New degrees of freedom in nonlinear metamaterials

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This is an overview of the recent achievements in exploiting novel degrees of freedom in metamaterial design, which enable sophisticated nonlinear coupling mechanisms and bring enhancement to nonlinear behaviour. One of the novel paradigms makes use of mechanical feedback, achieved by embedding electromagnetic resonators within elastic medium or engineering explicit elastic links between them, such as rotational feedback. These designs provide broad-band self-adjustable resonances, self-oscillations, chaotic regimes, nonlinear chirality and spontaneous chiral symmetry breaking. With this respect, a range of implementations has been analysed, from flexible helices for microwaves to artificial electrostriction in optics. Another concept benefits from multi-frequency operation, where the properties in completely distinct frequency ranges become entangled through specific metamaterial design — for example, direct optical coupling can be introduced between microwave resonators, providing an independent interaction channel. It was also found that hyperbolic metamaterials can bring notable benefits to classical nonlinear processes by imposing unusual phase matching solutions, with a rich choice of matching combinations. Finally, the boundary structure of metamaterials add yet another possibility to control their properties. Overall, the recent progress in these topics suggests a very positive outlook into the future of nonlinear metamaterials.

1 Introduction The immense technological progress of past centuries has brought forth a spectacular variety of artificial materials, which are absolutely essential at all scales from major industry to simple household. However, even man-made materials rely on conventional materials in a sense that they are built up with natural atoms. The next logical, or hierarchical [1], step in matter organisation is to use conventional materials to create the building blocks for effective material properties at a larger scale [2,3]. This is achieved with metamaterials — artificial subwavelength structures aimed at achieving unusual properties, not readily found in nature [4]. Metamaterials are assembled from artificial structural units which effectively play the role of "atoms" and acquire unusual properties by virtue of both the specific characteristics of those sub-components and their collective response [5,6].

Over the past decade, metamaterial research has developed into a vivid, rapidly developing area, involving various aspects of physics, electrical engineering, chemistry and nanotechnology, and having fruitful outcomes towards applications in electromagnetics, photonics, acoustics and other disciplines related to wave propagation [7–9].

Metamaterials emerged at the beginning of this century [10], and have gained their initial popularity by promising a route towards negative refraction, a spectacular phenomenon [11–13] not available in conventional materials, which offers a way towards super-resolution in imaging [14–22], electromagnetic, acoustic or thermal cloaking [23–30], and many other amazing possibilities [31–40].

Initial development of metamaterials commenced in the domain of electromagnetics and departed from microwave and radio frequency range, where the large size of possible "meta-atoms" allowed easy and precise fabrication. This area is important for radio-engineering and mobile communications [41–44], as well as many important applications such as metamaterial lenses [45–47], waveguides [48–50], detectors [51,52] or metasurfaces [53,54] to improve magnetic resonance imaging. Further development was mostly driven by the quest for bringing metamaterials into optics [55], which seems to yield fruitful nonlinearities not only on a traditional plasmonic track [56–65] and fish-nets [66–72] but also with all-dielectric designs [73–80].

On the line of scaling-down, the research passed the challenges of sub-mm waves and THz frequencies, one of the most difficult and unexplored domains in electromagnetics [81]. In this range, metamaterials offer exciting opportunities for perfect absorbers, emitters, fibers, switches, modulators [82–88], also available with superconducting [89, 90] or flexible [91–94] implementations.

Reaching the visible range still remains one of the greatest challenges for metamaterials [95, 96]. In view of the great promise for enhanced nonlinearity [97], fabrication of optical metamaterials for controlling structured light [98–101] requires precise patterning at nanoscale, which raises tough tasks for nanotechnology. As it is typical for metamaterials, in the optical range, an enhanced collective response is useful for fruitful applications [102–104]. Controllable and reproducible patterning for producing arrays of identical elements with nanometer precision is increasingly complicated for "top-down" approaches, such as nanolithography, direct laser writing, etc., [105–108], particularly for three-dimensional arrays. For this reason, "bottom-up" methods, such as self-assembly, are actively explored [109–113], however such structures are prone to a deteriorating effect of disorder in the lattice [114, 115], element properties [116], or both [117], which is also crucial for nonlinear response [118, 119].

