Photonic crystal: energy-related applications

Zhuo Ye  
*Iowa State University, zye@iastate.edu*

Joong Mok Park  
*Iowa State University, joongmok@iastate.edu*

Kristen P. Constant  
*Iowa State University, constant@iastate.edu*

Tae Guen Kim  
*Korea University*

Kai-Ming Ho  
*Iowa State University, kmh@ameslab.gov*

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Zhuo Ye
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Zhuo Ye, a,b Joong-Mok Park, a,b Kristen Constant, a,c Tae-Geun Kim, d and Kai-Ming Ho a,b

aU.S. Department of Energy, Ames Laboratory, Ames, Iowa 50011
zye@iastate.edu; kmh@cmpgroup.ameslab.gov

bIowa State University, Department of Physics and Astronomy, Ames, Iowa 50011

cIowa State University, Department of Materials Science and Engineering, Ames, Iowa 50011

dKorea University, School of Electric Engineering, Seoul 136-701, Republic of Korea

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Keywords: photonic bandgap materials; soft lithography; solar cells; light-emitting devices.

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1 Introduction

Photonic crystals (PCs) are structures with dielectric properties that vary periodically in space. They are optical analogs to semi-conductors. The periodic dielectric nanostructures of PCs affect photons in a similar way as the periodic potential in a crystal affects electrons. The concept of PCs was first introduced by Yablonovitch and John in 1987, having been inspired by natural crystals. Initially, researchers attempted to fabricate proof-of-concept structures by trial and error, fabricating various periodic structures and experimentally determining whether a photonic band gap was present. In 1990, Ho and coworkers introduced the plane-wave expansion method to solve Maxwell’s equations resulting in the theoretical prediction that a diamond structure would have a full band gap. Since then, several PC structures have been proposed and fabricated, and numerous fabrication techniques and numerical simulation methods have been introduced to advance this fast-developing field of research.

Long before the fabrication of synthetic PCs, biological systems consisting of PC architectures were known to produce numerous striking optical phenomena. An example is the “structural color” present in some butterfly wings, bird feathers, and gem opals. However, replication of such natural periodic structures is not easy. The challenge lies in the very small feature-length scale, which is only several hundred nanometers. First beginning with large PCs designed to function in the microwave range, researchers eventually succeeded in fabricating structures at infrared (IR) and visible scales. Economically efficient and robust mass-production techniques are being sought to make feasible various commercial applications.

PCs have numerous potential applications, many of which are in the area of optical communications. However, the last decade has seen the emergence of novel structures combining photonic crystal properties with other functionalities that have potential in energy-related fields such as lighting and solar energy harvesting. One-dimensional PCs have found applications in photovoltaic cells in the form of metal gratings and Bragg stacks. These behave as solar harvesting enhancers through increasing the optical absorption in the active layer, or as spectral filters in multiple-junction solar cells and thermal photovoltaic (TPV) cells. Two-dimensional PCs have demonstrated potential in lighting, either as high-efficiency light sources or as patterned surface textures to enhance outcoupling in light-emitting diodes. Three-dimensional PCs have
the potential to provide a full band gap, and have found applications in optoelectronics\textsuperscript{17} and TPVs.\textsuperscript{18}

In this paper, we focus on some of our recent studies concerning energy applications of PCs. We begin by reviewing a promising fabrication technique for photonic-crystal structures: soft lithography.\textsuperscript{19–22} Soft lithography uses elastomeric stamps, molds, and photomasks to fabricate or replicate structures. It has a number of advantages including low cost, greater design flexibility, and high resolution. Most importantly, soft lithography is especially suitable in building PC structures for use in energy applications. Different from all-optical-network applications that require compaction and integration in an optical circuit where most elements are made from Si or III–V semiconductors, energy-related applications favor flexible materials like polymers and low-cost, large-area fabrication techniques. In an ideal case, the PC structure can be lifted off the substrate on which it is fabricated and attached onto another substrate with arbitrary shape. After introducing soft-lithography techniques, we will review four resulting energy applications in part 3 including: 1. A polarized metallic thermal emitter in which the emission wavelength is controlled and emissivity enhancement is achieved. 2. Transparent structures for use as electrodes as replacements of conventional indium tin oxide (ITO) electrodes or heat mirrors. 3. Microlens arrays for enhanced outcoupling in light-emitting devices. 4. Structures for enhanced light absorption in photovoltaic cells. All of these applications can be realized using the soft-lithography technique discussed in part 2.

