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14. ABSTRACT We report the realization of a new class of artificial media termed Photonic Hypercrystals that combine the large photonic density of states in hyperbolic metamaterials with the light scattering efficiency of photonic crystals. This new class of artificial photonic media is used to demonstrate enhancement in spontaneous emission rate by 20x and out coupling enhancement by 100x from quantum dots embedded inside the medium. Furthermore, we also show enhancement in spontaneous emission from two-dimensional semiconductors placed in the near field of the hypercrystal. This new class of artificial media overcomes the major issue of out coupling from hyperbolic media.					
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Report Title

Final Report: ENHANCED LIGHT EMITTERS BASED ON METAMATERIALS

ABSTRACT

We report the realization of a new class of artificial media termed Photonic Hypercrystals that combine the large photonic density of states in hyperbolic metamaterials with the light scattering efficiency of photonic crystals. This new class of artificial photonic media is used to demonstrate enhancement in spontaneous emission rate by 20x and out coupling enhancement by 100x from quantum dots embedded inside the medium. Furthermore, we also show enhancement in spontaneous emission from two-dimensional semiconductors placed in the near field of the hypercrystal. This new class of artificial media overcomes the major issue of out-coupling from hyperbolic media and thus presents a viable approach towards using the large density of photonic states in hyperbolic media for practical applications such as ultrafast LEDs.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
04/01/2017	5 Tal Galfsky, Zheng Sun, Christopher R. Conside, Cheng-Tse Chou, Wei-Chun Ko, Yi-Hsien Lee, Evgenii E. Narimanov, and Vinod M. Menon. Broadband Enhancement of Spontaneous Emission in Two-Dimensional Semiconductors Using Photonic Hypercrystals, Nano Letters, (): 4940. doi:
04/01/2017	1 T. GALFSKY, H. N. S. KRISHNAMOORTHY, W. NEWMAN, E. E. NARIMANOV, Z. JACOB AND V. M. MENON. Active hyperbolic metamaterials: enhanced spontaneous emission and light extraction, Optica, (01 2015): 0. doi: 10.1364/OPTICA.2.000062
04/01/2017	6 Tal Galfsky, Jie Gu, Evgenii Narimanov and Vinod Menon. Photonic hypercrystals: New media for control of light matter interactions, Proceedings of the National Academy of Science, (): . doi:
04/04/2017	7 Tal Galfsky, Zheng Sun, Zubin Jacob, and Vinod Menon. Preferential emission into epsilon near zero metamaterial, Optics Materials Express, (): 2878. doi:
TOTAL:	4

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

1. Photonic hypercrystals: new media for control of light-matter interaction, Tal Galfsky, Evgenii Narimanov, and Vinod Menon, SPIE Nanoscience and Engineering, August 23, 2016.

Number of Presentations: 1.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
04/04/2017	10 Tal Galfsky, Evgenii Narimanov and Vinod Meno. Enhanced spontaneous emission in photonic hypercrystals, Frontiers in Optics. 19-OCT-15, San Jose, California. : ,
04/04/2017	8 Tal Galfsky, Evgenii Narimanov, and Vinod Meon. Photonic hypercrystals for controlled enhancement of radiation from quantum emitters, CLEO: QELS_Fundamental Science. 06-JUN-16, San Jose, California. : ,
04/04/2017	9 Tal Galfsky, Zheng Sun, Evgenii Narimanov, and Vinod Menon. Broadband enhancement of light-matter interaction in 2D semiconductors by photonic hypercrystals, CLEO: Science and Innovations. 06-JUN-16, San Jose, California. : ,
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Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

<u>Received</u>	<u>Paper</u>
04/04/2017	3.00 Zheng Sun, Tal Galfsky, Zubin Jacob, and Vinod Menon. Preferential emission into epsilon-near-zero metamaterial, CLEO: QELS_Fundamental Science. 11-MAY-15, San Jose, California. : ,
04/04/2017	4.00 Tal Galfsky, Ward Newman, Zubin Jacob, Evgenii Narimanov, Vinod Menon. Simultaneous enhancement of decay rate and light extraction from active hyperbolic metamaterial, CLEO: QELS_Fundamental Science. 11-MAY-15, San Jose, California. : ,
TOTAL:	2

