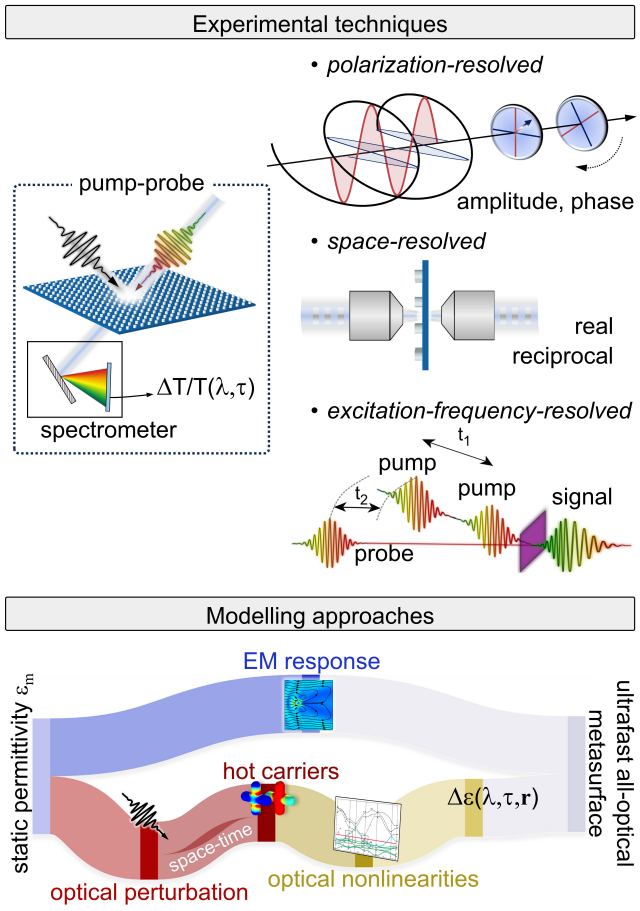


Figure 2: **Advanced methods for the study of ultrafast all-optical metasurfaces.** Sketches of the typical experimental (top) and modelling (bottom) techniques at the basis of the study and the exploitation of ultrafast all-optical metasurfaces.

2.2 Modelling Approaches

The tantalizing opportunities and new research frontiers of ultrafast all-optical metasurfaces rely on the understanding of the rich landscape of nonequilibrium mechanisms which preside over the connections between light pulses, the charge carriers in the material, the near- and far-field optical response of the metastructure. Complementing experimental investigations with modelling tools capable of describing the ultrafast photophysics of relevant systems is paramount. Accurate simulations have the twofold advantage of (i) producing outcomes directly comparable to experimental data, to dissect and disentangle experimental observations; and (ii) providing predictive results, to design optimal platforms and guide experiments.

In the most general scenario, modelling the ultrafast optical phenomena taking place in photoexcited metasurfaces involves a series of conceptual steps, schematically depicted in Fig. 2 (bottom) and further detailed in the Supporting Information Section S2. The foremost information to begin with is the material composing the meta-atoms or, more precisely, its optical properties in unperturbed conditions, which are fully determined by its static permittivity ε_m. Once ε_m is known, the geometrical characteristics of the metasurface establish its optical response in the linear regime, both in the near (electromagnetic fields distribution, induced currents, etc.) and in the far field (optical observables as transmittivity or reflectivity). Analytical or numerical full-vectorial techniques can be utilized to reproduce the metamaterial linear electromagnetic (EM) behaviour, by accounting for the shape and size of the meta-atoms, the array periodicity and its planar spatial arrangement.

While it is relatively straightforward to numerically characterise static metasurfaces in the linear regime, modelling the transient nonlinear response of an ultrafast all-optical metasurface is more elaborate. This demands to determine, with (at least) temporal and spectral resolution, the dynamic modulations of the material permittivity $\Delta\varepsilon$, which in turn ensue

from a cascade of phenomena triggered by the photoexcitation with an ultrashort light pulse. Accordingly, some key ingredients need to be included in such models:

- the pulsed optical perturbation, i.e. how the photon energy of the exciting (pump) pulse is absorbed by the metasurface to transiently bring the system out of equilibrium;

- the energy distribution of the photogenerated electron-hole pairs and their ultrafast relaxation dynamics (possibly with spatial resolution across each meta-atom): the most rigorous description involves first-principles calculations,[59–61] yet a promising alternative is to employ rate-equation (two/three-temperature) models,[62,63] whose simple formulation based on a classical thermodynamics approach (the carrier dynamics is treated as a flow of energy across relaxation channels, in analogy to the thermalisation process of thermal reservoirs), is especially general, hence suitable for a large variety of materials;

- the material optical nonlinearities, i.e. how the nonequilibrium short-lived electronic states affect the permittivity of the nanostructured material: this connection is realised by the electronic band structure, since modifying the energy occupancy distribution of carriers amounts to depleting/filling states with respect to the unperturbed case, which entails a change in the allowed optical transitions. This translates into a modulation of the material permittivity $\Delta\varepsilon$ over a broad range of (probe) photon energies, whose calculation can be accomplished via a semi-classical formalism for light-matter interaction;

- the resulting ultrafast broadband modulation as a function of time (the pump-probe delay) and (probe) wavelength of the metasurface optical response: the nonequilibrium EM behaviour is computed considering a modified time-dependent permittivity $\varepsilon(\lambda, \tau, \mathbf{r}) = \varepsilon_m(\lambda) + \Delta\varepsilon(\lambda, \tau, \mathbf{r})$.

Considering the state-of-the-art techniques to model ultrafast all-optical metasurfaces, some open challenges and future frontiers could be envisioned:

- quantifying the optical nonlinearities in new materials, by analysing their band structure and identifying the optical transitions relevant to $\Delta\varepsilon$, so as to expand the portfolio when designing metasurfaces.

- filling the gap between rigorous first-principles and simplified rate-equation (thermodynamic) models in treating the photogeneration and relaxation of hot carriers. Towards this direction, we spotlight the recently reported Quantum Two-Temperature Model,[23,64] which retains the rate-equation formalism, yet estimates the carrier generation rate based on refined quantum arguments.