2 Soft-Lithography Technique for Photonic Crystal Fabrication

Many efforts have focused on fabrication of high-quality, low-loss 2-D or 3-D PC structures, especially on the optical scale. 2-D PCs have found commercial applications in the form of photonic-crystal fibers,\textsuperscript{23} which are fabricated using fiber-drawing techniques similar to those developed for communication fibers. However, the 3-D counterparts are still far from commercialization. Various techniques have been explored for fabricating 3-D PCs, including the electron-beam lithography,\textsuperscript{24} holographic lithography,\textsuperscript{25} self-assembly,\textsuperscript{26} micromanipulation,\textsuperscript{27} and auto-cloning.\textsuperscript{28} However, these fabrication methods are either slow, costly, or lack robustness for low-cost mass production. Another drawback of using these techniques is the difficulty in the incorporation of microcavities or waveguides in a controllable way.

In this paper, we will emphasize a relatively new technique: soft-lithography, and specifically, microtransfer molding (\(\mu\)TM).\textsuperscript{29} \(\mu\)TM has a number of advantages, including low cost, potential for nonperiodic 3-D structures, compatibility with a wide range of materials, and flexibility in design. Our group has developed an advanced technique called two-polymer \(\mu\)TM\textsuperscript{30–33} to create high-quality layer-by-layer PC structures on an optical scale. In this technique two different UV-curable prepolymer B are used, one as a filler (prepolymer A) and another as an adhesive (prepolymer B), to build a 3-D structure layer by layer (the schematic procedure is shown in Fig. 1). The two-polymer \(\mu\)TM has the advantages of high resolution, low cost, no etching required, and no need for clean room facilities. Additionally, by applying two different prepolymer as filler and adhesive, we obtained both high bonding strength and low capillary wicking, reducing the distortion from the liquid meniscus and allowing us to achieve high fidelity for the fabricated structures. A 12-layer woodpile structure fabricated by the two-polymer \(\mu\)TM is shown in Fig. 2.\textsuperscript{30} The area of the fabricated structure is 4 by 4 mm and the orientation of each layer is perpendicular to the one below. This layer-by-layer woodpile PC was first proposed and fabricated at microwave scale in the 1990s,\textsuperscript{6} then in the IR\textsuperscript{34} and most recently on an optical scale.\textsuperscript{30} Such structures have attracted much interest because it has a full band gap and can also be built by layer-by-layer semiconductor processing.\textsuperscript{34,35}

Using the multilayer polymer structure as shown in Fig. 2, it is possible to create metallic PCs. Converting a polymer template to a metallic PC is readily accomplished as described in Ref. 31, where a nickel woodpile PC was fabricated from a multilayer polymer template by backfilling nickel through electrodeposition until a homogeneous overlayer was formed. The backfilled structure was peeled off from the conducting substrate, and the polymer template embedded within was chemically removed (the schematic procedure is shown in Fig. 3). The metallic PCs, specifically in such a woodpile pattern, have attracted much attention because they can
serve as efficient thermal emitters and photovoltaic devices through tailoring of the absorption and thermal emission spectrum.\textsuperscript{35,36} In Sec. 3, this application will be addressed in more detail.

3 Energy-Related Applications

PCs are used in a diverse range of applications, from PC fibers\textsuperscript{23} to highly reflective mirrors in laser cavities.\textsuperscript{37} In this work, however, we shall focus only on selected energy-related applications.

3.1 Metallic Photonic Crystals as Thermal Emitters

The characteristic property of PCs is the photonic band gap (PBG). Photons with wavelengths in the PBG are prohibited from propagating in PCs. If PCs are made of materials with large absorption coefficients, like metals, the absorption is suppressed for light of wavelengths in the band gap. As absorption and emission processes have the same physical origin, we can manipulate the emission spectrum as well as the absorption spectrum. In a TPV cell, a thermal emitter replaces the sun as the radiation source, from which the thermal radiation is converted into electricity. As

![Fig. 1 Schematic of two-polymer μTM procedure. WAD: wet-and-drag. Reproduced from Ref. 30.](image1)

![Fig. 2 SEM images of a 12-layer PC microstructure. Reproduced from Ref. 30.](image2)
the portion of photons with energy smaller than the electronic band gap is wasted, it is desirable
to have a narrow-band spectrum with its radiation energy slightly above the electronic band
gap.\textsuperscript{18} A full PBG can be used to suppress radiation below the electronic band gap. At the
same time, emission can be enhanced due to a high density of states at a narrow band near
the edge of PBG.

