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ABSTRACT

Optical metasurfaces have recently emerged as the game changer in light manipulation and opened up new perspectives in many subfields of optics and photonics. Recent developments in nonlocal metasurfaces, in which the nanoscale building blocks respond to the incoming light collectively rather than as individual objects, are especially promising for enhancing and controlling the nonlinear optical phenomena. In this article, we provide a brief overview of the basic principles of nonlocal metasurfaces in the context of their nonlinear optical functionalities. We discuss the origin and the regimes of the nonlocal response, covering the aspects of multiple scattering, radiation damping, quality factor, local-field enhancement, and temporal dynamics. Some important aspects are illustrated by computational examples. We also give our personal viewpoint on the selected ideas and research directions in nonlocal and nonlinear metasurfaces, including the role of spatial symmetry in nonlocal interactions, the effects of phase and momentum matching in frequency conversion, as well as the possibilities offered by new material platforms and novel concepts, such as bound states in the continuum, parity–time symmetry, and time-variant metasurfaces.

I. INTRODUCTION

In the past few decades, controlled use of light has revolutionized science and technology, having a broad impact on our daily life, especially in connection with telecommunications and information technologies. However, continuous technological advancements have created increasing demands for light manipulation at ever smaller spatial and temporal scales.[1] Nonlinear optical effects, such as frequency conversion, all-optical modulation, and generation of non-classical light, are highly desirable in many applications of optical science, but they are very challenging to observe and use in miniaturized optical devices. Since traditional optical materials have already reached their limits in conventional applications, the need for innovation in materials design, such as metamaterials, has emerged.[2] Metamaterials can exhibit optical properties not found in natural materials, such as optical magnetism[3,4] and negative refraction.[5,6] They are typically composed of subwavelength building blocks—the so-called meta-atoms. For the visible and near-infrared spectral ranges, the dimensions of these structures are in the nanoscale, making it difficult to fabricate 3D optical metamaterials. Therefore, the research has moved from 3D toward more feasible 2D structures—metasurfaces.[7]

Currently, after nearly two decades of developments, optical metasurfaces can provide essentially arbitrary control over the amplitude, phase, and polarization of light, serving as ultracompact elements for holographic devices, optical sources and detectors, sensors, imaging systems, and components for optical communication and information processing.[8–14] In nonlinear optics, metasurfaces provide a platform for frequency conversion, production of entangled photon pairs, all-optical switching and modulation, phase conjugation, optical limiting, saturable absorption, etc.[15–20] However, despite numerous experimental demonstrations, nonlinear metasurfaces are still far from wide commercial use, and many of their intriguing fundamental properties and potential functions have not yet been fully explored.

Recent developments in the area of nonlocal metasurfaces are particularly interesting. It has been shown that collective nonlinear optical response of metasurfaces can be tailored by spatially extended resonances. The nonlocal optical modes associated with these resonances enter an advanced level of sophistication and make use of the concepts of parity–time ($\mathcal{PT}$) symmetry,[21–23] bound states in the continuum (BICs),[24,25] and topological robustness.[26,27] Nontrivial engineering of collective resonances can also involve effects that are unique to nonlinear optical interactions, such as nonlinear geometric phase,[28] nonreciprocal parametric gain and loss,[29] exotic symmetries of nonlinear susceptibility tensors,[30] bistability,[31] and time-varying optical properties.[32] Combining them with novel material platforms, such as multi-quantum well nanostructures,[33] Dirac/Weyl semimetals,[34] quantum dot-2D material hybrid systems,[35] and epsilon-near-zero materials,[36] could ultimately yield nonlinear metasurfaces of unprecedented properties and superior performance. In this Perspective, we highlight selected hot topics in the field of nonlinear metasurfaces related to their possibly nonlocal optical responses. We provide a brief overview of the basic concepts underlying the optical properties of such metasurfaces and introduce their most important nonlinear functionalities. The discussions contain our personal viewpoints on the state of the art and the most interesting current and future research directions in the realm of nonlinear nonlocal metasurfaces.

II. METASURFACES—BASIC PRINCIPLES

The nonlinear optical functionalities of photonic structures are tailored by the choice of the constituent materials determined by their nonlinear susceptibilities. However, micro- and nanostructuring affects both the linear and nonlinear optical properties such that the nonlinear response can become largely governed by the linear response.[37,38] In periodic metasurfaces, the main factors that determine their optical responses are the polarizabilities and spatial arrangement of the meta-atoms. In this section, we provide a brief overview of the main concepts underlying the nonlocal optical response of periodic metasurfaces.

A. General classification

In simple terms, metasurfaces can be classified as either diffractive or subdiffractive, depending on the distance between the meta-atoms relative to the wavelength. On the other hand, their local or nonlocal character is determined by the strength of multiple scattering between nearby meta-atoms, which depends on the polarizability of the meta-atoms and surface confinement of the scattered light. This simplified classification is graphically summarized in Fig. 1. Similar classifications of metasurfaces have been proposed also previously, e.g., in the recent review by Overvig and Alú,[39] where, in contrast to our classification, diffractive metasurfaces that are local are not considered. Here, we prefer to consider the nonlocality and the ability of the structure to diffract light as two independent degrees of freedom. For example, nonlocal/collective excitations, such as Fourier lattice resonances, can be realized in multipartite or even aperiodic structures,[40] which at least in the conventional sense are not diffractive. Moreover, some diffractive structures can respond to the incident light either locally or nonlocally depending on the excitation conditions, as we explain further in the upcoming sections.

B. Polarizability of meta-atoms

In general, the optical polarizability of a particle, $\boldsymbol{\alpha}$, can be a complex tensorial quantity, with possible contributions of magnetoelectric coupling,[41] higher-order multipolar terms,[42] optical gain and loss,[43] or even temporal modulation.[44] Among these contributions, the higher-order multipoles have been a subject of extensive research related to tailoring and enhancing of the nonlinear optical properties of nanostructured materials through localized plasmonic[45–47] and Mie resonances.[48–52]

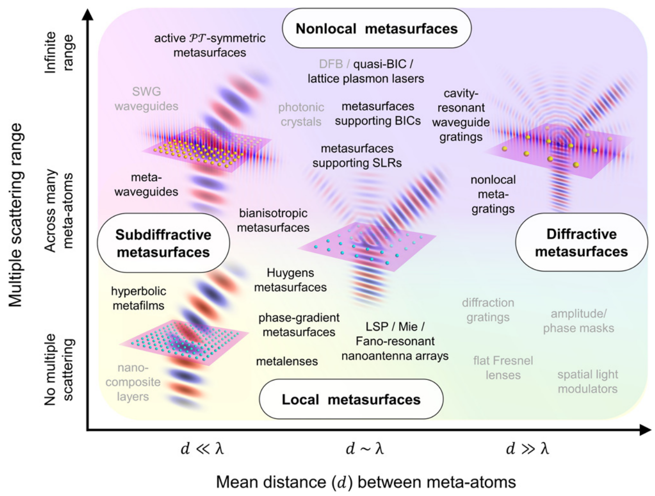

FIG. 1. Different regimes of optical response of periodic metasurfaces, depending on the mean distance (d) between meta-atoms (subdiffractive–diffractive) and the range of multiple in-plane scattering between meta-atoms (local/nonlocal). In perfectly local metasurfaces, each meta-atom can be treated as an independent source of scattered optical fields. Nonlocal metasurfaces are distinguished by the presence of in-plane propagating modes that contribute to the far-field scattering. The limit of full lateral coupling between the meta-atoms in an infinite lattice (maximum nonlocality) can be reached in metasurfaces with gain, e.g., in $\mathcal{PT}$-symmetric metasurfaces and plasmonic lattice lasers. Diffractive metasurfaces with the meta-atomic separation on the order of λ or larger can couple light to multiple diffraction orders. In contrast, subdiffractive metasurfaces typically produce only one output beam, the propagation direction of which can in general differ from that of the incident beam. We note that the period of the structure (Λ) can be larger than d. In such cases, the metasurface contains more than just one meta-atom per unit cell and may operate in either subdiffractive or diffractive regime, depending on the conditions of light incidence. We provide some examples of metasurfaces corresponding to the above regimes. Examples in gray are usually not called metasurfaces, although they follow similar principles.