- pursuing a self-consistent description of the pump absorption in time-domain. With this aim, some (partly approximated) approaches have been recently reported for isolated nanoparticles,[65,66] and an ITO-based metasurface.[67] Perfecting these approaches is of critical relevance, as they are expected to be essential for modelling ultrafast all-optical Floquet metamaterials, in which the optical modulations induced by a fs-periodic drive could not be treated quasi-statically.

- overcoming the semi-classical formulation of rate-equation models to treat coherent optical phenomena resulting from the ultrafast photoexcitation. This is anticipated to advance the study of the nonlinear processes of light emission, or the coupling mechanisms in hybrid metasurfaces.

3. Challenges and New Frontiers of Research

3.1 Control of Light with Nonlinear Metasurfaces

Combining metasurfaces with femtosecond laser pulses emerges as an ideal approach to moulding the state of light dynamically on ultrafast timescales and in highly tailorable architectures. The use of ultrafast pulses to tune the attributes of light via the transient nonlinearities of nanostructured materials has been already explored, and demonstrated to be a powerful technique. For instance, this all-optical strategy has laid the foundation for the field of ultrafast active plasmonics, which originally involved propagating surface plasmon polaritons (typically on metallic films or waveguides) to achieve optical modulation.[68–74] Beyond noble metal surfaces, the same concept has been subsequently applied to thin films of semi-metals or ENZ materials,[75,76] and implemented in more advanced configurations such as microcavities[77,78] or colloidal nanoparticle ensembles,[79,80] contributing to an ample literature on the topic.[81] None of these active nanostructures, however, offers the same degrees of freedom to confine and engineer nanoscale light-matter interactions as flat optics does. This exceptional feature of metasurfaces confers on them unique temporal, spatial and spectral tunability, setting the stage for addressing some of the open challenges of the ultrafast all-optical control of light, as for instance:

- producing different, slower and faster switching speeds within the same platform
- choosing dynamically the system modulation rate via an external light stimulus
- triggering distinct responses by spectral tuning of the excitation, so to interleave different optical control functions in the same device
- suppressing slower, detrimental contributions which hinder the switching speed
- developing compact, lightweight architectures to be conveniently integrated with well-established platforms (e.g. electronics and semiconductor photonics)

- pushing the spectral limits of all-optical modulators, e.g. to the extreme ultraviolet[82]
- carving the interaction conditions at the subwavelength scale, which is unattainable without the control over the configurational characteristics (e.g. spatial orientation) of the nanoresonators (i.e., without an ordered arrangement)
- imparting spatial profiles to the temporal modulation
- tailoring light on demand beyond mere changes in amplitude (which is the most conventional yet least powerful approach), so as to access the reversible, ultrafast manipulation of further properties such as phase, polarisation, or even the linear, spin angular (connected to circular polarisations), and orbital angular momentum of photons.

To date, some promising approaches have been proposed. Among the most established platforms, plasmonic materials have been widely explored (see e.g. refs.[83–87] and references therein). Despite the remarkable results, plasmonic metasurfaces inherently pose some limitations to the speed (due to the characteristic times of carrier relaxation, typically several ps) and the efficiency (due to the metal resistive losses) of the optical modulation. To circumvent such shortcomings, more sophisticated platforms and ideas have been devised. Figure 3 reviews some of the most noteworthy advancements achieved towards this direction: Fig. 3a reports the expedited control over the phase of visible light via electron transport;[88] Fig. 3b illustrates an approach based on carrier diffusion to beat the speed limits imposed by carrier relaxation;[89] in Fig. 3c diffraction management is demonstrated with an all-dielectric metasurface;[90] Fig. 3d presents results on photoinduced transient chirality.[91]

A first possible route to expand the limits set by the intrinsic nonlinearities of plasmonic meta-atoms is to integrate different materials within the same metasurface. The resulting system exhibits extra spectral and temporal tuning opportunities, as the interplay of distinct materials can be advantageously engineered. This is the rationale behind the platform

proposed in ref.[88] to expedite the manipulation of visible light (Fig. 3a). The system,[92] investigated by means of ultrafast polarisation-resolved pump-probe spectroscopy (pump pulse duration 150 fs), angle-resolved static ellipsometry and time-resolved polarimetry, consists of an array of Au nanorods lying on a thin film of the ENZ material ITO, possessing a polarisation-selective high-Q extended resonance. In the ultrafast regime, the plasmonic hot carriers govern the nonlinear optical modulation, but ITO possibly acts as electron acceptor. When carriers have enough energy to be injected from Au to ITO, an extra relaxation pathway opens, resulting in a faster decay of the differential reflection ($\sim$200 fs instead of $\sim$7.5 ps). By monitoring the dynamic state of the output light, linear polarisation transiently evolves into elliptical due to a large ($>20^\circ$) photoinduced phase difference between the field components with a $\sim$1 ps recovery time, much faster than what observed when carrier injection at the Au/ITO interface does not occur ($\sim$13 ps).

In passing, we note that metasurfaces combining plasmonic metals and trasparent conductive oxides have also been proven to support increased nonlinearities and efficient conditions to modulate light in the near-infrared, thanks to the peculiar features of ENZ materials.[93–97]

An alternative strategy to circumvent the speed constraints of plasmonics consists in breaking the paradigm of driving the modulation via the relaxation of hot carriers. Towards this direction, the electron spatio-temporal diffusion within the single meta-atoms (i.e., at the intra-particle level) has emerged as a promising choice.[98–102] An implementation of this concept used a square array of Au isotropic nanocrosses to manipulate the light polarisation with sub-ps recovery time.[89] Resonant pumping with an ultrashort (sub-30-fs) pulse polarised along one of the symmetric arms of the crosses promotes a non-uniform spatial distribution of nonequilibrium carriers within each meta-atom. In turn, the carrier-mediated inhomogeneous permittivity modulation opens an optical symmetry-breaking window, whose fingerprint is a transient anisotropy that can be revealed in a polarisation-resolved pump-probe experiment. By collecting the probe light transmitted along the directions parallel and

perpendicular to the pump polarisation, the difference of the two signals (identically zero in unperturbed conditions) measures the photoinduced anisotropy, while its lifetime informs on the optical symmetry recovery time. The background-free experiments, in excellent agreement with a model for the carriers' local homogenisation, show a broadband linear dichroism vanishing in $<$1 ps, long before carrier relaxation (Fig. 3b). As the process is reversible and fully all-optical, the approach allows for tuning the anisotropy on demand by optical means. The hot-carrier pattern can be shaped by rotation of the pump polarisation, offering a way to change the output polarisation with the exciting pulse, up to a sign reversal of the transient dichroism.[103]

