RESEARCH NEWS

Amorphous Photonic Crystals with Only Short-Range Order

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Distinct from conventional photonic crystals with both short- and long-range order, amorphous photonic crystals that possess only short-range order show interesting optical responses owing to their unique structural features. Amorphous photonic crystals exhibit unique light scattering and transport, which lead to a variety of interesting phenomena such as isotropic photonic bandgaps or pseudogaps, noniridescent structural colors, and light localization. Recent experimental and theoretical advances in the study of amorphous photonic crystals are summarized, focusing on their unique optical properties, artificial fabrication, bionspiration, and potential applications.

1. Introduction

As a state of matter, amorphous solids differ from their crystalline counterparts in lacking long-range order of the positions of atoms, which leads to many unusual electronic, mechanical, and lattice dynamical properties.[1] For instance, electronic pseudogaps near Fermi levels in amorphous metals[2] and tail states in amorphous semiconductors[3] appear due to short-range order, giving rise to unusual thermal stability and electronic transport properties. Disorder in amorphous solids may even lead to interesting localization of electronic states.[4]

Analogous to crystalline solids, optical materials can be arranged in a periodic way to form so-called photonic crystals (PCs),[5,6] which have received great attention over the past decades. For light propagations in PCs, multiple Bragg scatterings are expected, which give rise to complicated photonic band structures. Between photonic bands, a photonic bandgap (PBG) may appear. The existence of complicated photonic band structures and PBGs in PCs offers unprecedented opportunities in the control of light propagation, leading to a variety of novel applications ranging from photonics to optoelectronics.[7]

Inspired by amorphous solids, in recent years there has been considerable interest in amorphous PCs (APCs) that possess only short-range order.[8–18] Due to their unique structural features, APCs show many interesting and unique optical properties, representing a novel kind of optical media despite being much less studied and understood than conventional PCs due to the difficulties in both fabrication and theoretical treatment. Herein, we focus on the recent progress in the study of APCs.

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2. Optical Properties

Generally, photonic structures can be classified into three categories according to their structural arrangements: ordered structures with both short- and long-range order such as PCs, APCs with only short-range order, and random structures with neither short- nor long-range order. Their structural differences can be distinguished by analyzing the spatial correlation function, Fourier power spectrum, or radial distribution function. For example, in an APC consisting of random-close-packed spheres as shown in **Figure 1**A the radial distribution function shows obvious peaks at small distances. However, at large distances the radial distribution function is almost a constant, which implies that this structure is without long-range order but has short-range order. The ringlike pattern in the Fourier power spectrum indicates the short-range order and the isotropy of the APC as well. The unique structural features of APCs may considerably influence their optical properties.

2.1. PBG Formation

A hallmark of PCs is the existence of complicated photonic band structures and PBGs which depend on both short- and long-range order.[7] In the field of PCs, there has been a quest for large PBGs.[7] In APCs, photonic band structures do not exist since there is no long-range order. The question of whether or not a PBG can exist in APCs arises naturally.

In a calculation of 2D APCs consisting of high-refractive-index cylinders,[8] a PBG was found for one polarization. No PBG exists for the other polarization. Inspired by amorphous semiconductors, optical properties of a Si-rod-connected APC with an amorphous-diamond symmetry were studied theoretically.[13] A PBG was found, which can be clearly seen from the calculated photonic density of states (PDOS) shown in Figure 1B. These results suggest that long-range order is not a necessary condition for the formation of PBGs.

It is known that the connectivity of inclusions in PCs also plays an important role in the formation of PBGs;[19,20] this should also be the case for APCs. In 2D APCs, it was found

