Nonlinear optics in metamaterials

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ABSTRACT

Nonlinear optics focuses on the phenomena that arise when intense light interacts with matter. At low intensities, optical response of a material scales linearly with the amplitude of the electric field. However, at high intensities, light–matter interactions become more complicated leading to such fascinating nonlinear effects as self-focusing, soliton propagation, and high-harmonic generation. Although the field of nonlinear optics was developing for many decades since the invention of lasers, nonlinear optical materials available to date are still limited by either slow materials’ response time in such phenomena as saturable absorption, photorefractive effect, and thermal nonlinear phenomena, or by relatively low and generally band-limited nonlinear susceptibilities responsible for ultra-fast nonlinear processes. The emergence of metamaterials is likely to revolutionize nonlinear optics, enabling to low-power, compact, and ultra-fast applications of nonlinear optical phenomena. We review a subset of recent progress in theoretical and experimental studies of nonlinear optical properties and phenomena enabled by the unique properties of engineered optical media. We discuss new regimes of nonlinear optical interactions enabled by judiciously designed metamaterials; review fundamental limit of nonlinear optical phenomena and several metamaterial-based approaches to enhancing nonlinearities at optical frequencies; and consider engineered nonlinear properties of soft matter-based nonlinear media.

KEYWORDS

Optical nonlinearities; Nonlinear metamaterials; Phase matching; Modulation instability

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1. Introduction

For years, researchers have been exploring ways of creating materials with a large, fast, and broadband nonlinear response. Some progress in this direction has been achieved in the microwave frequency range. Designing and fabricating such materials at higher frequencies would revolutionize nonlinear optics, leading to low-power, compact, and ultra-fast applications of nonlinear optical phenomena. The emergence of metamaterials (MMs) has a potential to provide a breakthrough in the development of such materials. By selecting proper building block materials and rationally designing them on a scale of nanometers, it should be possible to create composite electromagnetic media with properties significantly different from their individual constituents and suitable for the realization of new functionalities.

Over the past decade, optical MMs have been predicted and demonstrated to enable a number of unique linear optical properties. For instance, their dielectric permittivity, magnetic permeability, and refractive index that can be designed to be positive, negative or even zero at any selected frequency by properly adjusting the dimensions, periodicity, and other properties of the so-called meta-atoms (the unit cells of MMs) [1–18]. Therefore, it can be expected that a similar approach can be used transform the nonlinear properties of MMs. Figure 1(a) shows several examples of nonlinear meta-atoms proposed and demonstrated for MM-enhanced nonlinear optical interactions [19–22]. The nonlinear optical response of a material is generated by the collective response of the basic elements comprising it and therefore, is limited by the properties of the conventional atoms or molecules. While some of these limits are fundamental in character, depending, for example,

![Figure 1](image-url)

**Figure 1.** (a) Nonlinear meta-atoms proposed and demonstrated for MMs-enhanced nonlinear optics [19–22]. (b) First hyperpolarizability vs. wavelength of the E10 transition.

Notes: The data points appearing below the dashed line are the experimental data. The solid line labeled shows the prediction of the three-level model [23,25] and the dashed–dotted line corresponds to the highest value calculated directly from many Hamiltonian models [28].
on the characteristics of the energy levels involved or on the matrix elements of transitions that give rise to the nonlinearity, others can be manipulated to optimize particular interactions.

The question of materials’ largest possible nonlinear optical response has drawn the attention of researchers for many years \([23–29]\). The properties of nonlinear materials are better understood when discussed with reference to the theory of nonlinear phenomena and desirable functionalities. Four main criteria that a material should satisfy in order to enable efficient optical interactions and their applications are sufficient nonlinearity, optical transparency, proper phase-matching conditions, and sufficient resistance to optical damage by intense optical irradiation. Microscopically, the nonlinear terms arise through several mechanisms such as electronic, atomic (including molecular motions), electrostatic, and thermal processes. Among these, the electronic process can provide the fastest response with fs–ps time scales, which is usually needed for optical processing, switching, and logic.