While more sophisticated approaches can speed up the modulation of plasmonic all-optical platforms, none of them enables to reduce the intrinsic, large absorption losses of metals. Switching to alternative material platforms is mandatory for this challenge, which can be addressed with high-index semiconductors, representing excellent candidates to develop more power-efficient architectures.[34,104–106] Among the illustrative examples, the all-optical tuning of Mie resonances in an all-dielectric metasurface has been shown to enable the ultrafast manipulation of light diffraction with rather moderate fluences.[90] In this demonstration (Fig. 3c), a GaAs metasurface is designed to support both dipolar and quadrupolar high-Q Mie modes, whose radiations are destructively combined so as to suppress, in unperturbed conditions, the array first diffraction order.[107–109] By exciting the system with an ultrashort (duration 85 fs) pulse of 215 μJ/cm^2 fluence, the ultrafast photo-injection of free carriers takes place, and produces a decrease of refractive index, blue shifting the multipole resonances. This allows to couple radiation and transiently turn 'on' the otherwise latent diffraction orders. Broadband pump-probe experiments and simulations demonstrate the diffraction switching with a 10 ps recovery time. At higher fluence pumping, thermo-optic effects arise and prolong the metasurface response modulation, deteriorating the switching rate.

Note that, although direct-gap semiconductors are typically favourable in terms of amplitude of the carrier-mediated modulation,[110–113] also indirect-gap materials represent an advantageous choice. In particular, Si[114–116] and Si-rich nitride[117] are especially attractive, since they are fully CMOS-compatible and operate in the visible. In the ultrafast regime, Si-based metasurfaces have been proven to offer an efficient avenue to manipulate light.[118–120]

A further property of light whose manipulation garners great interest is chirality. Attaining reversible high-speed chirality switching and developing platforms with on-demand invertible chiral states would have a broad impact across physics, chemistry and biology, advancing e.g. the techniques for the dynamic detection of chiral biomolecules, or the study of light-matter interactions in opto-spintronic systems, such as 2D materials and van der Waals heterostructures. In this regard, a strategy has been proposed to reconfigure the chiral state of light on ultrafast timescales by photoinducing chiroptical effects in an otherwise achiral metasurface.[121] In a recent implementation (Fig. 3d), an array of symmetric Au split-ring resonators lying on Au nanostripes is used to achieve complete handedness inversion in the visible, whose speed (dictated by the carrier intra-particle diffusion) outperforms the conventional approaches.[91] Upon ultrafast illumination, the photogenerated hot carriers, with spatial patterns determined by the exciting pulse polarisation, modify anisotropically the material permittivity, breaking the optical mirror symmetry prior to homogenisation. Broadband ultrafast pump-probe spectroscopy (pump pulse of $\sim$90 fs) and transient Stokes polarimetry are employed to demonstrate ultrafast photoinduced ($<$5 ps) circular dichroism and optical rotatory dispersion, whose sign is controlled by adjusting the pump polarisation. Finally, the near-field chirality is shown to produce a chiral response also in the far field, thanks to the space dependence introduced by hot carriers in the optical nonlinearities.

The selection of works presented above provides, with no claim of exhaustiveness, an outline of original strategies for the high-speed control of light via photoinduced nonlineari-

ties in ultrafast all-optical metasurfaces beyond the conventional techniques. Note that such transient nonlinear effects can be also superposed to static nonlinear properties, which is the concept underpinning several promising demonstrations of the ultrafast all-optical control over nonlinear higher-harmonics generation processes in metasurfaces.[122–126] Several open questions remain, yet exciting application prospects can be envisaged. For instance, renewed approaches to the ultrafast optical manipulation could advance the development of all-optical switches that, serving as the optical analogue of transistors, represent the starting block for integrated nanophotonics. Controlling light by light could allow for processing information at higher speeds and operating at lower energy consumptions. In addition, the possibilities granted by metasurfaces to mould light open new ways of carrying streams of information, possibly leading to critical technological progresses such as: (i) higher transmission rates, by packing and multiplexing bits of information more densely onto the optical signal; (ii) more reliable data networks, by encoding the information in more stable properties, so as to retrieve it with high accuracy even under lower signal-to-noise operation conditions.