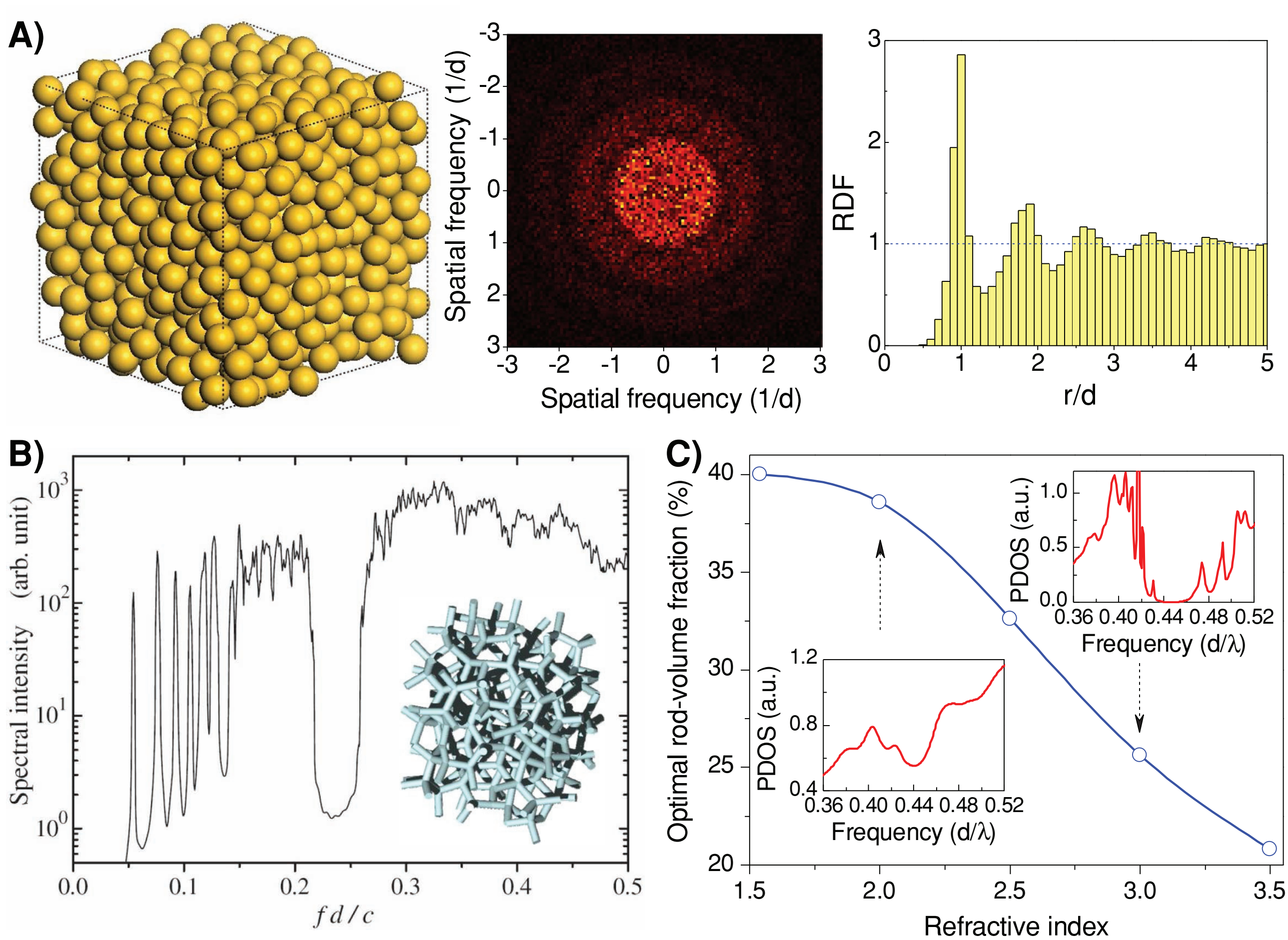

Figure 1. A) Schematic of a 3D APC consisting of random-close-packed spheres (left), its Fourier power spectrum (middle), and radial distribution function (right), where *d* is the sphere diameter. B) Calculated PDOS of a Si-rod-connected amorphous-diamond-structured APC (inset), where *f* is frequency, *d* is the average rod length, and *c* is the speed of light in a vacuum. Reproduced with permission.[13] Copyright 2008, The American Physical Society. C) Calculated optimal rod-filling fraction for a rod-connected amorphous-diamond-structured APC as a function of the refractive index of the rods. Insets show the PDOS for rods with a refractive index of 2 (lower left) and 3 (upper right). Frequency is in reduced units of d/λ, where *d* is the average rod length and λ is the wavelength. Reproduced with permission.[21] Copyright 2012, Proceedings of the National Academy of Sciences of United States of America.

that connected inclusions are more favorable for opening up a PBG than isolated inclusions for both polarizations.[14] Calculations for 3D APCs, such as jammed (i.e., random-close-packed) spheres, inverted jammed-sphere structures, and tetrahedral networks, also confirm this conclusion.[15]

In addition to structural order and connectivity, other factors such as the refractive index and filling fraction of inclusions may influence PBGs in APCs. For example in a rod-connected amorphous-diamond-structured APC with a small refractive-index contrast, there exists only a photonic pseudogap[21] with nonzero PDOS (Figure 1C), different from a conventional PBG within which the PDOS is zero. Upon increasing the refractive-index contrast, however, there is an interesting transition from a photonic pseudogap to a PBG.