About 15 years ago, Kuzyk predicted that there are fundamental limits to the off-resonant, electronic, nonlinear optical response \([23,25]\). Today, the theory of the fundamental limits of nonlinear optics is a powerful tool for experimentalists seeking to create molecules and materials with large responses. Figure 1(b) is an updated version of an original plot in Kuzyk’s paper \([28]\), showing the value of the first hyperpolarizability versus wavelength, associated with the \(E_{10}\)-level transition. The dots are experimental measurements representing all of the known measurements at that time. This figure clearly indicates that the experimental data fall below an apparent or empirical boundary suggesting that there may be many opportunities to design better nonlinear materials, if we understand which properties of the conventional molecules determine their hyperpolarizability. In particular, engineering nonlinear properties beyond those available in nature may be feasible by judiciously designing their quantum, geometric, and topological properties.

In parallel with the development of fundamental quantum models, several semi-empirical relationships have been proposed to estimate the nonlinear response from its linear counterpart \([30–37]\). Since MMs gain their unique linear properties from their structural design, it is not obvious whether any generalized rules relating linear and nonlinear properties can be established. The initial studies of the second-order susceptibility of a plasmonic metasurface, suggest that while the direct application of Miller’s rule may not provide accurate estimates, nonlinear scattering theory approach agrees well with experimental results \([37]\).

While the systematic approach to designing nonlinear materials at optical frequencies guided by the theoretical models is still in its early stage, rather a significant progress has been made in other spectral ranges. In 1991, long before the MMs field emerged, Kalinin and Shtykov suggested the construction of an artificial nonlinear medium from dipoles with nonlinear inclusions \([38]\). Later, Lapine et al. theoretically considered the possibility of creating a magnetic nonlinear MM by inserting a diode into each resonant conductive element, which can be represented by linear RLC contours \([39]\). Zharov et al. have investigated the nonlinear response
of a two-dimensional negative index material (NIM) periodic structure consisting of arrays of metallic wires and split-ring resonators (SRRs) embedded into a nonlinear dielectric and operating at microwave frequencies [40]. Two important contributions to the nonlinear properties of this structure have been identified. The first one originates from an intensity-dependent part of the effective dielectric permittivity of the nonlinear dielectric host material. The second contribution originates from the periodic structures of resonators, since the SRR capacitance depends on the strength of the local electric field in a narrow slot and results in the intensity-dependent effective magnetic permeability. The magnetic nonlinearity in this case has been found to be much stronger than the nonlinearity in the dielectric properties, owing to the field enhancement in the SRRs. At optical frequencies, in the related field of plasmonics, to date, significant efforts have been devoted to the enhancement of nonlinear optical response using various plasmonic nanostructures [41–51], including metallic bow tie nanoantennas, dimers, trimers, and other structures sometimes referred to as plasmonic MMs. While these first steps demonstrate the feasibility of locally enhancing the nonlinear response, it is important to note that a majority of these studies essentially exploited strong local field enhancements enabled by plasmonic nanostructures rather than actual rational design of the nonlinear optical response of the MM. There are several limitations associated with such an approach [47]. First, the majority of approaches rely on strong local field enhancement in metal-dielectric nanostructures leading to very large local effective nonlinear refractive index, while strong absorption in the metals limits the attainable nonlinear phase shift (in the case of self- and cross-phase modulation effects) and frequency conversion efficiency (in the case of four-wave mixing) at very short distances. It should be noted that local field enhancements are not unique to MMs and have been previously realized in many other structures, including photonic bandgap structures, micro-cavities, and ring resonators [52–54]. Moreover, despite many impressive demonstrations of various high-Q resonantly enhanced structures, careful analysis suggests that they are not as promising as was initially hoped for nonlinear optical processes requiring high efficiency such as wavelength conversion or switching [47]. Therefore, in the recent years MMs researchers have been looking for more sophisticated strategies for designing nonlinear properties of optical materials at will.

Nowadays, nonlinear optics in metamaterials and metasurfaces is advancing in many exciting directions, and therefore, in this short review paper, we had to limit ourselves to discussion only a subset of this very broad field. For a more comprehensive review of the fascinating field of nonlinear metamaterials we refer readers to Ref. [55]. In the following sections, we discuss recent theoretical predictions and experimental demonstrations of new regimes of nonlinear optical interactions enabled by judiciously designed MMs (Section 2), several MM-based approaches to enhancing nonlinearities at optical frequencies (Section 3), and engineered nonlinear properties of soft matter-based nonlinear media (Section 4). In Section 5, we summarize
recent progress in nonlinear light–matter interactions in engineered optical media and discuss remaining questions in this fascinating and rapidly developing field.