In the simplest case, a meta-atom can be considered as a point electric dipole with a moment $\mathbf{p} = \boldsymbol{\alpha} \cdot \mathbf{E}_{\text{in}}$ induced by an incoming field (where we used a convention in which ε_0—required for the compliance with the SI units—is already included in $\boldsymbol{\alpha}$). The dipole radiates a secondary field $\mathbf{E}_{\text{sc}}(\mathbf{r}') = \mathbf{G}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{p}(\mathbf{r})$ shaped by the dyadic Green's function $\mathbf{G}(\mathbf{r}, \mathbf{r}')$. Since the energy is radiated away, $\boldsymbol{\alpha}$ is always complex, even if meta-atoms are made of a lossless dielectric (unless the radiative loss is exactly compensated for by gain). This imposes fundamental constraints on light manipulation by nanoscatterers, e.g., limiting their magnetoelectric response.[53]

In fact, higher-order multipoles are always excited in meta-atoms either by near-fields of neighboring meta-atoms or due to phase delays of the excitation field when it propagates through spatially extended meta-atoms. Usually, however, higher-order multipoles only weakly contribute to light scattering. The most significant of them are electric quadrupoles and magnetic dipoles, of which the latter are excited by the electric rather than magnetic field at optical frequencies. The magnetic response of meta-atoms can formally be described by introducing their magnetic, magnetoelectric, and electromagnetic polarizabilities ($\boldsymbol{\alpha}_{mm}$, $\boldsymbol{\alpha}_{me}$, and $\boldsymbol{\alpha}_{em}$, respectively), as for bianisotropic metamaterials and metasurfaces.[54] The electric and magnetic dipole moments $\mathbf{p}$ and $\mathbf{m}$ are in this case given by

$$\mathbf{p} = \boldsymbol{\alpha}_{ee} \cdot \mathbf{E} + \boldsymbol{\alpha}_{em} \cdot \mathbf{H}, \quad \mathbf{m} = \boldsymbol{\alpha}_{me} \cdot \mathbf{E} + \boldsymbol{\alpha}_{mm} \cdot \mathbf{H}, \tag{1}$$

where $\boldsymbol{\alpha}_{ee}$ is the electric polarizability denoted above by $\boldsymbol{\alpha}$. Bianisotropy can result from a nonlocal response within each unit cell of the structure or from the meta-atomic excitations by the fields of neighboring meta-atoms, manifesting the phenomenon of spatial dispersion. Since usually, structural units of metamaterials and metasurfaces have non-trivial geometries, and for example, are fabricated on a substrate, spatial dispersion leads to the dependence of the response on the light propagation direction.[55–58] This type of nonlocality, however, is effectively localized, since contribution of neighboring meta-atoms to the excitation weakens with the distance to them. In contrast, metasurfaces that we call nonlocal in this work have an effectively much larger range of nonlocality, because, in them, coupling between meta-atoms is provided by optical modes that are guided or diffracted along the metasurface. Higher-order multipoles can also be excited in such a metasurface, but they are not required for nonlocality. Furthermore, since higher-order multipoles are rather dark compared to electric dipoles, we can ignore them for simplicity of the description. In the next sections, multiple scattering of meta-atoms is described using the electric-dipole approximation.

C. Local response

If multiple scattering between meta-atoms is negligible (e.g., due to small scattering cross sections), then each meta-atom can be regarded as an independent source of a secondary field $\mathbf{E}_{sc}$. In this case, the optical response of the metasurface is governed by the far-field interference of $\mathbf{E}_{sc}$ (Huygens principle). This approximation is commonly used in designing subdiffractive metasurfaces that behave as homogeneous phase-shifting interfaces obeying a modified Snell's law.[59]

D. Diffractive and subdiffractive metasurfaces

In the diffractive regime, the periodicity of the structures enables constructive interference of $\mathbf{E}_{sc}$ in certain directions, which produces diffraction orders. In the parameter space spanned by frequency ω and in-plane momentum $\mathbf{k}_{||}$, the diffraction orders emerge at Rayleigh anomalies governed by condition $|\mathbf{k}_{||} + \mathbf{K}| = n\omega/c$, where $\mathbf{K}$ represents the reciprocal lattice vectors and n is the refractive index of the embedding medium. This can be illustrated using the repeated zone scheme (Fig. 2), in which the dispersion cones $|\mathbf{k}_{||}| = n\omega/c$ are repeated at the reciprocal lattice nodes $\mathbf{K}$, forming the backbone of photonic band structure, known as the empty-lattice approximation.[60,61]

Rayleigh anomalies are associated with a resonant response due to propagation of diffracted waves along the metasurface plane. Under such conditions, it is impossible to neglect multiple scattering between meta-atoms, which makes the optical response inherently nonlocal.[39] However, diffractive optical elements (such as diffraction gratings) may, in general, operate away from Rayleigh anomalies. In such a case, local response and paraxial approximation are sufficient to describe their optical properties.

In addition to Rayleigh anomalies, resonant response can result from the propagation and multiple scattering of surface waves. A well-known example is associated with excitation of surface plasmon polaritons (SPPs) in periodic metal structures, which gives rise to the so-called Wood anomalies.[62] Resonant excitation of SPP standing waves is responsible for the famous extraordinary optical transmission through periodic arrays of holes in metal films.[63] Due to the phase contribution from the complex-valued SPP propagation constants, Wood anomalies are often characterized by asymmetric spectra. They can also be distinguished from Rayleigh anomalies by their distinct polarization response: as opposed to waves scattered by nanoparticles in a dielectric medium, SPPs are launched along the metal surface in the direction parallel to the incident polarization.[30,64]

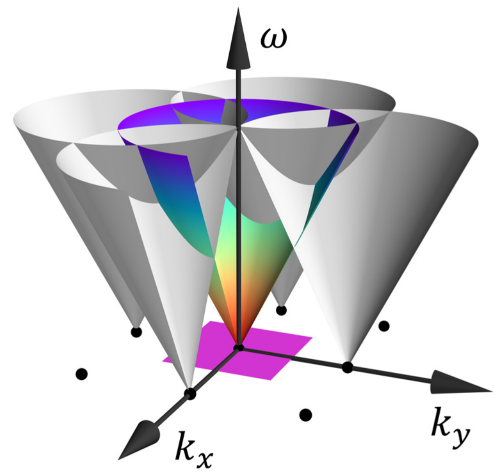

FIG. 2. Empty lattice model—the backbone of photonic band structure of periodic metasurfaces. The magenta square marks the first Brillouin zone—the unit cell of the momentum space—spanning over $|k_x|, |k_y| \leq \pi/\Lambda$. Black points represent the reciprocal lattice nodes, while the cones represent the dispersion of plane waves in the embedding medium. The rainbow cone is the light cone, while the gray cones correspond to higher diffraction orders. Inside the light cone, the in-plane momentum is smaller than the momentum of freely propagating waves, meaning that the wave propagates at an angle to the plane of the metasurface. Some of the gray cones intersect with the light cone, allowing to couple to higher diffraction orders, if frequency ω is sufficiently high.

E. Multiple scattering and collective resonances

Multiple scattering may become significant if $|\boldsymbol{\alpha}|$ is large. For example, the scattering cross sections of resonant plasmonic nanoparticles may extend beyond the unit cell even in diffractive lattices. The coupling between meta-atoms can also be enhanced at Rayleigh anomalies, Wood anomalies, or through guided modes in waveguide structures.[65,66] A nonlocal interaction between meta-atoms can be regarded as an effective modification of their local electric-dipole response,[67]

$$\mathbf{p}_i = \boldsymbol{\alpha}_i \cdot \mathbf{E}_{\text{in}} + \boldsymbol{\alpha}_i \cdot \sum_{j \neq i} \mathbf{G}_{ij} \cdot \mathbf{p}_j = \boldsymbol{\alpha}_{\text{eff},i} \cdot \mathbf{E}_{\text{in}}. \tag{2}$$

In the above expression, the dipole moment of meta-atom i is induced by $\mathbf{E}_{\text{in}}$ and by the fields scattered by all other particles j. These scattered fields modify the polarizability of meta-atom i with respect to $\mathbf{E}_{\text{in}}$ from $\boldsymbol{\alpha}_i$ to $\boldsymbol{\alpha}_{\text{eff},i}$. In periodic metasurfaces, effective polarizabilities of meta-atoms are the same (in accordance with Bloch's theorem), but they depend on the in-plane momentum $\mathbf{k}_{||}$ and the corresponding phase difference between the neighboring unit cells,

$$\boldsymbol{\alpha}_{\text{eff}}(\mathbf{k}_{||}) = \left[\boldsymbol{\alpha}^{-1} - \mathbf{G}_{\text{lattice}}(\mathbf{k}_{||})\right]^{-1}. \quad (3)$$

In general, lattices may contain many (identical or different) meta-atoms per unit cell, which turns Eq. (3) into a matrix equation. In such a case, $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}_{\text{eff}}$ are block-diagonal matrices, and $\mathbf{G}_{\text{lattice}}$—the so-called lattice Green's function—describes the interactions between meta-atoms within the same sublattice (diagonal blocks) and between different sublattices (off-diagonal blocks).[43,68,69] Such complex metasurfaces can be designed to operate in either diffractive or subdiffractive regime, especially when the distance between meta-atoms d is smaller than λ, while the lattice period Λ is larger than λ.