In Ref.\textsuperscript{18}, Lin and coworkers reported a woodpile tungsten PC, which was fabricated with
a modified silicon process and was used to tailor the absorption spectrum (Fig. 4). This tungsten
PC possesses a complete band gap at wavelengths $\lambda \geq 3 \mu m$, and the thermal emission
was suppressed in this PBG regime. The resulting emission spectrum at a temperature of

\begin{figure}
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\includegraphics[width=\textwidth]{fig3.png}
\caption{Schematic procedure of converting a polymer template to a metallic PC.}
\end{figure}

\begin{figure}
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\includegraphics[width=\textwidth]{fig4.png}
\caption{(a) A SEM view of a 3D tungsten photonic-crystal. Within each layer, the rod width is 0.5 \(\mu m\) and the rod-to-rod spacing 1.5 \(\mu m\). (b) The measured photonic-crystal emission power at a bias of 3, 4, 5, and 6.5 V, respectively. The effective temperature, average over the entire sample, is $\sim 1190$, 1320, 1440, and 1535 K, respectively. At $\sim 1535$ K, the emission peaks at $\lambda \sim 1.5 \mu m$ and has a full width at half maximum of 0.9 \(\mu m\). The dashed blue line is a blackbody radiation curve. The electronic band gap wavelength of GaSb is also shown as a red line. Reproduced from Ref. 18.}
\end{figure}
~1535 K peaked at $\lambda \sim 1.5 \mu m$ with a narrow spectral width [Fig. 4(b)]. By comparing with a blackbody radiation curve indicated by a dashed line, a much narrower emission peak due to the PBG effect was observed, which implied a better match of the emission spectrum to the electronic band gap of a photovoltaic cell. This effective radiation spectrum could lead to an optical-to-electric conversion efficiency of $\sim 34\%$.18

Metallic PC thermal emitters can also be designed to have an emission spectrum that is strongly dependent on polarization when the intrinsic symmetry of PC structure is broken. Polarization-engineered thermal emitters have the potential to achieve higher conversion efficiency. For instance, in a photovoltaic cell with incorporated metal nanostructures, the optical absorption may be enhanced only in one polarization.38 Then it is preferable to have a thermal emitter with enhancement in the same polarization. To obtain a polarization-sensitive emission spectrum, we explored two variations of nickel woodpile-like PCs consisting of two layers of 1. differently structured nickel gratings [Fig. 5(a)–5(c)],32 and 2. various mutual angles [Fig. 6(a)–6(c)].33 The measured emissivity showed strong polarization dependence [Figs. 5(d)–5(f) and 6(d)–6(f)]. The strong absorption (emission) for a certain polarization was attributed to the greatly reduced group velocity in the vicinity of the photonic band edge for that polarization. The significantly reduced group velocity increased the interaction of light with nickel and enhanced the intrinsic absorption. The identification of the mechanism underlying this polarization sensitivity can help us design such polarized thermal emitters. Furthermore, as these polarized thermal emitters do not rely on angle-sensitive phenomena such as refraction or diffraction, they can be used for a wide range of emission angles.

3.2 Transparent Electrodes as Replacements for ITO and Heat Mirrors

Transparent conducting electrodes have been widely used in optoelectronic devices requiring both high optical transmittance and electrical conductivity such as flat screen displays, light
emitting diodes (LEDs) and solar cells. Among them, metal oxides (especially indium, indium tin, or zinc) have been commonly used as transparent electrodes (TEs) due to high optical transmission in the visible range and good electrical conductivity. However, due to their brittle nature and high demands by industry, alternative electrodes\textsuperscript{39,40} have been widely studied including unpatterned thin metal films,\textsuperscript{41} metal nanowire networks,\textsuperscript{42} metal nanowire grating,\textsuperscript{43–45} carbon nanotubes,\textsuperscript{46} and graphene.\textsuperscript{40} Most materials studied as TEs cannot satisfy high-transmission and high-conductivity requirements at the same time because the transmittance exponentially decreases with the thickness of the materials $\propto e^{-\alpha l}$. Either optical transmission or electric conductivity is sacrificed when maximizing the other. A recent study showed that metallic 1-D grating or 2-D mesh photonic-crystal structures can match or exceed the performance of indium tin oxide (ITO), commonly used as TEs.\textsuperscript{43–45} Furthermore a new photonic-crystal architecture of high-aspect metal grating or mesh structures was proposed and demonstrated to decouple the trade off between high optical transmittance and high electrical conductivity.\textsuperscript{47,48} In addition, such metallic PCs have high reflectance in IR which can be used as heat-blocking mirrors. IR reflection prevents unnecessary heating of the devices and improves device performance and lifetime.

