Photonic hypercrystals for control of light–matter interactions

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Photonic crystals (PCs) have emerged as one of the most widely used platforms for controlling light–matter interaction in solid-state systems. They rely on Bragg scattering from wavelength-sized periodic modulation in the dielectric environment for manipulating the electromagnetic field. A complementary approach to manipulate light–matter interaction is offered by artificial media known as metamaterials that rely on the average response of deep-subwavelength unit cells. Here we demonstrate a class of artificial photonic media termed “photonic hypercrystals” (PHCs) that combine the large broadband photonic density of states provided by hyperbolic metamaterials with the light-scattering efficiency of PCs. Enhanced radiative rate (20%–40%) and light outcoupling (100×) from PHCs embedded with quantum dots is observed. Such designer photonic media with complete control over the optical properties provide a platform for broadband control of light–matter interaction.

metamaterial | light–matter interaction | photonic crystal | cavity QED | quantum dots

Until recently, artificial optical media were represented by two different classes of composite materials—metamaterials and photonic crystals (PCs). The former rely on the averaged polarization of deep-subwavelength-sized unit cells (metaatoms) to control the electromagnetic response of the composite. In contrast, PCs rely on periodic modification of the dielectric constant at the wavelength-sized scale resulting in electromagnetic response arising from Bragg scattering of the propagating electromagnetic field. Very recently, a class of photonic media termed “photonic hypercrystals” (PHCs) was proposed. PHCs do not belong to either of the aforementioned classes as they have unit cells that are subwavelength in dimension and yet their electromagnetic response is qualitatively different from the expected average behavior seen in metamaterials. This fundamental difference results in a number of nontrivial electromagnetic properties of the hypercrystals such as the simultaneous enhancement of spontaneous emission rate and outcoupling over wide spectral bandwidth, to Dirac physics and singularities which are markedly distinct from realizations using macroscopic optical systems such as resonators. More recently, passive structures akin to a PHC have been realized via self-assembly and also shown to have magnetic hyperbolic dispersion in optical frequencies. Here we report the realization of an active PHC embedded with colloidal quantum dots (QDs) that show both enhanced spontaneous emission rate (20×) and outcoupling (100×) arising from the hyperbolic dispersion of the metamaterial and enhanced light outcoupling due to the 2D lattice structure.

Metamaterials designed to have hyperbolic dispersion have been used extensively to control spontaneous emission (6–10) and are subsequently known as hyperbolic metamaterials (HMMs). These metamaterials are characterized by the hyperbolic shape of their isofrequency surface due to the emergence of high-momentum wave-vector states known as high-κ modes (11). An artificial material supporting high-κ modes can be realized using alternating metal/dielectric layers with deep-subwavelength thickness (11, 12). A schematic of such an HMM composed of silver (Ag) and alumina (Al₂O₃) is shown in Fig. 1A. An overview of the modal dispersion in Fig. 1B for a seven-period structure. The system possesses high-κ plasmonic bands for κ₅ > 1.10⁷m⁻¹, as well as two surface-plasmon-polariton (SPP) modes close to the ω = cκ light line (dashed). The existence of high-κ modes provides multiple decay channels for dipole emitters placed inside or on top of the HMM, thereby enhancing the local photonic density of states (LPDOS) and increasing the rate of spontaneous emission (13). Due to the nonresonant nature of the LPDOS enhancement, HMMs are an ideal platform for applications that require broadband control of light–matter interaction.

Despite the attractiveness of the aforementioned features, the use of HMMs in realizing practical devices has been hindered due to the fact that the high-κ modes lie below the light line; hence, the coupled radiation cannot propagate to the far field and is eventually dissipated through ohmic loss. The situation is demonstrated in Fig. 1C by the high reflectivity of the metamaterial, where the simulated reflectivity of a plane wave is plotted as a function of frequency and source angle. It is seen that HMM structures are omnidirectionally highly reflective, indicating that no coupling to high-κ modes can be achieved from free space, or reciprocally, high-κ modes cannot propagate into free space. Indeed, for embedded dipole emitters, typically less than 1% of the total power can escape to the far field. To alleviate this issue, different approaches such as diffraction gratings and nanopatterning have been used to achieve moderate enhancement in light extraction/coupling efficiency from light emitters embedded in HMMs (14–17).