2. Novel regimes of nonlinear light–matter interactions in metamaterials

Since the early days of nonlinear optics, it was realized that phase matching is a critical requirement for many coherent nonlinear optical processes. The phase mismatch between the waves propagating originates from material dispersion and results in a lack of optical momentum conservation between different photons. The phase mismatch prevents the constructive interference of the nonlinear fields and as a result, leads to the reduction of the efficiency of nonlinear optical interactions. In conventional optical materials, the most common approaches to obtaining efficient nonlinear interactions include birefringence phase matching, angle phase matching, and quasi-phase matching. Metamaterials open new possibilities for the realization of efficient nonlinear optical interactions such as wavelength conversion and parametric amplification effects.

For instance, phase mismatch-free nonlinear generation was predicted and experimentally demonstrated in a zero index optical metamaterials [56]. In particular, it was shown that the zero index eliminates the need for phase matching, allowing efficient nonlinear generation in both forward and backward directions. Indeed, in the microscopic picture of nonlinear wave interactions, each point of the wavefront can be considered as a source that accumulates a phase that is proportional to the refractive index as it propagates in a finite refractive index medium. For maximum efficiency of nonlinear interactions these sources have to add up coherently. However, in a zero index material, the emission from all these sources acquire no phase as they propagate, thus ensuring constructive interference and an increase of the signal in both directions of propagation.

On the other hand, many new regimes of nonlinear interactions were predicted in NIMs, including backward phase matching, unconventional Manley–Rowe relations, spatially distributed nonlinear feedback, and cavity-less optical parametrical oscillations [57–77]. These unusual properties potentially enable such novel functionalities as quadratic nonlinear mirror and second-harmonic generation (SHG)-based lenses. Historically, SHG, one of the first nonlinear optical phenomena that was observed in laboratory experiment [78], gave rise to intensive studies of other nonlinear wave-mixing processes, including spontaneous parametric down conversion that is of a particular importance for quantum optics [79]. As with many other nonlinear processes, the SHG relies on phase matching between waves at fundamental and doubled frequencies. Over the past several decades, many phase-matching approaches and geometries have been proposed aiming at increasing conversion efficiency of the fundamental field (FF) into the second-harmonic (SH) one. In particular, it was pointed out a long time ago [80] that backward phase matching of collinear waves would be desirable but is difficult
to realize in the visible and near-infrared frequency range. One approach to the realization of backward phase matching relies on using quasi-phase matching [81]. Negative index materials offers another way for the realization of backward phase matching owing to their inherent properties, including (i) opposite directionality of Poynting vector and wave vector and (ii) frequency-dependent material parameters such that the same material could have negative refractive index in a certain range of frequencies and positive refractive index at other frequencies.

In order to understand the details of SHG process in NIMs, we consider a material that possesses negative refractive index at the FF frequency ω and positive refractive index at the second-harmonic SH frequency 2ω. The electric fields of the FF and that of the SH waves are taken in the following form, respectively. The process of SHG in the NIM in a slowly varying envelope and phase approximation can be written in the following form

\[
\begin{align*}
\left(-\frac{\partial}{\partial z} + \frac{1}{v_\omega} \frac{\partial}{\partial t}\right)A_\omega &= \frac{2\pi\omega^2 \mu_\omega}{c^2 k_\omega} P_{NL}(\omega) \exp(-ik_\omega z) \\
\left(-\frac{\partial}{\partial z} + \frac{1}{v_{2\omega}} \frac{\partial}{\partial t}\right)A_{2\omega} &= \frac{2\pi(2\omega)^2 \mu_{2\omega}}{c^2 k_{2\omega}} P_{NL}(2\omega) \exp(-ik_{2\omega} z)
\end{align*}
\]