Most recently, spectacular advances of DNA-origami [120] have been applied towards metamaterial design, with simple arrangements with metallic nanoparticles have been already demonstrated [121–123], some of which are even capable of photochemically controllable reconfiguration [124]. These techniques may eventually enable artificial molecular machines with adjustable effective dielectric properties [125], which would be more robust across changing experimental conditions than natural ones [126].

Naturally, the idea of metamaterials was quite fruitful for other domains in physics [127], concerned with wave propagation, in particular, in acoustics, with prospects made for cloaking [128, 129], imaging [130, 131] and even seismic applications [132]. Although the development of acoustic metamaterials was greatly inspired by the ideas born in the electromagnetic domain, these two areas were separately developing. The prospects for a useful merging of these two directions were recently highlighted with magnetoelastic metamaterials [133]. This route opens wide horizons for cross-disciplinary scenarios, where acoustic and electromagnetic waves, otherwise weakly interacting, can be strongly coupled to enable novel tuning and switching schemes, efficient sensing and unusual nonlinear properties. For a successful combination of mechanical and electromagnetic phenomena, good progress has been made on the track of flexible metamaterial substrates [91, 92], flexible "meta-atoms" [134–137] and mechanically reconfigurable systems [138–140].

With the ongoing development in understanding and computational procedures related to mutual coupling between closely positioned resonators [141–143], analysis of dense metamaterials will be greatly facilitated. It is also important to account for noise issues [144] and spatial dispersion [145], quite relevant in a range of practical situations [146–153], also when analysing nonlinear metamaterials [154–156].

It is helpful to point out a number of useful reviews published in relevant areas, covering a great range of topics: effective medium modelling [157, 158]; active and tunable metamaterials [159, 139]; nonlinearities in plasmonics [160], photonics [161], and metamaterials [162]; ring-resonator-media [163] and wire media [164]; optical nanoantennas [165]; superconducting metamaterials [166, 167]; "metasurfaces" [168]; fabrication [169, 170]; as well as also metamaterials development beyond electromagnetic domain [127].

Given that the entire development of nonlinear metamaterials was reviewed recently [162], this feature article focusses on the most up-to-date achievements specifically related to the use of new degrees of freedom in metamaterial design.

2 Mechanical degrees of freedom

2.1 Flexible lattice Enabling a mechanical, or structural, degree of freedom in metamaterials opened wide opportunities for merging electromagnetic and acoustic response within one functional structure. The basic idea is to depart from a fixed metamaterial lattice and let the individual elements displace from their original positions. The resulting change in the lattice can produce dramatic effects, because metamaterial properties strongly depend on the lattice constants and symmetry [171] when the mutual interaction between individual elements is significant. The effect was readily employed for structural tuning [138], and has been quite fruitful for a range of implementations at different frequency ranges[139, 172].

The next step is a metamaterial that acts on itself, with the feedback relying on electromagnetic forces. This has

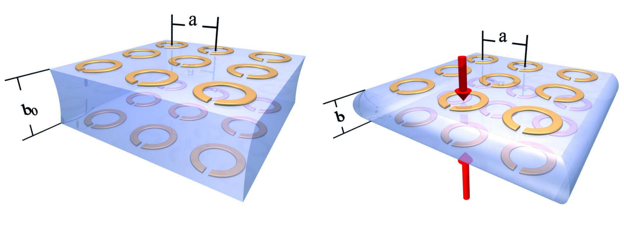

Figure 1 Conceptual layout of a fragment of magnetoelastic metamaterial, with fixed lattice constant a and variable lattice constant b. The vertical spacing b is exaggerated for clarity; it is essential that $b \ll a$. Reproduced with permission from Ref. [133].

