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# (INVITED) Roadmap on perovskite nanophotonics

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ABSTRACT

Perovskite nanophotonics is a rapidly emerging field that combines research in synthesis of materials with novel properties and in photonics design strategies. Starting from early pioneering works on halide perovskite compounds that showed great potential across optoelectronics and photonics applications, the field is ready to blossom by combining recent advances in synthetic material design, the development of bottom-up or top-down nanostructuring approaches and new concepts in nanohophotonic engineering of light matter interaction at the nanoscale, with a chance of having real impact on current and future technologies. This roadmap is a collective outlook from pioneers in the field of perovskite nanophotonics that encompasses a number of the emerging research areas with the aim of identifying current and future challenges and highlighting the most promising research directions. It will be of interest and serve as a reference to a wide audience of physicists, chemists and engineers with interest in perovskite nanophotonics.

## 1. Forewords

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### 1.1. Perovskite nanophotonics: introduction

Since the first pioneering works in the 80's-90's, metal halide perovskites have made great strides in optoelectronics and photonics, demonstrating excellent performances in applications ranging from

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photovoltaic and photodetectors to light-emitting devices, lasers and scintillators. With the progressing of the field, this varied material platform is becoming more and more established, and it is now possible to synthetically design materials with tunable bandgap in the UV–Vis–NIR range preserving excellent charge transport, high luminescence efficiency and optimal morphologies, with an ease which is hardly met by other traditional organic and inorganic semiconductors. Nanostructuring by either bottom-up or top-down approaches has further empowered this materials platform, yielding improved performance of existing devices (e.g. ultralow-threshold lasers, tunable displays) or enabling the realization of new nanophotonic technologies for chiral and phase-change metasurfaces and single-photon sources for quantum photonics. At the forefront of the field, halide perovskites are also holding great promises for the development of the next-generation of small footprint, on-chip actuators and modulators thanks to their electro-optic and electro-absorption properties. The possibility to form both active and passive nanoscale components jointly with the easily scalable fabrication makes these materials highly attractive for the realization of perovskite-based photonic integrated circuits (PICs), rivalling III-V semiconductors which require epitaxial growth or bonding for low-cost applications. Perovskite nanophotonics is advancing very rapidly, but to further push the boundaries of this field several challenges need to be addressed at the intersection between chemistry, physics, materials science and engineering. This roadmap presents the recent advances in perovskite nanophotonics, providing a perspective on the future directions needed to improve the material's synthetic strategies, metamaterials design, all-optical devices based on polaritonic effects, perovskite integrated photonic platforms and quantum nanophotonic technologies.

### 1.2. Roadmap and current progress in perovskite nanophotonics

Here, we briefly overview the sections of this Roadmap which touch upon the most relevant topics of the field.

Metal halide perovskites are ionic semiconductors with exceptionally high synthetic versatility. Since their composition and crystal structure are intimately connected to their optoelectronic properties, synthetic design is crucial to meet the specific requirements for nanophotonic applications. The first section by Daniele Cortecchia discusses the synthetic challenges to reduce trap density and increase radiative recombination efficiency, stability and tunability of perovskites by exploiting lower dimensional structures, inorganic doping and optoelectronically active functional organic cations.

Patterning and integration techniques allow the realization of micro and nanostructured perovskites beyond the reach of conventional synthetic techniques, enhancing the efficiency of traditional photovoltaic and light emitting devices and enabling new applications like dynamic color display, flat lenses and ultrafast optical switching. The section by Kaiyang Wang, Shumin Xiao and Qinghai Song covers the state-of-the-art of the bottom-up and top-down approaches for the fabrication of in-plain patterned areas and 3D perovskite superstructures for applications going from single-photon sources to nonlinear optical devices.

Photonic integrated circuits (PIC) are a key technology for applications in communication, sensing, diagnostic and quantum computing. Here, integration of light sources which are efficient, tunable and easily scalable is a critical requirement. Anna Lena Schall-Giesecke, Piotr J. Cegielski and Max. C. Lemme give an overview of how the unique properties of metal halide perovskites make them valuable optical gain and lasing media for replacement of current state-of-the-art systems based on III-V or II-VI material-based semiconductors.

In the past decade, polariton physics has demonstrated spectacular effects such as superfluidity and topology-related phenomena. Recently, strong nonlinearities induced by the polarization field of the exciton component have been demonstrated in low-dimensional perovskites, making them highly attractive for the study of exciton-polaritons. The section by Dario Gerace and Daniele Sanvitto discusses the main trends of the field from room temperature polariton condensation to the development of efficient thresholdless lasers.

Metamaterials allow the realization of extreme light confinement and strong light-matter interaction through proper periodic nanostructuring of the material at the subwavelength scale. The section by Jingyi Tian and coworkers illustrates how the outstanding optoelectronic properties of halide perovskites are ideally suited for the realization of perovskite metamaterials with both monolithic dielectric and hybridized plasmonic designs, chiral metasurfaces, ultracompact vortex microlasers and dynamically switchable phase-change metasurfaces.

Metaphotonics is attracting great interest for the creation of efficient light sources and nanolasers employing optically induced Mie resonances of dielectric nanoparticles used as "meta-atoms". Pavel A. Tonkaev, Sergey V. Makarov and Yuri S. Kivshar discuss in this section the main fabrication strategies (synthesis of single-particle nanolasers, ion and electron beam lithography and high-throughput nanoimprint techniques) that will push the next developments of halide perovskites towards advanced and scalable metaphotonic light sources.

Current photonic technologies heavily rely on integrated optical modulators used to encode electrical signals onto guided optical fields by operating on their intensity, phase and polarization. The section by Oscar A. Jimenez Gordillo and Andrea Melloni highlights how the recent demonstration of electro-optic and electro-absorption in metal halide perovskites make them excellent candidates to meet the ever-increasing demand for small footprint devices providing faster, efficient, and high-performing optical modulation for the next-generation integrated photonic platforms.

Reconfigurable nanophotonic structures where the material's properties are tuned on demand through external stimuli are highly attractive for displays, adaptive optics and data recording. Anatoly P. Pushkarev and Sergey V. Makarov examine the structural flexibility of halide perovskites allowing the occurrence of phase transitions with marked impact on their optoelectronic properties. This results in dramatic changes in the material upon application of external electric field or chemical processing opening new horizons for creation of tunable optotelectronic devices.

Finally, single photon generation is the key towards future quantum devices and revolutionary qubits for quantum networks. Mariangela D'Amato, Emmanuel Lhuillier and Alberto Bramati review the outstanding advantages held by perovskite nanocrystals as single photon sources in comparison to other physical systems like single defects in nanodiamonds and epitaxial quantum dots. The challenges ahead to improve their photostability and enable their effective integration with photonic interfaces such as waveguides and fiber optics are also discussed.

### 1.3. Concluding remarks

Perovskite photonics encompasses several exciting applications of halide perovskite materials beyond conventional photovoltaic and light-emitting devices. The possibility to tune their transport, emission and dielectric properties by synthetic chemistry, superstructural engineering and nanopatterning make halide perovskites a unique platform for the development of integrated light sources and electro-optic modulators, tunable and reconfigurable metaoptics, as well as polaritonic and quantum nanophotonic devices. In our opinion, the future of perovskite photonics is in the fundamental progress with tailored materials synthesis and large-area nanofabrication techniques, which will enable a new generation of low-cost, highly functional devices working in both classical and quantum regime. At the same time, progress with existing light-emitting and photovoltaic perovskite technologies will establish a reliable platform for translation of the emerging photonic technologies into practical devices and their commercialization. The interest and potential of this field may be gauged by the large number of research groups working at the forefront of perovskite photonics, many of which are represented by the Authors of this roadmap, and the creation of

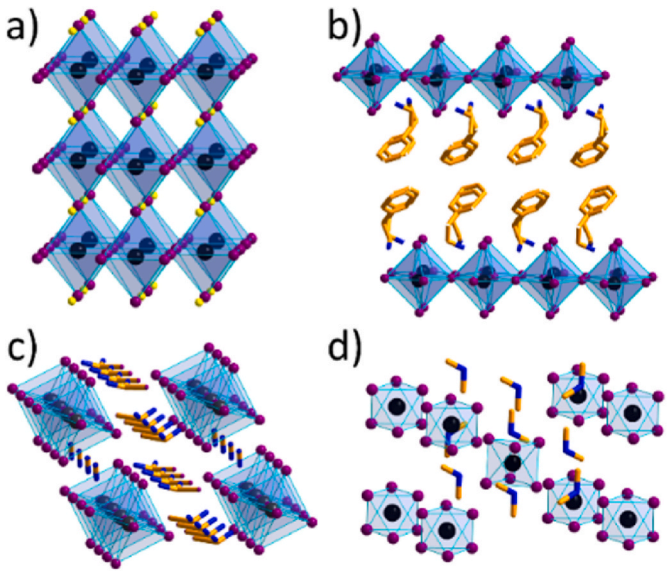


**Fig. 1.** Structural diversity in metal halide perovskites with different dimensionalities: a) 3D, b) 2D, c) 1D and d) 0D. Reproduced with permission [2] Copyright 2019, Royal Society of Chemistry.

dedicated collaborative research consortia, such as the Horizon 2020 Marie Curie Innovative Training Network on PERovskite SEmiconductors for PHOtoNics (PERSEPHONe) to which several of these Authors are affiliated.

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## 2. Engineering metal halide perovskite materials for nanophotonics

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### 2.1. State of the art

Metal halide perovskites (MHPs) have rapidly set new benchmarks in fields ranging from photovoltaic to light emitting devices and lasers thanks to their exceptional optoelectronic properties. Their easy processability from both vapor and solution-based techniques, either in form of bulk films or colloidal nanocrystals, makes them highly appealing for combination with silicon photonics and on-chip integration. The unrivalled synthetic versatility allows a nearly continuous modulation of the material's bandgap and correspondingly of its luminescence, covering the entire UV–Vis–NIR range up to 1000 nm with a single material platform, holding great promises for nanophotonic technologies. The standard structure, forming a continuous three-dimensional network of corner-sharing metal halide octahedra, carries the general formula $AMX_3$, where M is a divalent metal (typically $Sn^{2+}$ and $Pb^{2+}$), X is a halide and A is a small inorganic or organic cation (usually carrying an ammonium tethering unit). A huge structural richness arises from the addition of cations which are larger than the cubooctahedral perovskite sites, originating low-dimensional perovskites. A typical case is that of layered perovskites, self-assembled multi-quantum-wells structures consisting of alternating organic and inorganic layers where the dimensionality of the inorganic part can be tuned by synthetic design, from single-layered 2D perovskites to the extreme case of 3D perovskites (Fig. 1a and b) [1]. Depending on the steric hindrance, the coordination capabilities and the intermolecular interactions of the templating cations, the inorganic framework can further be disrupted, even promoting new geometric patterns of the inorganic motif (e.g. edge and face-sharing MX6 octahedra or alternative coordination polyhedra) resulting in lower dimensional structures like bulk assemblies of 1D core-shell quantum wires and 0D molecular clusters (Fig. 1c and d) [2]. In these materials, the optoelectronic properties are determined not only by the chemical composition, but also by the structural and electronic dimensionality and structural deformations (e.g. tilting and distortion of the octahedral networks) [3]. In fact, hydrogen bonding between the ammonium group of the organic cations and the metal halide framework, and the strong conformational disorder induced by the interpenetrating molecular networks based on ionic bonds may induce strong lattice distortions modifying the electronic landscape of the semiconductor [4]. Such peculiarities have a great potential as they widen the functionality the material can provide upon educated design, which then is translated into the functionalities of relative devices.