Here, z is the propagation coordinate, t is time, \(A_\omega\) and \(A_{2\omega}\) are the slowly varying amplitudes of the waves, \(k_\omega^2 = (\omega/c)^2 \varepsilon(\omega) \mu(\omega)\) are the wave vectors at frequencies ω and 2ω, respectively, \(\varepsilon(\omega)\) and \(\mu(\omega)\) are the dielectric permittivity and magnetic permeabilities at FF(SH) frequencies, respectively, c is the speed of light in free space, \(v_\omega\) (\(v_{2\omega}\)) is the group velocities, and \(P_{NL}\) is the nonlinear polarizabilities. Considering the continuous wave case and using the symmetry properties of the tensor \(\chi^{(2)}[82,83]\), Equation (1) can be written in the following form

\[
\begin{align*}
\frac{\partial A_\omega}{\partial z} &= -i\frac{2K\omega^2 \mu_\omega}{c^2 k_\omega} A_{2\omega} A_\omega^* \exp\left[-i\Delta k z\right], \\
\frac{\partial A_{2\omega}}{\partial z} &= i\frac{4K\omega^2 \mu_{2\omega}}{c^2 k_{2\omega}} A_{2\omega}^2 \exp\left[i\Delta k z\right],
\end{align*}
\]

where \(K = \frac{2\pi}{c^2} \chi^{(2)}(2\omega) = \frac{\pi}{c^2} \chi^{(2)}(\omega)\), \(\chi^{(2)}\) is the effective nonlinear susceptibility and \(\Delta k = k_{2\omega} - 2k_\omega\) is the phase mismatch.

From Equation (2) we find

\[
\frac{k_\omega}{\mu_\omega} \frac{d|A_\omega|^2}{dz} - \frac{k_{2\omega}}{2\mu_{2\omega}} \frac{d|A_{2\omega}|^2}{dz} = 0.
\]

Assuming that the phase-matching condition \(k_{2\omega} = 2k_\omega\) is satisfied (implying that \(\mu_\omega = -\mu_{2\omega}\)), the spatially invariant Manley–Rowe relations take the form

\[
|A_\omega|^2 - |A_{2\omega}|^2 = C^2 = \text{const.}
\]
In a conventional positive index material (PIM), the Manley–Rowe relations require that the sum of the squared amplitudes is constant. This unusual form of Manley–Rowe relations (4) in NIMs is the result of the fact that the Poynting vectors for the fundamental and the second harmonic are antiparallel, while their wave vectors are parallel. Equation (4) shows that the incoming radiation at the fundamental frequency can be converted to the second-harmonic frequency propagating in opposite direction with efficiency approaching 100%. Thus, the NIM slab acts as a nonlinear mirror.

Backward SHG and the predicted nonlinear mirror-like behavior were experimentally demonstrated at microwave wavelengths using the experimental setup shown in Figure 2(a) [77]. The nonlinear MM was realized using the varactor-loaded split-ring resonators placed in an aluminum waveguide (Figure 2(b)). SHG was studied in three configurations, including the reflected SH phase matching in a negative index spectral range, transmitted SH quasi-phase matching, and simultaneous quasi-phase matching of both the reflected and transmitted SH waves near a zero index spectral range. Additionally, experimental measurements of three- and four-wave mixing phenomena in an artificially structured nonlinear magnetic metacrystal at microwave frequencies have been reported [76]. However, applying a similar approach to experimental realization of the backward

Figure 2. Phase-matching conditions for second-harmonic generation in nonlinear optical media [84]: (upper plot) conventional forward phase matching, and (lower plot) backward phase matching enabled by NIMs. (b) Image of NIM based on a waveguide loaded with four identical sections of varactor-loaded split-ring resonator [77]. Inset shows an enlarged view of unit cell. (c) Experimental design for backward phase matching in a plasmonic waveguide with a double-layered dielectric core [84]. (d) Second-harmonic generation with optical vortices in NIMs: the helical wavefront for the FF and SH for an incident Laguerre–Gaussian beam with topological charge –1.

Notes: Arrows show the direction of rotation of the wavefront for the FF in PIM (top left) and in NIM (top right), for the SH (bottom) [85].
phase-matched nonlinear processes at optical frequencies is challenging due to the difficulties in fabrication of sufficiently large NIMs and designing proper refractive index at both the fundamental and harmonic frequencies simultaneously in bulk engineered media.