TOTAL:**TOTAL:****TOTAL:**

Ultrafast LED based on hyperbolic metasurface

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Tal Galfsky	1.00	
FTE Equivalent:	1.00	
Total Number:	1	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	
Jared Day	0.50	
FTE Equivalent:	0.50	
Total Number:	1	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Vinod Menon	0.90	
FTE Equivalent:	0.90	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Charles Cohen	0.00	Physics
FTE Equivalent:	0.00	
Total Number:	1	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 1.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 1.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 1.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:..... 0.00

Names of Personnel receiving masters degrees

<u>NAME</u>
Total Number:

Names of personnel receiving PhDs

<u>NAME</u> Tal Galfsky Total Number:	1
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Names of other research staff

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

5 Ultrafast LED based on Hyperbolic Metasurface (HMS ULED)

Patent Filed in US? (5d-1) Y

Patent Filed in Foreign Countries? (5d-2) N

Was the assignment forwarded to the contracting officer? (5e) N

Foreign Countries of application (5g-2):

5a: Tal Galfsky

5f-1a: CUNY

5f-c: 160 Convent Ave.

New York NY 10031

5a: Vinod Menon

5f-1a: CUNY

5f-c: 160 Convent Ave.

New York NY 10031

Scientific Progress

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Technology Transfer

None

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Agreement Number:	W911NF1510019
Proposal Title:	ENHANCED LIGHT EMITTERS BASED ON METAMATERIALS
Report Period Begin Date:	12/08/2014
Report Period End Date:	07/07/2016

Abstract: We report the realization of a new class of artificial media termed **Photonic Hypercrystals** that combine the large photonic density of states in hyperbolic metamaterials with the light scattering efficiency of photonic crystals. This new class of artificial photonic media is used to demonstrate enhancement in spontaneous emission rate by 20x and out coupling enhancement by 100x from quantum dots embedded inside the medium. Furthermore, we also show enhancement in spontaneous emission from two-dimensional semiconductors placed in the near field of the hypercrystal. This new class of artificial media overcomes the major issue of out-coupling from hyperbolic media and thus presents a viable approach towards using the large density of photonic states in hyperbolic media for practical applications such as ultrafast LEDs.

Introduction: Metamaterials designed to have hyperbolic dispersion have been used extensively to control spontaneous emission over wide spectral bandwidth in a variety of quantum emitters ranging from quantum dots (QDs) to nitrogen vacancy centers in diamond and even to control thermal emission¹⁻⁵. These metamaterials are characterized by the hyperbolic shape of their iso-frequency surface due to the emergence of a number of large wave-vector (high- k) states⁶. They can be thought of as extremely anisotropic materials where the dielectric constants along the principal axis have opposite sign. This kind of extreme anisotropy can be designed for a chosen frequency range by selecting materials with negative and positive dielectric permittivity and layering them to form a superstrate where each unit cell is of deep subwavelength thickness^{6,7}. The existence of multiple plasmonic bands provides multiple decay channels for a dipole emitter placed inside or in the near-field of a HMM, thereby enhancing the local photonic density of states (LPDOS) and increasing the rate of spontaneous emission (Fig. 1). Due to the non-resonant nature of the LPDOS enhancement, HMMs are an ideal platform for applications that require broadband control of light-matter interaction such as light emitting diodes, nonlinear optical switches and solar cells.