### 2.2. Current and future challenges

From a material perspective, the growing interest for photonic applications is pointing out a series of limitations that need to be addressed. I) The metal halide lattice is susceptible to the formation of various types of defects that reduce the material's performance by inducing detrimental trap-mediated non-radiative losses. II) The standard 3D perovskite structure is characterized by a very low exciton binding energy (9–60 meV) promoting the generation of free charges; while being extremely beneficial for photovoltaic, this also results in slow bimolecular electron-hole recombination in turn limiting the luminescence yield of the material. III) The material's stability remains a major issue of the field, frustrated by degradation mechanisms triggered by several factors that include moisture sensitivity, oxidation, heat and electric field, fostering the perovskite decomposition, phase-segregation phenomena and ionic diffusion. Solving these issues is fundamental not only to achieve long-term stability, but it is also critical to enable the operation of key-enabling technologies such as the long sought-after perovskite electrically-pumped lasers. IV) The overwhelming predominance of lead-based materials poses serious environmental and toxicity concerns, and can potentially hamper the actual implementation and commercialization of perovskite-based devices. V) The need to accurately tune the perovskite luminescence energy and bandwidth for nanophotonic applications requires a more complex structural tunability than that typically required for photovoltaic, for which the development of low-dimensional perovskites will have a critical role.

### 2.3. Advances in science and technology to meet the challenges

Defect passivation strategies employing small molecules and formation of perovskite nanocrystals have been successful to mitigate the effects of ionic defects and remarkably improve the radiative recombination efficiency of 3D perovskites. On the other hand, low-dimensional systems represent promising alternatives to further improve the luminescence efficiency, thanks to their extremely high exciton binding energy which allows the formation of stable excitons and favours fast radiative decay. At the same time, their layered architecture can grant increased environmental stability, block detrimental ionic diffusion, and relax geometrical constraints giving access to a wider range of lead-free compositions. Among these, Ruddlesden-Popper perovskites have been employed to design self-assembled multidimensional systems exploiting

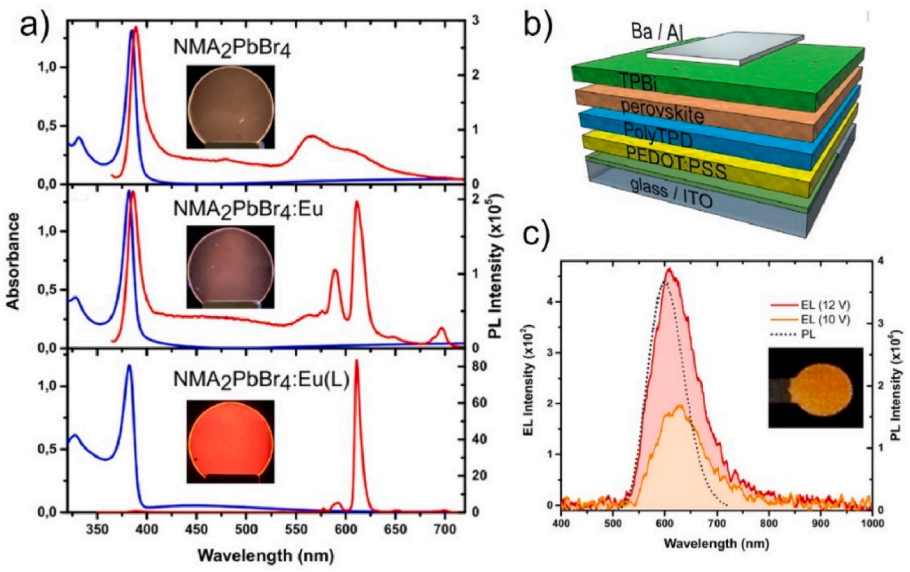


**Fig. 2.** a) Tuning of the narrowband emission in 2D perovskites through $Eu^{3+}$ doping. The figure compares the optical properties of the pristine perovskite $NMA_2PbBr_4$ (NMA = 1-naphtylmethylammonium) (upper panel), the B-site substitutional doping with $Eu^{3+}$ ions (medium panel) and the doping with the europium complex Eu(L) = $Eu(tta)_4P(Ph)_4$ (lower panel). Reproduced with permission [6]. b) Example of light-emitting diode structure employing the $Mn^{2+}$-doped perovskite $NMA_2PbBr_4$ as emissive layer and c) corresponding electroluminescence spectrum, showing the broadband orange emission characteristic of the $Mn^{2+}$ $^4T_1 \rightarrow {}^6A_1$ transition. Reproduced with permission [7]. Copyright 2019, Elsevier. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

energy funnelling mechanisms to achieve high photoluminescence quantum efficiencies surpassing 60% [1]; however, as the radiative recombination still relies on the phases with highest dimensionalities, they cannot fully exploit the richness of properties of low dimensional phases. Phase-pure 2D structures with close-to-ideal coordination geometries can in fact be exploited for their quantum and dielectric confinement to obtain narrowband emission (FWHM down to 20 nm) and exceptionally high colour purity arising from their characteristic excitonic recombination [4]. Alternatively, it is possible to synthetically design highly distorted systems where the interaction between the photogenerated excitons and perovskite ionic lattice leads to the formation of self-trapped excitonic states, that ultimately result in ultra-broadband emission (FWHM >250 nm) [3,5]. The exciton self-trapping efficiency is greatly increased in 1D and 0D structures, which can originate ultrabroadband luminescence with PLQYs approaching unity [2]. Inorganic doping is also emerging as a powerful strategy to tune and enhance the perovskite luminescence by exploiting the unique properties of d and f-block metals (Fig. 2) [6,7]; it allows to selectively achieve narrow and broadband UV–Vis luminescence with exceptionally high quantum yields and can be exploited to extend the emission range even further in the NIR, beyond the limits of the undoped materials, with the use of elements such as $Yb^{3+}$ and $Er^{3+}$. On the front of lead free-perovskites, while $Sn^{2+}$-based materials represent one of the most established and high-performing platforms [8,9], new breakthroughs can be expected from the growing class of double perovskites $A^{I}_2M^{I}M'^{III}X_6$ (and their low-dimensional counterparts), opening up the use of mixtures of mono- and trivalent elements (e.g. In, Ag, Bi, Sb, Na) greatly enlarging the compositional library and properties tunability.

Despite the surge of interest, single layered perovskites often show an unexpectedly lower PLQY compared to the parental 3D MHPs, while their ability to sustain coherent emission remains unclear and highly debated. The reason behind this behaviour is still largely unknown, and has tentatively been attributed to high defect densities, presence of dark states and exciton thermal quenching [8]. Moreover, poor charge transport properties are ubiquitous to all low-dimensional perovskites, deriving from the disruption of the metal-halide framework with electrically insulating cations, frustrating their implementation in electrically-driven devices [2]. To take full advantage of the perovskite structural diversity, future works will need to elucidate the role of exciton-phonon interactions to determine the exciton quenching behaviour and how we can synthetically improve the radiative recombination efficiency by controlling the structural rigidity, also supported by advanced defect engineering strategies. The development of optoelectronically active organic cations, capable to positively contribute to the optical and transport properties of the material, will also be a crucial step in the development of a new class truly functional perovskites with improved optoelectronic properties and stability.

### 2.4. Concluding remarks

While the field of 3D perovskites is reaching maturity, the full potential of their parental low-dimensional phases has yet to be unleashed, and they will likely play a crucial role in improving the material's performance and stability, expand its tunability and reduce toxicity. The marked structural dependence of their optoelectronic properties opens the possibility to design tunable emitters with unparalleled colour purity, useful for wide colour gamut displays and lab-on-a-chip technologies, while the development of phase-change perovskite metamaterials and applications in all-optical switching photonic devices can be envisioned [10]. On the other hand, ultrabroadband and white light emitters are extremely attractive as active metamaterials, as their emission could be enhanced at selected spectral regions achieving different luminescence properties from a single material. Beyond applications in the visible range, typical of MHPs, it is expected that inorganic doping will allow their exploitations also at telecommunication wavelengths, opening a completely new frontier for their applications. Given the extreme structural and compositional versatility of MHPs, synthetic chemistry will play a crucial role in the creation of new materials with enhanced functionalities for nanophotonics.

### 2.5. Acknowledgements

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## 3. Superstructural tunability by nanostructuring

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### 3.1. State of the art

Metal halide perovskites, as an emerging class of photovoltaic materials, have sparked tremendous attention due to the prominent material properties, such as solution processability, large optical absorption,

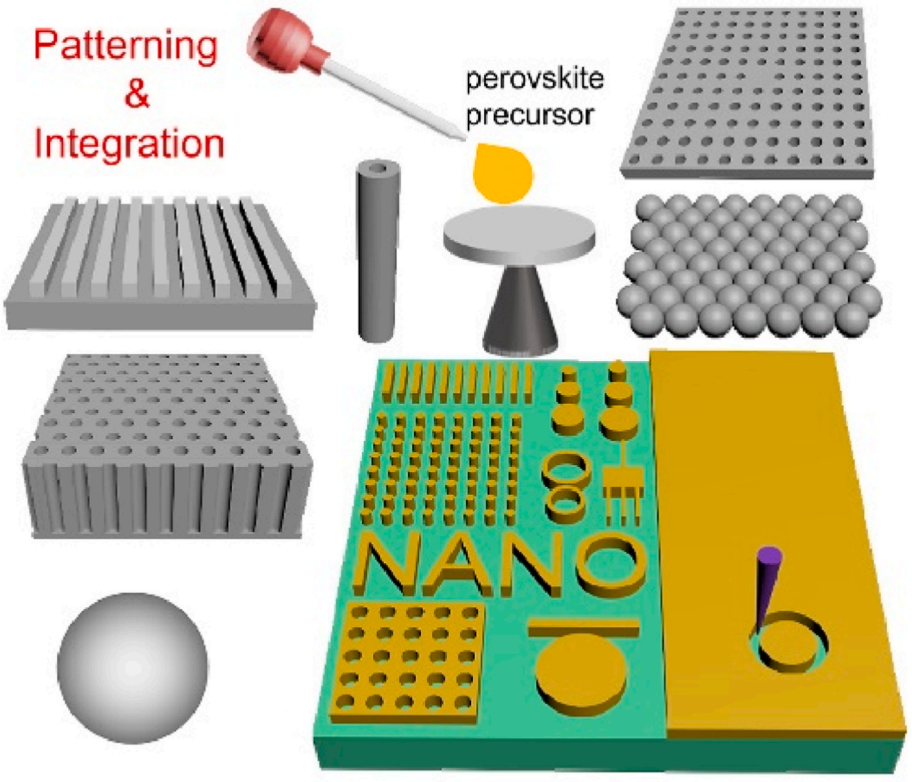


**Fig. 3.** Schematic diagram showing the patterning and integration techniques for micro- and nanostructured perovskites. Reproduced with permission [11]. Copyright 2020, Wiley-VCH.

long and balanced charge carrier diffusion length, high defect tolerance, bandgap tunability, and low nonradiative recombination rate, etc. The potential applications have been expanded from solar cells to light emitting devices (LEDs & lasers) and photodetectors/scintillators. Perovskites possess inherent merits in the chemical and structural diversity owing to the facile composition engineering and synthesis control. Beyond the chemical treatment, versatile patterning and integration techniques (see Fig. 3) have been developed to realize micro- and nanostructured perovskites, which can be classified into bottom-up synthesis and top-down fabrication [11]. Benefiting from the solution processability, pattern transfer could be facilitated by spin/drop coating the perovskite film on the pre-patterned nanostructured substrate, thereby forming a hybrid configuration. Such integration scheme is also suitable for perovskites prepared via chemical vapor deposition. Space-confined self-assembly method could produce wafer-scale perovskite microcrystal array with designed distribution on the pre-treated substrate. However, the size and orientation of perovskite crystal vary randomly as a result of the intrinsic self-assembly process. Besides, microsphere opal assisted deposition could serve as an efficient route to obtain 3D photonic crystal nanostructure made of perovskites. The above-mentioned bottom-up synthesis techniques share certain similarities, e.g. nanostructured scaffolds are prepared ahead of perovskite growth; the patterning process is additive without any perovskite removal procedure. In contrast, the top-down fabrication technique is a subtractive and post-synthetic process that peels off unnecessary parts of perovskite to sculpt desirable nanostructures. Given that perovskites are vulnerable to polar solvent, etching gas and energetic ions/electrons, conventional semiconductor manufacturing techniques, including photo/electron-beam/X-ray/laser lithography with subsequent etching, nanoimprint, inkjet printing, focused ion beam (FIB) milling and direct laser writing, have been modified to reduce adverse impact on the nanostructured perovskite. Based on the well-developed patterning techniques, micro- and nanostructured perovskites with various morphologies (shape, size and dimension) on demand have been realized in either high throughput or high resolution. To assess the effectiveness of patterning techniques applied to structure perovskites, several criterions could be considered, specifically, cost, complexity, scalability, homogeneity/uniformity, resolution, integrity and reproducibility. Typically, the selection of structuring techniques depends on specific application scene and compromise has to be made to guarantee the effectiveness.