Recently, Cai et al. proposed that instead of engineering refractive indices across different wavelength ranges in bulk optical MMs, the index-matching relation of $n_{2\omega} = -n_{\omega}$ can be achieved using two distinct modes supported in a plasmonic waveguide. Indeed, when nonlinear wave interactions occur in a waveguide configuration, the phase-matching condition for SHG requires the propagation constants of the FF and SH waves to be identical. This condition can be satisfied by exploiting the dispersion characteristics of different waveguide modes. In particular, backward phase matching was realized by optimizing the material and geometry parameters of the plasmonic waveguide such that the real parts of the mode refractive indices were 3.4 and −3.4 for the fundamental and the harmonic waves, respectively, as shown in Figure 2(c). In this demonstration Instead of the regular $\chi^{(2)}$ response intrinsic to non-centrosymmetric crystals and interfaces, the effective second-order nonlinearity was produced by the interplay between the third-order nonlinear susceptibility $\chi^{(3)}$ and the applied control field [84]. In addition, the 30-nm-thick dielectric spacer is split into two halves, 15 nm each, made of Si$_3$N$_4$ and HfO$_2$ (Figure 2(c)). These two materials have similar linear refractive indices, but very different electrical conductivity and nonlinear properties. As a result, the electrically induced $\chi^{(2)}$ nonlinearity was effectively absent in one half of the dielectric channel.

Until recently, a majority of studies of linear and nonlinear phenomena in MMs focused on simple light beams carrying linear momentum only. However, a combination of unprecedented materials design flexibility facilitated by MM technology and unique properties of structured light beams, containing phase or polarization singularities was shown to result in unusual properties of SHG with optical beams carrying the orbital angular momentum (OAM) in NIMs, as shown in Figure 2(d). In particular, it was demonstrated that the Laguerre–Gaussian beam launched from the positive linear material changes its helicity in the NIM, while the generated SH wave propagates backward with simultaneously doubled frequency, doubled OAM, and reversed rotation direction of the wavefront. These results maybe generalized to the case of efficient parametric wavelength conversion of OAM light that is of a particular importance for high-dimensional communication systems, quantum information processing, and optical manipulation on nanoscale [85].

3. Enhanced optical nonlinearities in metamaterials

As discussed in the introduction, many previously demonstrated approaches to achieving enhanced nonlinearity in MMs were based on the resonantly enhanced fields by the MMs unit cells (or meta-atoms), such as strong field enhancements
occurring in the capacitive regions of the SRR. Then, a nonlinear material or element placed in the vicinity of such local field enhancement leads to the enhanced local and effective nonlinear response of the composite medium. While this approach indeed leads to the increase of effective nonlinearity through the local field enhancements inside each meta-atom, such resonant enhancement usually comes at the expense of increased loss or decreased coherence length, which limits its use for ultra-fast, low-power switching and wavelength conversion [47, 86]. It is important to realize that for all-optical switching or wavelength conversion applications, a large value of nonlinear index of refraction is not enough. Instead, it is the maximum amount of phase shift that can be attained over a distance that is less than one absorption length determines the efficiency or the figure of merit (FOM) for nonlinear materials developed for such applications.

Two processes can limit this FOM: linear optical absorption and saturation of the nonlinearity. Therefore, efficient all-optical switching applications require materials with a large nonlinear refractive index, but relatively small linear and nonlinear absorption loss. A majority of nonlinear optical interactions in these materials, including self-phase modulations, cross-phase modulation, and four-wave mixing, rely on phase shift, $\Phi = \gamma P L$, induced by either the signal itself or by another control signal. Here, the nonlinear coefficient $\gamma$ depends on the nonlinear refractive index $n_2$, the effective mode area $A_{\text{eff}}$, the peak pulse power $P$, and the interaction length $L$ as $\gamma = \beta n_2 / c A_{\text{eff}}$. Then, the FOM can be defined as $\text{FOM} = n_2 / \beta \lambda$, where $\beta$ is the absorption coefficient, characterizes the nonlinear phase shift achievable over one absorption length. The difficulty in nonlinear optics is therefore finding a material that has both a large Kerr nonlinear coefficient and a small absorption coefficient. Another possibility is to use materials with relatively low nonlinearity, low absorption, but long propagation length. The best-known example of such media to date is optical fiber [87, 88]. The drawback of optical fiber-based nonlinear devices is obviously excessively large size (length) making it difficult to integrate them on a chip. Other nonlinear materials that have been discussed in a context of optical signal processing applications include silicon [89] and chalcogenide glass [90]. However, the trade-offs in practical applications of these materials are still significantly long propagation lengths and non-negligible two photon absorption.