It should be mentioned that photonic pseudogaps or PBGs in APCs are direction-independent, which implies that APCs are isotropic optical media, different from conventional PCs. This difference is because in APCs there is no preferred orientation so that light is scattered evenly in all directions.

2.2. Defect States

If defects are introduced into PCs, defect states may appear which are essential for the control and confinement of light.[22–24] Similarly, one can introduce defects into APCs to obtain defect states. By introducing a defect into a 2D waveguide APC, a defect state residing in the PBG was found experimentally.[25] This defect state is strongly localized. If line defects are introduced in a 2D APC consisting of dielectric cylinders, waveguides and bends can also be formed[9] as in conventional PCs. By removing an arbitrarily selected rod in a Si-rod-connected amorphous-diamond-structured APC, defect states arise within the PBG, offering a light confinement as strong as in conventional PCs.[13]

2.3. Localization

Disorder in electron systems may lead to many exotic emergent phenomena such as Anderson localization.[4] Recently,

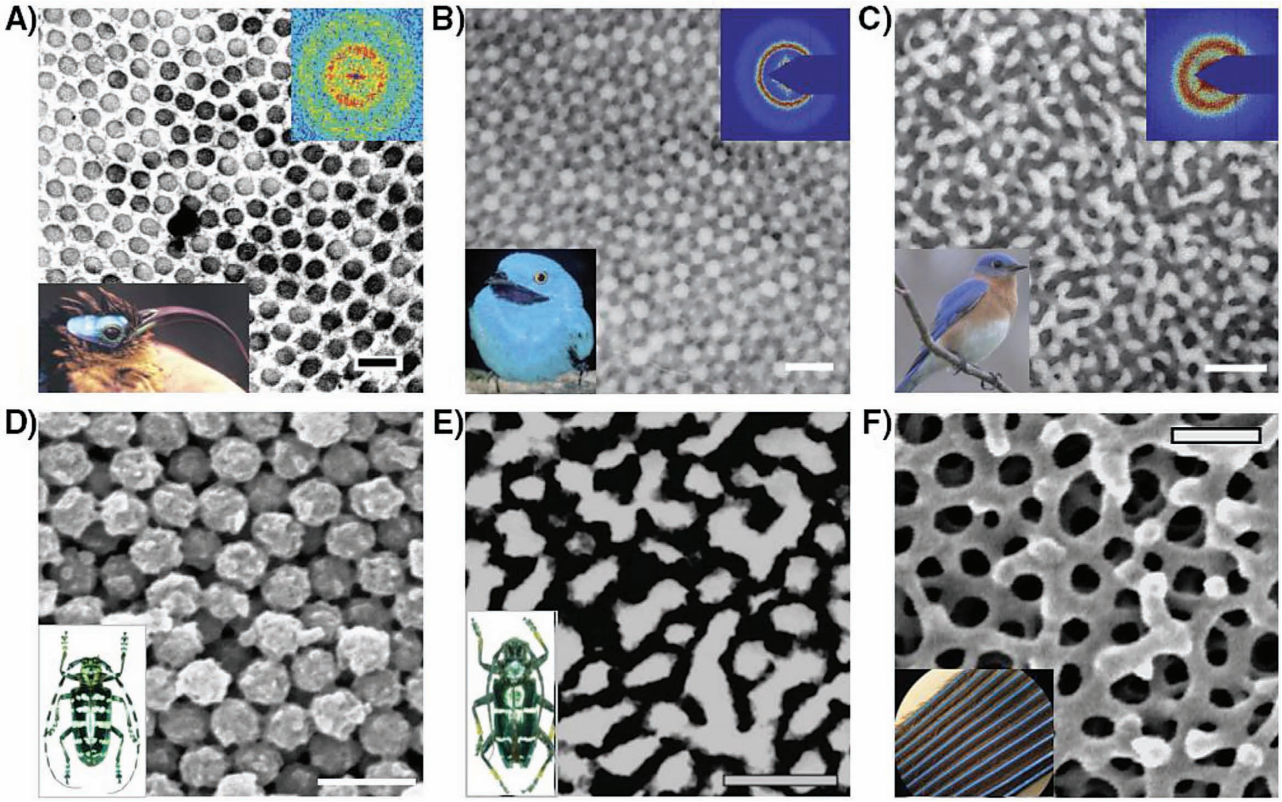

Figure 2. Cross-sectional micrographs of APCs found in the biological world. A) Transmission electron micrograph (TEM) of collagen arrays in the light-blue-colored caruncle of the asity *N. coruscans* (lower left corner). Inset (upper right) shows the corresponding Fourier power spectrum. Reproduced with permission.[33] Copyright 1999, The Company of Biologists Limited. B) TEM of a feather barb of the male plum-throated cotinga (lower left corner). C) TEM of a feather barb of the male eastern bluebird (lower left corner). Insets (upper right corner) in B) and C) show the small-angle X-ray scattering (SAXS) spectra. B) and C) are reproduced with permission.[34] Copyright 2009, The Royal Society of Chemistry. D) Scanning electron micrograph (SEM) of a green scale of the longhorn beetle *A. graafi* (lower left corner). Reproduced with permission.[35] Copyright 2010, The Optical Society of America. E) TEM of a scale of the longhorn beetle *S. mirabilis* (lower left corner). Reproduced with permission.[36] Copyright 2011, The American Physical Society. F) SEM of a blue feather barb of the scarlet macaw. Inset (lower left corner) shows micrograph of the feathers. Reproduced with permission.[21] Copyright 2012, Proceedings of the National Academy of Sciences of the United States of America. Scale bars: A) 200 nm, and B) to F) 500 nm.