An important deviation from the description of nonlocal metasurfaces as perfectly periodic systems comes from the finite size effects, which should be carefully considered in any practical implementation. With any (finite) number of meta-atoms, the periodic boundary conditions of the scattered fields break down. Thus, the overall amplitude and phase of the field scattered by each meta-atom will become dependent on its position in the lattice.[70,71]

In subdiffractive metasurfaces, multiple scattering between periodically arranged meta-atoms may give rise to collective modes that are confined within the metasurface due to a momentum mismatch with freely propagating waves. The properties of such modes are strongly dependent on the properties of $\boldsymbol{\alpha}$,[72] which distinguishes them from the conventional Bloch modes of photonic crystals resulting from spatial modulation of the refractive index.

In the diffractive regime, the metasurface modes and the Rayleigh anomalies appear in the light cone. As a result, the "empty lattice" backbone (Fig. 2) is imprinted on the $(\mathbf{k}_{||}, \omega)$-distribution of $\boldsymbol{\alpha}_{\text{eff}}$. This gives rise to the dispersive surface lattice resonances (SLRs)[73] often characterized by asymmetric Fano-like spectral lineshapes.[74,75] Near the intersections of Rayleigh anomalies, which occur at high-symmetry points in the momentum space, SLRs form band edges—standing waves, as in optical resonators, with enhanced density of states (van Hove singularity).[76] SPPs and Wood anomalies give rise to analogous features.[77] Such band edges serve as an efficient platform for distributed feedback lasing,[78] strong light–matter coupling,[79] slow-light-related phenomena,[80,81] and nonlinear optical effects.[30,82]

F. Band folding—A computational example

The properties of SLRs and their interactions with external light fields can be engineered by the symmetry of the lattice and meta-atoms. This can be used for controlling the radiation loss and optimizing the Q factors or the local field enhancement, depending on the desired purpose. One of the simplest approaches to engineer such properties is the band folding.[69,78,83]

To give an illustrative example, we consider a subdiffractive lattice of silver nanoparticles embedded in a slab waveguide (Fig. 3). Initially, in the given frequency range, all high-Q eigenmodes of the lattice are outside the light cone [Fig. 3(a)]. These modes can be "folded" into the light cone [Fig. 3(b)] by distorting the lattice geometry along one of the lattice vectors, leading to doubling of the period and rendering the lattice diffractive.

Using such a system for nonlinear optical applications requires optimization of the local field enhancement under external illumination. A larger Q factor usually means a larger field enhancement. However, coupling of the modes to external radiation causes the Q factor to decrease. In this case, a desired compromise can be achieved by fine-tuning the lattice geometry [Fig. 3(c)]. Similar approaches are used to engineer and study bound states in the continuum (BICs). Perfect BICs are turned into quasi-BICs[84,85] by controllable geometry distortions, such that the coupling to the incident and outgoing waves is adjusted not to reduce the Q factor significantly.[86]

G. Resonant response in time domain

Nonlocal resonant response of metasurfaces is often described and tailored in the frequency domain. This approach has many advantages, such as the inherent simplicity of implementing the periodic boundary conditions, which we illustrated in the computational example in Fig. 3. However, the physical picture can be more complicated, especially when it comes to ultrafast nonlinear optical interactions.

In the time domain, the local fields $E_{\text{local}}(t)$ inside a resonant system can be expressed as a convolution of the temporal impulse response function of the system $G(t)$ and the driving fields $E_{\text{drive}}(t)$,

$$E_{\text{local}}(t) = G(t) * E_{\text{drive}}(t). \quad (4)$$

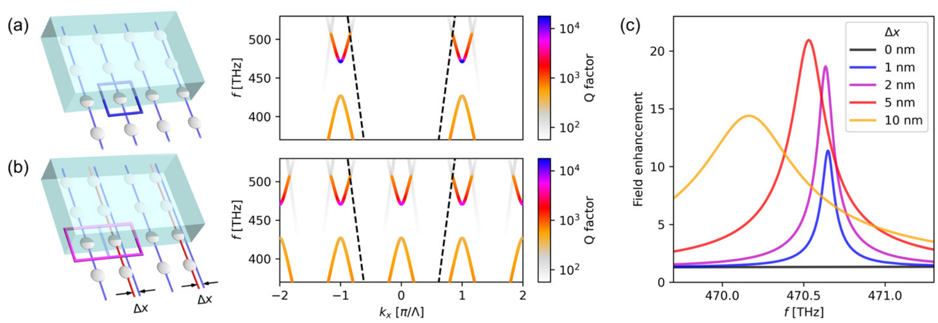

FIG. 3. Simple example of radiation loss management through band folding. The approach is based on doubling the period of the structure, which, in the present case, is a square array of silver nanoellipsoids (height 20 nm, diameter 80 nm) in a slab waveguide made of glass ($n_{\text{core}} = 1.5$, $n_{\text{cladding}} = 1$, core thickness 200 nm). (a) and (b) Lattice schematic (left) and photonic band structure (right) of the TE-polarized modes calculated using COMSOL Multiphysics (TM-polarized modes are not shown). The light cone is indicated by the black dashed lines, and the Q factors are encoded in the colors of the photonic bands. (a) A subdiffractive array with a period of 250 nm. The band edges are outside the light cone. (b) Diffractive array obtained by shifting every second column of the array by $\Delta x = 10$ nm. As a result, the band edges are "folded" into the center of the light cone. (c) Spectral dependence of the average field enhancement inside the slab for the upper band edge under illumination by a normally incident plane wave. The highest field enhancement is obtained at $\Delta x = 5$ nm (red curve), showing a compromise between the coupling efficiency and the Q factor reduction.

In the case of a simple underdamped harmonic oscillator, $G(t)$ can be expressed as[87,88]

$$G(t) \propto H(t)e^{-\gamma t}\sin(\omega' t), \quad (5)$$

where $H(t)$ is the Heaviside step function, γ is the damping rate, and $\omega' = \sqrt{\omega^2 - \gamma^2}$, where ω is the resonance frequency. The Q factor of such a resonance is equal to $\omega/2\gamma$. To illustrate the resonant response, we can assume that $E_{\mathrm{drive}}(t)$ is a Gaussian pulse,

$$E_{\mathrm{drive}}(t) \propto e^{-t^2/\tau^2}\cos(\omega t), \quad (6)$$

with pulse duration τ and frequency equal to the resonant frequency ω. The frequency-domain description of such a system is useful only if the pulse duration is much longer than the decay time. This scenario is illustrated in Fig. 4(a). The local field amplitude builds up by constructive interference with the driving field, and the peak local field enhancement is directly proportional to the Q factor.

A different situation is shown in Fig. 4(b), where a high-Q resonance is excited by a very short driving pulse. In this case, the local field amplitude has a short time to accumulate before it starts to decay. It is clear that further increase in the Q factor will only extend the decay time, but it will not enhance the local field any further.

This tendency is summarized in Fig. 4(c) that shows the peak amplitude of $E_{\mathrm{local}}(t)$ as a function of the Q factor for a fixed duration τ of the driving pulse. After the initial linear increase, the curve flattens as the excitation decay time becomes much longer than τ. This trend should be taken into account in designing high-Q resonant systems for nonlinear optical applications using ultrashort pulses. The efficiency of an n^{th}-order nonlinear effect scales with the n^{th} power of the local field amplitude, but ultrashort pulses may be too short for sufficient buildup of the electric field of a high-Q resonance. This is why high-Q resonances often lead to surprisingly modest enhancements of nonlinear effects when using ultrashort laser pulses. On the other hand, high Q factors can be efficiently utilized for enhancing nonlinear optical effects under continuous-wave excitation, e.g., in continuous-wave SHG,[89] surface enhanced Raman spectroscopy (SERS),[90] and non-instantaneous bistable optical systems.[31]

Tailoring the time-domain optical response of nanostructures is one of the current challenges in the design of resonant systems for nonlinear optical applications.[91,92] In practice, the temporal response may be much more complex than in Eqs. (4) and (5), for example, due to the interference between different resonant modes,[93,94] especially when nonlocal interactions come into play.[95,96] In the case of nonlocal metasurfaces, the time-domain description must include the in-plane retardation and finite-size-related effects. On top of that, the dynamics of the material response (such as the electronic excitations in metals) must be considered[97] at the time scales that can be comparable to the individual optical cycle.