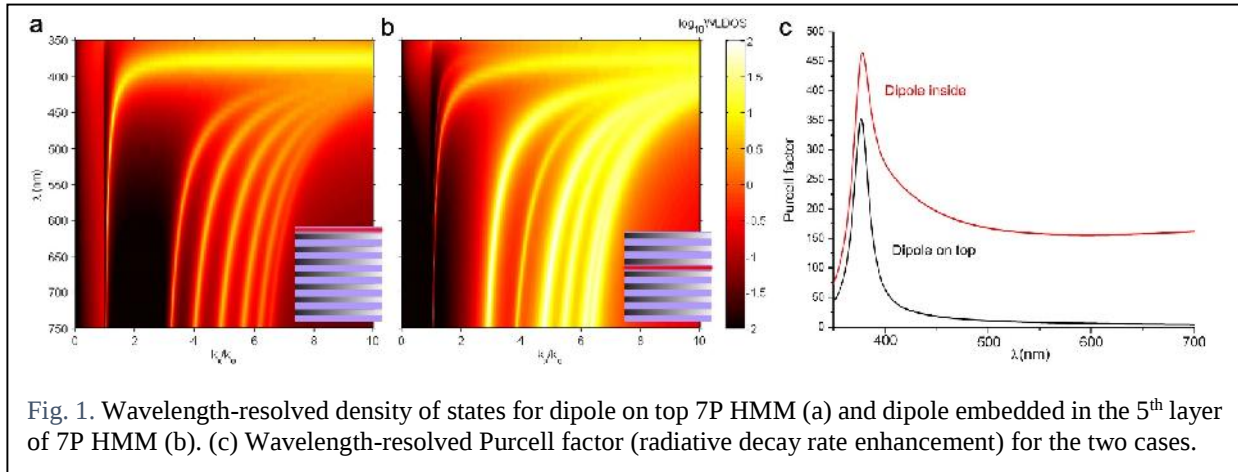


Fig. 1. Wavelength-resolved density of states for dipole on top 7P HMM (a) and dipole embedded in the 5th layer of 7P HMM (b). (c) Wavelength-resolved Purcell factor (radiative decay rate enhancement) for the two cases.

Despite the aforementioned highly attractive feature, the use of HMMs in realizing practical devices has not been easy due to the fact that nearly all the radiation is coupled to the high- k modes that lie below the light line and hence cannot propagate to the far-field and is eventually dissipated through ohmic loss. It is seen that the HMM structures are omni-directionally highly reflective, indicating that no coupling to high- k modes can be achieved from free-space, or reciprocally, high- k modes cannot propagate into free-space without a scattering mechanism. Indeed, for dipole emitters embedded inside the structure typically less than a hundredth of the total power can escape to the far-field⁸. This is a major roadblock for developing practical light emitters based on HMMs. To alleviate this issue, different groups have used approaches such as high-contrast gratings and nano-patterning to achieve modest enhancements in light extraction/coupling efficiency from HMMs embedded with light emitters (active HMMs)^{8–11}. Through a previous ARO grant, we developed the Bulls-Eye grating structure that partially overcame the out-coupling issue⁸.

Project Description: An alternate and highly promising approach to out-couple light from such active HMMs is the use of the recently proposed concept of “photonic-hypercrystals” (PHC) where the large density of states offered by HMMs is combined with the efficient Bragg scattering of high- k modes in photonic crystals¹². Schematic of the two-dimensional hypercrystal is shown in Fig. 2a and consists of a 2D triangular lattice of holes with periodicity designed for the wavelength range of interest. The result of introducing such a lattice periodicity is that the high- k modes fold and lies above the light line as shown in Fig. 2b.