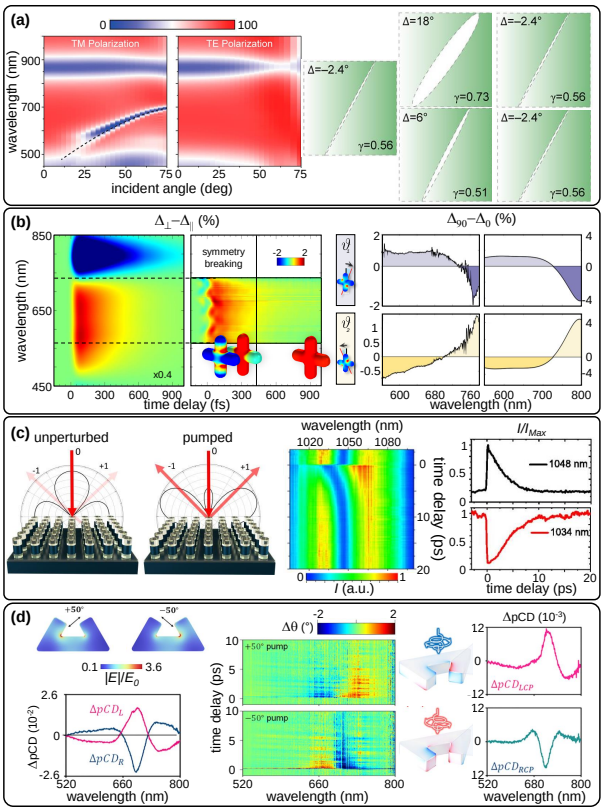


Figure 3: **Ultrafast control of light with nonlinear metasurfaces. a) Manipulating light phase with a gold metasurface lying on a ENZ (ITO) thin film.** From left to right: simulated polarisation-sensitive static response resolved in angle; measured static and transient (short and long time delays) polarisation ellipses for the on- (top) and off-resonant (bottom) ultrafast pump at a selected wavelength. Adapted with permission from ref.[88] Copyright 2019, American Chemical Society. **b) Sub-ps polarisation control via spatio-temporal hot carriers.** From left to right: simulated and measured sub-ps broadband linear dichroism photoinduced in the Au nanocross metasurface. The transient symmetry-breaking window is highlighted, and the corresponding spatio-temporal patterns of the nonlinear permittivity modulation (imaginary part at a selected exemplary wavelength) are depicted; simulated and measured dichroic spectra (100-fs delay) for a pair of pump pulses with polarisation differing by 90°. Adapted with permission from refs.[89,103] Copyright 2020, Springer Nature, and copyright 2022 Author(s), licensed under a CC-BY Creative Common Attributions 4.0 License. **c) Low-fluence ultrafast diffraction switching.** From left to right: illustration of the ultrafast all-optical diffraction switching; measured pump-probe map of the first diffraction order (non-normalised) and temporal traces of the intensity at selected wavelengths for off-to-on (top) and on-to-off (bottom) switching; measured spectra for higher fluence pumping. Adapted with permission from ref.[90] Copyright 2021, AIP Publishing. **d) Light-enabled control of chirality in achiral metasurfaces.** From left to right: simulated near fields, measured (by isolating the photoinduced term only) differential circular dichroism and measured optical rotation upon pumping with mirror-inverted linear polarisations; measured differential circular dichroism upon pumping with either left or right circular polarisations. Adapted with permission from ref.[91] Copyright 2023 Author(s), licensed under a CC-BY Creative Commons Attributions 4.0 Licence.

3.2 Enhancement of Light-Matter Interactions with Hybrid Metasurfaces

Besides the all-optical structuring of light in the far field, metasurfaces provide a flexible platform for manipulating light-matter interactions at the nanoscale. Indeed, metasurfaces support intense local electromagnetic fields which can be shaped in phase and amplitude at the deep subwavelength scale, and finely engineered to tailor near-field interactions.[127] In these terms, metasurfaces can be thought of as on-demand optical resonators, and used in cavity-like architectures in interaction with emitters or molecules, creating 'hybrid' systems. Such configuration has lately emerged as an extraordinary route to access weak- and strong-coupling regimes, opening a host of opportunities in photocatalysis, biochemical sensing, and polariton chemistry.[128]

Pushing the current state-of-the-art with the aim of probing in real time dynamic light-matter interactions at the nano- and molecular scale calls for developing active hybrid architectures responsive to femtosecond-laser stimuli. The interest in this new frontier holds especially true for the regime of strong light-matter coupling, an exceptional hybridisation condition between optical modes and photoexcited states in matter, which is attracting growing attention in various contexts (we refer the readers to previous reviews for a detailed description[129–133]). In this framework, ultrafast all-optical metasurfaces functioning as 'metacavities' feature some critical advantages:

- intense near fields and strong light confinement in the proximity of meta-atoms, which contribute to enhancing the (linear and nonlinear) interactions of photons and materials.

- unparalleled degrees of freedom for tailoring resonances' spectral position, near-field modal profiles and modes' (cavity) lifetime, towards a highly-tunable coupling.

- unique ease of input/output coupling due to their character of 'open' cavities, in stark

contrast with more conventional Fabry-Pérot microcavities.

- the use of resonances supported by all-optical metasurfaces offers the possibility of actively controlling dynamic photoinduced processes via the ultrafast modulation of the optical properties of the meta-atoms; this allows for manipulating the ultrafast response of the hybrid system in its light component, in addition to the matter one.

To date, several configurations have been explored, relying on either plasmonic or all-dielectric metasurfaces coupled to excitonic 2D materials, quantum dots, and organic molecules. Without any intent of exhaustiveness, Figure 4 showcases some intriguing examples: in Fig. 4a an enhancement of all-optical switching performances is achieved in a hybridised plasmonic/photonic grating,[134] Fig. 4b illustrates the ultrafast all-optical control of strongly-coupled intersubband polaritonic meta-atoms,[135] Fig. 4c presents the ultrafast modulation of the exciton/plasmon coupling in a TMD/Au hybrid nanostructure,[136] Fig. 4d reports the enhanced transport of exciton-polaritons in a silver metasurface strongly coupled to 2D perovskites.[137]

The spectral overlap between local (mediated by the single nanoresonator) and nonlocal (due to collective effects within the ensemble) modes of the metasurface to create hybridised states is the condition achieved in ref.[134] The work employs a Au nanostripe array embedded in a dielectric slab to access the strong coupling between the nanostripes' localised surface plasmon and a high-Q Rayleigh anomaly (Fig. 4a). By varying the incidence angle, and accordingly the light in-plane momentum, the extended resonance properties are readily tuned, and two polariton branches arise, mixing the local and nonlocal characters of the two modes. Ultrafast angle-resolved pump-probe spectroscopy is used to track the temporal dynamics of the light interaction with the hybrid system upon hot carrier photoexcitation, and to retrieve the transient modulation of the angular optical dispersion. The results indicate an enhancement of the switching performances (larger modulation contrast) at the early stages

(<2 ps) of the dynamics near the polaritons' avoided crossing, interpreted in terms of the influence of the coupled modes on the polariton bands. Moreover, the hybrid metasurface affords to tailor the dispersion of the pump-probe signal relaxation time, suggesting an extra post-fabrication way of controlling the switching speeds by adjusting the modes' coupling strength.