III. NONLINEAR AND NONLOCAL METASURFACES

Conventionally, nonlinear effects have been realized in bulk materials with non-negligible higher-order susceptibilities. Under proper conditions, such as phase matching between pumping light and its harmonics, intrinsically weak nonlinear effects can accumulate over long propagation distances inside the material, resulting in strong nonlinear signals.[99]

In addition to free-space optical systems, waveguiding platforms, such as nonlinear fibers,[100] resonant waveguide structures,[101,102] and (nano)waveguides[103] have been utilized for nonlinear optics.[104] The main advantage of these platforms is that confinement of light in one or two dimensions can enhance light–matter interaction and the associated nonlinear effects. For the effects to become sufficiently strong, light should propagate over a sufficiently long distance, which limits the applicability of the approach, for instance, in flat optics.

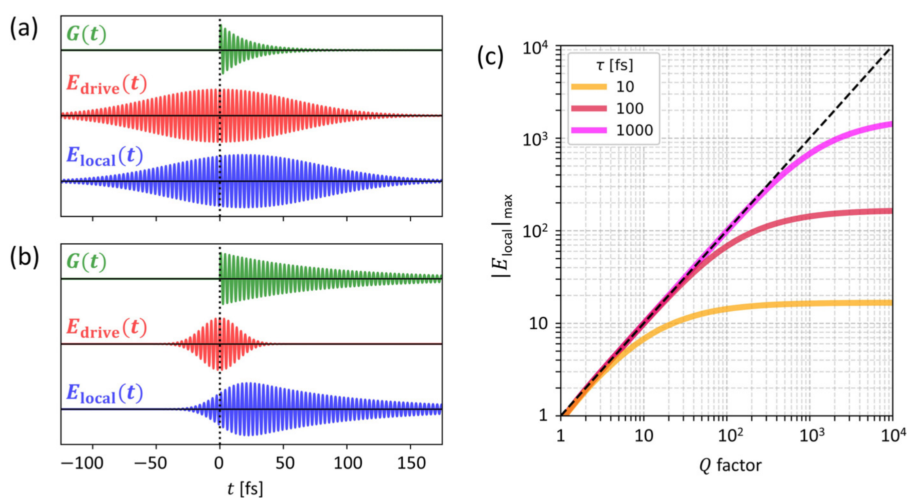

FIG. 4. Resonant response in the time domain. (a) Excitation of a low-Q resonance ($Q = 20$) by a long pulse ($\tau = 80$ fs). (b) Excitation of a high-Q resonance ($Q = 80$) by a short pulse ($\tau = 20$ fs). In both (a) and (b), the green curves represent the impulse response function, the red curves the temporal profile of the driving optical field, and the blue curves the profile of the local fields. (c) Peak enhancement of the local field amplitude as a function of the Q factor for optical pulses with τ equal to 10 fs (orange), 100 fs (dark red), and 1000 fs (magenta). Excitation by longer pulses leads to more efficient buildup of the local field amplitude at a high-Q resonance. As the resonance decay time extends beyond the incident pulse duration, the peak enhancement factor starts to deviate from the linear dependence on the Q factor (black dashed line). A perfect linear dependence would only be seen under a continuous-wave excitation.

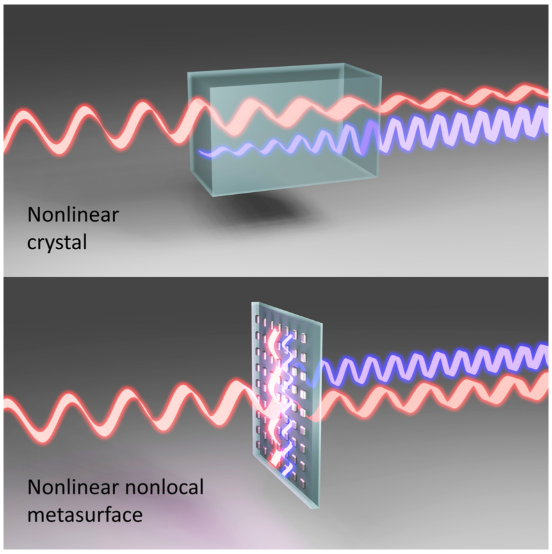

FIG. 5. Illustration of the second-harmonic generation in a nonlinear crystal (top) and a nonlinear nonlocal metasurface (bottom). In the first case, the phase matching allows the power transfer from the fundamental (red) to the second-harmonic beam (blue) to be accumulated during propagation through the nonlinear medium. However, due to refractive index dispersion in solids, phase matching can be achieved only in birefringent or periodically poled crystals, excluding many nonlinear materials, e.g., those with zinc blende structure.[98] In the metasurface, the nonlinear process is distributed in the transverse rather than the longitudinal direction, enhancing the frequency conversion by collective resonances and circumventing the phase-matching requirements.

One of the main interest toward metasurfaces in the context of nonlinear optics is to establish a mechanism for strong light–matter interactions in optically thin systems, wherein such effects would otherwise be negligible. This can be achieved by coupling light to waves that propagate in the plane perpendicular to its principal propagation direction (see Fig. 5). Periodically structured materials have shown enhanced and controllable nonlinear generation of light in exotic frequency ranges, such as XUV[105] and THz.[106] On the other hand, there is a growing interest in exploring and testing novel concepts related to nonlocal resonant interactions in nonlinear metasurfaces, some of which could potentially lead to superior functionalities of optical devices. Here, we provide an overview and Perspective of selected topics from this broad and active research area.

A. Inversion symmetry breaking

Symmetry has always been a critical factor in nonlinear optics, especially for even-order nonlinear effects, such as second-harmonic generation (SHG), requiring the nonlinear media to be noncentrosymmetric. Indeed, noncentrosymmetric dielectric and semiconductor materials, such as GaAs, offer an efficient platform for nonlinear optical metasurfaces.[52,108] On the other hand, efficient second-order nonlinear metasurfaces can be constructed from noncentrosymmetric structures made of centrosymmetric materials, e.g., plasmonic metals or high-index dielectrics suitable for supporting resonant optical modes. In such cases, the inversion symmetry can be broken by the shape of the meta-atoms, which is a common strategy to achieve efficient SHG from plasmonic nanostructures.[109,110] Similar artificial second-order susceptibility has been achieved by noncentrosymmetric stacking of different centrosymmetric materials into so-called ABC nanolaminates.[111]

Among many ideas to be explored for breaking the inversion symmetry, nonlocal metasurfaces could be a platform of the so-called accidental mode degeneracies, where two or more collective modes can be simultaneously excited. If such degenerate modes have non-matching symmetries, their simultaneous excitation can reshape the local fields into highly asymmetric patterns, enhancing the buildup of an effective second-order nonlinearity. This could also involve coupling between local multipolar excitations, such as localized dark resonances,[107] taking advantage of strong local field gradients associated with the multipoles to further enhance the effects of symmetry breaking.[112,113]

B. Matching mode symmetry and phase

Large enhancement of the frequency-conversion efficiency enabled by the local and nonlocal resonances (see Fig. 6) allows the metasurfaces to be much thinner than conventional nonlinear crystals. As a result, the use of metasurfaces can largely circumvent the phase matching requirements inherent to bulk nonlinear media. However, the phase matching becomes an important factor when metasurfaces are stacked into a multilayer structure.[114] Another aspect to be considered is the phase matching in the transverse (in-plane) direction (k-matching), especially if the frequency conversion involves nonlocal resonances at off-normal incidence angles. Both the transverse and longitudinal phase matching could be facilitated by the phase gradients[115] and band folding.[78] The efficiency of the nonlinear frequency conversion in nanostructured systems depends also on the spatiotemporal overlaps of the optical modes at the input and output frequencies.[38] In nonlocal metasurfaces, the overlaps are additionally affected by the symmetries of the periodic lattice (e.g., odd/even modes and orthogonally polarized modes[116]). In such cases, designs with controllable symmetry-breaking perturbations could lead to improved mode overlap.