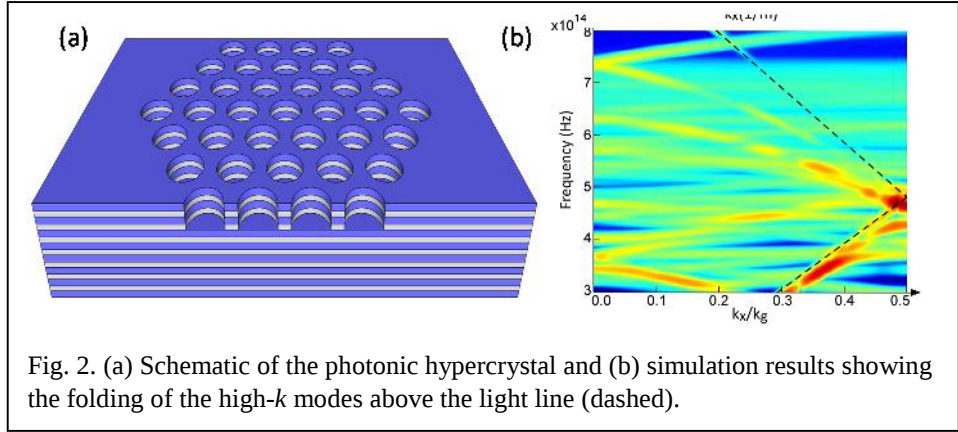


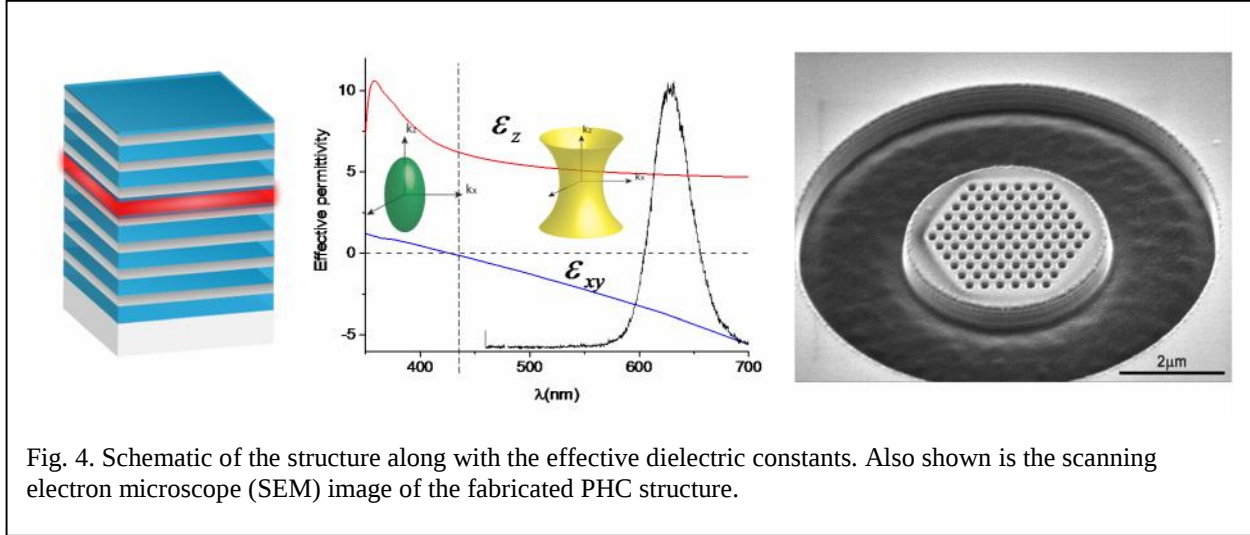
Fig. 2. (a) Schematic of the photonic hypercrystal and (b) simulation results showing the folding of the high- k modes above the light line (dashed).

In addition to the enhancement in out-coupling, the PHC still preserves the increase in the PDOS and hence the Purcell enhancement of spontaneous emission. This was verified through finite difference time domain simulations of Purcell enhancement. As shown in Fig. 3, the Purcell enhancement expected between the HMM and the PHC is comparable. Thus unlike in the microcavity/ photonic crystal cavity where the Purcell enhancement drops significantly when the out-coupling is increased, here we do not see this change because the out-coupling using the 2D lattice is only a small perturbation on the density of states.