Recently, Khurgin et al. [91] showed that the most general reason for relatively small nonlinearity in optical materials is simply the fact that fast ‘electronic’ nonlinearity that originates from a small displacement of electronic cloud confined by the binding potential, while the potential itself stays unchanged. In contrast, thermal nonlinearity that is related to the changes in the distance between neighboring atoms as the temperature increases (i.e. the change in the binding potential itself) leads to much larger changes of refractive index. This nonlinearity is conventionally considered to be too slow for any practical applications for ultra-fast all-optical switching. However, as it was pointed out in [91] the intrinsic speed of thermal nonlinearity determined by the speed of relative motion of ions can be
as fast as a few hundred femtoseconds. The reason for the conventionally measured thermal nonlinearity of a bulk material being slow is the slow heat dissipation, when the heated region is relatively large (on the micrometer scale). Remarkably, it was shown that when optical fields are concentrated on the scale of few tens of nanometers, the speed of the thermo-optical effects approaches picosecond scale and leads to phase shifts sufficient for all optical switching on the ultra-fast scale. This intriguing prediction is awaiting experimental demonstration.

Next, we discuss two recent experimental demonstrations of large nonlinear response facilitated by engineered MM structures attained using two fundamentally different approaches. Back in 1980s and 1990s, it was shown that intersubband transitions in \( n \)-doped multi-quantum well semiconductor heterostructures enable one of the largest known nonlinear optical responses in condensed matter systems. By controlling the widths of wells and barriers in these structures, one can tailor the transition energy and dipole moments between electron subbands in order to maximize the quantum mechanical expression for a particular nonlinear process. However, this nonlinear response is limited to light with electric field polarized normal to the semiconductor layers [92]. Recent progress in the field of optical metasurfaces [93–98], two-dimensional analogues of MMs, offers a solution to this problem. Compared to conventional optical elements, which rely on long propagation distances, these devices facilitate strong light–matter interaction on a subwavelength scale, allowing abrupt changes of beam parameters. Moreover, metasurfaces, unlike bulk metamaterials, do not require complicated fabrication techniques, which makes them very promising for integration on a photonic chip and well suited for mass production. Combining quantum electronic engineering of intersubband nonlinearities with electromagnetic engineering of plasmonic nanoresonators, ultra-thin, planarized, highly nonlinear optical 400-nm-thick metasurface with nonlinear susceptibility of greater than \( 5 \times 10^4 \) pm/V for SHG at 8 microns was demonstrated (Figure 3).

Another approach to engineering of strongly nonlinear MMs was demonstrated by structuring three centrosymmetric dielectrics on a nanoscale to realize

![Figure 3. Nonlinear metasurface structure [92]: (a) Conduction band diagram of one period of an In\(_{0.53}\)Ga\(_{0.47}\)As/Al\(_{0.48}\)In\(_{0.52}\)As coupled quantum well structure designed for giant nonlinear response for SHG. (b) Schematic of the metasurface design and operation. (c) TEM image of an ABC-stack made by iterating \( M = 25 \) times the deposition of \( N = 12 \) cycles of each of the materials A = Al\(_2\)O\(_3\), B = TiO\(_2\), and C = HfO\(_2\) [99].](image-url)
nanolaminates exhibiting second-order nonlinearity that none of the constituent components had. Thin layers of A = Al₂O₃, B = TiO₂, and C = HfO₂ were arranged into a non-centrosymmetric ABC-stack such that the individual surface nonlinearities originating at the boundary of neighboring materials do not sum up to zero, as shown in Figure 3(c) [99]. Following this approach, artificial unidimensional crystals with the main component of their nonlinear susceptibility tensor of ~5 pm/V, which are comparable to well-established materials, were reported [100]. These dielectric nonlinear MMs were grown by atomic layer deposition making them compatible with standard fabrication technologies. The nonlinearity of the nanolaminates can be increased in several ways. For example, it was suggested that quaternary samples can achieve higher nonlinearities than any ternary sample utilizing the same ingredients. Other material combination and optimized fabrication process are likely to enable larger nonlinearities and higher density of interfaces, respectively.