C. Nonlocal polarization effects

In the context of the required symmetry matching, nonlocal interactions at the fundamental and nonlinearly generated frequencies can be enabled by the symmetry of the local nonlinear susceptibility tensor. In the case of SHG, every $\boldsymbol{\chi}^{(2)}$ tensor can be decomposed into the dipolar and octupolar contributions, which are related to the rotational symmetry of the system.[117] In the systems with threefold rotational symmetry in the xy plane, the octupolar contribution becomes dominant, with the $\boldsymbol{\chi}^{(2)}$-tensor symmetry governed by relation $\chi^{(2)}_{yyy} = -\chi^{(2)}_{yxx}$, (with subscript permutation). This leads to a peculiar SHG polarization response, such as SHG having an orthogonal polarization with respect to the fundamental beam. These polarization effects have been demonstrated in triangular plasmonic nanostructures,[110,118,119] suggesting the possibility to control the coupling and nonlocal response of nanostructure assemblies and lattices.[30,120,121] Another well-known SHG polarization effect is the giant nonlinear chiroptical response,[122] which can also be enabled by nonlocal interactions.[123] Moreover, engineering the polarization properties through the effective $\boldsymbol{\chi}^{(2)}$ can be very useful from the point of view of entangled photon-pair generation via spontaneous parametric downconversion.[20] Future research directions in tailoring the polarization

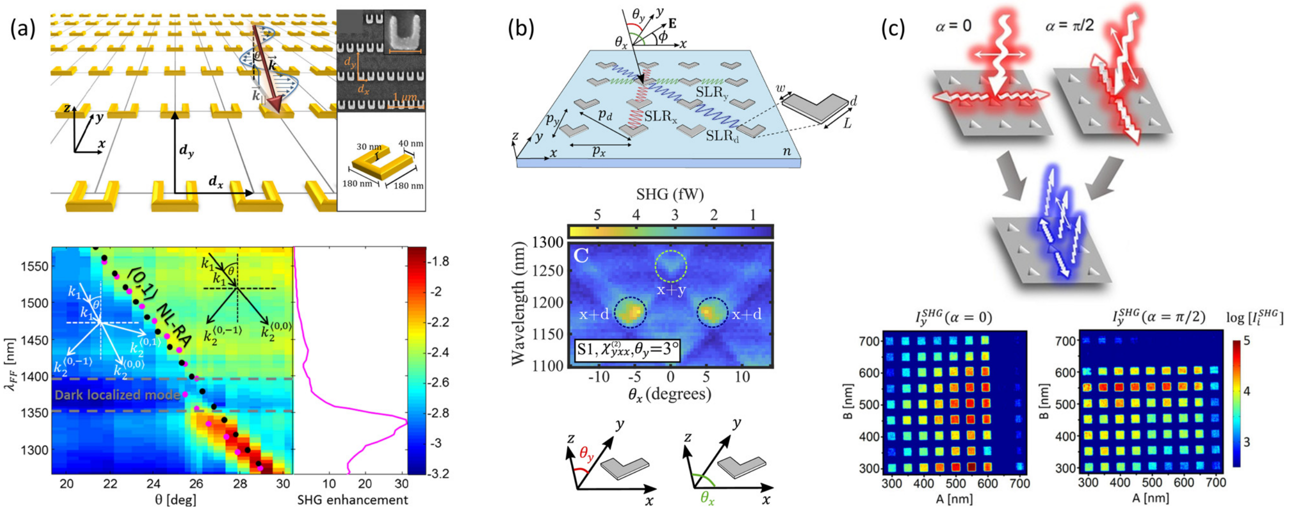

FIG. 6. Examples of nonlocal interactions governing the resonant enhancement of second-harmonic generation in periodic lattices with broken symmetry. (a) Enhancement of SHG due to SLR and Rayleigh anomalies near their anticrossing with a dark localized resonance.[107] Reprinted with permission from Michaeli *et al.*, Phys. Rev. Lett. **118**(24), 243904 (2017). Copyright 2017 American Physical Society. (b) SHG enhancement using collective resonances excited along different lattice directions.[82] Reprinted with permission from Stolt *et al.*, Opt. Express **30**(3), 3620–3631 (2022). Copyright 2022 Optica Publishing Group. (c) SHG enhancement due to collective resonances in a surface-plasmon-polaritonic (SPP) crystal at fundamental and second-harmonic frequencies, coupled through $\chi^{(2)}$ tensor constrained by the local threefold rotational symmetry. The SPP bandgap and band edge standing-wave resonances are revealed by variation of the lattice periods (A and B) at a fixed fundamental frequency.[30] Reprinted with permission from Kolkowski *et al.*, Laser Photonics Rev. **10**(2), 287–298 (2016). Copyright 2016 by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

properties of nonlinear nonlocal metasurfaces may be based on the connections between the polarization states of light and the topology of optical modes,[124,125] involving concepts such as BICs and non-Hermitian photonic bands, which are discussed in the further sections of this article.

D. Shaping the nonlinear emission

Engineering the metasurface geometry allows the nonlinear emission to be shaped in ways analogous to those used for linearly scattered light,[18] creating nonlinear versions of beam-deflecting metasurfaces,[130] metalenses,[115] meta-holograms,[131] orbital-angular-momentum (OAM) beams,[132] etc., as well as controlling the nonlinear emission processes themselves, e.g., for efficient generation of THz radiation.[106] However, most of the current designs rely on the local response of the meta-atoms, in particular, their localized plasmonic/Mie resonances. In nonlocal metasurfaces, collective resonances could be used to engineer phase gradients even in the absence of resonant meta-atoms. For example, dispersive character of the collective resonances can be employed for angular color separation in various frequency-mixing schemes, similar to what has been achieved in local linear metasurfaces.[133] Nonlocal interactions could also be used to enhance more sophisticated functionalities, including the aforementioned nonlinear beam shaping, dynamically controllable generation of nonlinear holograms and images,[134] and enhancement of phase conjugation for nonlinear imaging beyond the diffraction limit.[135] In the broader perspective, metasurfaces with spatial and/or angular dispersion,[136] such as diffraction-compensating meta-waveguides[137] and components adjusting the group velocity,[138] could also be used to control the directionality and propagation speed of nonlinear emission, especially in integrated photonic systems.

E. Bound states in the continuum

In periodic scattering systems, the symmetry can lead to complete elimination of radiation losses. This can happen through various mechanisms, e.g., by forcing the outgoing radiation to interfere destructively in the far field,[139] or by creating a polarization vortex.[140] The resulting optical states are known as the bound states in the continuum (BICs)[24,25]—highly nonlocal collective resonances that can be efficiently confined within periodic scattering systems despite being momentum-matched to freely propagating waves. Breaking the symmetry by small perturbation converts BICs into quasi-BICs, enabling their controllable excitation[84,85] and outcoupling.[71,141,142] This makes quasi-BICs highly promising for direct generation of THz waves,[128] entangled photon-pair creation,[129,143] as well as second- and higher-order harmonics, which has recently been demonstrated using various material platforms (see Fig. 7).[89,126,127,144–146]

However, direct frequency conversion schemes cannot fully exploit the most unique properties of BICs resulting from their topological nature. By merging multiple BICs in momentum space, exceptionally robust cancelation of radiation can be manifested.[26,142,147] Although in the regime of linear optical effects, such perfectly dark merging BICs are completely inaccessible by external propagating optical fields, they could be used to enhance cascaded frequency mixing processes[148,149] by providing resonant field enhancement at intermediate frequencies.