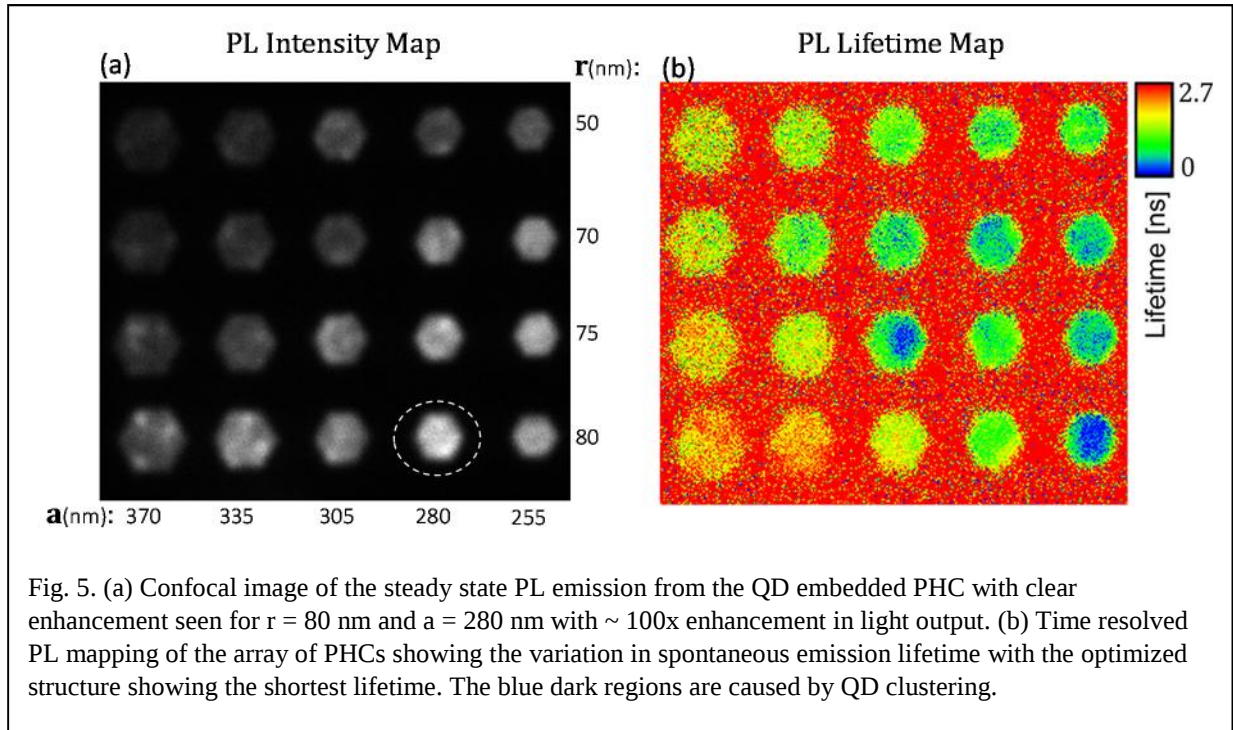
Following the design of the PHC structure, we fabricated a seven period structure consisting of alternating layers of silver (Ag) and Alumina (Al_2O_3) with average thickness of 15 nm and a 2nm thick germanium (Ge) layer acting as the wetting layer for the silver layer deposition. The layers were deposited

Fig. 3. Purcell enhancement as a function of wavelength for the HMM versus the PHC. Clearly the PHC does not distort the Purcell enhancement when compared to the HMM.

using electron beam evaporation. Schematic of the layered structure along with the simulated effective dielectric constant of the 7 period structure is shown in Fig. 4. The emission spectrum of the CdSe/ZnS colloidal core/shell quantum dots used in the experiments is also shown. The structure is designed so that the emission lies in the hyperbolic dispersion regime. The quantum dot layers are embedded inside the structure as shown in the schematic drawing. Also shown in Fig. 4 is a scanning electron microscope image of the fabricated PHC structure. Here following the fabrication of the multilayered structure, the 2D triangular lattice is defined on the structure using focused ion beam (FIB) etching. The holes are milled through the top two periods stopping prior to the quantum dot layer.



Photoluminescence (PL) measurements from the active PHCs were carried out using a home built confocal microscope that allows us to perform fluorescence-lifetime imaging microscopy (FLIM).