Significance

Light–matter interaction lies at the heart of several fundamental phenomena and technological applications ranging from photovoltaics to lasers. Current approaches to control this interaction such as optical cavities, photonic crystals, and metamaterials either rely on frequency resonance mechanisms which limit the bandwidth or suffer from poor light-coupling issues. Here we report a class of artificial media: photonic hypercrystals to control light–matter interactions. Both bandwidth and outcoupling limitations are overcome using hypercrystals. This characteristics is demonstrated through simultaneous enhancement of spontaneous emission rate (20×) and outcoupling (100×) from quantum dots embedded in the hypercrystal. This platform for broadband control of light–matter interaction will push the boundaries of applications such as ultrafast light-emitting diodes, photovoltaics, and quantum informatics.

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In the present work, we use the PHC concept to exploit LPDOS enhancement offered by the hyperbolic dispersion while simultaneously enhancing light outcoupling as well. A schematic of a 2D PHC is shown in Fig. 1D, where a periodic subwavelength lattice of holes is patterned onto an HMM. The periodicity folds the high-\( k \) modes inside the light line, as shown through finite difference time domain (FDTD) simulations in Fig. 1E. This enables efficient outcoupling of the high-\( k \) states that were previously below the light line. The corresponding reflectivity map (Fig. 1F) of the PHC now shows multiple regions with near-zero reflectivity, indicating broadband and wide-angle light coupling to the structure. It is the complex interaction between multiple folded modes in the PHC that results in the enhanced outcoupling that is observed in this paper. Equally important is the fact that the LPDOS in the HMM is not significantly altered by the structural modification. This is shown in SI Appendix, section 3 through 3D FDTD calculations for the Purcell factor of an emitter embedded in PHC and HMM.

To optimize the design of a PHC for outcoupling of light from the embedded quantum emitters, 2D and 3D FDTD simulations are used (see Methods for details). A parametric sweep is performed over the pitch and lattice constant of the PHC with the goal of maximizing the output power for a dipole embedded inside the HMM. Fig. 2A shows a color map which plots the portion of outcoupled power into a 60° half-angle (corresponds to 0.8-N.A.) objective used in the experiments) for a wavelength of 630 nm, which corresponds to the emission peak of the QDs used in the experiment (see SI Appendix, section 1, QD emission spectrum). We see that for this wavelength, maximum outcoupling is achieved for lattice spacing of \( 280-300 \) nm and pitch of 0.42, corresponding to a hole radius of \( \sim 60 \) nm. In Fig. 2B we show the wavelength-resolved outcoupling contrast ratio for PHCs with \( a = 280 \) and 370 nm and a pitch of 0.42. The outcoupling contrast is simply defined as the power transmitted to the far field by an emitter embedded in PHC versus the same emitter embedded in HMM. We observe that for a wide spectral range between 575 and 700 nm the outcoupling contrast exceeds 100.

The HMM structure used in the present work consists of seven periods of Ag (\( \varepsilon < 0 \)) and Al\(_2\)O\(_3\) (\( \varepsilon > 0 \)) with average thickness of 15 nm with colloidal CdSe/ZnS QDs embedded inside the fifth dielectric layer. Very thin germanium (~1 nm) is used as a seed layer before the deposition of Ag for achieving ultrasmooth, consistent thin films (18). In Fig. 3A a cross-sectional transmission electron microscope image of the structure shows the alternating layers along with the embedded QD layer. The effective dielectric constants of this structure are shown in SI Appendix, Fig. S1A. The emission wavelength of the QDs (630-nm center of emission) is specifically chosen to be in the hyperbolic dispersion range (SI Appendix, Fig. S1B). The location of the QD layer has been carefully selected to provide maximum coupling to plasmonic modes while keeping a distance of \( \sim 6 \) nm from the nearest metallic layer to avoid quenching (13). To realize the PHCs, focused ion-beam milling is used to etch hexagonal arrays of holes into the HMM structure with varying lattice constant \( a \) and radii \( r \). A scanning electron microscope image of the array of PHCs is shown in Fig. 3B along with a magnified image of one of the PHCs (Inset). The holes are milled through the top two silver layers and terminated before the QD layer.