much effort has been devoted to disorder-induced light localization and even Anderson localization in optical systems such as APCs.[6,25–28] Anderson localized states that show exponential decay profiles were experimentally demonstrated in 2D disordered photonic lattices.[25,26] Besides the existence of a PBG in a rod-connected amorphous-diamond-structured APC, interesting light diffusion and localization were found.[27] Within the passbands, light propagations are diffusive, which implies weak localization. At the PBG edge, significant light scattering occurs, which suggests the possibility of Anderson localization. Instead of those near the PBG edges, localized states were also found inside photonic pseudogaps in 2D APCs with a low refractive-index contrast.[28]

3. Fabrications, Bioinspiration, and Applications

State-of-art fabrication techniques can be used in the artificial fabrication of APCs. Interestingly, various APCs have been revealed in the biological world from which we may get valuable inspiration. The interesting structural and optical properties of APCs could potentially offer many interesting applications.

3.1. APCs Occuring in the Biological World

Living organisms have already exploited photonic structures to produce striking structural coloration[29] since the Cambrian period. A variety of ordered photonic structures that produce iridescent colors have been revealed, including thin films, multilayers, diffraction gratings, and PCs, found in birds, insects, sea animals, and even in plants.[17,29–32]

In addition to ordered structures in the biological world, there exist APCs that can produce noniridescent structural colors,[21,33–37] as shown in **Figure 2**. The caruncles of some birds show noniridescent blue or green colors, which arise from a 2D APC.[33] Structural characterization revealed that the dermis of the caruncles consists of a thick layer of collagen of a few hundred microns and the parallel collagen fibers form a quasiordered array (Figure 2A).

In the plum-throated cotinga (*Cotinga maynana*), the back feather barbs contain a 3D APC consisting of nearly random-close-packed spherical air cavities (Figure 2B), sometimes with small interconnections.[34] This 3D APC gives rise to a vivid noniridescent turquoise-blue color. A similar 3D APC is also found in the scales of the longhorn beetle *Anoplophora graafi*

(Figure 2D).[35] Lying lapped over each other in regular order on the beetle elytra (anterior wings), each needlelike scale has a distinct color and the scale colors vary from blue, green, yellow, to red. Structural characterization revealed that the scales possess a 3D APC consisting of random-close-packed chitin nanoparticles. Different scale color is due to different chitin-nanoparticle sizes, eventually leading to bright greenish white lateral stripes on the elytra by color mixing.

In the eastern bluebird (*Sialia sialis*), on the other hand, a spinodal-decomposition-like APC was found in the back feather barbs,[34] which leads to a noniridescent blue structural color (Figure 2C). A similar structure was also found in the scales of the longhorn beetle *Sphingnotus mirabilis* (Figure 2E).[36] This spinodal-decomposition-like APC can produce a noniridescent shallow cyan color.