An efficient alternative to couple optical modes to matter excitations in a flat-optical cavity is to produce hybrid meta-atoms, interleaving both the light and the matter component in each unit cell. This can be done by directly nanostructuring excitonic materials (as TMDs or perovskites) to constitute the meta-atoms,[138–141] or by embedding distinct materials in each nanoresonator. The latter approach is at the basis of the THz polaritonic metasurface presented in ref.,[135] whose unit cell consists of Au patch antennas strongly coupled to the intersubband transition of a multi-quantum-well (MQW) heterostructure (Fig. 4b). The system exhibits a giant third-order nonlinearity, enhanced by the hybrid architecture and regulated by the photoinduced depletion of the MQW ground state, which reduces the intersubband oscillator strength, and accordingly the coupling to the plasmonic antenna optical mode. In the frequency domain, this translates into the transition from two resonant peaks (arising from polaritonic splitting) in unperturbed condition to a single peak upon MQW absorption saturation. Such nonequilibrium mechanism can be driven all-optically in the ultrafast regime by photoexciting the metasurface with intense pump pulses (∼1.6 ps duration), which transiently modify the MQW-plasmon coupling constant. A recovery time of <2 ps is observed, demonstrating the ultrafast all-optical control over the polariton branches, and suggesting a new approach to manipulate light-matter interactions in hybrid meta-atoms.[142]

Hybrid ultrafast metasurfaces can also consist of nanoresonator arrays covered by films of the photoactive material poised to interact with the meta-atoms' optical modes. In this

configuration, seminal results were obtained with molecular aggregates.[143] More recently, in the implementation reported in ref.,[136] a plasmonic metasurface is integrated with a WS_2 monolayer, enabling to couple a plasmonic mode to the TMD exciton (Fig. 4c). The signature of the coupling is a Fano-type interference manifesting itself as a resonance in the steady-state spectra, while ultrafast photoexcitation produces the instantaneous switch-off of the Fano resonance, with a recovery starting in the sub-100-fs range. Such dynamics is qualitatively interpreted as due to the transient modulation of the exciton/plasmon coupling strength. The photoinduced carrier injection into the TMD excited state and subsequent decrease in ground-state absorption reduce (increase) the oscillator strength (damping rate) of the excitonic resonance (most likely dominating over plasmon modulation). The coupling with the plasmonic resonance is thus degraded at the onset of the hybrid system ultrafast response, and recovered upon exciton radiative decay to the plasmon within the first hundred fs, namely before other relaxation pathways, and faster than in pristine WS_2.

Furthermore, the open character of ultrafast metacavities creates the ideal conditions to inspect dynamic light-matter interactions altered by the strong coupling with spatio-temporal resolution, a dramatic advantage for the study of transport phenomena, as demonstrated in ref.[137] In this work, a high-Q ($\sim$100, mode lifetime $\sim$500 fs) surface lattice resonance (SLR) across a plasmonic array is strongly coupled to the exciton of a 2D lead halide perovskite (Fig. 4d). Angle-dependent TA microscopy is employed to directly image the ultrafast exciton transport. In the strong coupling regime, an enhancement is observed by more than one order of magnitude compared to the pristine perovskite film, with an estimated migration distance increase from $\sim$10 nm to $\sim$300 nm in 10 ps. To further ascertain the role of the light/matter hybridised nature of the SLR/exciton polaritons, measurements are repeated for two objective numerical apertures (NAs). Changing the collection angle, and accordingly the probe momentum, translates into tuning the photonic fraction of the polariton, hence the exciton effective mass. A faster transport is obtained on ps timescales

by restricting the momenta with a ∼35% smaller NA, corresponding to an estimated Hopfield coefficient, measuring the degree of exciton-photon mixing in the polariton, ∼1.2 times higher.

Beyond the results here presented, the flexibility of flat-optical designs allows the efficient coupling to an extremely large variety of materials, which expands significantly the accessible ultrafast photophysical properties of the hybrid systems to be characterised and exploited.[144–149] In addition, the use of all-optical active meta-atoms sets the stage for developing open cavities with reconfigurable optical modes able to overcome the intrinsic limits of conventional, typically closed and/or too slow[150–153] architectures to access dynamic light-matter interactions, with implications from high-speed signal processing to the control of photophysical material functionalities. Major contributions are envisaged, opening up to opportunities encompassing all-optical switching, in-situ probing of polariton-mediated ultrafast reactive pathways, and real-time manipulation of ultrafast processes.