F. Non-Hermitian and $\mathcal{PT}$-symmetric metasurfaces

The concept of parity-time ($\mathcal{PT}$) symmetry is based on the fact that the interaction between modes having different complex eigenvalues (encoded in a non-Hermitian Hamiltonian) can give rise to supermodes whose eigenvalues are real.[155] In optical systems, this may

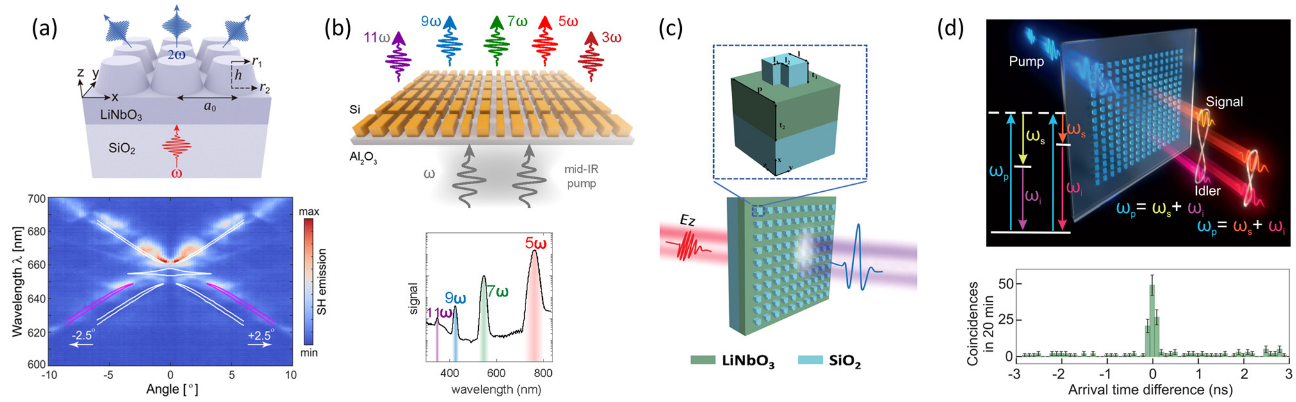

FIG. 7. Examples of nonlinear optical effects enhanced by quasi-bound states in the continuum in all-dielectric metasurfaces. (a) Second-harmonic generation enhancement around BIC in the frequency-momentum space.[126] Reprinted with permission from Zhang *et al.*, Laser Photonics Rev. **16**, 2200031 (2022). Copyright 2022 Wiley-VCH GmbH. (b) Odd-order high-harmonic generation enhanced by a quasi-BIC.[127] Reprinted with permission from Zograf *et al.*, ACS Photonics **9**(2), 567–574 (2022). Copyright 2022 American Chemical Society. (c) THz generation enhanced by a quasi-BIC.[128] Reprinted with permission from Hu *et al.*, Adv. Opt. Mater. **10**, 2200193 (2022). Copyright 2022 Wiley-VCH GmbH. (d) Entangled photon-pair creation using spontaneous parametric downconversion in a nonlinear metasurface with a quasi-BIC.[129] Reprinted with permission from Santiago-Cruz *et al.*, Science **377**(6609), 991–995 (2022). Copyright 2022 The American Association for the Advancement of Science.

correspond to coupling between resonances that differ in the imaginary parts of their eigenfrequencies, e.g., exhibiting loss and gain, or different losses. Due to unique optical effects associated with $\mathcal{PT}$ symmetry,[156,157] it has recently emerged as a novel approach to tailor the optical response of metasurfaces.[22] In the field of periodic nonlocal metasurfaces and other similar systems, such as photonic crystals, this approach is established as non-Hermitian photonic band engineering.[158] Here, the dispersion of collective resonances is shaped through formation of exceptional points and flat bands in the photonic band structure.[23,43,159,160] Controlling the polarization properties of the scattered light is one of the most natural functionalities of such metasurfaces due to the inherent chirality enforced by the topological structure of the exceptional points.[69,161–163] Nonlocal metasurfaces based on resonant scatterers (supporting either plasmonic or Mie resonances) constitute a promising platform for non-Hermitian band engineering due to large radiative damping that can be controlled by symmetry and used to provide large imaginary parts of the eigenfrequencies of nonlocal resonances. It is straightforward to apply the linear optical functionalities of non-Hermitian metasurfaces in engineering their nonlinear optical response. For example, exceptional points embedded in the photonic band structure[163] can be used to shape the polarization of nonlinear emission, whereas BICs in the middle of non-Hermitian flat bands[160] can be used as high-Q resonances to boost the nonlinear effects.

G. Metasurfaces with parametric gain

A combination of nonlinear optics and non-Hermitian physics can lead to qualitatively new phenomena. One of the most intriguing examples are metasurfaces in which gain and loss are provided by stimulated parametric downconversion.[29] Such a nonlinear process can be considered as a type of temporal harmonic modulation of the scattering properties of the medium[153] (see Fig. 8). Its non-Hermitian character is evident through the presence of momentum band gaps,[171] which are the temporal counterparts of the frequency band gaps.[172] Here, the temporal analogy of the band edges is represented by the exceptional points connected by the flat bands that span across the momentum gaps.

Parametrically driven metasurfaces can be used to realize various optical functions, such as negative refraction through nonlinear phase conjugation,[135] nonreciprocal propagation by emulating the Faraday rotation,[165,166] dynamically controlled coherent perfect absorption,[173] and parametric oscillation.[151] Future research directions could also include shaping the nonlinear emission through the so-called supercollimation and superprismatic effect, using the non-Hermitian flat bands controlled by the pump beam.[23] Nonlinear wavefront shaping could also involve parametric-gain-induced phase singularities resulting from spectral intersection of Rayleigh anomalies with the gain region in the frequency-momentum space.[174]

The presence of parametric gain could also enable realization of nonreciprocal metasurfaces acting as parametric amplifiers or oscillators[170,175] (see Fig. 9), similar to the recently demonstrated topological insulator lasers.[168,169,176] The nontrivial topological character of such metasurfaces could result from the inherent nonreciprocity encoded in the phase relation between the pump and the nonlinearly amplified optical fields.[167] The phase locking associated with stimulated parametric gain can also be used to create metasurfaces acting as parametrons,[177] offering ultrafast all-optical modulation and switching, as well as generation of phase-squeezed non-classical light by spontaneous downconversion.[129,170]

H. Actively driven time-variant metasurfaces

Parametrically driven metasurfaces described in the previous section can be regarded as a part of a more general family of time-variant metasurfaces (Fig. 8), in which the temporal change of optical properties may be anything from abrupt change to a gradual tuning that happens over many optical cycles. Abrupt changes can be considered as "time interfaces," playing the role of "temporal meta-atoms" that can be coupled to each other by temporally nonlocal interactions.[150] Such systems can support temporal analogues of spatial phenomena, such as reflection, refraction, and diffraction. This may enable formation of nontrivial topological phases[164,172,178] that are photonic counterparts of externally driven electronic Floquet systems.[179,180] Similarly, time-variant