Fig. 4A shows an angle-resolved white-light reflectivity image obtained by Fourier space (\( k \)-space) imaging (details in SI Appendix, section 4) of the PHC with \( a = 280 \) nm and \( r = 80 \) nm. The reflectivity is normalized with respect to the HMM substrate using the following formula:

\[
R_p = \frac{(R_{\text{HMM}} - R_{\text{PHC}})}{R_{\text{HMM}}}. \quad [1]
\]

\( R_p \) is termed partial reflectivity, \( R_{\text{HMM}} \) is reflectivity from HMM, and \( R_{\text{PHC}} \) is reflectivity from PHC. We can see that the response...
of the 280-nm-period PHC (Fig. 4A) extends in a narrow range of angles from 580 to 750 nm allowing for increased outcoupling in that range. The range overlaps the emission spectrum of the embedded QDs. The larger-period PHCs do not exhibit this narrow band of transmission (see SI Appendix, section 3 for k-space image of larger period and real-space reflectivity graphs).

Photoluminescence (PL) measurements from the active PHCs were carried out using a home-built confocal microscope for fluorescence-lifetime imaging microscopy (FLIM), a technique which allows simultaneous mapping of intensity and lifetime for every pixel of the fluorescence image, thus providing spatial, temporal, and steady-state emission properties of the active PHC structure. Shown in Fig. 4B is a PL intensity image of the array of PHCs. We see a clear dependence of the emission intensity on lattice constants and radii of the holes with the maximum emission intensity observed for \( a = 280 \text{ nm} \) and radius \( r = 80 \text{ nm} \). Fig. 4C shows the steady-state emission spectra of the HMM and the PHC. A 100x enhancement factor in emission intensity is observed due to the outcoupling of the high-\( k \) states by the PHC structure. Another important attribute is that the outcoupling efficiency is a broadband effect. Unlike a conventional PC defect cavity that relies on narrow-band resonance, the PHC enhances and outcouples the entire spectral range of the QDs.

Shown in Fig. 4D are the time-resolved emission traces for QDs on glass compared with QDs in HMM and PHC. The spontaneous emission lifetimes of QDs in HMM and PHC are strongly modified with respect to QDs on glass by factors of 9 and 20, respectively. The reason for the observed difference between PHC and HMM is that the outcoupled light in PHC is preferentially composed of emission from the vertically oriented dipoles of the QD film which are better coupled to the transverse magnetic polarized high-\( k \) modes and therefore experience larger enhancement and better out-coupling to the far field.

To further substantiate that the collective modes of the hypercrystal are responsible for the enhanced emission from the PHC, we investigated structures where the lattice spacing was large enough (600 nm) for each of the holes to act as an individual scatterer. This structure shows a small enhancement factor of 7x as shown in Fig. 5A, in contrast to the PHC structure with lattice constant of 280 nm where strong coupling between the fields around the nanoholes leads to outcoupling efficiency.
of 100× (Fig. 5B). The result is consistent with our simulations (Fig. 1E) showing the appearance of coupled modes in the PHC structure which are responsible for the large outcoupling enhancement.

In summary, we have demonstrated a class of artificial photonic medium: the PHC. We show how one can independently engineer the LPDOS using the hyperbolic dispersion of the metamaterial and the light outcoupling using subwavelength-sized
lattice geometry. Enhancement in light extraction by a factor of 100 and spontaneous emission rate by a factor of 20 is seen from QDs embedded in the PHC structure. Furthermore, our PHC structures show broadband enhancement in light coupling and spontaneous emission rate, thus providing an ideal platform for applications that require nonresonant control over light-matter interaction such as in solar cells, ultrafast light-emitting diodes, and single-photon sources.