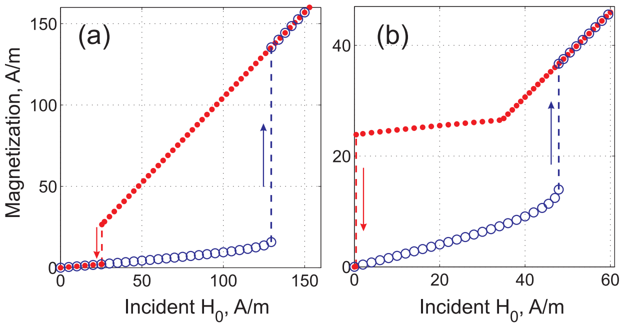

Figure 2 Example of nonlinear magnetisation hysteresis in magnetoelastic metamaterials, depending on the incident field H_0, with two different frequencies in panels (a) and (b).

been first achieved in so-called magnetoelastic metamaterials [133], with the general principle as follows (Fig. 1). Under excitation with incident electromagnetic waves, currents are induced in the resonators (these can be split-ring resonators [173] or capacitively loaded rings [3]). Due to the subwavelength regime of a metamaterial, the currents are in phase for closely positioned resonators, and therefore the arising Ampère forces uniformly act towards attraction of the rings; the effect is a time-averaged attraction, as the currents oscillate. When positioned in a flexible, elastic host medium, the resonators will displace due to these forces, until balanced against the elastic feedback of the host material. This, however, changes the lattice constant in the metamaterial, and therefore the frequency of its electromagnetic resonance shifts; as the resonators become closer to each other, the resonance frequency decreases. In turn, this affects the amplitude of the currents excited at the supplied frequency, so the induced currents change and thus the forces change too. Eventually, the new equilibrium is determined through a complex nonlinear feedback which involves a balance between the distance-dependent elastic force, and the electromagnetic force which also depends on distance as well as on the frequency and intensity of the incident field.

The resulting nonlinear behaviour is quite rich, with a possibility of hysteresis when either amplitude (Fig. 2) or frequency of the incident wave are varied, and the emergence of quasi-inaccessible meta-stable states [133].

In view of the challenges with realising a very soft feedback, required for this type of designs, a plausible experimental approach was relying on gravitational restoring force using a perforated substrate [174].

2.2 Variable near-field coupling However exciting magnetoelastic metamaterials may appear, it must be admitted that their practical realisation is rather challenging, since the Ampère forces are weak and either extreme intensity or impractically soft materials would be required for a functional implementation. This makes a feasible experimental system extremely sensitive to quite a subtle influence from the environment, so that even a gravitational feedback can be employed [174].

For this reason, a different approach was developed, which exploits another mechanical degree of freedom, rotation (Fig. 3). For such designs, elastic connection is introduced along the axis of two split-ring resonators, advancing the logic of broadside coupling [175] so that the two rings can rotate around their common axis with respect to each other. In this scenario [176], electromagnetic forces are strongest in the vicinity of the gaps of the resonators, where the charges are concentrated. The torque of these forces with respect to rotation axis has a great advantage over the torque of the elastic feedback which occurs at the axis and thus has a much shorter arm with respect to rotation. As a result, even relatively weak electromagnetic forces are sufficient to initiate mutual rotation. The rotation however changes mutual orientation of the gaps in the two split rings, which affects the pattern of the electromagnetic modes excited in the pair [142], and shifts the frequency of the resonance. This creates the required nonlinear feedback and results in a hysteresis-like behaviour, with remarkable angles of rotation achieved in experiment [176]. Note that, as far as rotation is concerned, electromagnetic forces act with a net time-averaged effect, as the characteristic mechanical frequencies are orders of magnitude smaller than the electromagnetic frequencies.

Adding further freedom to the above system, by including a third ring resonator elastically connected to rotate with respect to the other two [177], results in a complex dynamic response. In this scenario, slow mutual rotation of the three rings with respect to each other around their common axis is induced by electromagnetic waves. The motion takes a form of self-oscillations, yielding continuous variation of the electromagnetic parameters of the system [177]. For certain parameters and power levels, periodic pattern of mutual rotation ceases and chaotic behaviour is observed [177]. Quite remarkably, self-oscillations in this system turn out to be robust against viscous damping: albeit slow, mutual rotation will continue as long as electromagnetic energy is supplied [177].