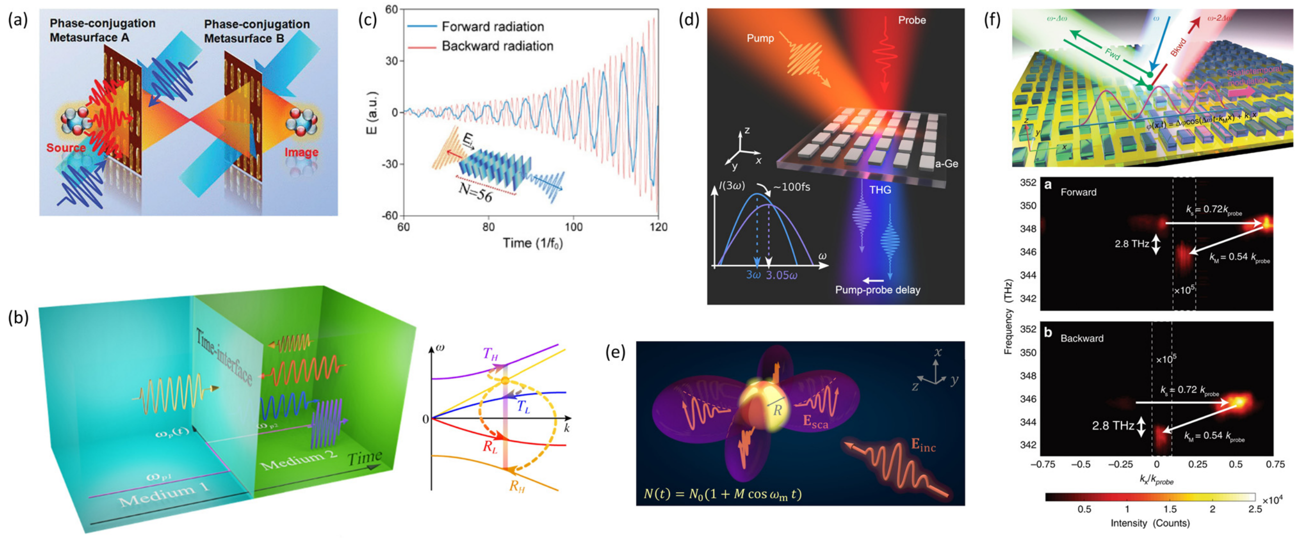

FIG. 8. Examples of time-variant nonlocal nonlinear optical systems. (a) Nonlinear metasurfaces, in which phase conjugation is enhanced by SLRs, can be used to realize perfect volumetric imaging.[135] Reprinted with permission from P.-Y. Chen and A. Alù, Nano Lett. 11(12), 5514–5518 (2011). Copyright 2011 American Chemical Society. (b) Illustration of the concept of a time interface in the case of a temporally nonlocal medium (a temporal analogue of the spatial nonlocality). Such a medium corresponds to a dispersive medium, in which the plasma frequency changes in time. The temporal scattering process (illustrated on the right-hand side) results in parametric generation of light at new frequencies.[150] (c) A spatiotemporal photonic crystal can support parametric amplification leading to parametric oscillation. This can occur inside momentum bandgaps—a temporal analogy of the frequency band gaps of the usual (spatial) photonic crystals.[151] Reprinted with permission from Lee *et al.*, Photonics Res. **9**(2), 142–150 (2021). Copyright 2021 Chinese Laser Press. (d) A rapid change of the refractive index inside a high-Q resonant metasurface induces a spectral shift of the third-harmonic generation.[152] Reprinted with permission from Zubyuk *et al.*, ACS Photonics **9**(2), 493–502 (2022). Copyright 2022 American Chemical Society. (e) Illustration of a Mie-resonant time-variant scatterer whose directional scattering characteristics and parametric amplification can be dynamically controlled through the pump field.[153] Reprinted with permission from Asadchy *et al.*, Phys. Rev. Appl. **18**(5), 054065 (2022). Copyright 2022 American Physical Society. (f) Demonstration of nonreciprocal scattering by a time-variant metasurface[154] Reprinted with permission from Guo *et al.*, Light Sci. Appl. **8**(1), 123 (2019). Copyright 2019 Nature.

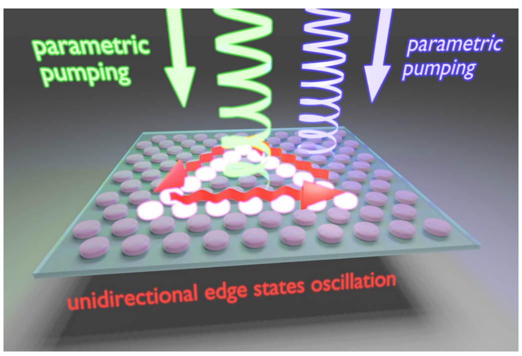

FIG. 9. Illustration of the concept of a topological metasurface-based optical parametric oscillator (OPO). In this example, the cavity for parametric oscillation is formed by unidirectional edge states[164] (red arrows) resulting from a nonreciprocal orbital angular momentum bias,[165,166] which is induced by circularly polarized pump beams (green and blue). At the same time, parametric pumping gives rise to unidirectional gain along the edges.[167] Such a parametric oscillator would be similar to the kind of topological lasers realized in gyrotropic photonic crystals,[168] except that the nonreciprocal character of the cavity would be enabled by the source of gain (i.e., the parametric pumping) itself. Similar to topological lasers,[169] topological metasurface OPOs could show superior performance and robustness against defects, e.g., in their quantum phase-squeezing characteristics.[170]

periodic systems can support artificial gauge fields for photons[181] that can effectively provide truly nonreciprocal response.[165,166,182]

Time-variant metasurfaces are inherently nonlinear, as their realization requires fast modulation of the refractive index or meta-atom polarizability,[44,153] e.g., via the Kerr nonlinearity. These metasurfaces are also a natural platform for supporting nonlinear optical functions (see Fig. 8), such as ultrafast beam steering,[173,183] nonlinearly enabled optical isolation,[154] parametric amplification,[167,184] and dynamically controllable frequency conversion,[152,185,186] including frequency shifting that occurs through Doppler or Raman-like mechanisms.[32] Time-variant metasurfaces are currently at the forefront of the research on nonlinear nonlocal metasurfaces.

I. Efficiency optimization by coherent control

As discussed earlier, the efficiency of nonlinear optical processes, such as frequency conversion, is strongly dependent on the temporal overlap between the driving pulses and the excitation in the system. This aspect of light–matter interaction is managed by coherent control of the temporal profile of the incident pulses[92,187,188] (to match the system's response) or by modification of the system parameters (to match the incident pulses). The latter can be achieved by tailoring the overall dynamics of the resonant response due to micro- and nanostructuring and the transient material-related effects (e.g., excitation of hot electrons in metal nanoparticles).[97] In metasurfaces, this strategy could take advantage of the phase interplay between local and collective resonances, exploring various types of hybrid temporal dynamics, which have recently been studied in the case of interacting local resonances.[93–96]

J. Nonlocal interactions mediated by exotic surface waves

While collective resonances, such as SLRs, are inherently confined to the metasurface plane, the nonlocal interactions between meta-atoms can be mediated by waves that are confined by other means. Such independent confinement mechanisms often enable a significant increase in the Q factors by limiting the out-of-plane scattering losses. Regarding nonlinear metasurfaces, the most straightforward optical states to be used as nonlocal interaction carriers are guided modes of a slab waveguide[65,66] and surface plasmon polaritons (SPPs).[30,120,121] Other types of confined waves, such as Dyakonov surface waves,[189,190] Bloch surface waves,[191] or gain-loss surface waves,[192] have, to date, received comparatively little attention in the context of multiple scattering and nonlinear optical effects. This stems from their elusive character and strong sensitivity to the environment, putting at question their robustness against perturbations. On the other hand, this type of limitation could be overcome by topologically robust surface waves, such as those existing at the surface of 3D topological photonic crystals.[193,194] The potential of exotic surface waves in tailoring the nonlocal interactions and nonlinear effects remains to be explored.

K. Emerging material platforms

Apart from structural engineering, one strategy to boost the efficiency of nonlinear effects is to employ alternative and innovative material platforms.[195] Here, the most challenging aspect is often the lack of established nanopatterning protocols, which hinders the use of emerging materials directly as metasurface building blocks. However, these limitations can be circumvented by combining them with metasurfaces fabricated using traditional materials, such as high-index dielectrics and plasmonic metals. A well-known example of realizing this strategy is a nonlinear metasurface that combines plasmonic nanoparticle arrays with multi-quantum-well nanostructures supporting intersubband transitions. In this case, the coupling between photonic and electronic resonances has led to a record-high second-order nonlinearity in the mid-IR spectral range.[33]

Other material platforms that have recently shown outstanding second-order nonlinear optical properties are Dirac and Weyl semimetals[34] (Fig. 10) and 2D materials resonantly coupled to semiconductor quantum dots.[35] Realization of nonlocal metasurfaces based on these platforms is yet to be demonstrated. With high-Q collective resonances, and possibly also with other concepts described in the previous sections, one may expect further enhancement of the effective second-order susceptibility and unprecedented performance of the related nonlinear effects, such as SHG, optical rectification, THz generation, and photon-pair creation.

In the field of all-optical switching, the nonlinear refractive index modulation has greatly benefited from using epsilon-near-zero (ENZ) materials.[196,197] These materials have enabled the first implementation of time-variant optical elements and their use for optical phase conjugation.[198] However, the nonlinear response of these materials is not instantaneous, as it is related to the dynamics of the excitations in the medium.[36] Nevertheless, ENZ materials remain one of the hot topics in nonlinear optics, especially in the context of tailoring the nonlocal interactions and temporal characteristics of optical media.[199]

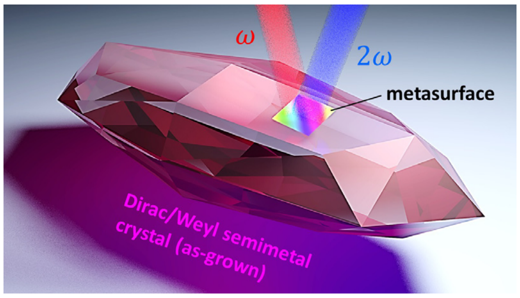

FIG. 10. Topological materials, such as Dirac and Weyl semimetals, exhibit the highest second-order susceptibility for bulk media in the near-infrared spectral range, resulting from their nontrivial electronic band structure.[34] Dirac and Weyl semimetals could be the future materials for highly nonlinear metasurfaces. Such metasurfaces could be fabricated simply by depositing a usual plasmonic or dielectric nanoparticle array on the surface of a high-quality semimetal crystal, as proposed in the illustration.