**Methods**

**Fabrication.** HMM substrates were grown by a Kurt Lesker PVD 75 electron-beam evaporation system on top of precleaned glass substrates with surface roughness <1 nm. The pressure inside the vacuum chamber was kept to ∼5 × 10⁻⁶ torr throughout the process. As a part of the calibration and characterization process, thin films of Al₂O₃ and Ag with Ge seed were grown and optically characterized by a J.A. Woollam M-2000 ellipsometer. The measured optical constants (included in SI Appendix) were used in our analytical calculations and FDTD simulations.

**PL and Lifetime Measurements.** Intensity and lifetime measurements were taken on a modified Olympus IX-81 microscope using an FLIM technique. The QDs are pumped by a 440-nm pulsed diode laser at 40-MHz repetition rate with 90-ps pulse width. The emission from the QDs is spectrally separated from the laser by a Semrock RazorEdge 532 long-pass filter and detected by an Avalanche photodiode coupled to a PicoHarp 300 time analyzer. This setup allows us to detect the intensity and the lifetime at any spot on the sample. The sample is mounted on a piezo-controlled stage with a maximum scan area of 80 μm × 80 μm. SymphoTime 64 software is used to obtain intensity and lifetime maps of the sample.

**FDTD Simulations.** Simulations model the quantum emitter as an electric dipole oriented parallel or perpendicular to the metal-dielectric layers of the HMM/PHC with an emission spectrum extending from 0.3 to 1.2 μm. As a first step to PHC design, 2D FDTD simulations are carried out with a double parameter sweep (hole radius and periodicity) to maximize the outcoupled power for the wavelength corresponding to the emission maximum of the QDs. Full 3D simulations are also carried out to obtain meaningful values of the hole radius and periodicity to ensure a realistic design. The simulations also calculate the Purcell factor of the dipole and the spatial distribution of the electric field, both as a function of wavelength (see SI Appendix, section 3).

For details on bandstructure and reflectivity simulations, see SI Appendix, section 2.

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SUPPORTING INFORMATION

Photonic Hypercrystals: new media for control of light-matter interaction

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S1. Effective permittivity and emission spectrum of CdSe/ZnS core-shell quantum dots

A metal-dielectric stack can be validly approximated by a homogenous anisotropic medium when the unit cell thickness is smaller than $\lambda/10$ and if it consists of at least 4 periods (1). In such case the parallel (x- and y-directions) and perpendicular (z-direction) permittivities of the medium with respect to the layers are given by

$$\varepsilon_x = \varepsilon_y = \rho \varepsilon_m + (1 - \rho) \varepsilon_d, \quad \text{(S1)}$$

$$\varepsilon_z = \frac{\varepsilon_m \varepsilon_d}{\rho \varepsilon_d + (1 - \rho) \varepsilon_m}, \quad \text{(S2)}$$

where $\varepsilon_m$ and $\varepsilon_d$ are the complex permittivities of the metal and dielectric respectively, $\rho$ is the fill fraction of the metal in the structure

$$\rho = t_m / (t_m + t_d) \quad \text{(S3)}$$

and $t_m$ and $t_d$ are the layer thickness of the metal and dielectric layers in a single unit cell respectively. For convenience, since $\varepsilon_x = \varepsilon_y$, we define $\varepsilon_x = \varepsilon = \varepsilon_y$ as the permittivity component any in-plane direction (parallel to the layers). In our fabricated structure $\rho \sim 0.5$ which places the transition point for $\varepsilon_z$ at wavelength $\lambda \sim 380 \text{ nm}$.

Figure S1.a plots the real and imaginary components of the effective permittivity tensor in the xy-plane and the z-direction (see inset for schematic of the structure with direction arrows). The effective permittivity of the metamaterial was calculated using the optical constants of thin film Ag and Al\textsubscript{2}O\textsubscript{3} obtain by ellipsometry measurements (using J.A. Woollam M-2000) on control samples prepared by electro-beam deposition. On the top right a schematic of the iso-frequency contour shows a hyperboloid shape which occurs when, $\varepsilon_{xy} < 0$ and $\varepsilon_z > 0$. The emission of the embedded quantum dots is chosen in order to operate in the deeply hyperbolic regime. In figure S1.b the emission spectrum of the CdSe/ZnS core-shell quantum dots is plotted.
Figure S1. (a) Real and imaginary components of the effective permittivity tensor in the xy and z directions (as seen in schematic). The upper right inset sketches the shape of the iso-frequency contour for $\varepsilon_{xy} < 0$ and $\varepsilon_z > 0$. (b) Emission spectrum of CdSe/ZnS core-shell quantum dots used in the experiments.