### 3.2. *Current and future challenges*

Nanostructured perovskites have not only improved the performance of photovoltaic and optoelectronic devices, but also prompted novel optical applications, such as dynamic color display [12], flat lens [13] and ultrafast optical switching [14]. Despite the impressive progress made based on the nanostructured perovskites, challenges still remain in the following aspects. i) At the current stage, polycrystalline thin films with relatively good coverage and controllable thickness are mostly employed to implement scalable nanofabrication techniques. Nevertheless, the resultant nanostructured perovskites suffer severely from defects widely distributed at the grain boundaries and on rough surfaces. ii) The developed patterning techniques are limited to the in-plain structure or two dimensions. There is still plenty of room in the third dimension to be explored. iii) Extra loss, degradation and even decomposition have been introduced to the structured perovskites during the patterning process, discounting the corresponding overall enhancement

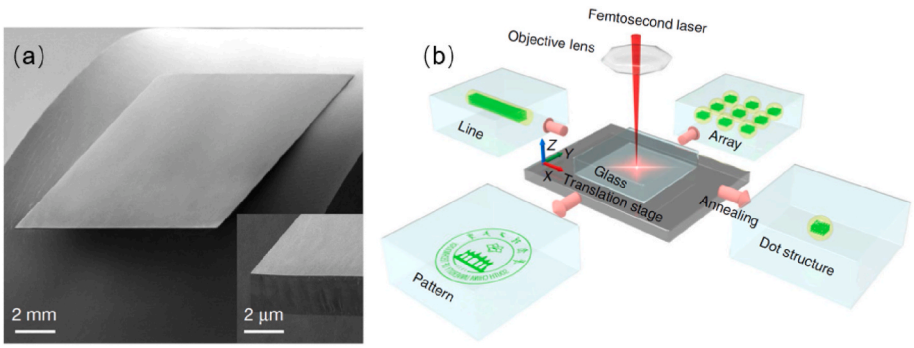


**Fig. 4.** a) SEM images showing a single-crystal $MAPbI_3$ thin film where inset shows the zoom-in image of the cross section. Reproduced with permission [18]. Copyright 2020, Springer Nature. b) Schematic diagram showing the 3D femtosecond laser writing process. Reproduced with permission [19]. Copyright 2019, Springer Nature.

effect. In particular, the most vulnerable properties involve surface roughness (during the process of etching and direct laser writing [15]), photoluminescence intensity and wavelength (upon exposure to energetic ions in FIB [16] and halide exchange in plasma etching [17]).

### 3.3. Advances in science and technology to meet the challenges

i) Single-crystal perovskites possess ultralow trap states and excellent uniformity/homogeneity, which hold great potential in the field of constructing devices and exploring basic photophysics. However, the as-grown single crystals are either too bulky to integrate into devices or too small to conduct scalable nanofabrication. Recent progress in the synthesis of wafer-scale single-crystal thin film have revolutionized this field. Wafer-scale single-crystal hybrid perovskites with precise thickness control down to 600 nm have been demonstrated on arbitrary substrates (Fig. 4a) [18], shining light on the patterning/integration techniques and even commercialization. ii) 3D patterning has been realized via direct laser writing of perovskite quantum dot in transparent glass materials (Fig. 4b) [19] and guiding evaporation-induced perovskite crystallization in mid-air [20]. Moreover, based on the in-plane 2D patterning techniques, the stacked heterostructures comprising different perovskites with varied bandgaps in the vertical direction could also achieve 3D nanostructures. The overall resolution could be assured via efficient combination of the top-down fabrication and atomically aligned bottom-up synthesis. iii) To address the additional loss issue, the most direct and effective way is to update the fabrication techniques, such as adjusting the intensity and exposure time in laser direct writing process and choosing proper protective and reactive gas in etching recipe. As an alternative way, the etchless patterning method could eliminate the etching process induced adverse impact, which relies on the subtle design on the electromagnetic field distribution [21].

### 3.4. Concluding remarks

The versatile patterning and integration techniques have endowed perovskites with superstructural tunability. More importantly, the micro- and nanostructured perovskites have boosted the corresponding device performance greatly. With the development of nanofabrication techniques, more complicated and novel applications could be expected in perovskites, e.g. single-photon source by incorporating perovskite quantum dot into resonators with ultra-high quality factor; nonlinear optical devices employing the intrinsic high optical nonlinearities of perovskites with designed nonlinearity enhancement nanostructures; eventually, real all-perovskite system by integrating the power source, modulation elements and detection capability into a unit.

## 4. Monolithically integrated lasers

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### 4.1. State of the art

The integration of light sources is one of the critical steps for implementing full functionalities on-chip. Nowadays, a wide range of applications such as communication, sensing, and medical diagnostic rely on photonic integrated circuits (PIC). Applications in the field of optical quantum computing and quantum sensors require high-quality chips in the future.

The publication rate of novel lasers based on metal halide perovskite has increased tremendously within the last five years. Nanocrystals, Thin-film, nanowires, and other types of microlasers have been demonstrated.

State-of-the-art III-V or II-VI material-based systems are produced by employing expensive epitaxy. One significant advantage of metal halide perovskites is that they can be solution-processed on any relevant material for photonic integrated circuits (PICs). Thus, perovskite lasers offer the possibility of cost-effective processing.

The need for electrically pumped lasers has increased dramatically due to the growing importance of miniaturization and multiplication of the application potential of PICs in displays, healthcare, communications, and sensing.

Lasers for PICs need to be low-cost and reliable, and the integration into silicon process technology needs to be sufficiently accessible. The photonic integration within the last decade mainly focused on III-V material-based lasers both as research and development topics. So far, mass-manufacturable lasers high gain laser directly deposited laser on 300 mm wafers remains a challenge, and heterogeneous or hybrid integration processes lead the applications scenarios.

Being direct bandgap semiconductors, metal halide perovskites offer many properties of ideal gain materials: such as strong absorption, high photoluminescence quantum yield, and low levels of charge trapping defects. Moreover, the non-radiative Auger recombination is slow under intense excitation conditions. Light emission of the perovskite types lies in the range between 400 and approx. 950 nm, and thus these materials can be used for the integration of light sources into a silicon-based platform for photonic integrated circuits such as silicon nitride. Since the silicon nitride waveguides can be deposited on top of CMOS circuitry, integration of light source, waveguide, and CMOS control circuit becomes possible.

Compared to organic materials, perovskites have to dominate bright emissions after excitation, even in the triplet state (i.e., for $CsPbI_3$).

### 4.2. Current and future challenges

Most of the early work on metal halide perovskite laser was based on nanocrystals since their crystallinity offers ideal conditions proving low optical loss and thus the possibility of overcoming the lasing threshold.

By improving the material, thin-film lasers, Fabry-Pérot cavities, and other microstructured devices became more relevant. By combining a perovskite gain medium which a cavity, the feedback structure is used to obtain lasing. Usually, the smaller the laser becomes, the more suffering from loss occurs, leading to higher lasing thresholds. The polycrystallinity of annealed perovskite material can lead to scattering and thus loss in the gain material and cavity. The development of top-down and bottom-up structures that emit light and can be used in laser structures has been a focus topic of research during the later years. Meanwhile, perovskite LEDs could be developed, reaching quantum efficiencies useable for first applications.

Many examples of perovskite lasers that have been optically pumped have been demonstrated so far. Continuous-wave operation at room temperature marks an essential step toward electrically pumped lasers in the future. Long-lived triplet excitons seem to be one of the intrinsic sources of the so-called lasing death phenomenon [22].

Due to the hygroscopic properties of perovskites, severe stability problems occur if encapsulation is incomplete. In addition, the injection of many charge carriers often heats the material above the destruction threshold. Nevertheless, properties such as the decrease in Auger recombination give hope for a useful application in strongly pumped regimes.

At the same time, geometries must be found for monolithic integration in which the generated laser light from the perovskite is coupled efficiently enough into the resonator structure also to produce low laser thresholds. It must also be ensured that photonic waveguides, which are usually made of materials with low refractive indices (for example, silicon nitrides or other dielectric materials whose refractive index is also around 2.0), can be coupled as the perovskite.

### 4.3. Advances in science and technology to meet the challenges

The development of reliable top-down processing methods has brought a step in the crucial direction for the applicability of perovskites in the future [23]. The usability of standard lithography techniques thus enables small structures such as waveguides, micro disks, or other microstructures. This way, structures can be created whose geometry can be optimized for the laser concept. Furthermore, the reproducibility of the optical functionality of the layers is of crucial importance. Here, too, great progress has been made in the past, but after each change in the material composition, the suitability for reproducible manufacturing techniques must be proven again.

### 4.4. Material engineering will play an essential role in the future of monolithic lasers

Surface roughness and internal crystallinity must be considered when choosing the gain material. By using hot-pressed perovskites, the optical properties of the materials used could be increased significantly, and the laser threshold could be lowered considerably in some cases [24].

The exchange of metalammonium into formamidinium lead halide (FAPbX3) offers better stability since FA, and the halide ions are strongly bound. Shaping the perovskite structure from 3D to quasi 2D materials also deepened the understanding of triplet exciton processes [22].

### 4.5. Concluding remarks

The route to stable and efficient highly integrated perovskite lasers in the near future is still filled with obstacles.

Regimes at high excitation need to be studied in more detail to understand the underlying mechanism in interface layers as well as in the perovskite material.

Also, the question of using non-toxic gain material needs to be answered in the future even though chips usually only contain small portions of lead and can be sealed after fabrication.

By choosing stable, inorganic contact materials and especially electron and hole transport layers, which can endure high current densities of around $10^{18}$ $cm^{-3}$, monolithic perovskite layers may be devices of highly integrated photonic integrated circuits.

## 5. Exciton-polaritons and photonic crystals in layered perovskites

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### 5.1. State of the art

Polariton physics is a relatively new field of research that has shown spectacular effects in these past 10 years–from superfluidity to topology-related phenomena. In particular, new exotic materials such as three-dimensional (3D) and two-dimensional (2D) layered perovskites have shown unprecedented properties once coupled to photonic modes in nanostructured devices such as microcavities, waveguides, and photonic crystals. In fact, controlling radiation-matter interaction in these materials through the suitable engineering of the photonic density of states with a periodic texturing has become an increasingly successful tool. Exciton-polaritons are the hybrid excitations of electromagnetic radiation and optically active elementary transitions in semiconductors and insulators given by bound electron-hole pairs. In perovskites, the exciton binding energy is much larger than in inorganic semiconductors, and the oscillator strength of such a transition is typically giving rise to a strong radiation-matter coupling energy (in the hundreds meV range) that exceeds losses of the oscillating fields, even at room temperature. Polaritonic eigenmodes thus possess an energy-wave vector dispersion inheriting the light effective mass of photons in the material, and possibly the strong nonlinearities induced by the polarization field of the exciton component [25]. The latter have been shown to be remarkably larger in 2D perovskites than in organic compounds [26].