Another development in the systems with rotational degree of freedom was the discovery of spontaneous chi-

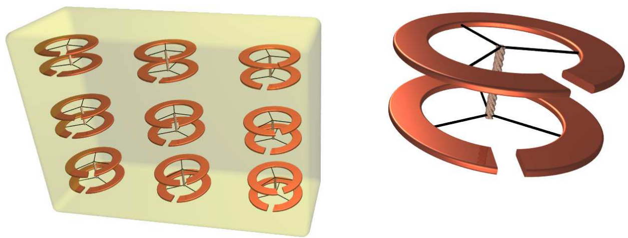

Figure 3 Metamaterial with the elements where two ring resonators are connected via elastic feedback, allowing them to rotate around their common axis (picture courtesy I. V. Shadrivov).

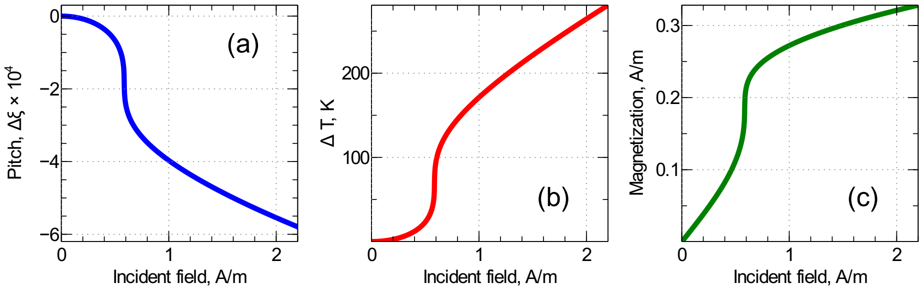

Figure 4 Mechanical and thermal feedback over magnetic response in flexible helices: variation in the relative helix pitch (a), temperature (b) and magnetisation (c) depending on the magnetic field of the incident wave.

ral symmetry breaking [178]. This can be observed in a racemic mixture of chiral resonator pairs with elastic feedback, where equal fractions of such pairs with initially opposite gap angles are taken. The two subsystems of the resonators initially have opposite chirality, resulting in zero total chirality. However, interaction with electromagnetic waves causes mutual rotation within the pairs, and it happens that, upon a certain power threshold, the original symmetry breaks: the two subsystems experience different changes in the angle between the gaps, so that the gap is increased in one and decreased in the other subsystem. Their chirality coefficients change accordingly, and fail to compensate each other, so the entire metamaterial acquires a chiral response [178].

2.3 Flexible resonators In the examples discussed above, the underlying idea was that a new degree of freedom is introduced on the level of mutual interaction of the resonators, in the from of either nonlinear mutual inductance in an entire lattice [133], or near-field coupling between two or more resonators within a “meta-molecule” [176, 177].

An alternative possibility is to introduce a mechanical degree of freedom within an electromagnetic resonator itself. The most straightforward example is a flexible conducting helix [179]. Such a particle is a chiral electromagnetic resonator, popular for metamaterials [180–182], but at the same time it is a mechanical spring. Electromagnetic waves induce oscillating current along the windings of a helix, which result in a time-averaged attractive force between the windings and lead to contraction of the helix. This reduces the pitch of the helix and thus its resonance frequency [183], providing a nonlinear feedback qualitatively similar to that in magnetoelastic metamaterials.

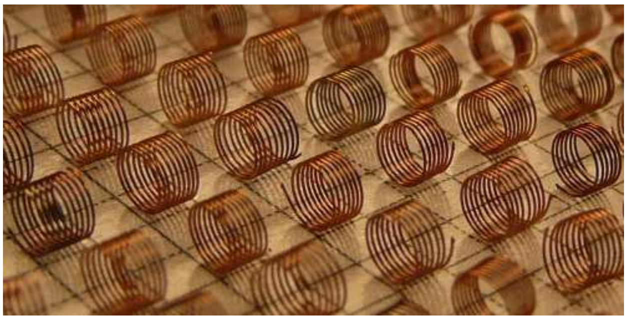

Figure 5 Experimental array of flexible helices for achieving nonlinear chirality (photograph courtesy A. P. Slobozhanyuk).

Quite nicely, there is also a thermal effect due to heating with the strong current required to generate measurable forces. This effect contributes in the same way, causing a decrease in the resonance frequency via its thermal expansion (Fig. 4). Thus, the system of flexible helices comprises a system with a complex nonlinear thermo-magneto-mechanical effect [179].