PCs with a diamond symmetry were demonstrated to be the most promising candidates to open up a large PBG, among which the rod-connected diamond-structured PCs offer the champion PBGs.[20] Rod-connected amorphous-diamond-structured APCs also offer excellent PBGs.[13,15,21] Interestingly, such APCs exist already in the biological world, found in the blue feather barbs of the scarlet macaw as shown in Figure 2F.[21]

Noniridescent structural coloration by APCs has fascinated scientists for a long time. Incoherent scatterings from individual scatterers such as Rayleigh and Tyndall or Mie scatterings were hypothesized over 100 years ago. But experimental results such as reflection spectra cannot be explained based on incoherent scattering. The hypothesis of coherent scatterings was proposed later. Fourier power analyses (inset of Figure 2A) and SAXS (insets of Figures 2B and 2C) revealed the short-range order of natural APCs, confirming the coherent-scattering hypothesis.[34,37] Recent calculations of the PDOS of APCs such as rod-connected amorphous-diamond-structured,[21] random-close-packed,[35] and spinodal-decomposition-like APCs[36] uncovered the ultimate origin of the noniridescent structural coloration, namely, from isotropic photonic pseudogaps. Due to the depletion of PDOS, strong reflections for wavelengths within photonic pseudogaps are expected, which lead to noniridescent structrual colors.

3.2. Artificial Fabrication

To fabricate APCs, both top-down and bottom-up methods have been used. With top-down methods such as lithography, 2D APCs can be fabricated as for conventional 2D PCs. However, top-down methods have limitations on the fabrications of 3D APCs especially in the visible and near-infrared regimes. Bottom-up methods and replications from templates have thus frequently been used to fabricate 3D APCs.[38–43]

One of the most commonly used bottom-up methods is the self-assembly of colloids in suspensions. Based on phase-transition behaviors in colloidal systems, amorphous soft glassy colloidal gel with only short-range order was successfully fabricated.[38–40] Obtained samples show homogeneous and angle-independent structural colors as shown in the upper panel of **Figure 3**A. When the concentration of monodisperse colloids is low in a liquid suspension, the colloids can move freely, forming a fluidlike state (B and C bottles in the lower panel of Figure 3A). When the colloidal content increases, the electrostatic interactions among colloids increase and the suspension forms an equilibrium crystalline state. As the concentration increases further and exceeds a certain critical point, the viscosity of the suspension is so large that the relaxation time for forming crystalline arrangements approaches infinity. At this stage, the system forms a metastable glassy state and the colloidal arrangements possesses only short-range order (D, E, and F bottles in the lower panel of Figure 3A).

Artificial APCs composed of dried colloids were also successfully fabricated by mixing two sizes of colloids which can suppress the tendency for formation of long-range order during the self-assembly process,[41,42] as shown in Figure 3B. Obtained samples of the bidisperse dried colloids show noniridescent structural colors.[41] The measured SAXS spectrum displays ringlike profiles, which indicate short-range order of the obtained APC samples.

Another self-assembly method, i.e., phase separation, may also be used to fabricate APCs.[44] Basically, there are two types of phase separation: nucleation and growth, and spinodal decomposition. In an immiscible polymer blend, for example, phase separation via nucleation and growth may give rise to a morphology of isolated sphere droplets dispersed in a matrix. In contrast, spinodal decompostion may lead to a co-continuous morphology consisting of continuous and interconnected phases. Nature may already adopt such phase separation to produce 3D APCs. Based on the morphological similarities, it was conjectured[34] that random-close-packed and disordered bicontinuous 3D APCs found in some bird feather barbs may be self-assembled by phase separation, driven by the polymerization of keratin from the cellular cytoplasm via nucleation and growth, and spinodal decompostion.

As shown in the previous subsection, nature provides delicate APCs which can be served as templates to convert into inorganic structures. In the peach-faced lovebird (*Agapornis Roseicollis*), the blue feather barbs possess a 3D APC consisting of disordered bicontinuous random network of keratin backbones. By using the feather barbs as hard templates, SiO_2 and TiO_2 3D APCs were replicated with a sol-gel method,[43] as shown in Figure 3C. Inverted SiO_2 and TiO_2 APCs display bright noniridescent structural colors. The corresponding Fourier power spectra display a uniform ringlike pattern, which implies short-range order of the fabricated oxide structures. Faithful replication was further confirmed by SEM observations.