Layered crystalline perovskites generally consist of an inorganic lead halide sandwiched between organic layers, as sketched in Fig. 5a. Since the lowest-energy excitonic transitions are associated with the inorganic sheet, while the organic part is believed to behave as a potential barrier, these structures share strong similarities with inorganic semiconductor multiple quantum wells (multi-QW). They are particularly appealing since they combine the easy and cheap manufacturing of organics with the robust optical properties of the 3D perovskites, whose band gap can also be tuned by just changing the inorganic precursor during growth. This is all very promising since it opens the way to investigating and exploiting exciton-polariton properties at room temperature [25]. In fact, polarization-dependent polariton effects in thin films of halide perovskites unravelled peculiar symmetry properties of the excitonic eigenstates, such as the out-of-plane component of the oscillator strength that is typically absent in other 2D layered materials with radiative excitonic response [27]. By suitably exploiting intrinsic properties of 2D perovskite crystals, such as their polarization anisotropies and excitonic response to external magnetic fields, it has been possible to induce a nontrivial topological property, such as a non-zero Berry curvature associated with the polariton dispersion [28].

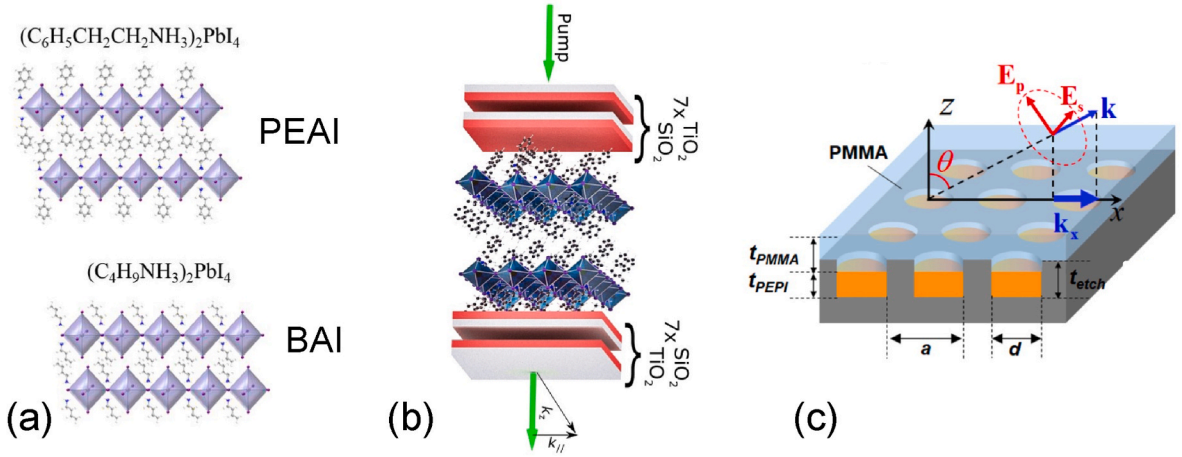


**Fig. 5.** a) Two example of layered lead-halide perovskite multi-QW structures with organic barriers, namely phenethylammonium lead iodide (PEAI) and butylammonium lead iodide (BAI) perovskites, respectively; b) planar microcavity structure with the active perovskite multi-layer sandwiched between two Bragg mirrors; Reproduced with permission [26]. Copyright 2019, AAAS. c) 2D photonic crystal lattice with active perovskite multilayers within the backbone (glass) voids, created by nano-imprint lithography for example Reproduced with permission Copyright 2020,[31] American Chemical Society.

### 5.2. *Perspectives and challenges*

The exploration and exploitation of exciton-polaritons in 2D perovskite metamaterials is still in its infancy, and much has to be done in terms of either materials research or design and fabrication. In general, the presence of a hybrid organic/inorganic structure provides an ideal platform for developing novel and cost-effective multifunctional materials, since the crystalline architectures can be synthetically fine-tuned to provide a huge range of semiconductors with different properties, thus paving the way for the design and fabrication of all-optical devices based on polaritonic effects. In this respect, we hereby emphasize some of the most appealing perspectives and the upcoming challenges in the near future.

Exciton-polaritons are particularly suited to investigate the physics of weakly interacting Bose gases out-of thermal equilibrium. In this respect the intrinsic nonlinearity of perovskite polaritons has been measured in microcavity samples (see Fig. 5b) and found to be orders of magnitude higher than the corresponding value in fully organic polaritons [26], which holds great promises for, e.g., all-optical switching, routing and gating in perovskite polariton devices working at room temperature [29]. Moreover, tailoring the photonic dispersion allows to explore even more exotic phenomena [30]. A possible way of imposing a photonic crystal potential is sketched in Fig. 5c, yielding large tunability of the exciton-polariton dispersion just depending on the lattice parameter [31]. This adds a further knob to the control of light propagation and confinement in these materials, such as group velocity dispersion, vertical emission profile and quality factor of resonant modes, which can hardly be engineered in conventional microcavity samples, as recently shown by a tunable phase change perovskite microlaser through emission into a bound state in the continuum (BIC) created by a periodic texturing of the active material [32]. Furthermore, such BICs are extremely interesting for polariton-related applications. As an example, it has recently been shown that strong light coupling of perovskite excitons to optical BICs allow to impart a topological charge to the emitted polariton field, as evidenced at room temperature by measuring a polarization vortex in the far-field [33]. These results are particularly promising in view of showing room temperature polariton condensation into states that are completely decoupled from the radiative continuum, in analogy to recent reports on fully inorganic samples at cryogenic temperatures [34], a direction to be definitely pursued in perovskite materials towards the realization of low-cost, energy efficient thresholdless lasers.

Polariton nonlinearities are well known to represent a relevant resource for device applications [25]. Enhancing such nonlinearities through suitable materials engineering, such as exploiting the dipolar effect, or charge saturation is a possible route. In this respect, recent work on controlling the 2D crystalline layers thickness and the organic barriers surrounding them has been successfully reported [35], as sketched in Fig. 6a. This is particularly promising in view of engineering the excitonic properties, their confinement and, applying an in-plane

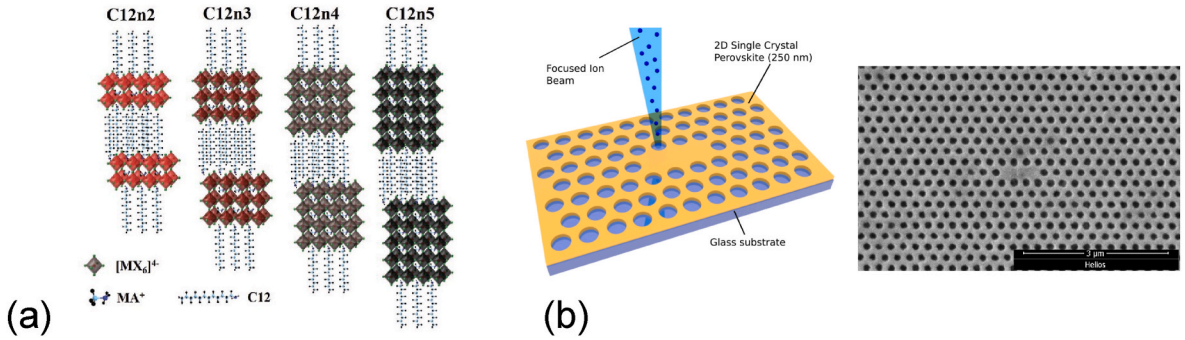


**Fig. 6.** a) Sketch of 2D layered organic/inorganic structures with varying number of inorganic layers; here, $MA^+$ is a methylammonium cation, while $[MX_6]^{4-}$ octahedra are typically composed of a metallic cation (e.g., $Pb^{2+}$) and a halide ion (e.g., $Cl^-$, $Br^-$, $I^-$); Reproduced with permission [35]. b) Schematic illustration of direct FIB patterning of a thin 2D layered perovskite flake, and SEM picture (courtesy of G. Adamo and L. De Marco) showing a photonic crystal L3 cavity realized on a PEAI single crystal (about 500 nm thick) in a joint collaboration between the groups of D. Gerace (University of Pavia), D. Sanvitto (CNR-Nanotec, Lecce), and C. Soci (NTU, Singapore).

field, even their dipolar character. As an alternative route to strong enhancement of nonlinear effects, we envision polariton confinement down to scales on the order of its wavelength. Ultimately, full three dimensional polariton confinement could even lead to nonlinearities that are relevant at the level of few quanta of excitations [36]. Realizing such a paradigm for perovskite exciton-polaritons could foster a totally open and unexplored quantum photonics approach based on low-cost materials and room temperature operation. In addition to recently explored confinement strategies [30], a possible route lies in creating point-defect cavities in a photonic crystal lattice, for which it would be helpful to increase the index contrast of the periodic modulation. An example based on directly patterning the photonic lattice into the 2D layered perovskite by Focused Ion Beam (FIB) lithography is shown in Fig. 6b, where a photonic crystal defect cavity has been realized in an exfoliated single PEAI crystal (see SEM image in Fig. 6b). Improving the optical quality of such structures may be considered a significant but typically high gain challenge to be undertaken in the near future.

### 5.3. Acknowledgements

We acknowledge Luisa De Marco for useful discussions and a critical reading of the contribution. Italian MUR through PRIN project "INPhoPOL" is gratefully acknowledged.

## 6. Perovskite metamaterials and metasurfaces

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### 6.1. State of the art

Halide perovskites offer a brand-new and advanced photonic materials platform that merges high luminescence quantum yield, compositionally tunable emission spectrum, outstanding optoelectronic properties, phase-change tunable optical characteristics and high refractive index [37]. This allows realizing extreme light confinement and strong light-matter interaction with excellent radiative properties by using metasurface/metamaterial designs consisting of periodic arrangements of subwavelength perovskite nanostructures. They hold high promise for achieving passive/active amplitude, polarization and wavefront manipulation of the transmitted/reflected light or their own luminescence.

Perovskite metasurfaces/metamaterials have initially been used for (reflective) color display and (active) luminescence enhancement [38,39]. Both monolithic dielectric designs (Fig. 7a) [38] and hybridized plasmonic designs [39] have been adopted. The first examples of a monolithic perovskite metasurface were one-dimensional nanogratings (Fig. 7a). The incident light is confined as various optical resonances inside the ridges of the nanogratings, leading to spectrally designed reflection that covers a broad color gamut. Meanwhile, by coupling the perovskite excitonic emission to the optical resonances, a dramatic enhancement of luminescence can be obtained due to Purcell effect.

Apart from amplitude manipulation, perovskite metasurfaces/metamaterials also offer extra degree of freedom for light manipulation, i.e., polarization and wavefront control, which is still in its infancy. High degree of linear polarization of photoluminescence was obtained through an anisotropic hyperbolic metasurface design, with alternative perovskites and gold constitutes [40]. Another hallmark of polarization control, circular dichroism (CD) and emission of circularly polarized light, has mainly been realized by chemically incorporating chiral ligands into perovskites with a limited degree of circular polarization (DOP) and CD. Creating metasurfaces with chiral meta-atoms or arrangements is an effective strategy to impart strong extrinsic chirality to perovskites. Recently, gammadion-shaped $MAPbI_3$ metasurfaces with strong asymmetric responses to circularly polarized incident light were demonstrated [41], yielding an extremely high CD of ~0.5 (Fig. 7b). Apart from achieving giant passive optical activity, obtaining luminescence with well-defined directionality and high DOP by superstructuring is also very appealing, considering that luminescence from perovskite film is normally omnidirectional and unpolarized. By breaking in-plane inversion symmetry to mimic the two-dimensional transition metal dichalcogenides, a monolithic halide perovskite metasurface emitting highly directional chiral photoluminescence, a manifestation of the optical Rashba effect (Fig. 7c), was demonstrated in a recent work [42]. This metasurface yielded a DOP of 60%, more than one order of magnitude higher than state of art chiral perovskites, highly directional chiral photoluminescence, with different handedness directed into opposite hemispheres of the far field, and four-fold enhancement of photoluminescence quantum yield compared with bare perovskite films, due to the Purcell effect.