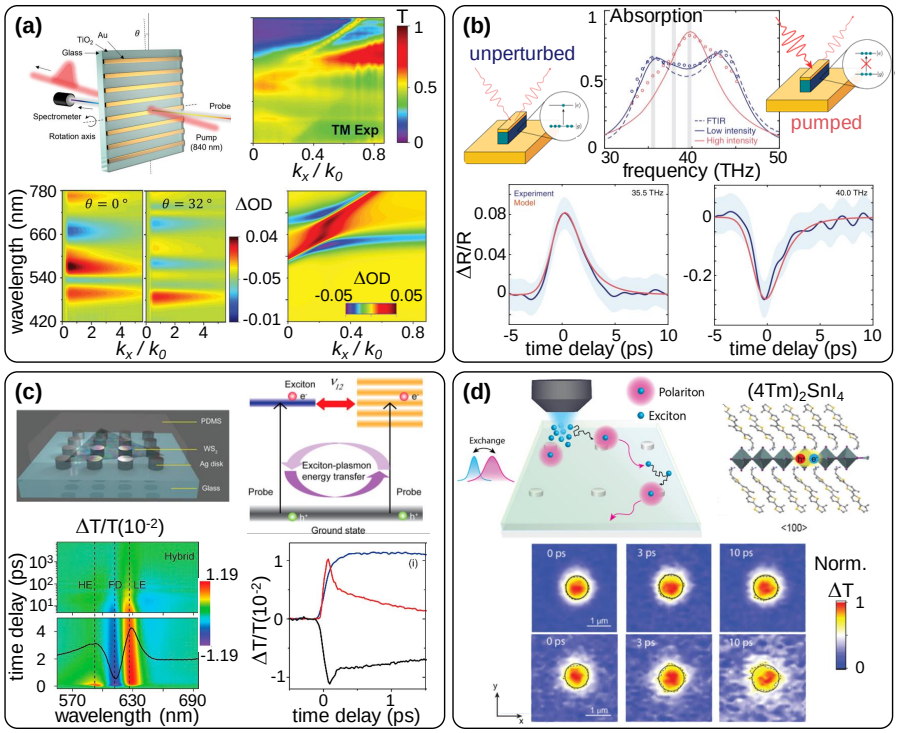


Figure 4: **Ultrafast enhanced light-matter interactions with hybrid metasurfaces. a) Enhancement of all-optical switching in hybrid plasmonic/photonic metasurface.** Clockwise: illustration of the hybrid metasurface, with a schematic of the experimental set-up; measured angle-resolved static transmission of TM-polarised light, for which polaritons arise upon increasing the angle of incidence; simulated transient angular dispersion via a simplified, qualitative model of the photoinduced modulation driving the signal enhancement near the anti-crossing point; measured pump-probe maps of differential optical density upon increasing the probe angle of incidence Adapted with permission from ref.[134] Copyright 2023, Wiley Publishing. **b) Modulating strong coupling in a hybrid polaritonic metasurface.** Schematic of the operation principle of the hybrid metasurface, which undergoes a transition from strong coupling to weak coupling upon photoexcitation, due to the light-induced absorption saturation in the multi-quantum well; ultrafast (degenerate-)pump-probe dynamics at two selected wavelengths. Adapted with permission from ref.[135] Copyright 2021, Optica Publishing. **c) Ultrafast coupling modulation in a TMD/plasmonic metasurface.** Anticlockwise: illustration of the hybrid metasurface; measured pump-probe map of differential transmission; ultrafast temporal traces at three selected wavelengths; sketch of the operation principle for intermediate exciton/plasmon coupling modulation. Adapted with permission from ref.[136] Copyright 2021, American Chemical Society. **d) Polariton-enhanced ultrafast exciton transport in a strongly-coupled hybrid 2D perovskite/plasmonic metasurface.** Illustration of the polariton-assisted transport of excitons within the hybrid metacavity, and sketch of the lead halide perovskites used; transient absorption microscopy images at selected pump-probe time delays (0, 3, 10 ps) of the exciton diffusion in the pristine perovskite (top) and in the hybrid system (bottom). Adapted with permission from ref.[137] Copyright 2023, American Chemical Society.

3.3 Frequency conversion with time-variant metasurfaces

The all-optical control of light discussed in Section 3.1 is empowered by the ultrafast changes of permittivity that femtosecond laser pulses impart to the metasurface, leveraging the third-order nonlinearities of the material constituting the meta-atoms. The optical signal interrogating the nanostructure after these variations experiences a nonequilibrium state of the metasurface, whose optical response is different from the one exhibited in unperturbed conditions. In this picture, the metasurface can be thought of as an ultrafast reconfigurable linear optical filter, commuting at high speed between two distinct static or quasi-static spectral shapes. However, beyond this configuration, a further operation mode can be envisaged for all-optically modulated metasurfaces, by exploiting the short time interval during which the meta-atoms are commuting and the corresponding temporal boundaries created upon ultrafast illumination. Given the critical role of the temporal evolution of their response, metasurfaces operating in such nonequilibrium regime are referred to as 'time-variant'. Completely new functionalities can be accessed, for which the peculiar flexibility of flat-optic platforms in engineering resonances and modal profiles represents a crucial advantage. Here we restrict our overview to examples of frequency conversion not relying on conventional nonlinear generation processes, commonly referred to as 'linear frequency conversion'.[43,154–156] This time-dependent mechanism, contrary to what observed in intrinsic nonlinear media, does not involve directly nonlinear optical susceptibilities nor harmonics (in well-spaced integer multiples) of the fundamental frequency, but it rather exploits the frequency mixing and spectral modifications (in the form of sidebands, possibly spread out in bandwidth when the modulation frequency approaches the impinging radiation frequency) arising when light propagates through a medium with a rapidly varying refractive index.[157] In this context, Figure 5 summarises some illustrative results: Fig. 5a and 5b apply ultrafast micro/nano-cavity modulations to the linear frequency conversion of Terahertz and infrared light;[158,159] Fig. 5c reports ultrafast changes of a harmonic-generation process driven by concurrent time-varying mechanisms;[160] Fig. 5d obtains nonreciprocal reflection via ultrafast space-time phase mod-

ulations.[161]

Among the pioneering works,[158] time-dependent linear frequency (up- and down-) conversion has been achieved with a Terahertz metasurface made of Au split-ring resonators (SRRs) on a semi-insulating GaAs substrate (Fig. 5a). In unperturbed conditions, the system exhibits two non-degenerate extinction resonances, at 0.62 THz and 1.24 THz, belonging to the two SRRs in the unit cell. When illuminated with an ultrashort (50 fs duration) pulse at 800 nm, hot carriers are photogenerated in the GaAs and the two SRRs are dynamically short-circuited, with subsequent sudden merging of their resonances. This nonperturbative transition is extremely fast, hence it contains a broad spectrum of THz frequency components. Especially, when this abrupt process takes place during the arrival of an input narrow-band THz pulse, it promotes the efficient transfer of the new THz components from the time-varying metasurface to the output THz pulse, which as a result undergoes a substantial spectral broadening.

In a different manifestation of an analogous mechanism,[159] an all-dielectric GaAs metasurface supporting a high-Q ($\sim$500) Fano-like resonance at 970 nm was employed to demonstrate time-dependent linear frequency conversion in the infrared (Fig. 5b). Ultrafast (80 fs) pump pulses at 800 nm photoexcite hot carriers in the meta-atoms, modifying the GaAs permittivity in both its real and imaginary part. The former, of negative sign, causes a blue shift of the Fano resonance, whereas the photoinduced extra losses translate into a decrease of the mode Q-factor. When a broadband infrared probe pulse impinges on the metasurface slightly prior to the pump arrival but within the resonance lifetime ($\sim$500 fs), it experiences a medium whose refractive index is varying throughout the interaction. The outcome is a generation of new spectral components (varying with the pump-probe delay and manifesting as ultrafast interference fringes) in the 940-970 nm wavelength range of the reflected probe bandwidth. In passing, note that the effect has origins similar to that of the coherent arte-

facts in ultrafast infrared spectroscopy of vibrational transitions in chemical systems and semiconductors.