3.3. Applications

As discussed previously, APCs possess many interesting and unique optical and structural features. A variety of novel applications such as in photonics, displays, solar energy, textiles, and paint can be envisioned.

One of the interesting optical properties of APCs is the production of noniridescent structural colors. Obviously, APCs can be a new kind of colorant, potentially used, for example, in cosmetics, paint products, printing, and fabrics.[45–47] Over conventional pigment-based colorants, APCs are superior in many aspects such as high brightness and saturation, colorfastness if structures remain unchanged, tunability of hue if

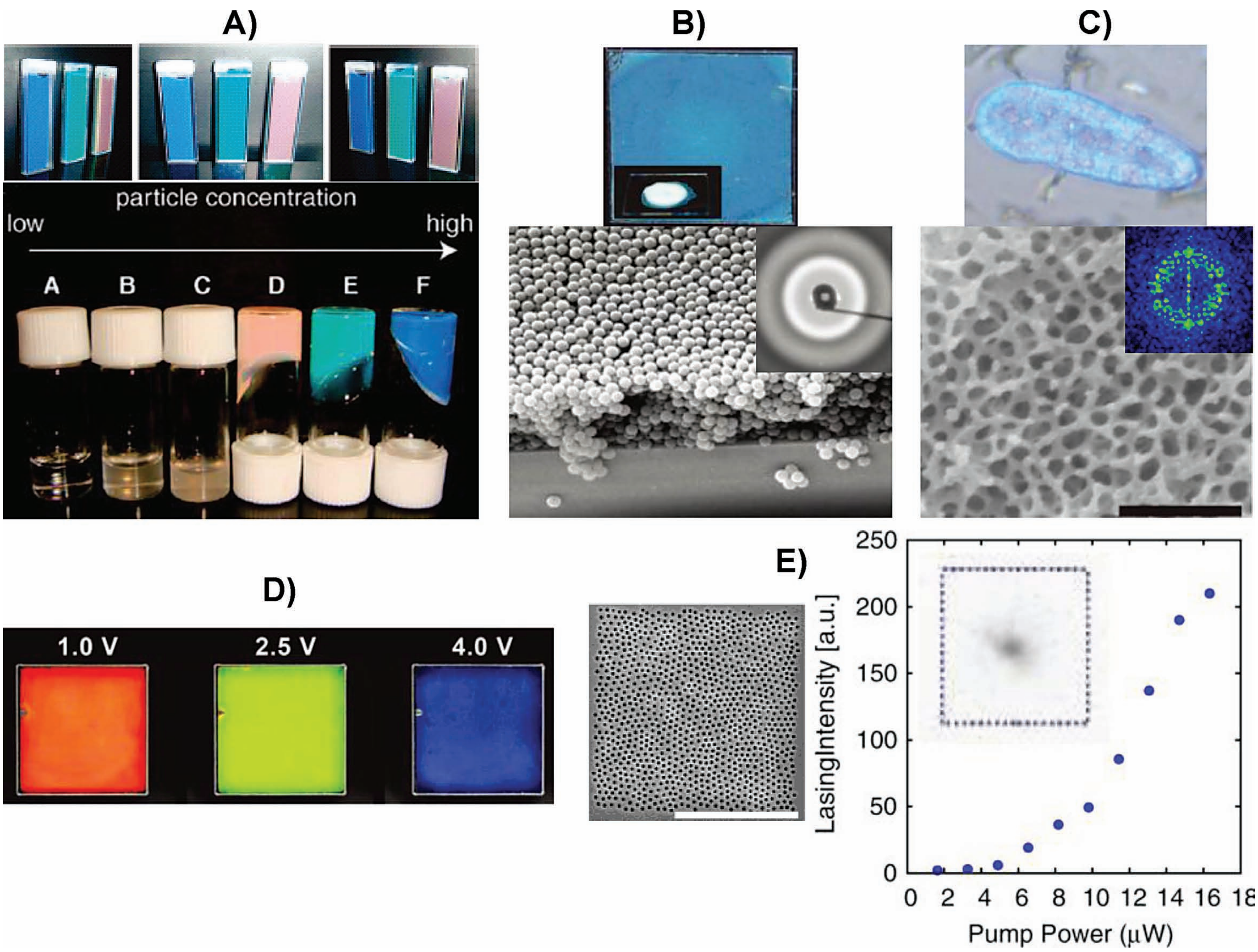

Figure 3. A) Upper panels, photographs of gel colloidal suspensions with different polymer contents at different viewing angles. Reproduced with permission.[38] Copyright 2009, American Chemical Society. Lower panel, photograph of soft glassy colloidal gel with different colloidal concentrations. Reproduced with permission.[40] Copyright 2010, American Chemical Society. B) Upper panel, photograph of a dried colloidal APC film with mixed 226- and 271-nm colloids. Lower panel, SEM of the APC film. Inset shows the SAXS spectrum. Reproduced with permission.[41] C) Upper panel, microphotograph of the transverse cross-section of a SiO_2 disordered bicontinuous APC, replicated from the feather barbs of the peach-faced lovebird. Lower panel, SEM of the fabricated SiO_2 APC. Scale bar, 1 μm. Inset shows the corresponding 2D Fourier power spectrum. Reproduced with permission.[43] Copyright 2010, The Royal Society of Chemistry. D) Photographs of electrically tunable structural-color display pixels at different applied voltages. Reproduced with permission.[49] E) Left panel, SEM of a GaAs film perforated with an amorphous array of holes for lasing action. Scale bar, 5 μm. Right panel, measured emission intensity at 930 nm as a function of the pump power. Inset shows a photograph of the lasing mode image. Reproduced with permission.[53] Copyright 2011, American Physical Society.

refractive-index environments or structures are changed, and environmental-friendliness since no toxic chemicals are needed.

If structural colors produced by APCs can be tuned dynamically in the visible spectrum by some external measures, these tunable APCs can be used as photonic pixels for display.[48,49] Inspired by structural coloration in natural APCs, electrically tunable full-color display pixels were obtained based on 3D APCs composed of a $Fe_3O_4@SiO_2$ core-shell colloidal suspension,[49] as shown in Figure 3D. Pixel colors can be switched very fast by applying an electrical voltage due to the sensitive electrophoretic responses of the core-shell colloids.