Since perovskites have high refractive index, they can function as small footprint cavities with high radiative quality factors, a feature that makes them an ideal platform for realizing ultracompact low threshold microlasers. Recently, a vortex microlaser based on perovskite metasurfaces with bound states in the continuum design was introduced and showed ultrafast all-optical switching between vortex- and linearly

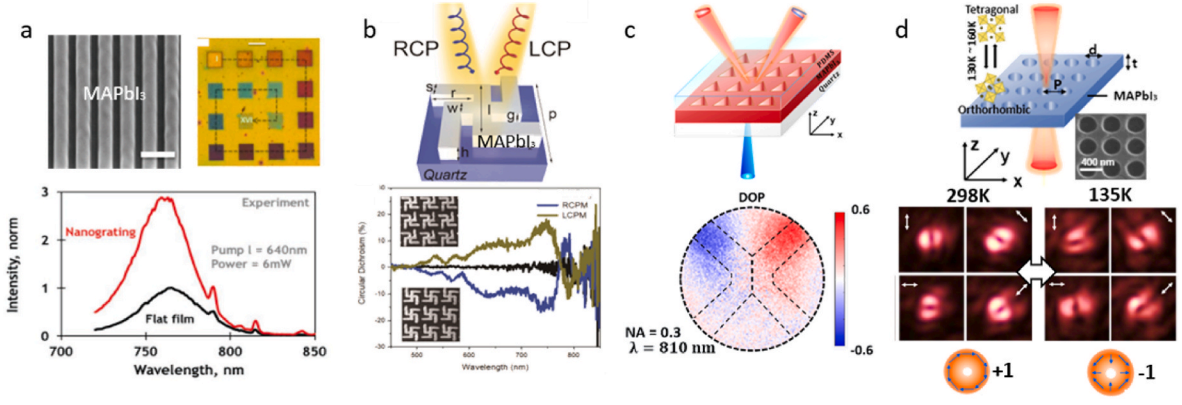


**Fig. 7.** a) Perovskite nanograting metasurfaces with a broad color gamut and enhanced photoluminescence. Reproduced with permission [38]. Copyright 2017, Wiley-VCH b) Giant circular dichroism of perovskite chiral metasurfaces. Reproduced with permission [41]. c) Perovskite metasurfaces with broken in-plane inversion symmetry for directional control of enhanced chiral luminescence. Reproduced with permission [42]. Copyright 2022, Wiley-VCH d) A fundamentally new type of optically pumped tunable semiconductor laser based on a phase-change perovskite metasurface with bistable switching of lasing beams carrying different topological charges. Reproduced with permission [32]. Copyright 2022, Wiley-VCH. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

polarized-beam lasing, at room temperature [14]. A fundamentally new type of tunable semiconductor laser, based on a phase-change perovskite metasurface that acts simultaneously as tunable gain medium and optical cavity, was demonstrated soon after (Fig. 7d) [32]. The laser showed distinct hysteretic behavior yielding unique spectral and spatial optical bistability around the phase-transition temperature, where it sustained dynamic switching of different polarization vortex lasing beams carrying opposite topological charges, underpinned by multiple BICs.

### 6.2. Current and future challenges

Although perovskite metasurfaces/metamaterials show great promise for both passive and active light manipulation, they are still in their infancy and many challenges lie ahead for large scale deployment.

1) Stability is a known bottle neck for perovskite-based devices. Their instability and intolerance to air, moisture, light, many solvents and the associated standard fabrication techniques significantly limit their extensive deployment.
2) Only relatively simple physics and design principles from the field of optical metasurface/metamaterials have been applied to perovskite metamaterials/metasurfaces, thus far. Therefore their potential as monolithic platform for light generation, manipulation, and detection for applications for lasers, light modulators and photodetectors has not been fully harnessed yet.
3) So far, only optically pumped lasers have been realized using perovskites as gain media. Although continuous wave pumped lasers have recently been realized at room temperature [22], great strides are still needed to tackle the problems, such as Joule heating, that hinder the realization of an electrically pumped perovskite laser.
4) Reconfigurable perovskite metasurfaces might pose further challenges. The introduction of tunability into perovskite metasurfaces/metamaterials might limit performance by reducing the device lifetime or working bandwidth and result in complex control circuitry.

### 6.3. Advances in science and technology to meet the challenges

Following up on the aforementioned challenges, we witness the rise of scientific studies and technological remedies aiming at addressing them, which make us confident that solutions enabling the practical realization of perovskite metasurface/metamaterial devices are on the horizon.

1) In view of recent progress in improvement of stability of perovskite-based photovoltaics, the immunity of the perovskite metasurfaces/metamaterials to air, moisture, etc. can be strengthened by doping, element substitution, interface engineering or encapsulation in air-isolated and water-proof matrix [45].
2) The design principles of perovskite metasurfaces/metamaterials can be enriched benefiting from the rapid development of optical metasurfaces and naophotonics. Novel physics such as topological photonics [46] and BIC can be applied to improve the performance and expand the functionalities of perovskite metamaterials/metasurfaces towards an all-perovskite monolithic platform for light generation, manipulation, and detection.
3) Thanks to the development of new perovskite compounds showing high tolerance to joule heating, metasurfaces can be directly patterned onto perovskite light-emitting devices (e.g. light-emitting transistors,LETs) for enhanced light-matter interactions. Here, a strong Purcell enhancement can be achieved through design of high-quality factor resonators and high carrier density is attainable without generating too much heat [47,48]. This may pave the way to electrically driven perovskite lasers.
4) Reconfigurable perovskite metasurfaces can be realized by exerting optical, electrical, and thermal stimuli. One ingenious way may be to achieve all-optical spatial control by influencing the material gain and loss distribution with extra light pump, which though has little impact on the spectral tunability [14]. Another way is to take advantage of the phase-change nature of perovskites, which show tunable refractive index and optical gain simultaneously through phase transition for realizing both spectral and spatial tunability [32, 49].

### 6.4. Concluding remarks

Despite various challenges, the robust progress in theoretical principles, materials technology, and fabrication techniques of perovskite metasurfaces/metamaterials offer a rosy perspective to surpass the limits of passive platforms with emergence of new capabilities for the exploration of novel physical phenomena. Applications include optical communications, optical memories, optical sensors and embedded LIDAR systems, towards an all-perovskite monolithic platform for light generation, manipulation and detection.

### 6.5. Acknowledgements

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## 7. Active resonant perovskite metaphotonics

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### 7.1. State of the art

Metaphotonics is a new direction in subwavelength photonics based on the metamaterials concept [50]. The main idea of metaphotonics is to use the physics of optically induced Mie resonances of dielectric nanoparticles (employed as “meta-atoms” in metamaterials structures and metasurfaces) for creating optical metasystems with novel functionalities derived from their magnetic response. The growing interest in metaphotonics is related to the unique opportunities for creation of novel efficient light sources and nanolasers. Recently, halide perovskites have attracted a lot of attention from various research communities owing to their outstanding optical and electronic properties. Due to their relatively high refractive index ($n > 2$), halide perovskites can be used as a platform for Mie-resonant nanophotonics in the optical range [51], and these materials are ready to make the next step of development toward the metaphotonics [52].

### 7.2. Current and future challenges

Despite the halide perovskites is a class of materials with high defect tolerance, meaning that they can demonstrate high luminescence quantum yield and charge carriers mobility even being created by a simple chemical synthesis [53], they are quite sensitive to the environmental conditions (e.g., to oxygen and humidity in the atmosphere) [54]. Also, polar liquids (water, alcohols, etc.) virtually dissolve the perovskites, making it problematic to use standard nanolithography procedures. Thus, one of the main challenges in perovskite metaphotonics is to develop advanced methods of metasurfaces fabrication. A successful solution of these problems would allow the creation of highly efficient light-emitting metaphotonic structures for linear and nonlinear optics.

### 7.3. Advances in science and technology to meet the challenges

The most straightforward approach is the direct chemical synthesis of halide perovskite resonant nanoparticles on a substrate. Fig. 8a shows an example of the perovskite single-particle nanolaser [55] grown chemically on a sapphire substrate for low defect concentration. In this design, the high-quality single-crystal $CsPbBr_3$ perovskite has an optimized size (the smallest cube is just 310 nm) supporting 3rd order Mie mode (magnetic octupole), which is enough to demonstrate lasing generation upon optical pump at room temperature. Indeed, the high crystal quality and pronounced exciton result in high gain of the material achieving the values of the order $10^4$ $cm^{-1}$ at room temperature.

In order to scale-up the technology of metaatom fabrication and create high-quality metasurfaces, focus ion beam lithography can be applied. Fig. 8b shows perovskite metasurface assembled as bullet-type nanoparticles [56]. Due to the interference of electric and magnetic dipoles, the energy of the incident wave is mainly scattered in the forward direction and causes broadband antireflection. In particular, reflectivity was decreased from 30% down to a few percent in a significant part of the visible spectrum (see photo in Fig. 8b). In addition, as a result of the support of the resonant Mie mode and the Purcell factor, the photoluminescence intensity of the perovskite metasurface (containing less material) was even 10% higher in comparison with the initial film. However, this method of nanolithography requires additional post-processing for material healing after bombarding the perovskite film by heavy Ga ions leading to defects formation and reduction of gain.

To avoid such perovskite damage by the ions, one can apply electron-beam lithography, which also allows for high-precision nanofabrication for formation of subwavelength optical nanostructures. Fig. 9a shows perovskite metasurface created by application of this method [57]. Strong coupling with the metasurface mode and high localization of the electromagnetic field of the excitation wave allows for significant enhancement of nonlinear two-photon photoluminescence, which becomes comparable with the linear one. Moreover, the threshold of the two-photon amplified spontaneous emission (inflection point on the upper part of the curves) is only 2.7 times higher than that for the linear one. In addition, the enhancement in the perovskite metasurface is not limited in the two-photon process. The intensity of the three-photon process is also comparable to the linear one and the threshold of three-photon induced amplified spontaneous emission is 30 times of the corresponding process for linear photoexcitation.

The most high-throughput method is nanoimprint lithography which allows to create perovskite metasurfaces [58] and has high potential for metasurfaces scaling-up with roll-to-roll fabrication technology. Fig. 9b illustrates two-photon luminescence from this kind of imprinted metasurfaces consisting of nanoholes and nanostripes. Maintenance of the magnetic Mie resonances at excitation wavelength results in enhancement of electric field in the structures. During the multiphoton excitation linear losses are low that allows to excite resonances with high Q-factors, whereas high absorption occurs only in hotspots. In addition, supporting mode at emission wavelength causes acceleration of photoluminescence due to the Purcell effect. All these factors lead to the enhancement of nonlinear photoluminescence intensity up to 70 times in perovskite metasurface compared to pristine film.