In general, these time-dependent linear frequency-synthesis effects can be also optically driven in parallel with more conventional nonlinear generation processes,[162,163] as reported in ref.[160] for the third-harmonic generation (THG) in amorphous germanium (a-Ge) metasurfaces (Fig. 5c). In this implementation, the designed meta-atoms support a magnetic quadrupolar Mie resonance at 1640 nm, enhancing THG of visible light (540 nm). In the nonequilibrium regime, the metasurface is dynamically controlled via the hot carriers photogenerated by ultrashort (60 fs, 800 nm) pump pulses throughout the interaction with a probe pulse, acting as the fundamental in the THG. As such, the frequency conversion due to the meta-atoms' time-varying permittivity does not only affect the spectral bandwidth of the transmitted probe (ultrafast fringes appear around 1640 nm), but it also leads to a sizeable blue shift and broadening of the THG spectrum at very short, negative pump-probe delays.

The interaction regime described above involves a fast commutation of the material permittivity with respect to the probe pulse envelope, giving rise to a continuum of high temporal frequencies in the time-varying metasurface. An alternative approach using instead monochromatic modulation has been exploited to demonstrate not only frequency conversion, but also nonreciprocal reflection from a gradient metasurface.[161] The idea is to implement a travelling-wave permittivity modulation along the metasurface via Kerr effect, breaking the time-reversal symmetry. Practically, the interference fringes generated by two counter-propagating infrared pump pulses with few nm detuning, and strongly enhanced by the resonant meta-atoms, induce such changes of permittivity (Fig. 5d). A probe light can then experience both frequency up-/down-conversion (due to the pumps' detuning) and beam deflection (due to the pumps' momentum mismatch), akin to what happens in electro-optic

modulators, but at much higher modulation frequencies (here 2.8 THz). The superimposed static linear phase gradient of the metasurface selects, via in-plane momentum conservation, either the up- or down-conversion for the propagating diffracted beams (depending on the direction of the travelling-wave modulation), which in turn violates reciprocity.

In the representative works outlined above, the timescale over which the active metasurface response is varying is longer than the cycle of the signal carrier. In principle, a novel regime can be accessed if the switching event becomes much faster than half of the signal carrier period, provided that the modulation amplitude remains large (ideally close to 1). This identifies a new genus of 'Floquet metamaterials',[164] which operate upon a periodic temporal drive and feature a nonequilibrium response dynamics approaching the timescale of the involved signals, potentially unsealing extraordinary ways to control light and enabling unconventional wave phenomena.[165] Borrowing concepts from solid–state theory, periodically modulated systems can be interpreted to possess an extra synthetic dimension, introduced by the periodic dependence on time.[166–168] From the experimental standpoint, ultrafast all-optical metasurfaces are among the most promising candidates to tailor spatio-temporal modulations faster than the periodic drive and access the Floquet regime.

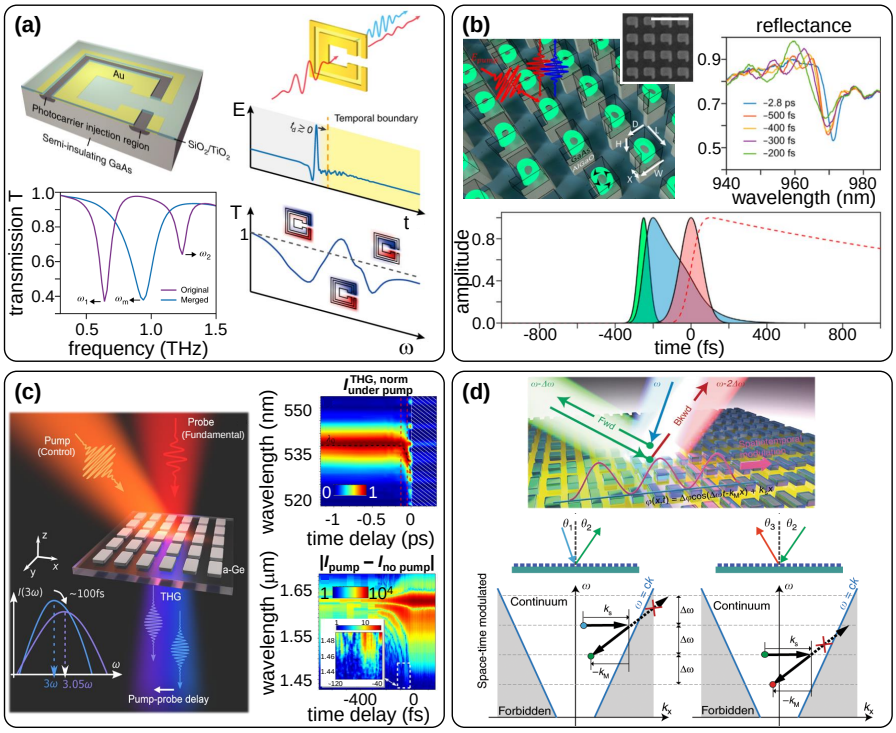


Figure 5: **Ultrafast frequency conversion with time-variant metasurfaces. a) THz-frequency conversion** Clockwise: schematic of the meta-atom; illustration (conceptual scheme, timing diagram, and transmission from top to bottom) of the operation principle of the time-variant metasurface, where a new frequency component is radiated from an oscillating merged mode; simulated transmission in unperturbed and photoexcited conditions. Adapted with permission from ref.[158] Copyright 2018, Springer Nature. **b) Infrared-frequency conversion.** Clockwise: illustration of the GaAs-based metasurface; transient reflectance spectra from the photoexcited metasurface at selected pump-probe delays; simulated temporal dynamics of the pulses interaction driving the frequency conversion: the normalized field envelopes of the probe pulse (green), the pump pulse (red), and the resonance mode (blue) are depicted. Adapted with permission from ref.[159] Copyright 2020, American Chemical Society. **c) Third-harmonic generation modulation via time-variant frequency conversion.** Illustration of amorphous germanium-based metasurface, and the schematic of the operation mechanism for the dynamical up-conversion of the third-harmonic signal, following the ultrafast photoinduced evolution of the resonance; normalized spectra of the third-harmonic signal generated by the probe pulse under ultrafast pump excitation (top), and difference (logarithmic scale) between probe spectra with or without pump illumination (bottom). Adapted with permission from ref.[160] Copyright 2022, American Chemical Society. **d) Nonreciprocity via photoinduced space-time phase modulations.** Illustration of the concept of space-time phase modulated metasurface, where a travelling phase sinusoidal modulation is superposed on the designed phase gradient, which allows for achieving nonreciprocal reflection; schematic of the asymmetric forward (left) and backward (right) reflections and related dispersion diagram, showing that the metasurface provides additional momentum in the x-direction as well as converts the light frequency. Adapted with permission from ref.[161] Copyright 2018, Springer Nature.