S2. FDTD simulations for bandstructure and reflection

FDTD simulations were performed using commercial software Lumerical™. For bandstructure simulations scattered broadband dipole sources are placed inside a corrugated or un-patterned HMM structure surrounded by simulation region with Bloch boundary conditions. Several energy monitors distributed in the simulation region recorded the energy spectrum from a sweep of the wave-vector on the boundary conditions. Fourier transform is performed on the recorded energy spectra to obtain the bandstructure as a function of wave-vector. Figure S2.a shows the simulation region with electric dipoles placed inside a 7P HMM with etched air holes of 300nm period. Figure S2.b shows the bandstructure of the corrugated film as a function of the normalized wave-vector, $k_x/k_0$.

Figure S2. (a) FDTD simulation region. The blue arrows are dipoles positioned inside the HMM. The yellow X’s are time monitors that measure energy as a function of simulation time. The faint grey square is an etched air grating. The distance between the horizontal green lines dictated the periodicity of the structure. (b) The simulated bandstructure of an HMM with 300nm periodic hole array. The hole diameter is 150nm.
The bandstructure simulations were supplemented by reflection simulations for various substrates. In this simulation a plane wave source is placed below the substrate at a given angle and the amount of power reflected from the structure is measured by a power monitor placed behind the plane wave source (see Fig. S3.a). The simulation uses Bloch boundary conditions to match the source angle. One crucial element to consider when running FDTD simulations of this kind is the change in source angle as a function of wavelength \(^6\). Since the source is broadband there’s a variation in the injection angle as a function of wavelength. Higher wavelengths get injected at higher angles than the source angle for the center wavelength of the injected spectrum. If the source is tilted beyond a certain angle, the injection angle for higher frequencies can surpass 90 degrees which results in injection of evanescent waves into the simulation region \(^6\). We avoid this problem by keeping the source angle below that critical angle. By doing this the reflection curve is only obtained up to a certain angle which decreases as a function of wavelength. The evanescent region in our reflection figures is marked by a black semi-circle. In addition to the reflection simulations in the paper body, we present here simulations for a single silver film of 500nm thickness etched with 900nm period (Fig. S3.b) air-grating and 300nm (Fig. S3.d). These simulations were performed as an accuracy check for this method. Overlaid on the two reflection maps is the grating order dispersion for these two patterns, obtained by equating the grating equation to the wave-vector of a surface-plasmon-polariton \((k_{SPP})\).

\[
k_{SPP} = k_0 \sin(\theta) + m \cdot k_g,
\]

where \(k_0 = 2\pi / \lambda\), and \(k_g = 2\pi / a\), \(a\) is the grating period, \(m\) is the grating order. \(k_{SPP}\) was obtained from a simple Drude model for the SPP dispersion.

Figure S3. (a) Reflection simulation region. The source angle is swept from 0 to 45 degrees. The grey rectangle is an etched air hole with 450nm diameter. Periodicity is set to 900nm. (b) Reflection map of air-hole array of 900nm periodicity. Grating orders are overlaid on the image.
No fitting function was applied. (c) Screen grab of simulation with 300nm periodic array, hole diameter of 150nm. (d) Reflection map of air-hole array with 300nm grating. Only the $m=-1$ grating order can couple the SPP mode to free-space.