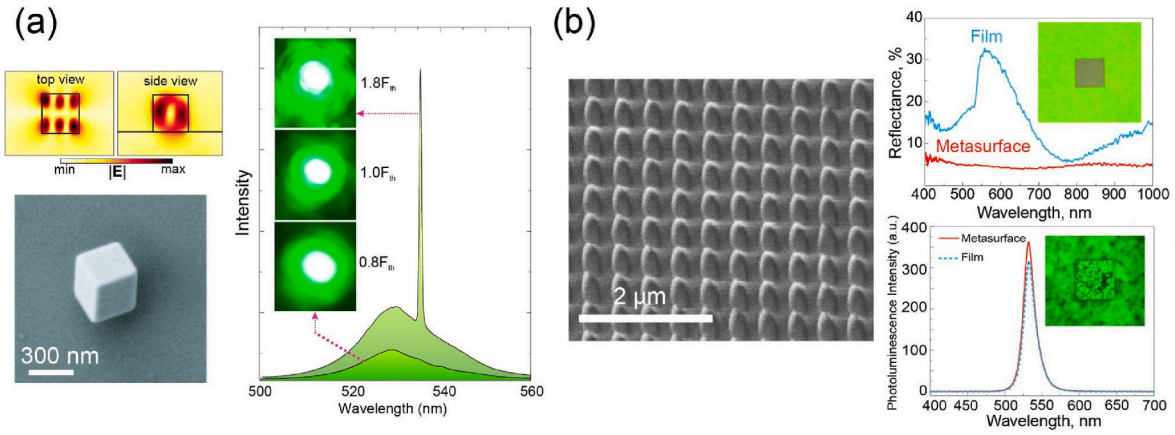


**Fig. 8. Linearly excited active perovskite meta-structures.** a) Cubic perovskite nanolaser with SEM image, near-field structure, and luminescence spectra below and above the lasing threshold. Reproduced with permission. [55]. Copyright 2020, American Chemical Society b) Anti-reflective perovskite metasurface with its reflection and photoluminescence properties. Reproduced with permission. [56]. Copyright 2020, Wiley-VCH.

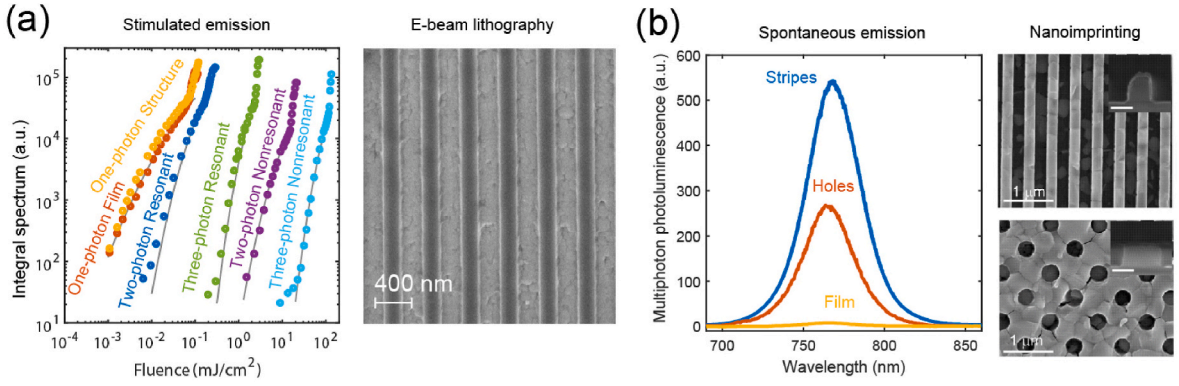


**Fig. 9. Nonlinearly active perovskite metastructures.** a) Perovskite metasurfaces created by electron-beam lithography with the dependence of photoluminescence signal on the incident fluence. Reproduced with permission. [57]. Copyright 2021, American Chemical Society. b) Nanoimprinted perovskite metasurfaces and nonlinearly excited multiphoton photoluminescence. Reproduced with permission. [58]. Copyright 2017, American Chemical Society.

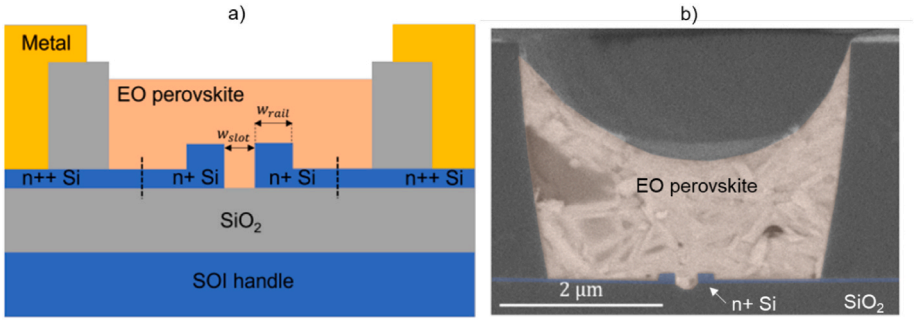


**Fig. 10.** a) Schematic of a Si slot waveguide cross-section filled with electro-optic (EO) perovskite material. b) False colored SEM of a fabricated device cross-section. The EO perovskite material fills in the Si slot waveguide's central gap. Riproduced with permission. [67].

### *7.4. Concluding remarks*

Recent demonstrations of perovskite metaphotonic structures have shown a high potential for the realization of subwavelength active nanophotonics for various light-emission applications: ranging from Mie-resonant ultracompact nanolasers to highly efficient IR light visualizers. Further decreasing the nanolaser size becomes possible by not only decreasing temperature, but also via optimization of perovskite material properties (e.g. defects passivation or exciton strength optimization) or optimization of the resonant mode quality factor in the nanocavity. We also envision further development in the direction of active topological perovskite metaphotonics, where the designs would pave the way for unique lasing properties in visible range. Moreover, we anticipate the demonstration of electrically driven metasurface-based perovskite LEDs and even lasers, where flat metaphotonic structures with high quality factors integrated with injecting electrodes should be designed via multi-physical smart optimization taking into account not only optical properties but also electro-physical ones.

### *7.5. Acknowledgements*

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## 8. Towards integrated perovskite electro-optic modulators

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### *8.1. State of the art*

Most of today's photonic applications depend heavily on integrated optical modulators, which are on-chip devices used to encode electrical signals onto guided optical fields. Modulators can operate on the intensity, phase, and polarization of the optical field, be analog or digital and, at the same time, there is an ever-increasing demand for small footprint devices providing faster, efficient, and high-performing modulation. The term modulator usually applies to devices operating at tens or hundreds of Gbit/s for optical communication or optical interconnects, whereas actuators is used for slow, energy efficient elements for programmable and reconfigurable circuits. The classical technologies exploit the Lithium Niobate electrooptic crystal, semiconductors as Indium Phosphide and silicon photonics. In recent years, the photonics community has been attracted by a hybrid materials approach, combining new materials with mature technologies such as silicon photonics, silicon nitride or other dielectrics with the aim to merge the best properties of both, even if facing technological challenges. Interestingly, during the past decade perovskites have been successfully proven as excellent optoelectronic materials that, in addition to their capability to be easily processed as a solution, makes them excellent candidates for both monolithic and hybrid integration on other technological platforms.

Electro-optical modulators can be classified by their working principle: 1) the electro-refractive effect, which generates a change on the material's refractive index when an electric filed is applied to it, being the effect linear (Pockels) or quadratic (Kerr); and 2) electro-absorption, which refers to a change in the optical absorption (band gap) [59]. It has been showed that perovskites exhibit both behaviours and are currently being explored for their application as modulators.

The modulation based on the linear Pockels effect requires non-centrosymmetric materials, but traditional metal halide perovskites are intrinsically centrosymmetric structures. Despite this, there are some alternative candidates [60]: by means of density functional theory, it was found that germanium halide perovskites have a strong linear electro-optic (LEO) response ($CsGeI_3$ has a predicted LEO coefficient of 47 pm·V$^{-1}$ at a wavelength of 1550 nm, which is larger than lithium niobate's of 31 pm·V$^{-1}$) [61]. Experimentally, LEO modulation was first demonstrated in layered $PMA_2PbCl_4$ perovskite crystals by Gao et al., [62] where they also determined the LEO coefficient to be 1.4 pm·V$^{-1}$, which opens the door for the exploration of this kind of layered structures for electro-optic modulation. The same group also demonstrated the LEO response of metal-free halide perovskites ($(MDABCO)NH_4I_3$ crystals). By introducing ammonium vacancies, they observed a LEO coefficient of 14 pm·V$^{-1}$ at a modulation frequency of 80 kHz, the highest measured among these materials [63]. A strong LEO coefficient of 20 pm·V$^{-1}$ has also been experimentally shown in organic $(DCl)(NH_4)(BF_4)_3$ crystal perovskites. Their large LEO behaviour arises from their high polarizability, which can be chemically tailored [64].

The Kerr effect is not symmetry dependent and arises from the material's nonlinear refractive index $n_2$ (which in turn is related to the real part of the material's third-order susceptibility, $\chi^{(3)}$). The imaginary part of $\chi^{(3)}$ depends on the material's nonlinear absorption coefficient $\beta$. Both parameters have been widely reported for a variety of hybrid, inorganic and Ruddlesden-Popper perovskites, and condensed in the review article by Zhou et al. [60] They also report the value of a figure of merit (FOM) that is a compromise between the co-existence of $n_2$ and $\beta$ for the design of non-linear devices. For Kerr based electro-optic phase modulation, a large FOM is desired, and the values reported for $n_2$ are equivalent to conventional semiconductors, which indicates that this effect could be exploited for electro-optical modulation in perovskites.

Electro-absorption modulators based on the quantum-confined Stark effect (QCSE) are well established and have excellent characteristics but are expensive to produce. Walters et al. [65] engineered layered perovskites to have QCSE behaviour and showed large absorption coefficient changes of 70 cm$^{-1}$ for 5.6 V µm$^{-1}$ applied fields in hybrid perovskite nanoplatelets. This modulation performance is on the same level of inorganic epitaxial semiconductors with the advantage of being fabricated via cheap solution-processing, a promising avenue for

integrated perovskite modulators.

Even other electro-optic modulation mechanisms have been observed, such as a combination of ion movement and photon recycling [66], that could have applications in the visible spectrum. The class of perovskite materials is very vast and motivates an intense research activity of efficient, compact and low cost modulators and actuators.

### 8.2. Current and future challenges

The works highlighted in the above section suggest the potential of solution-processed perovskites as an active material for integrated on-chip actuators and modulators, and their application on optical communications and beyond [62,63,65]. But challenges abound, and these were concisely brought to light by McKiel's and his colleagues [67], who, in a heroic effort, showed the first integration of solution-processable halide perovskites onto a foundry fabricated active SiPh platform (Fig. 10). Working towards the aim of demonstrating a modulator, they identified the next challenges: 1) Identify how does the reported LEO coefficient of bulk perovskites compare to thin films of the same material. 2) To induce the Pockels effect, the on-chip deposited polycrystalline material dipoles must be aligned with respect to the applied electric field. This is achieved via a poling process. The community needs to work towards identifying the ideal poling conditions for perovskite thin films. 3) Improve the perovskite thin film quality and adhesion to the SiPh substrate. This could help to avoid the formation of air pockets during the deposition process, minimize scattering losses, and the shifting of material during the poling process. 4) Work towards the growth of perovskite single crystals directly on the desired regions of the SiPh chip to overcome the obstacles posed by polycrystalline films.

### 8.3. Advances in science and technology to meet the challenges

Integrated photonics is a mature technology and there's a plethora of successful demonstrations of highly performing hybrid waveguide-based modulator configurations that use 2D materials as active medium [68]. And even more, integration of perovskites for lasing on an integrated SiPh platform has been successfully shown in the past 5 years [23]. These same waveguide configurations and fabrication processes can be directly used with perovskites for the implementation of modulators leading to both active and passive devices integrated on the same chip. The planarization and chemical preparation of the photonics chip surface to accommodate thin layers of foreign materials are common and well-developed processes in nanofabrication facilities. To temporarily circumvent the poling problem, and without losing sight of the goal of the solution processing of perovskites on top of the photonics chips, one could directly place perovskite single crystals onto a waveguide with micro-transfer techniques, as proposed by Ref. [69].