Metamaterial arrays comprising flexible helices have been experimentally realised in microwave frequency range [135] and, given the rapid progress in micro- and nano-fabrication [184–188], may eventually be realised for higher frequencies.

Flexible resonators can be implemented in a number of other varieties, such as, for example, flexible cubes carrying electromagnetic resonators on their sides [189], where elastic deformation breaks the original symmetry and may induce chiral response.

2.4 Alternative resonant designs Mechanical freedom in metamaterial designs has found a fruitful continuation in the microwave [190] and optical [136, 137] frequency ranges. One of the possibilities for enhancement, as compared to Ampère forces, was found in the use of Coulomb forces within a capacitor, made up with flexible metallic films [190]. Oscillating electric charges, induced by external fields, cause a time-averaged attraction which bends the thin capacitor plates, and thus increases its capacitance, changing the resonance frequency of the associated contour. This mechanism is most suitable for radio, microwave or low THz ranges. However, in the near-IR and optical domains, the use of metals is discouraged because of the dissipation, so dielectric materials come into play. On this track, a design was suggested [136, 137] with flexible strings carrying dielectric blocks, so that the optical forces induced between the blocks at resonance, can act to deflect the strings, changing the distance between the blocks and providing a feedback onto their electromagnetic interaction. As an alternative, flexible resonators were realised [137] using a "complimentary split-ring" geometry, where the cut-out dielectric patches can bend out of plane under the influence of electromagnetic forces, thus changing the resonances and modes in the system.

Apart from the rich nonlinear physics of such systems, power-dependent self-tuning of the resonance is useful for nonlinear dispersion compensation [191], whereby linear parameters of the system can be trapped through nonlinear feedback, so that it is possible, for example, to maintain negative effective permeability in a wide frequency range as long as the appropriate level of electromagnetic energy is supplied [191].

Such activities were recently reviewed considering both microwave [192] and optical [140] frequency ranges. Continuous advances in the research on optical forces [193–198] makes a promising ground for future success in this research area. Further development of this direction is expected to bring fruitful outcomes, bridging metamaterials with the achievements of optomechanics [199].

2.5 Non-resonant phenomena Scaling magneto-elastic metamaterials or self-compressible helices towards optical range is rather challenging outside the all-dielectric track, since the dissipation in metals increases dramatically. Estimates show that an attempt to realise a measurable Ampère attraction in the near-IR range would result in thermal effects completely dominating the response, so the concept will not really work. However, coupling between electromagnetic and elastic phenomena is possible and useful if a non-resonant system is designed. Indeed, non-resonant metamaterials prove to be quite capable of enhancing material properties, for example, providing strong artificial diamagnetism [200] competitive with ideal metals [201].

In optics, non-resonant structures are useful, for example, in designing artificial electrostriction [202]. Metamaterials for artificial electrostriction can be as simple as an array of spheres embedded into a matrix of a different material. When such a lattice is compressed, the two constituent materials shrink to a different degree, and then the filling fraction and therefore the effective permittivity changes, providing artificial photoelastic effect. Reciprocally, applying electric fields to the structure induces forces and compresses the lattice, an effect of artificial electrostriction. The corresponding phenomena have been analysed in the long-wavelength limit by using dielectric mixing formalism, and an effective electrostriction coefficient was derived [202]. Quite remarkably, artificial electrostriction arises even if the constituent materials are both non-electrostrictive. On the other hand, it is possible to suppress electrostriction by designing an appropriate mixture [202].

Artificial electrostriction is a weak effect, however it proves useful [203] for applications in stimulated Brillouin scattering (SBS) [204], which can be enhanced as compared to conventional materials for SBS, and potentially enable sufficient SBS levels on a silicon platform, aiding for a smooth integration on a chip. Alternatively, SBS can be suppressed for those cases where it is an undesirable side effect in optical fibres.

An interesting particular case of artificial structures suitable for SBS improvement, is a mixture of dielectric and air. As an example, inverse opals based on silicon were proposed, and it turns out that acousto-optical properties of such opals are much better than those of silicon, opening a path towards all-silicon chip-compatible implementations [205].

Overall, acousto-optic interaction in metamaterials with artificial electrostriction is a rich and complex phenomenon, requiring a careful analysis and advanced numerical modelling [206].