**S3. FDTD Simulations of Purcell factor and electric field radiation for PHC and HMM**

Enhancement in a hyperbolic medium stems from modifying the effective medium in the vicinity of the dipole emitter which is embedded inside the hyperbolic metamaterial and is an extremely near-field effect. The periodic modulation of the surface which forms the PHC only acts a small perturbation to the effective medium seen by the dipole emitter. Fig. S4a is obtained by a full 3D FDTD simulation of a dipole emitter on top of a PHC or a HMM. As seen in the graph the calculated Purcell factor is nearly the same for both HMM and PHC. Fig. S4b shows the out-coupling effect from a PHC with the correct periodicity for the center emission wavelength of our QDs (635nm). The top panel shows the electric-field emitted by a dipole oriented vertically to the layers embedded in the PHC. A horizontal dipole shows a similar effect with a lesser degree of coupling to high-k modes due to the dipoles primarily TE-polarized emission (2, 3). The vertical patterns seen in the figure are the result of scattering from the holes etched in the HMM. It is an interference effect from a single coherent source. This effect cannot be observed in experiments due to the incoherent nature of spontaneous emission of a dipole ensemble. The bottom panel shows the same dipole in a HMM. Notice most of the radiation remains trapped inside the metamaterial.

![Figure S4](image.png)

Figure S4. (a) Purcell factor as a function of wavelength for HMM (black), PHC (red). (b) TOP: electric field emitted by a vertical dipole embedded in a PHC with $a=280\text{nm}$ periodicity; BOTTOM: electric field emitted by a vertical dipole embedded in a HMM at 635nm.
S4. k-space and real-space reflection measurements for PHCs

Fourier space (k-space) images of white-light reflection from the PHCs are taken by projecting the back plane of the microscope objective lens onto the monochromator with a CCD. The k-space is imaged on 1024 x 1024 pixels CCD camera (Pixis 1024b) installed on a Princeton Instruments monochromator which allows us to resolve one of the k-space axis (x or y) as a function of wavelength.

Figure S5a, b show a k-space image in the y plane of two PHCs with the same hole radius (80nm) and different periodicity \( a = 280nm \) and \( a = 370nm \) respectively. Both k-space images show reduced reflection at 450-575nm range however this is an artifact of the normalization of the measurement rather than an indication of in-coupling to modes in the material. The metamaterial is composed of silver layers, a material which becomes exponentially transparent to light in the 500nm-400nm band (see paper Fig. 1c). The PHC reflection is normalized with respect to the reflection of the HMM according to the following formula:

\[
R_p = \frac{(R_{\text{HMM}} - R_{\text{PHC}})}{R_{\text{HMM}}}
\]  

(S5)

We must keep in mind that the PHC is achieved by removing material from the first two periods of the HMM and therefore contains a lesser amount silver than the HMM which makes it more transparent in this wavelength range. This is apparent in the normalized k-space images. The region of lower reflection seen in Fig. S5b is due to increased transmission in this range and not due to in-coupling. In contrast in Fig. S5a we see a clear dependence on angle for the wavelength range between 600-800 nm which includes the emission range of the QDs. Other PHCs show similar k-space images which gradually transitions from the image shown in Fig. S5a to the one shown in Fig. S5b with increasing period.

The relationship between out-coupling efficiency and reflection of the PHCs is demonstrated in Fig. S5c,d. In Fig. S5a we plot the real-space reflection of all the PHCs with hole radius of 80nm. Real-space reflection is taken by projecting the image-plane of a desired PHC onto the entrance slit to a spectrometer (effectively integrating over all angles in the k-space image). We see that in the range of emission of the QDs (635nm ± 20nm) each PHC has a different reflection with the lowest one belonging to the PHC with periodicity, \( a = 280nm \). Notice that this is a broadband effect which cannot be achieved with a regular photonic crystal defect cavity. Fig. 5b shows the emission intensity from the array of PHCs along a cutline through the center of all PHCs (taken from paper Fig. 4b). The intensity relations between different PHCs match the relations in the reflection graph. \( 280 > 255 > 335 > 305 > 370 \).
Figure S5. White-light k-space reflection measurements of (a) PHC with $a = 280\,nm$ and (b) PHC with $a = 370\,nm$. (c) Real-space reflection measurements for row of PHCs with radius 80nm and varying periodicity (labeled on graph). (d) Intensity measured along a cut-line of the photoluminescence (PL) intensity map shown in paper in Fig. 4b. Both out-coupled intensity and in-coupled white light in reflection measurement have a one-to-one correspondence in that highest PL intensity corresponds to lowest reflection of a PHC.

REFERENCES