### 8.4. Concluding remarks

Their rich and unique opto-electronic properties, their ease of fabrication, and the recent demonstration of electro-optic and electro-absorption behaviours make perovskites a material worth to be explored for the next-generation of on-chip densely integrated actuators and modulators.

## 9. Phase change tuning and modulation in resonant halide perovskite nanostructures

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### 9.1. State of the art

Reconfigurable nanophotonic and metaoptical designs are of a high demand in such fields as displays, adaptive optics, data recording, and many others. The most popular approaches are related to electrically, optically, chemically, and thermally induced modifications in the materials of nanostructures [70]. Namely, external treatment changes structural properties or composition of the material resulting in variation of refractive index, absorption, or emission wavelength. Remarkably, there is a general trade-off between speed and strength of the modulation: the stronger the modulation, the slower it is. In halide perovskites, this empirical law still works. For example, the Kerr-like all-optical modulation goes faster than 1 ps but provides refractive index variation on the few-percent level [71]. On the other hand, much slower phase transitions, resulting in significant changes of band structure, can take several orders of magnitude longer timescale. Finally, the ionic nature of the halide perovskite allows for slow but dramatic changes in the material upon external electric field or chemical processing.

Identifying the crystallographic phases of metal halide (chloride, bromide, or iodide) perovskites, one has to keep in mind what defines the latter. A perovskite crystal lattice consists of corner-sharing $BX_6$ octahedra (B - metal cation, X – halogen anion) whose negative charge is compensated by large cations (A – cesium, methylammonium, or formamidinium cation) captured in cavities confined by every four octahedra. As a result, metal halide perovskites adopt $ABX_3$ (or equivalent) stoichiometry. Therefore, only a few crystallographic phases, namely, orthorhombic, tetragonal, and cubic ones (Fig. 11a), meet the basic description of the crystal structure. These phases characterize all the chemical compositions including mixed-halide, mixed-cationic at A site, and double cationic at B site as well as dimensions of metal halide perovskites including bulk crystals and polycrystalline films, 2D nanoplatelets, 1D nanowires, and 0D nanocrystals (see a phase-transition diagram for Pb(ii) and Sn(ii) halide perovskites in Fig. 11b). However, some ternary metal halides, in which A-site cation is either too small or too large for stabilizing the network of $BX_6$ octahedra, possess perovskite structure in specific conditions (e.g. high temperature, pressure, etc.). Among them, there are $CsPbI_3$ crystallizing in 1D edge-sharing octahedral lattice and $FAPbI_3$ showing a 1D face-sharing motif at ambient conditions. Annealing these often called "post-perovskites" at 200–300 °C transforms their phases to perovskite ones. Another case that should be pointed out is quasi-2D metal halides exhibiting Ruddlesden-Popper (RP), or Dion-Jacobson (DJ) crystallographic phases. Such materials have layered structure owing to large cationic organic species (ligands) that cannot reside in the perovskite cavity and, hence, create organic layers separating the metal halide layers. Generally, they have $A'_2A_{n-1}B_nX_{3n+1}$ (RP) and $A''A_{n-1}B_nX_{3n+1}$ (DJ) composition where ligands A′ and A″ have a positive charge of +1 and + 2, respectively. On the one hand, reducing n down to 1 gives $A'_2BX_4$ and $A''BX_4$ structures that cannot be classified as perovskites since they do not fit $ABX_3$ (or equivalent) stoichiometry. On the other hand, increasing n up to infinity gives a conventional perovskite structure. Thus, the question of how to classify the layered phases for $1<n\ll\infty$ is still under debate [72].

### 9.2. Current and future challenges

Apparently, the most fascinating feature of metal halide perovskites - the tunability of their optical bandgap in the 1.24–3.55 eV range (Fig. 11c) [73]. Although the tuning can be to a certain extent realized via substituting corresponding cations for A-site or B-site ones, the gradual change of optical properties can be reached by mixing $Cl^-$ and $Br^-$, or $Br^-$ and $I^-$ anions at X site. Unfortunately, mixed-halide compositions suffer from light- and electric field-induced phase separation (halide segregation), and stabilizing them is challenging so far. The major mechanism of the segregation is migration of thermally activated anions (or their vacancies) that could be invoked by the following

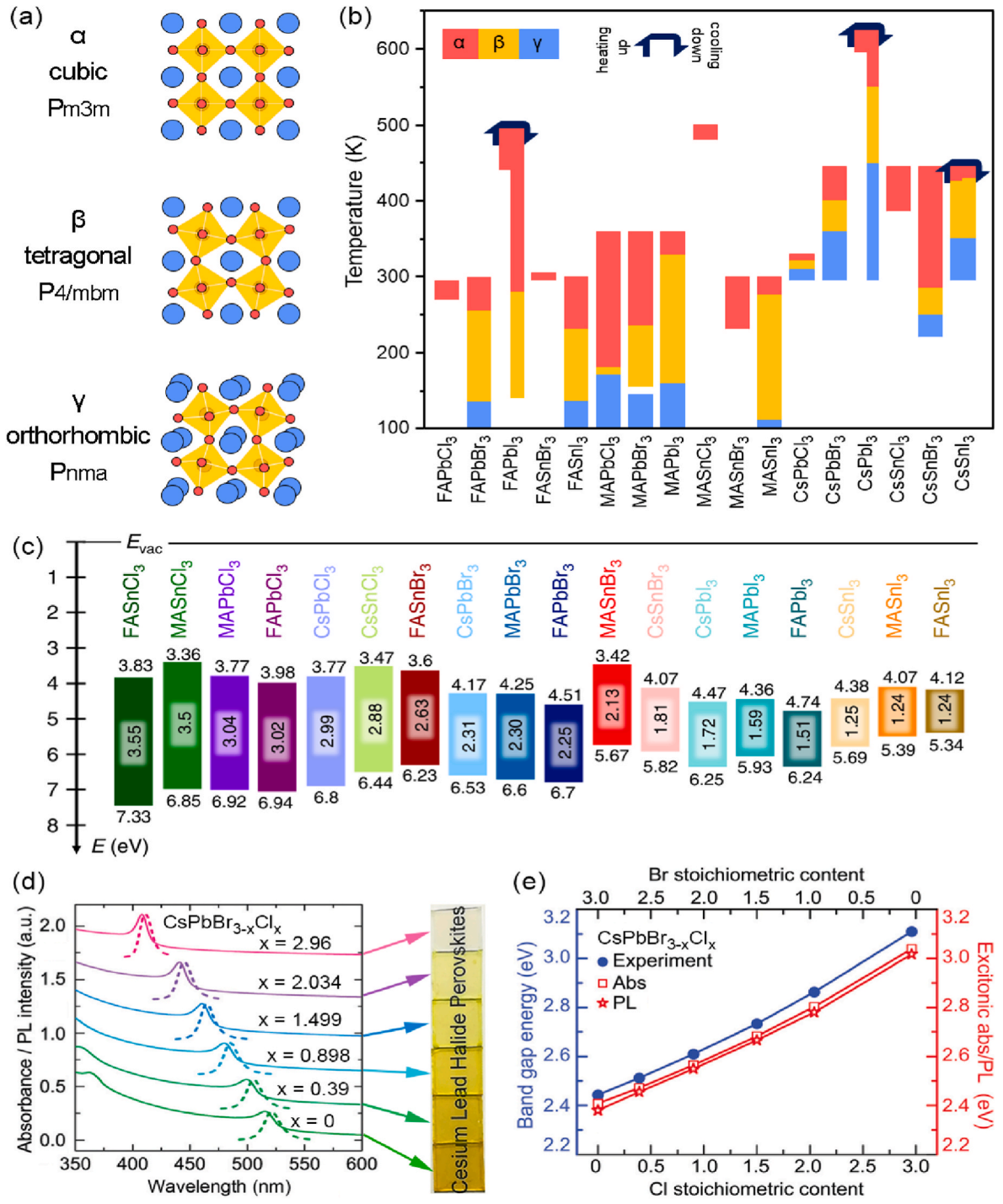


**Fig. 11.** a) illustration of cubic (α), tetragonal (β), and orthorhombic (γ) perovskite crystallographic phases. b) Perovskite phases for various ternary Pb(ii) and Sn(ii) halides at different temperatures. Note, for $FAPbI_3$, $CsPbI_3$, and $CsSnI_3$, phase-transition hysteresis is observed. Redrawn from Ref. [77] c) Energy values for bottom of the conduction band, top of the valence band, and band gap of Pb(ii) and Sn(ii) halide perovskites. Reproduced with permission. [73] d) Gradual tuning of absorption and photoluminescence spectra for mixed-halide $CsPbBr_{3-x}Cl_x$ thin films obtained by exposure of a tribromide film to HCl vapor at 120 °C. Side-image: a photograph of the samples. e) Experimentally derived band gap (blue circles), excitonic absorption (red squares) and emission (red stars) energies versus halides' stoichiometric content relationships for $CsPbBr_{3-x}Cl_x$ thin films. d) and e) adapted from Ref. [75]. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

phenomena: i) localization of the photogenerated polaron at the spontaneously formed the heaviest halogen-rich site where sufficient lattice strain occurs; ii) directional drift of halide ions having different mobility caused by non-homogeneous charge carrier generation rate profile through the thickness of a perovskite film; iii) minimization of the energy for generated holes that triggers the formation of the heaviest halogen-rich domains exhibiting a low energy valence band edge [74]. Besides the production of uniformly distributed photoexcitation for thin mixed-halide perovskite films, nano- and microstructures, the solution for the avoiding of the rest of the pointed out phenomena is a synthesis of single-crystals demonstrating a low number of halide vacancies along with reduced electron-phonon coupling owing to substituting $Cs^+$ for

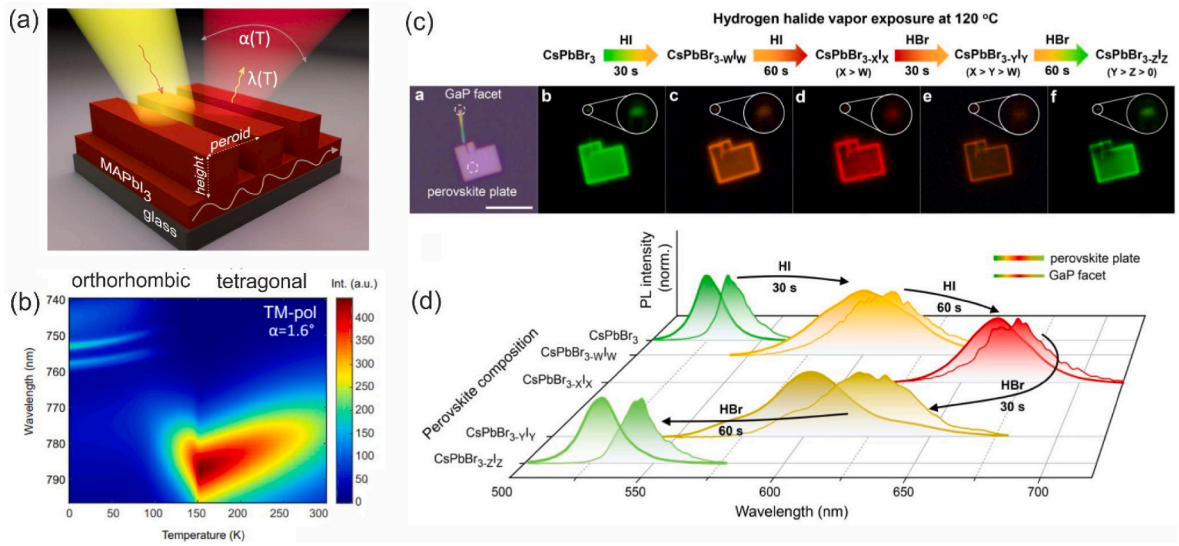


**Fig. 12.** a) Schematic illustration of the nanoimprinted perovskite grating and principle of temperature-dependent emission. Riproduced with permission [78]. Copyright 2018, Elsevier. b) PL temperature maps for a perovskite film with nanograting. c) Bright-field optical image of GaP NW integrated into $CsPbBr_3$ microplate (dashed circles confine the areas of the PL signal detection; scale bar is 10 μm). Fluorescent images of the same initial and iodine enriched GaP-$CsPbBr_{3-x}I_x$ ($0 < x < 3$) structures produced by subsequent exposure of the tribromide perovskite microplate to HI vapor. PL images of the iodine-doped device exposed to HBr vapor for different time at the same temperature. d) PL spectra of the structures (b–f) recorded from the microplate and NW end facet. Riproduced with permission [79]. Copyright 2020, American Chemical Society.

polar $MA^+$ and $FA^+$ cations. Such a synthesis cannot be realized by applying simple room-temperature wet chemical approaches to mixtures of various metal halides, because the latter have very different solubility in common organic solvents (DMF and DMSO) utilized for preparing perovskite precursor solution and, therefore, tend to crystallize in the form of pure-halide perovskites instead of mixed-halide ones.