3 Interaction of incompatible frequencies Quite a different approach which is nevertheless easily categorised as an introduction of a new degree of freedom to metamaterial design, comprises engineering of additional interaction channels between or within electromagnetic resonators. This implies having more than one link — perhaps of a different physical nature — between the interacting particles (Fig. 6, left). A pre-requisite to this research direction was the suggestion of K. Betzler and M. Gorkunov (2004) to embed photodiodes into split-ring resonators, so that these can be tuned by light [207]. This has been experimentally realised with an array of optically controllable resonators in various configurations [208], and enabled tunable beam steering applications and nonlinearity control [209].

On these grounds, it is now possible to provide an entirely self-tunable, intrinsically nonlinear response. This was realised by inclusion of light-emitting diodes (LEDs) into varactor-loaded split-ring resonators [210]. In the first design (Fig. 6, right), the resulting "meta-atom" contained two mutually orthogonal rings, one of them equipped with an LED and the other one with a photodiode (PD). The two resonators so positioned, do not feature direct electromagnetic coupling via magnetic fields, because of their orthogonality. However, with a sufficiently large incident power,

Figure 6 Conceptual representation (left) and microwave implementation (right) of an optical interaction channel between nonlinear split-ring resonators.

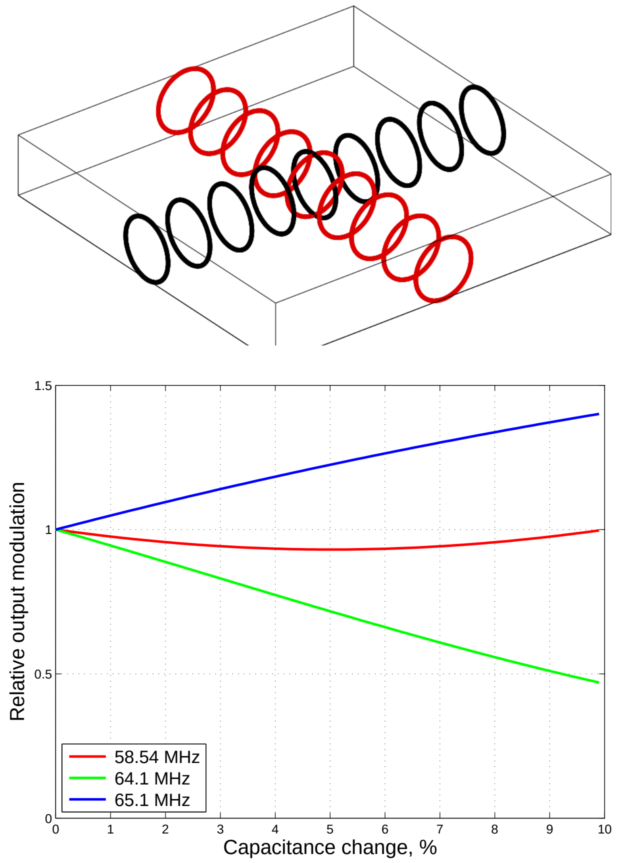

Figure 7 Schematics for a waveguide crossing with nonlinear traffic light, and its performance at three frequency channels.

LED in one of the rings is activated by the current rectified in the varactor, and shines light onto the PD in the other ring. The emerging photocurrent provides a biasing voltage onto the other varactor, and thus the net capacitance and hence the resonance frequency of the PD-ring measurably changes [210]. This makes an alternative, independent interaction channel between the two rings, that acts even in the absence of direct interaction.

This design is useful for implementation of nonlinear "traffic lights" where propagation over one waveguide is controlled by the power flowing through another waveguide, but not the other way round. Moreover, the resulting transmission control is frequency-selective, so that some of the frequency channels can be suppressed but other enhanced by the perpendicular traffic (Fig. 7).

4 Hyperbolic dispersion for nonlinear phase matching From the very beginning of metamaterials it has been anticipated that negative refraction provides novel opportunities for nonlinear processes [154].

Traditional techniques related to parametric amplification [211,212] or quasi-phase-matching with the help of metamaterial sub-structuring [213] or periodic modulation [214], were efficiently implemented. At the same time, interesting opportunities for nonlinear processes arise from the use of near-zero effective parameters [146], which has been shown helpful for phase-matching [215–218].