L. Hybrid structures

The nonlinear optical responses can be greatly enhanced by combining metasurfaces with other photonic structures, such as waveguides and cavities. Diffractive incoupling and outcoupling of free-space radiation to active waveguiding layers has been one of the major achievements in the realm of light-emitting metasurfaces.[13,200,201] Plasmonic lattices coupled to dielectric slab waveguides support a unique type of hybrid modes known as waveguide–plasmon polaritons.[65,66] Since the optical energy of such resonant excitations is shared between dielectric and plasmonic entities, they can exhibit very high Q-factors, as demonstrated in the example presented in Fig. 3. In slab waveguides, light undergoes multiple total internal reflections, which improves the light confinement. Replacing dielectric interfaces by strongly reflecting mirrors is a straightforward extension to Fabry–Pérot-cavity-embedded metasurfaces. In the form of metasurface etalons,[202] these structures can provide additional degrees of freedom to engineering of the collective resonances.[203] On the other hand, metasurfaces combined with large open multipass-type cavities can be useful in enhancing the overall nonlinear conversion efficiency, circumventing the challenges associated with 3D stacking and phase matching of multiple metasurfaces.[114] Incorporation of nonlinear metasurfaces into integrated photonic devices[204] is another promising research direction.

IV. OPPORTUNITIES AND CHALLENGES

Figure 11 summarizes the ideas related to future development of nonlinear nonlocal metasurfaces, including selected challenges and opportunities. Some of these challenges are similar to those associated with linear metasurfaces and their functions, which are discussed in detail in the recent reviews.[7,205,206]

Among the desired properties of nonlinear nonlocal metasurfaces (Fig. 11, green boxes on the left), frequency conversion is probably the

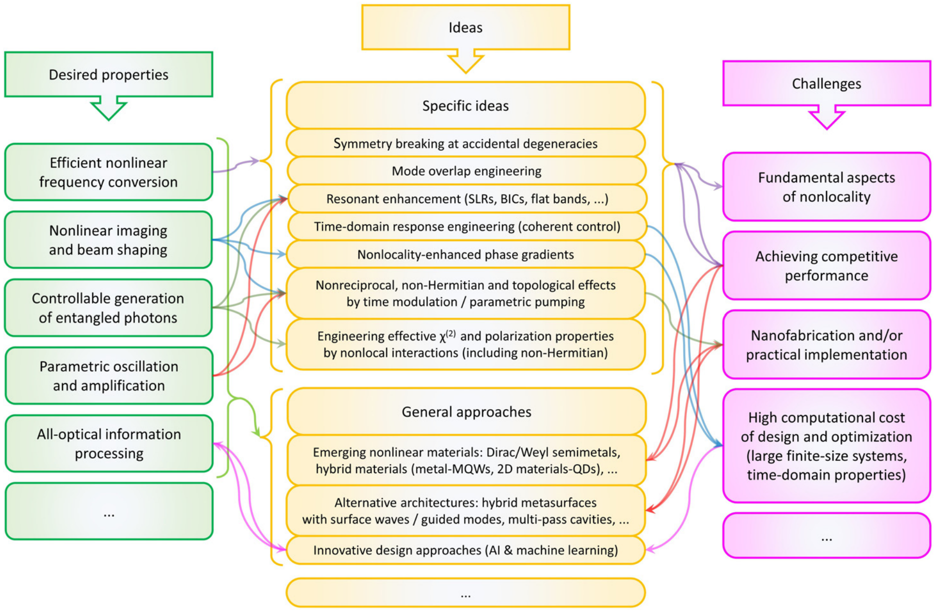

FIG. 11. Summary of the selected directions of current and future development of nonlinear nonlocal metasurfaces, including some of the specific ideas discussed throughout this article, and the associated challenges. Description is in the main text.

most investigated one. To achieve efficient frequency conversion, researchers have explored many ideas, including those listed in Fig. 11 (yellow boxes in the middle). Indeed, limited conversion efficiency in nonlinear metasurfaces is one of the biggest current challenges. The highest so-far reported conversion efficiencies have been in the range 0.001%–0.01%.[19,195] This leaves room and need for improvement, especially if one expects the metasurfaces to compete with conventional bulk optical elements. We believe that combining new materials and unconventional architectures with some of the specific ideas presented here will allow researchers to overcome this challenge, bringing frequency-converting nonlinear nonlocal metasurfaces closer to their commercial use. Among the possible alternative architectures, we consider open multi-pass cavities, stacked metasurfaces,[114] and hybrid metasurfaces (with nonlocality enhanced by guided modes and surface waves) as the most promising for enhancing the frequency conversion efficiency.

Some of the desired properties of nonlinear nonlocal metasurfaces presented in Fig. 11 are linked more specifically to certain ideas. For instance, nonlinear imaging and beam shaping can benefit mainly from resonant enhancement, phase gradients, and parametric phenomena. On the other hand, general approaches (yellow boxes at the bottom), such as emerging materials, alternative architectures, and novel design methods, can be used to achieve (or to improve) all nonlinearity-based metasurface functionalities. Here, a notable two-way connection (magenta arrows at the bottom left) is between the all-optical information processing and the design of metasurfaces using machine learning. This is because metasurfaces (and other photonic structures in general) can be designed using artificial neural networks,[205,207,208] while they can also be used to implement such networks.[209] In such a case, nonlinear metasurfaces could hinge on their capacity to perform massively parallel all-optical information processing,[210] while the nonlinearity could make them dynamically reprogrammable.

From the main challenges listed in Fig. 11 (magenta boxes on the right), probably the most critical are fundamental aspects of nonlocality (real-space vs momentum-space and time-domain vs frequency-domain engineering), as they determine the suitability of nonlocal properties in all applications.[206] For example, nonlinear optical functions requiring broadband and ultrafast operation may actually not benefit at all from spatial nonlocality (see Fig. 4). Here, a careful design is necessary to take advantage of the nonlocal properties, such as high-Q collective resonances.[211]

Other major challenges are related to the feasibility of practical realization of nonlinear nonlocal metasurfaces. This includes nanofabrication, which may be problematic for emerging nonlinear materials due to the lack of well-established nanopatterning methods for some of these materials. This is especially challenging in the case of second-order nonlinear materials (e.g., Weyl and Dirac semimetals), which require a high degree of crystallinity to maintain their nontrivial electronic properties and a high nonlinear optical susceptibility. Possible solutions may involve hybrid architectures, as proposed in Fig. 10. Another related challenge is the practical implementation of some of the novel concepts, especially related to time-variant and $\mathcal{PT}$-symmetric metasurfaces.[69] Whether unique optical properties enabled by such novel concepts can be accompanied by a decent efficiency remains an open question.

Finally, computational costs related to the design and optimization are a well-known issue, especially in the case of engineering the time-domain response and for large photonic structures. Here, it seems very likely that the artificial intelligence may soon become the game changer, replacing exact modeling in most of the practical design tasks, not only in the field of nonlinear nonlocal metasurfaces, but in optics and photonics in general.

V. CONCLUSION

In this work, we have provided an overview of nonlinear nonlocal metasurfaces, identifying various physical mechanisms that govern their nonlinear response. We presented some of the most interesting current trends in the field and discussed the possible future research directions. The collective nonlocal optical response enables strong light–matter interactions to take place even in optically thin systems, which is particularly promising for ever increasing number of applications where flat optical components are required. The ability to tailor the system response at the individual meta-atom level enables unprecedented control over the amplitude, polarization, and phase response of the nonlinear material. In addition, tailoring the collective meta-atomic response offers entirely new degrees of freedom in nonlinear manipulation of light. These degrees of freedom will likely lead to new avenues to be taken in the realm of nonlinear optics. Examples include, but are not limited to, topological effects, $\mathcal{PT}$-symmetry, temporal modulation, nonlinear geometric phases, and new nonlinear materials. Thus, nonlinear nonlocal metasurfaces have the potential not only to outperform their conventional counterparts but also to provide novel functionalities and exhibit new light–matter interaction mechanisms.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Radoslaw Kolkowski: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Software (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Tommi Hakala:** Conceptualization (equal); Funding acquisition (equal); Writing – original draft (equal); Writing – review & editing (equal). **Andriy Shevchenko:** Conceptualization (equal); Funding acquisition (equal); Methodology (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **Mikko J. Huttunen:** Conceptualization (equal); Funding acquisition (equal); Methodology (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data presented in Fig. 3 and 4 and their replication instructions are openly available at https://doi.org/10.23729/a049df84–20ef-4ac7-a0d2-ed9f42958125, Ref. 212.

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