The unusual properties of epsilon-near-zero and zero index materials have attracted a significant attention due to their promise for applications in nonlinear optics [101–109]. Recently, it was shown that indium tin oxide can acquire an ultrafast and large intensity-dependent refractive index in the region of the spectrum where the real part of its permittivity vanishes [107]. A reversible change in the real part of the refractive index of 0.72 ± 0.025, corresponding to 170% of the linear refractive index, was observed. In another study, a sixfold increase of the Kerr nonlinear refractive index in Al-doped ZnO (AZO) thin films at the epsilon-near-zero wavelength, corresponding to 1300 nm, was experimentally demonstrated [108].

Finally, another promising approach to the development of novel, strongly nonlinear materials is enabled by all-dielectric systems. As opposed to extensively studies plasmonic particles-based nonlinear optical materials that suffer from significant losses, all-dielectric structures enable strong nonlinear response with considerably reduced losses.

Recent progress of all-dielectric nanostructures gave rise to the emerging field of multipolar nonlinear nanophotonics [110–114]. In all-dielectric nanostructures, the nonlinear effects are enhanced due to multipolar Mie-type resonances and optically induced magnetic response potentially enabling nonlinear photonic devices operating beyond the diffraction limit. Such nanostructures can be built of isolated or clustered dielectric nanodisks [113,114]. For example, a strong enhancement of third-harmonic generation was demonstrated using both isolated and arrayed silicon nanodisks. Such silicon nanodisks, pumped by resonant laser radiation, considerably boosted the resonant third-harmonic response by a factor of up to 100 compared with the THG from bulk silicon [113].

4. Nonlinear optics in engineered colloidal media

Until recently, the majority of studies in the field of MMs have focused on solid-state nanostructures. However, the possibility of engineering of optical properties in
soft matter offers new degrees of freedom for designing optical polarizabilities and nonlinearities. While the first ideas of using interplay of optical forces [114] and nonlinear self-action effects [115] were proposed in the 70s and 80s; recently, it was demonstrated that colloidal suspensions can offer a promising platform for engineering polarizabilities and large, nonlinearities [116–124]. It was shown that the nonlinear response of colloidal suspensions can be tailored at will by judiciously engineering their optical polarizability [117]. Such systems provide new opportunities for fundamental studies and applications of such nonlinear effects as self-focusing and related beam break-up effects.

Propagation of an optical beam in a nanocolloidal system can be described by the following equation governing the electric field envelope $\varphi$ [121,122]

$$i \frac{\partial \varphi}{\partial Z} + \frac{1}{2k_b n_b} \nabla^2 \varphi + k_0 \left( n_b - n_p \right) V_p \rho_0 e^{i\frac{2\pi}{\lambda_0} |\varphi|^2} \varphi + \frac{i}{2} \sigma \rho_0 e^{i\frac{2\pi}{\lambda_0} |\varphi|^2} \varphi = 0,$$  \hspace{1cm} (5)

where $\alpha$ is the particle polarizability, $k_b T$ is the thermal energy, $n_p$ and $n_b$ are the particle and background refractive indices, respectively, $V_p$ is the volume of a particle, $\rho_0$ is the unperturbed particle concentration, $\sigma$ is the scattering cross-section, and $k_0 = 2\pi/\lambda_0$ is the wave number. The particle polarizability is given by

$$\alpha = 3V_p \varepsilon_0 n_b^2 \left( \frac{m^2 - 1}{m^2 + 2} \right),$$  \hspace{1cm} (6)

where $m = n_p/n_b$, is the ratio of refractive indices of the particle and that of the background material. From the seminal work by Ashkin, it is known that the optical gradient force defines dynamics of light–matter interactions in colloidal suspensions. The optical gradient force can be written as $\vec{F} = \alpha \nabla I/4$, where $I$ is the optical intensity and $\alpha$ is the particle polarizability. Considering a particle near the waist of a Gaussian beam, the formula shows that a particle with positive polarizability (PP) experiences a force pulling it toward the beam. On the other hand, negative polarizability (NP) particles experience a repelling force pointing outward, as illustrated in Figure 4(a).