Further advance was connected with the rapid development of hyperbolic metamaterials [219–229], which feature hyperbolic isofrequency contours and thereby support extremely large wave vectors. Unique dispersion of hyperbolic metamaterials proved useful for nonlinear processes [230–232]. In particular, the advantages of hyperbolic dispersion, and of combining that with the normal dispersion at other frequencies, brought a new degree of freedom to realise phase matching in layered structures [233], potentially enabling the use of non-conventional nonlinear dielectrics. It should be noted that in layered hyperbolic materials, optimal phase matching conditions imply oblique incidence setups, which may be challenging in practice.

A more robust approach would be in the use of wire media [164] to realise phase matching in the range of hyperbolic dispersion [234]. Technological advances in fabrication of the wire filaments with cladding [93], as well as wire media tunability [235], may greatly enrich the opportunities for harmonic generation. Overall, such designs tend to overcome the limitations of both the conventional birefringent and quasi phase matching, making harmonics generation possible with materials where traditional approaches fail.

Quite an amazing possibility in the context of hyperbolic dispersion is the proposal to create a temporary hyperbolic metamaterial directly in the air, by triggering an array of plasma filaments by strong electric discharge was recently pointed out [236]. Such structure can be used for imaging or focusing over the limited time of its stability [237] and potentially will bring transient nonlinear response into play.

Furthermore, hyperbolic metamaterials may also offer cross-disciplinary degrees of freedom, such as links to chemistry — for example, in the studies on enhanced surface wetting properties [238].

5 Surface effects and strong coupling One more opportunity which can be employed to provide a new degree of freedom, relies on the effect of metamaterial boundaries. Surface effects are well known in artificial structures [239–242], however in metamaterials, thanks to extremely strong mutual coupling and structural discreteness[243–247], the effect of boundaries is particularly pronounced [248] so that the observable response demonstrate rather slow convergence [249] to effective medium predictions [171] as the size of the structure grows. For anisotropic designs, frequent in nonlinear metamaterials, strong coupling is expected to bring further complications to spatially dispersive response [250].

Boundary effects have a remarkable effect in metamaterial applications, not only with regards to nonlinear processes [251], but also for linear applications such as magnetic resonance imaging, where the impact on the achievable resolution with negative-permeability lenses [46] largely depends on the boundary structure [252].

Another way to exploit the strong coupling is to use specifically designed geometrical arrangements, such as circular shape, to provide a secondary level of resonances [51] to magnetoinductive waves [253].

Optical techniques for probing near-fields [254–257] may suggest a promising move of those findings towards the higher frequency range.

6 Conclusions and outlook Unusual ideas on combining very different materials and response functions continue to inspire metamaterial research, and we can feel certain to see many interesting designs in the future, with increasing diversity of various designs, specifically tailored to relevant applications and frequency ranges.

For example, water turns out be a rising star for metamaterial design, thanks to the rich opportunities it offers with thermal and geometrical tunability [258]. Permittivity of water in the THz and microwave range significantly varies with temperature, which allows for highly tunable dielectric wire media [258]. Also, the shape of water droplets, or the geometry of its filling fraction in resonance cavities [259] can be easily manipulated. Enabling self-action in such structures may offer fruitful outcome for nonlinear physics.

Liquid conductors also appear promising for various reconfigurable and potentially nonlinear systems [260, 261]. At the same time, graphene, still on the peak of its popularity, tends to invade the metamaterial research track in various forms [262–266].

Quite recently, an idea to create an entirely artificial liquid crystal [267, 268] was realised [269] for THz frequency range, with elongated filaments carrying electromagnetic resonators embedded in a viscous liquid so that their orientation, and hence the anisotropy of the overall response, can be changed by external fields. This is also a fruitful design for self-action nonlinear effects. Apart from that, huge activity on combining natural liquid crystals with metamaterials [270–276] will also bring new phenomena into play [277].

Overall, significant technological advances of recent decades, with the emphasis on nanotechnology, photonics, smart materials, and metamaterials, have enabled a step forward in the development of man-made materials and structures. Future designs are destined to be based on cross-disciplinary solutions and smart designs, enriching metamaterial research with advanced multi-physics.

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