For making high-aspect-ratio TEs, thin metal films are coated on both the sides and the top of polyurethane (PU) gratings by angular depositions. The top films are selectively removed by Ar ion milling resulting in a 1.4-micron-tall and 40-nm-thick high-aspect-ratio metal grating structure. The PU gratings are fabricated using two polymer microtransfer molding methods.\textsuperscript{29} The high-aspect-ratio metallic TEs showed high transmittance (about 70% to 80%) in the visible range (Fig. 7) and better electrical properties (resistance $\approx 10$ ohm/cm) compared to commercial ITO-coated glass. The exposed high-aspect metal gratings can be encapsulated with PU which has a refractive index of 1.55, very close to that of glass. After encapsulation, the transmission is...
slightly increased and diffraction is reduced (Fig. 8), making the structures visibly transparent and suitable for display and imaging devices.\textsuperscript{48} To demonstrate ITO-free large-area and low-cost TEs, high-aspect metallic grating structures were fabricated using 50-by-50-cm nano-imprinted PU. The TEs showed high transmission even though the incidence angle is large up to 70 deg.\textsuperscript{49}

One more advantage of the metallic grating structure is heat reflection. Most thermal radiation sources, including the solar spectrum, have significant IR which is converted to heat after absorption. By reflecting unwanted IR, while transmitting only useful visible spectra, heat shielding from the radiation source is possible. Such a heat shield is called a hot mirror (or cold mirror in the reverse case) and improves efficiency in optoelectronic devices. Reference 50 suggested that the height of patterned metal mesh should be tall enough to reflect long-wavelengths light. To understand the heat-shielding effect, we used encapsulated TEs made with high-aspect-ratio gold and placed them front of thin-film Si solar cells. It is well known that the open-circuit voltage of solar cell decreases as temperature increases due to the absorption from the radiation source. To measure the temperature change on thin-film solar cells, two identical cells were placed side by side and the TE was mounted in front of one solar cell [Fig. 9(a)]. The temperatures of the solar cells were monitored by a thermocouple attached on the backside of the assembly. Figure 9(b) shows the temperatures change over time of the two cells, and Fig. 9(c) shows the difference between the two. Initial temperatures were 24 deg for both cells. After the lamp was on the temperatures slowly increased to 39 deg at time $t =$ 200 sec. Then a piece of plate glass with the same thickness of TE was placed in front of a cell (T1) at 300 sec. The temperature changed less than 1 deg by partial reflection.

\textbf{Fig. 7} Left: (a) and (b) SEM images of PU grating made with micro transfer molding and (c) and (d) high aspect metal TEs. Thin metal film coated on top of the PU gratings was removed by directional Ar ion milling but coated on the side walls were remained. Right: Average transmissions of TE made of gold and silver are close to ITO coated glass. Reproduced from Ref. 48.

\textbf{Fig. 8} Left upper corner of the large area transparent electrodes was encapsulated with PU showing clear image even at high angles.
from the lamp. The slight increase in the temperature was due to greenhouse effect. TE was placed in front of the cell (T1) at 600 sec and then moved to the other cell (T2) at 900 sec. The temperature of the solar cell covered by the TE was three degrees lower compared to the uncovered cell.

3.3 Enhanced Light Outcoupling in Light Emitting Devices

There is an innate limitation to light extraction from LEDs and organic LEDs (OLEDs) due to the interface between the light-emitting materials and air. As illustrated in Fig. 10, only a fraction of the generated light is extracted and the rest is trapped in the substrate (as glass mode) and in the light-emitting layer/ITO layer (as high index mode). Some of the trapped light is reabsorbed or emitted through edge emission. There have been many efforts to extract the trapped light by texturing the surface on the light emitting side, putting microlens arrays on the other side, or both. Both schemes are breaking total internal refractions, in the high index mode by texturing, or in the glass mode with the microlens array. They can be regular periodic structures or randomly roughened surfaces. In periodic structures, light is extracted in the surface plasmon mode or by refracting the ray as classical optics depending on the size, periodicity, and refractive index of the textured surface.

Recently, we reported a microlens array design for OLEDs that results in electroluminescence (EL) enhancement of $\sim 100\%$ in the forward direction. This was achieved with soft-lithography imprinting of very uniform 2-$\mu$m-pitch square arrays, embossed on the blank side of a ITO-coated glass. By placing a microlens array on the other side of OLEDs, trapped light inside the substrate can be extracted. In general, methods to enhance light extraction of LEDs and OLEDs can be classified into two categories. One is texturing the surface of the light-emitting layer to extract light trapped inside. The other is texturing the substrate (glass) surface to extract the light trapped in it. These methods include, for example, attaching a microlens array or introducing a randomly roughened surface. A typical roughened surface often results in sub-wavelength scattering at the air/substrate interface, and thus significantly reduces the transmission. On the other hand, microlens has large curvature (ranged from a few to a few hundred microns) and the use of it will not reduce the transmission ($>90\%$). The extraction enhancement

Fig. 9 (a) Experimental setup for heat blocking TEs for the thin film Si solar cell. (b) and (c) Temperature change on the solar cell.