In addition to their color-related applications, APCs may be used in photonics. By introducing point and line defects in APCs, defect states with frequency residing into PBGs could be attained, leading to microcavities, waveguides, and bends for light-trapping or -guiding.[9,13,25] As shown by calculations,[36] spinodal-decomposition APCs can offer broadband reflection and scattering due to their unique structural properties. These interesting features can be useful in improving the performance of optical devices. For example, polymer films of spinodal-decomposition APCs were demonstrated to be excellent antireflection coating materials.[50] By introducing a spinodal-decomposition-like APC structure, the extraction efficiency of light-emitting diodes can be greatly enhanced over a wide spectral range.[51] APCs can be also used to enhance material absorption as demonstrated in an amorphous Si film.[52] The overall absorption efficiency for the film perforated with an amorphous array of holes was found to be higher than those perforated with periodic and random hole arrays, or a bare film in a wide frequency range. These results suggest that APCs can also be used in solar cells to enhance light absorption.

Due to strong light localization, APCs can be used for lasing. As demonstrated in a GaAs membrane embedded with InAs quantum dots and perforated with an amorphous array of holes, lasing becomes most efficient at certain frequencies due to strongly enhanced light confinement by short-range order,[53] as shown in Figure 3E. This result indicates that lasing in nanostructures can be efficiently improved and manipulated by means of the short-range order.

4. Conclusions

We have reviewed recent progress in the study of APCs, including optical properties, fabrication, bioinspiration, and applications. Although significant progress has been made in recent years, there still remain many interesting problems that need in-depth studies, both theoretically and experimentally. For example, light transport especially in 3D APCs is not fully understood and Anderson localization of light in 3D APCs is not realized in the optical regime. As for fabrication, high-quality 3D APCs in the visible or infrared region are still difficult to obtain, which hinders their applications to a large degree. As used for fabricating 3D PCs,[54] direct laser writing could be tried to fabricate 3D APCs. In certain cases, high-refractive-index 3D APCs are highly desired since they may offer strong light localization. With Si colloids as building blocks,[55] Si-based 3D APCs may be fabricated. Additionally, natural APCs together with their way of light steering may inspire our design and fabrication of APCs.

As shown, APCs possess many interesting and unique optical properties such as isotropic PBGs or photonic pseudogaps, noniridescent structural-color productions, localized states by defects, and localization due to disorder, which result from their unique structural features such as short-range order. These interesting properties demonstrate that APCs are a new kind of optical materials and could have a variety of important applications such as in photonics, color-related technologies, displays, and solar cells.

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