4. Conclusions and Outlook

To conclude this Perspective, a final section summarises our vision on future intriguing directions for ultrafast all-optical metasurfaces, pushing the current state-of-the-art. Based on recent publications, we highlight in Table 1 some of the emerging challenges that we envision will be addressed by active metasurfaces operating in the ultrafast regime, potentially inspiring new ideas, crossing the boundaries of nanophotonics, ultrafast optics, physical chemistry and material sciences.

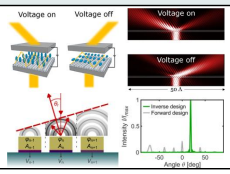
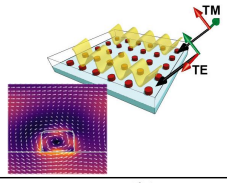
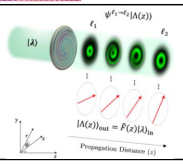
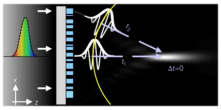
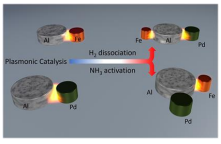
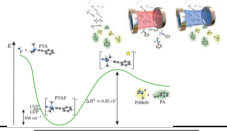
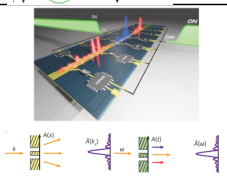
Challenges and perspectives for future applications			State-of-the-art
Temporal inverse design		Inverse design is establishing itself as a powerful tool in flat-optics. Enriching its principles and applications to optimise simultaneously the metasurface linear response and the dynamics[169,170] of the all-optical nonlinear modulations will push forward the strategies to tailor light-matter interactions on the ultrafast timescale.	171-177
High-Q resonances		Spectrally narrow resonances and long cavity lifetimes are desirable for far- and near-field all-optical applications, yet posing challenges, especially in the UV-visible. The design of all-dielectric nonlocal metasurfaces, combining low-loss materials and dispersive extended resonances hold great promise to face the challenge of achieving larger modulations and stronger coupling[178].	35, 179-183
OAM structuring		Imprinting an internal structure into light statically is routinely done with metasurfaces[184]. The ultrafast writing of OAM states by dynamically shaping the light wavefronts would open up to all-optical schemes manipulating topological light, with dramatic impact on optical communications and quantum encryption.	185-188
Active wavefront shaping		Combining the ability of metasurfaces to impart phase profiles on optical wavefronts[189] and the all-optical temporal modulation of the meta-atoms' properties, desired ultrafast phase gradients can be produced. Active platforms for the transient focusing of optical beams at ultrafast speed could be envisaged.	190,191
Light-driven chemistry		To address the formidable challenge of enhancing photocatalytic reaction rates, metasurfaces are emerging as ideal nanoreactors[192], whose rational spatial arrangement allows selectivity control. In the ultrafast regime, engineering electronic and thermal photoinduced processes could create optimal reaction pathways.	193-197
Tailoring photophysical dynamics		The far-reaching challenge of tailoring molecular dynamics on demand[198] can be addressed by employing ultrafast all-optical metasurfaces as the next generation of nanophotonic hybrid devices, with implications from light harvesting to photovoltaics.	149, 199, 200
Time interface engineering		Temporal reflection and broadband frequency translation via temporal interfaces has been reported for microwave signals in a transmission-line metamaterial[201]. Pushing this approach to the visible and infrared[202] is an outstanding challenge with the potential of revolutionizing photonics, and ultrafast all-optical metasurfaces could provide key contributions.	203-209

Table 1: **Challenges and perspectives for future applications of ultrafast all-optical metasurfaces. Temporal inverse design**. Adapted with permission from refs.[169,170] Copyright 2020 American Chemical Society. State-of-the-art: refs.;[171–177] **High-Q resonances**. Adapted with permission from ref.[178] Copyright 2020 American Chemical Society. State-of-the-art: refs.;[35,179–183] **OAM structuring**. Adapted with permission from ref.[184] Copyright 2021 Springer Nature. State-of.the-art: refs.;[185–188] **Active wavefront shaping**. Adapted with permission from ref.[189] Copyright 2018 Springer Nature. State-of-the-art: refs.;[190,191] **Light-driven chemistry**. Adapted with permission from ref.[192] Copyright 2022 American Chemical Society. State-of-the-art: refs.;[193–197] **Tailoring photophysical dynamics**. Adapted with permission from ref.[198] Copyright 2022 Springer Nature. State-of-the-art: refs.;[149,199,200] **Time interface engineering**. Adapted with permission from ref.[201,202] Copyright 2023 Springer Nature. State-of-the-art: refs.[203–209]

Associated content

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- The photophysics of ultrafast all-optical metasurfaces; Modelling ultrafast all-optical metasurfaces.

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Notes

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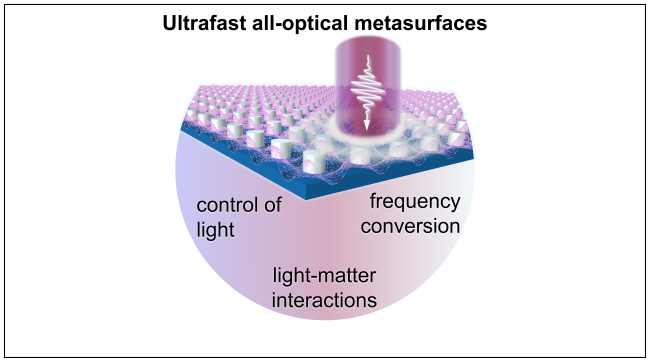
TOC Graphic


Ultrafast all-optical metasurfaces
control of light
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