Fig. 10 Diagram showing light extracted (ray I) and trapped inside of the substrate (ray II) and ITO/organic layers (ray III) by waveguide mode. Reproduced from Ref. 51.
with microlens array depends on the shape of each microlens, its periodicity, and the area covered. Larger microlens areas can extract all light in the substrate guiding mode. The microlens-covered pixels result in more light extraction even from areas outside the light emitting pixel area as in Fig. 11(a) and 11(b). By increasing the extraction area with the microlens array, the enhancement also increases [Fig. 11(c)]. This fabrication technique of the microlens array provides a low-cost means for device manufacturing. In addition, the use of microlenses provides a more diffuse light source that is advantageous for diffuse lighting purposes.

3.4 Enhanced Light Absorption in Organic Photovoltaic Cells

For practical solar-cell designs aimed at enhancing optical absorption in the active layer, it is essential to achieve a broad-band, polarization-insensitive absorption enhancement, since incoming and scattered sunlight is not preferentially polarized and includes both TE and TM polarizations. Metallic nanoparticle-based schemes can provide polarization-insensitive enhancement, but they do not offer conductive pathways and cannot replace ITO electrodes. Metallic grating-based designs do provide an alternative to ITO electrodes. However, such schemes either provide absorption enhancement in only one polarization, or provide no enhancement at any polarization because a significant part of incoming light is blocked by the metal structures.

In a recent study, we showed that the above-reviewed transparent electrode architecture reported in Ref. 48 not only matches and outperforms ITO in its transparency and conductivity, respectively, but can additionally enhance light absorption in organic photovoltaic (OPV) cells for both polarizations. A P3HT:PCBM-based OPV cell was simulated to illustrate the field enhancements produced by a high-aspect-ratio silver grating similar to the one reported in

![Fig. 11 Images of two OLED arrays with (a) green emitting Alq3 and (b) blue emitting DPVBi. The left side pixels in each image are under a microlens array and the right ones are reference pixels. The surrounding (rim) lines are the epoxy sealant used for OLED encapsulation. (c) EL spectra of the Alq3-based OLED with a PU microlens array measured with different apertures (d = 5, 10, 25 mm) of an integrating sphere. (d) EL spectra of a DPVBi-based OLED with microlenses measured with a 25 mm diameter integrating sphere aperture. The black lines in (c) and (d) are the reference spectra of nominally identical OLED pixels without the microlenses. Reproduced from Ref. 16.](http://photonicsforenergy.spiedigitallibrary.org/021012-9)
Ref. 48. A remarkable ten-folds-high peak light-absorption enhancement at specific wavelengths and a $\sim 23\%$ broadband enhancement were expected using the design, when compared to an ordinary cell using a 150-nm-thick ITO-coated glass as the transparent electrode. In this design, surface plasmon (SPs) excitations and effective coupling to cavity mode were utilized for the field enhancements for TM and TE polarizations, respectively (Fig. 12). Strong field enhancements in P3HT:PCBM were observed for both polarizations.

Our structure is superior to previous metallic-grating-based absorption-enhancement schemes because it provides geometric separation of electrical conductivity and optical absorption. The height of the metallic nanogratings can be increased to enhance conductivity, while minimally changing the absorption in the active layer below. Another advantage of our structure is that it provides a smooth top surface for device fabrication. Conventional grating design gives a rough surface, which can pose problems for solution processing of polymer layers.

The above examples regarding energy-related applications of PCs represent just a sampling of ongoing developments and exciting future directions. There are applications that we did not discuss here but are no less important, such as those in light emitting$^{14,15}$ (lasers and LEDs) based on the Purcell effect,$^{61}$ and in sunlight harvesting taking advantage of slow photons,$^{62}$ among others. In the last two decades, PCs have matured from an intellectual curiosity to a field with real applications. However, although potential applications emerge constantly, few have yielded quick and applicable results. An important goal is to combine PCs with existing devices to achieve high-efficiency light generating or harvesting schemes with low-cost and robust fabrication techniques. There is, however, still much effort required in order to meet this objective.

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