Light–matter interactions in colloidal systems can be significantly altered by changing particles’ polarizabilities, which depend on the particle and the background medium refractive indices. The particles with the refractive index larger than that of the background medium possess positive polarizability. When laser beam propagated in such a medium, these particles are attracted to the high-intensity region due to the radiation pressure. The particles with the refractive index lower than that of the background medium have negative polarizability and thus get repelled from the high-intensity part of the light beam in Figure 4(a).

Despite the initial belief that optical nonlinearity of colloidal suspensions is of the Kerr type, it was found to be exponential [117]. Moreover, this exponential nonlinearity can be saturable or supercritical with intensity, depending on the sign of
the particle polarizability in the suspension. The z-scan measurements showed that the PP nonlinearity exceeds the typical Kerr response because of the exponential character of the Boltzmann law involved in the particle distribution function. This super-critical nature of the nonlinearity leads to highly unstable propagation and catastrophic collapse in PP suspensions as shown in Figure 4(b). On the other hand, NP suspensions exhibit a saturable Kerr response preventing the collapse of the light beam and leading to longer propagation lengths (Figure 4(c)). It was first discussed in detail by El-Ganainy that in both cases (of positive and negative polarizability), the nonlinearity is of the self-focusing type. This rather unexpected conclusion can be understood as follows. In the case particles having a higher refractive index than the background medium, the polarizability of each particle is positive and thus the particles are attracted toward the high-intensity region, i.e. to the center of the beam, thus elevating the effective refractive index of the system. Obviously, this will increase the nonlinear scattering losses as well. On the other hand, particles having a lower refractive index than that of the background medium and hence a negative polarizability will be repelled away from the center of the beam, again effectively increasing the refractive index at the center. The important difference, however, is that in the case of NP-particles, the nonlinear losses decrease at the beam center due to the reduction in the particle concentration, thus increasing the transparency of the system. This difference in the type

Figure 4. (a) Schematic representation of the beam propagation in a colloidal suspension consisting of positive and negative polarizability particles. (b)–(c) Experimental demonstration of Gaussian beam propagation in different colloidal media [104]. (b) Positive polarizability case: catastrophic self-focusing collapse, (c) Negative polarizability case: enhance transmission, (d)–(f) Iso-intensity contours for a charge two vortex beam with initial radius $R = 20 \, \mu m$ and powers 6 W (d), 8 W (stable vortex) (e), and 18 W (f) [125].
of the exponential optical nonlinearity in these two cases was shown to have a significant impact on the dynamics of spatial soliton propagation in such media.

While a majority of previous studies investigated nonlinear propagation of Gaussian beams in colloidal suspensions [126], recently the break-up of an optical vortex beam due to the effect of azimuthal modulational instability (MI) was predicted. In particular, it was shown that different types of the exponential optical nonlinearity in the PP and NP cases lead to different MI gain for the same perturbation and to the formation of different spatial distributions of the necklace beams depending on the power of the incoming beam, as shown in Figure 4(d)–(f).

These results may be of significant importance to future studies of nonlinear propagation of structured light beams and filamentation in liquids, as well as in applications of light propagation in highly scattering biological and chemical colloidal media.

5. Summary and outlook

In summary, MMs and a broader class of engineered optical media offer several new ways of increasing nonlinear response and enabling new nonlinear optical phenomena. Future progress in the field of engineered nonlinear materials relies on a combination of the current advances in both classical and quantum theory of artificial nanostructures, patterns optimization, and understanding how topology and geometry affects the nonlinearities. Such materials would revolutionize nonlinear optics, enabling a new generation of nonlinear optical devices for optical and quantum communications, optical computing, and image processing. Although not discussed here in detail, another important functionality that is enabled by nonlinear metamaterials is tunable and reconfigurable metamaterials. For more details, we refer the reader to, for example, the following review and perspective articles [127–130].

Despite significant progress made in this field over the last few years, many questions that need careful theoretical and extensive experimental studies still remain. These include systematic studies of the physical reasons for the limited nonlinear response of existing natural and artificial structures; finding a path to overcome these limitations and create highly nonlinear materials with a good figure of merit; and finally, applying this knowledge to develop low-power, compact, and ultra-fast applications of nonlinear optical phenomena.

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