In our opinion, an anion exchange reaction between solid perovskite and hydrogen halide vapor could be a cornerstone for future inexpensive and large-scale technology of manufacturing advanced photonic designs and optoelectronic devices based on mixed-halide phases resistant to the segregation effect. It was shown that exposure of polycrystalline $CsPbBr_3$ thin films [75] and nanoparticles [76] to HCl vapor gives mixed-halide counterparts exhibiting the desired optical properties that can be tuned precisely in the 400–525 nm range (Fig. 11d and e) by taking control over the duration time for the exposure procedure. Similar anion exchange reaction was employed for the obtaining $CsPb(Br,I)_3$ polycrystalline film (PF) and single-crystal nanowire (NW) of nearly the same chemical composition. It was established that mixed-halide NW reveals better resistance to light-induced phase separation as compared to PF. Therefore, from this point of view, the future challenge could be the adapting of the described anion exchange reaction for the production of mixed-halide single-crystal films that can be further processed by using direct laser writing (DLW), nanoimprint lithography (NIL), or e-beam lithography to establish state-of-the-art optoelectronic applications operating in a wide spectral range.

### 9.3. Advances in science and technology to meet the challenges

As follows from previous discussions, halide perovskites and, thus, perovskite nanostructrures can exhibit relatively slow but extremely strong modulation of their optical properties upon variation of thermal or chemical conditions. Such relatively slow processes were recently applied for a number of advanced perovskite-based nanophotonic designs.

Standard 3D $MAPbI_3$ exhibits two main phase transitions: from cubic to the tragonal (in range $T = 300–400$ K) with relatively weak influence to optical properties, and from tetragonal to orthorhombic around $T = 150$ K with sharp blue-shift of the band gap by 0.2 eV. Thus, nanogratings written on $MAPbI_3$ demonstrated strong temperature sensitivity of outcoupled spontaneous emission [78] and lasing [32] around the $T = 150$ K (see Fig. 12a and b).

Since the anion exchange reaction can change band gap of the perovskites even stronger that temperature, it becomes possible to provide in situ and permanent tuning of the fabricated nano- and micro-scale photonic designs. For example, optical response of Mie resonances in $CsPbBr_3$ nanoparticles was gradually tunted by more than 0.5 eV in visible range after interaction of HCl acid vapor. Remarkably, that such strong tuning was reversible by applying HBr acid vapor to the nanoparticles already modified at the previous step [76]. Furthermore, perovskite $CsPbBr_3$ microlasers with integrated GaP nanowaveguide were also reversibly tuned by applying HI and HBr acids vapor [79]. To avoid the problem of phase segregation specific annealing protocols should be applied.

### 9.4. Concluding remarks

Recent achievements in phases changes in perovskite-based nanophotonic and metaoptical designs have opened new horizons for light control at nanoscale for creation of tunable optotelectronic devices. However, not all varieties of phase transitions have been exploited. For instance, we envision further progress in the direction of phase transitions between various quasi-2D to 3D phases, which can be achieved optically with high spatial resolution, being attractive for data recording applications. Also, combination of strongly, reversibly, wideband electrically-tunable perovskites with integrated metasurfaces properly designed for antireflective purposes can be very useful as photochromic coatings.

### 9.5. Acknowledgements

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## 10. Quantum nanophotonics devices based on perovskite single photon emitters

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### 10.1. State of the art

In the last two decades perovskites nanocrystals (NCs) have emerged as attractive material for light generation and optoelectronic applications thanks to their outstanding optical properties, such as wide tunable emission wavelength playing on their size and chemical composition, narrow bandwidth and high photoluminescence quantum yield (PLQY). Recently the interest in perovskite NCs is rapidly raising in the quantum optics community after the demonstration that they can generate quantum states of light in the quantum confinement regime [80]. The concept of quantum confinement is widely used for classical semiconductor nanostructures. These systems are considered quantum-confined when the electronic and optical properties are size dependent at the microscopic level and this happens when at least one dimension becomes lower than the exciton Bohr radius, with direct effects on the electronic band gap and the joint density of states (DOS). Similarly, for colloidal perovskite NCs with size smaller than the exciton Bohr radius, the quantum confinement induces an increase of the electronic bandgap and the exciton binding energy: the exciton experiences a significant spatial confinement of the wavefunction, and the photo-physics of the emitter is dominated by discrete hydrogen-like excitonic states, with the emergence of quantum properties. Confinement effects have been observed in both inorganic [81] and organic-inorganic [82] colloidal perovskites NCs proving that these emitters can be used as single photon sources.

The generation of single photons on demand has become a field of huge interest driven by a significant advance in quantum-information science - including quantum communication and cryptography, quantum simulation and quantum metrology - in which they are considered a building element of many future quantum devices as well as interesting qubits for quantum networks. The required single-photon source for such applications should produce light pulses with no more than one photon, in a pure quantum state and as efficiently as possible, showing a photo-stable emission without blinking or photo-bleaching and operating at room temperature [83]. In the last two decades, several physical systems have been studied in order to realize single photon sources ranging from cold atoms, ions and organic molecules to solid states emitters and color centers in diamonds. In this framework, perovskite NCs are promising nano-objects for quantum applications, showing a size-dependent high quality single photon emission also at room temperature [84], as shown in Fig. 13. Moreover, they present several advantages with respect to the competitors: they have a high PLQY up to 90% [81] and are easily synthesized by low-cost, well mastered wet chemistry techniques - differently from single defects in nanodiamonds and epitaxial quantum dots which require heavy fabrication facilities.

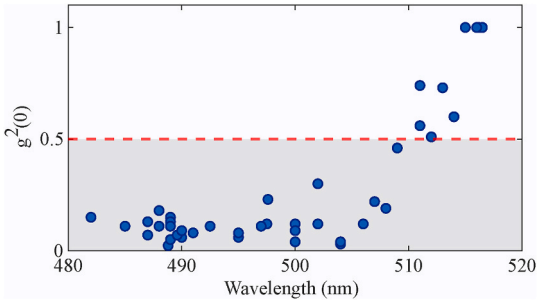


**Fig. 13. Quantum size effect on $CsPbBr_3$ NCs:** Experimental $g^2(0)$ values (blue dots) as a function of the central wavelength of the emission for a set of 40 $CsPbBr_3$ NCs. The threshold for antibunching is reported (red dashed line). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

### 10.2. Current and future challenges

Despite these interesting features, the use of perovskites NCs is limited by photo-instability effects as they usually bleach after prolonged illumination time and present fluctuations (blinking) in the photoluminescence between low and high intensity states. Overcoming these instabilities is a key challenge for the future development of this material. To reduce these problems several strategies are pursued going from polymer encapsulation [85] to surface passivation [86]. Another promising approach to reduce the blinking is to synthetize core/shell structured perovskites, capping the perovskite NCs into a protective semiconductor layer to enable the passivation of the surface defects [87]. Beside the improving of the photo-stability, the other challenge to address in order to boost the applications of perovskites in realistic devices is their integration with photonic interfaces such as waveguides and fiber optics.

### 10.3. Advances in science and technology to meet the challenges

A first approach to develop a high efficiency nanophotonic device is to couple the quantum emitter with a plasmonic nanogap in order to enhance the emission by means of surface plasmon cavity modes [88] or with external optical cavities including distributed Bragg reflector (DBR) and photonic crystal [89] to increase perovskite's optical response. Very recently a great deal of effort has focused into the coupling of perovskite NC of $CsPbBr_3$ with tapered nanofibers [90]. Tapered nanofibers are photonic waveguides fabricated starting from a standard single mode optical fiber and using taper fiber technology: the

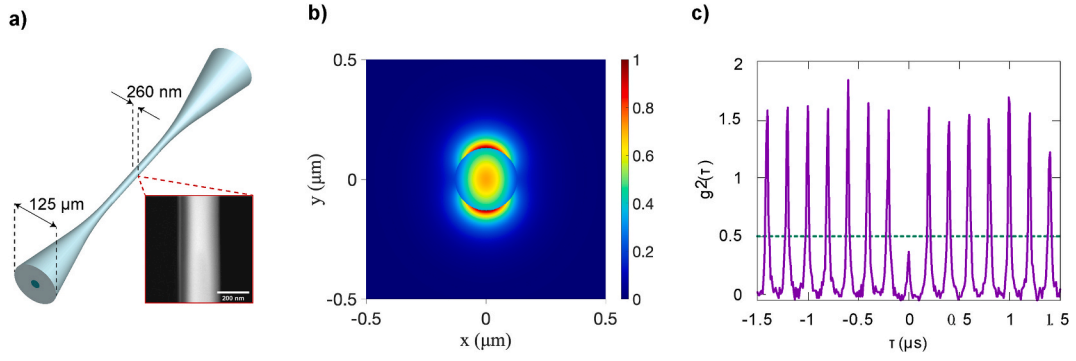


**Fig. 14. Coupling of a perovskite $CsPbBr_3$ NC with a tapered optical nanofiber:** a) Typical geometry of tapered optics nanofiber: the starting optical fiber has a diameter of 125 μm reaching a diameter of few hundreds of nanometers in the nanofiber zone. b) Simulation of the evanescent field's intensity for a nanofiber with a diameter of 260 nm. c) Second order correlation function of a single $CsPbBr_3$ NC emitting single photons into the nanofiber.

fiber is heated and pulled to a diameter of some hundreds of nanometers (Fig. 14a) keeping adiabatically the single-mode condition. This allows for a strong confinement of the electromagnetic field and intense evanescent field around the fiber (Fig. 14b), ensuring an efficient coupling to the fiber of the light emitted by a nano-object placed in the vicinity of the evanescent field. The deposition of a single perovskite NC is achieved by touching the nanofiber with a droplet of high-diluted solution. Then antibunching measurements are performed on the light collected via the nanofiber showing a very low value of the second-order autocorrelation function $g^2$ (0) = 0.24, as shown in Fig. 14c, which represents the probability to emit more than one photon per excitation laser pulse. These results demonstrate that a single perovskite nanocrystal is coupled to the nanofiber and emits single photons directly inside it.

### *10.4. Concluding remarks*

To conclude, perovskite NCs have shown several outstanding advantages with respect to the competitors quantum emitters, demonstrating a huge potential as on demand single photon sources. For their utilization as key components in quantum technologies-oriented applications, the research focus has moved to their challenging integration in photonics platforms, with the aim to realize innovative hybrid nanophotonic devices. Among the other perspectives, the community is striving for the implementation of nanofiber-based cavities like nanofiber Bragg grating (NFBG) by drilling periodic nano-grooves on a subwavelength-diameter fiber or the integration with plasmonic structures fabricated directly on the nanofiber, such as nanogap or nanoantennas, as exceptional enhancers of light−matter interactions to create bright integrated single photon sources that exploit the captivating properties of perovskites.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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