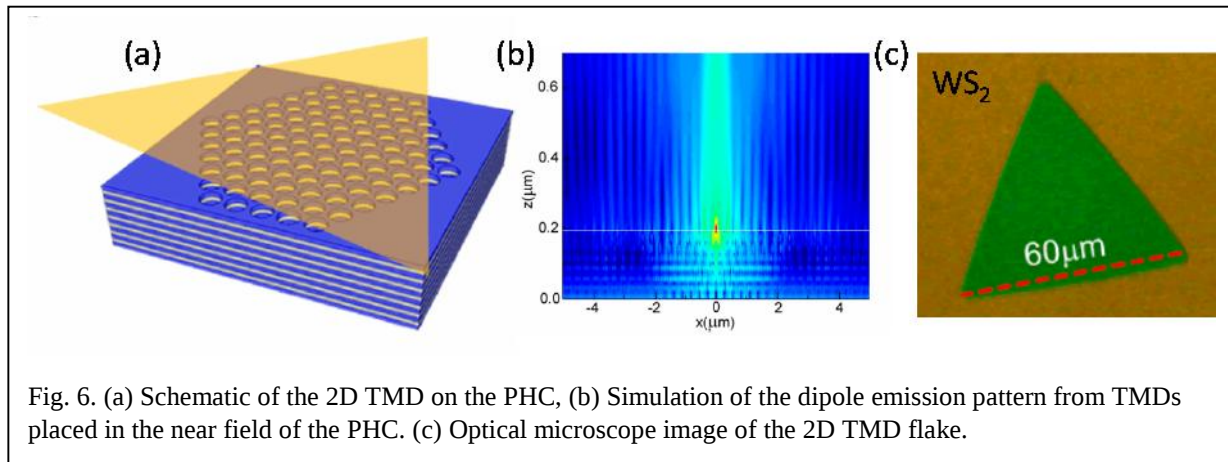


The technique allows simultaneous mapping of intensity and lifetime for every pixel of the fluorescence image thus providing spatial, temporal and steady state emission properties of the active PHC structure. Shown in Fig. 5a is the confocal PL intensity image of the array of PHCs. We see a clear dependence of the emission intensity on radii (r) and lattice constant (a) of the holes with the maximum emission intensity observed for the PHC structure with period, $a = 280$ nm and radius, $r = 80$ nm. Fig. 3b shows the time resolved emission of the QDs from the PHC structure obtained using the FLIM technique clearly showing lowest lifetime (with the exception of clustered region) for the array with same set of radii and lattice spacing. Factor of 100 enhancement in peak emission intensity is observed due to the out-coupling of the high- k states by the PHC structure. This is by far the largest out-coupling enhancement that has been observed from an active HMM with embedded emitters and clearly paves way for realizing practical light emitters having alleviated the issue of low out-coupling that plagues HMM structures.

Modification of spontaneous emission from 2D atomic crystals

Recently, 2D semiconductors of transition metal dichalcogenides (TMDs) have become highly attractive as a new class of optoelectronic material with unprecedented strength in its interaction with light. These TMDs in their monolayer limit become direct band gap and the light emission increases by a factor of three. However a very low quantum yield of $\sim 10^{-2}$ - 10^{-3} at 300K¹³ is a major hindrance in developing practical light emitting devices using these materials. Various approaches using photonic cavities¹⁴⁻¹⁶, plasmonic structures^{17,18} as well as chemical methods¹⁹ have been pursued to enhance the light emission from TMDs. Both photonic and plasmonic approaches rely on frequency resonances and hence are often bandwidth limited while the chemical approach is still in its infancy. Here we demonstrated broadband enhancement of spontaneous emission from two archetypical TMDs, Molybdenum-disulfide (MoS₂) and Tungsten-disulfide (WS₂) monolayers using PHCs that have hyperbolic dispersion.

Schematic of the PHC with the TMD monolayer flake on top (near field) is shown in Fig. 6 along with the simulated emission from in-plane dipole (akin to what is present in 2D TMDs). Also shown in Fig. 6 is an optical microscope image of a large monolayer flake of WS₂ grown via chemical vapor deposition in the group of Yi-Hsien Lee at National Tsing-Hua University, Taiwan.



Following the deposition of the 2D monolayer onto the PHC, we carried out steady state and time resolved PL measurements. Confocal scanning microscopy was used to image the emission intensity from the various locations on the 2D flake placed in the near field of the PHC. A clear bright spot corresponding to the PHC with optimized lattice parameters is shown in Fig. 7a along with the PL emission spectrum from the same monolayer sample placed on top of a HMM structure and on a SiO_2/Si substrate. The FLIM technique was once again used to study the time resolved luminescence properties. Fig. 7c shows the lifetime trace where, trace 1 corresponds to the lifetime of as grown WS_2 (~ 56 ps) and the trace 2 corresponds to that on the PHC structure (~ 7 ps) which is limited by the instrument response function. Once again we see simultaneous enhancement in emission intensity and the decrease in lifetime over the entire emission spectrum indicating the broadband Purcell enhancement of spontaneous emission from 2D materials using PHC. The PL intensity was increased by a factor of 50X compared to that on SiO_2/Si substrate while the lifetime decreased by a factor of ~ 7 . This approach presents a unique way to further the 2D optoelectronics field through the use of hyperbolic media.

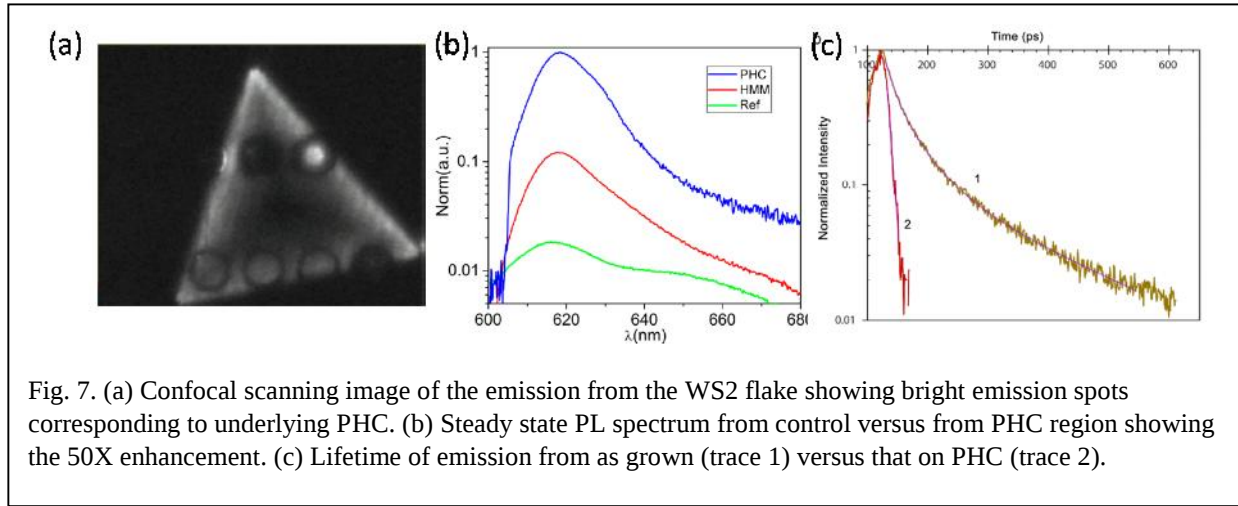
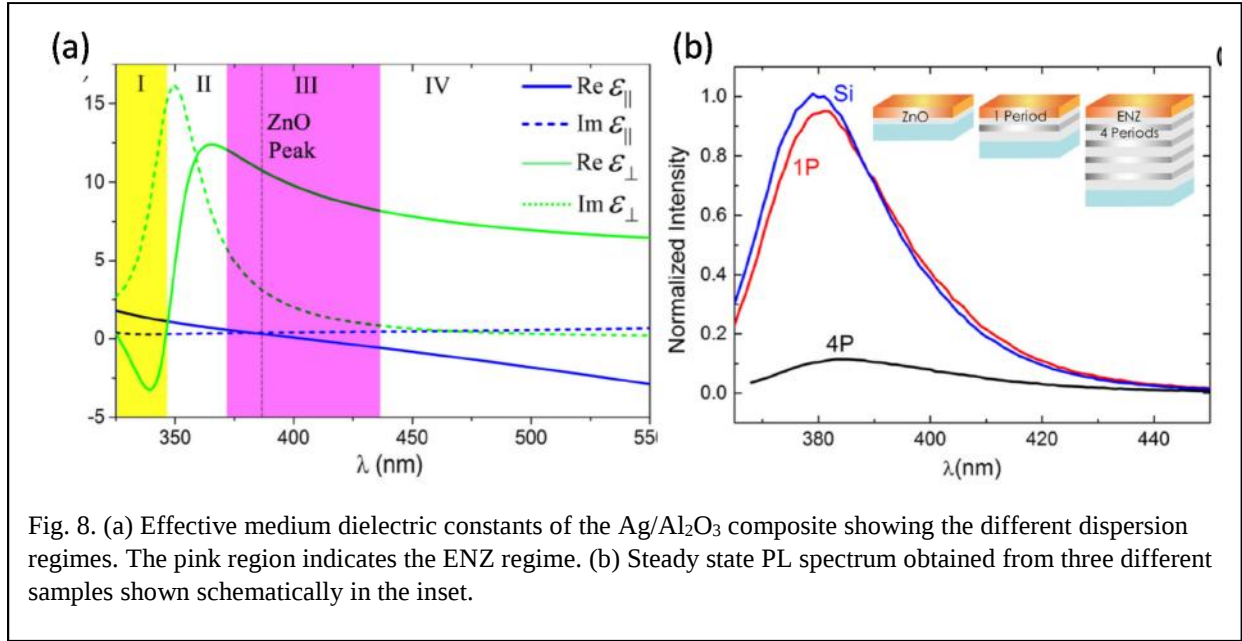


Fig. 7. (a) Confocal scanning image of the emission from the WS_2 flake showing bright emission spots corresponding to underlying PHC. (b) Steady state PL spectrum from control versus from PHC region showing the 50X enhancement. (c) Lifetime of emission from as grown (trace 1) versus that on PHC (trace 2).

Preferential Emission into Epsilon-Near-Zero (ENZ) medium:

In a closely related work, we demonstrated the effect of an ENZ substrate on the spontaneous emission properties of a semiconductor (ZnO) placed in its near field. ZnO grown via atomic layer deposition with emission maximum ~ 380 nm was used in these experiments. Alternating layers of Ag and Al_2O_3 were deposited as discussed previously to obtain the ENZ medium. The effective dielectric constants of the Ag/ Al_2O_3 composite is shown in Fig. 8a, where the different dispersion regimes such as type I hyperbolic (hyperbola of two surfaces), ENZ, and type II hyperbolic (hyperbola of one surface) are shown. The ENZ regime coincided with the emission of ZnO . Steady state PL experiments were carried out in the reflection geometry (pump and emission from the ZnO side). Shown in Fig. 8b is the PL emission observed from the three samples shown schematically in the inset. Clearly the emission in the forward direction is suppressed in the case of the ENZ metamaterial showing that the emission prefers to go into the ENZ medium rather than into air. This is due to the slow modes and large PDOS in the ENZ regime.



Summary: During the 18 month period, we successfully demonstrated the development of a new class of artificial medium: photonic hypercrystals that combine the large PDOS enhancement in hyperbolic metamaterials with the enhanced light scattering efficiency of photonic crystals through Bragg scattering. Using the PHC, we were able to demonstrate 100X enhancement in light out-coupling and 20X enhancement in emission rate using colloidal quantum dots as the active medium. We also demonstrated broadband enhancement in spontaneous emission from 2D monolayer of TMDs (WS_2) placed in the near field of a PHC where we observed 50X enhancement in emission intensity and 7X enhancement in spontaneous emission rate.

We also demonstrated preferential emission of spontaneous emission into ENZ metamaterial due to the presence of large PDOS as well as the slow light modes in the ENZ regime.

Future Plans: We are currently extending the above approaches for enhancing spontaneous emission using PHCs for realizing electrically pumped LED devices. Another direction being pursued is to realize PHC based nanocavities of sub-diffraction dimensions with reasonable Q factors compared to typical metamaterial or plasmonic cavities.

Publications from grant:

1. "Preferential emission into epsilon near zero metamaterial," T. Galfsky, Z. Sun, Z. Jacob and V. M. Menon, *Opt Materials Express* **5**, 2878 (2015)
2. "Broadband enhancement of spontaneous emission in 2D semiconductors using photonic hypercrystals," T. Galfsky, Z. Sun, C. R. Consideine, C-T Chou, W-C Ko, Y-H Lee, E. Narimanov and V. M. Menon, *Nano Lett.* **16**, 4940 (2016).
3. "Photonic Hypercrystals: New media or control of light-matter interactions," T. Galfsky, J. Gu, E. Narimanov and V. M. Menon, In Press – *PNAS* (2017).

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