BACK TO BASICS: History of photonic crystals and metamaterials

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We will review the history of photonic crystals and overview of the theoretical and experimental efforts in obtaining a photonic bandgap, a frequency band in three-dimensional dielectric structures in which electromagnetic (EM) waves are forbidden, is presented. Many experimental groups all over the world still employ this woodpile structure to fabricate PCs at optical wavelengths, waveguides, enhance nanocavities, and produce nanolasers with a low threshold limit. We have been focused on a new class of materials, the so-called metamaterials (MMs) or negative-index materials, which exhibit highly unusual electromagnetic properties and hold promise for new device applications. Metamaterials can be designed to exhibit both electric and magnetic resonances that can be separately tuned to occur in frequency bands from megahertz to terahertz frequencies, and hope-fully to the visible region of the EM spectrum.

Novel artificial materials (photonic crystals (PCs), MMs, nonlinear optical MMs, optical MMs containing gain media, graphene, chiral optical MMs and plasmonics) enable the realization of innovative electromagnetic (EM) properties unattainable in naturally existing materials. These materials, characterized here as MMs, have been in the foreground of scientific interest in the last ten years. There has been a truly amazing amount of innovation during the last few years and more is yet to come. Clearly, the field of PCs and MMs can develop breaking technologies for a plethora of applications, where control over light is a prominent ingredient – among them telecommunications, solar energy harvesting, biological and THz imaging and sensing, optical isolators, nanolasers, quantum emitters, wave sensors, switching and polarizers, and medical diagnostics. However, many serious obstacles must be overcome before the impressive possibilities of MMs, especially in the optical regime, become real applications.

History of photonic crystals: from diamond to woodpile structures

Shortly after the introduction of the concept of photonic band-gap (PBG) materials [1,2], our group at Iowa State/Ames Lab discovered the first diamond PBG structure that can exhibit a complete 3D photonic gap [1,2]. Complete gaps were found in the two diamond structures. However, this diamond dielectric structure is not easy to fabricate, especially at the micron and submicron length for infrared or optical devices. Therefore, it is important to find new periodic structures that possess full photonic gaps, but, at the same time, are easier to fabricate (see *Fig. 1*). We thus modified the

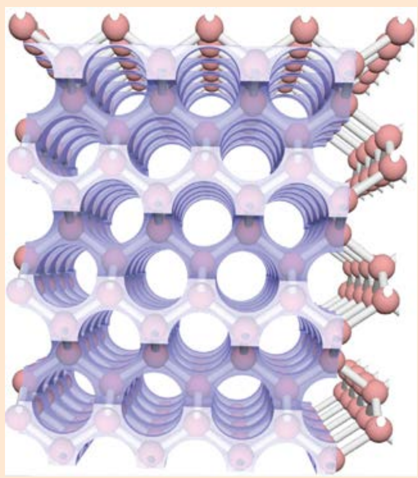

Figure 1. A "ball and the stick" model of the diamond structure, viewed from an angle such that the channels along a [110] direction are observable. Shrinking the size of the "balls", while keeping the "sticks" and "3-cylinder" diamond structure with "air-cylinder".

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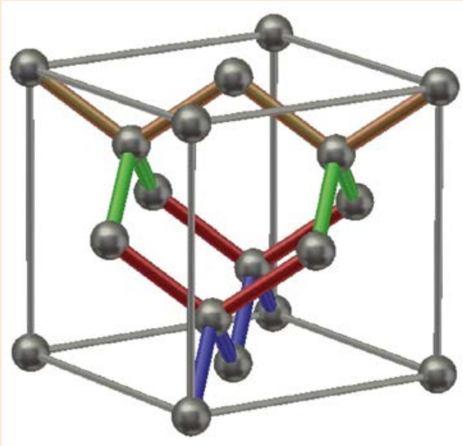

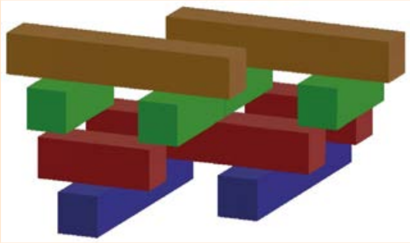

△ **Figure 3.** Short rods in the diamond structure are replaced by an equivalent large horizontal rod, which is a layer-by-layer structure or woodpile structure [1].

◁ **Figure 2.** Rod connected diamond structure with rods colored along the vertical direction.

diamond structure to obtain other photonic crystal structures easier to fabricate—hence, the introduction of a layer-by-layer (or woodpile) version of diamond. *Figure 2* shows the rod-connected diamond structure with rods colored along the [100] vertical direction. *Figure 3* presents the woodpile design – short rods in the diamond structure are replaced by an equivalent large horizontal rod.

The layer-by-layer (woodpile) structure was first fabricated in the microwave regime by stacking alumina cylinders that demonstrated a full 3D photonic bandgap at 12-14 GHz. We fabricated this woodpile structure and demonstrated a full 3D photonic bandgap at 100 and 500 GHz. The woodpile structure was then fabricated by microelectronic fabrication technology (see *Fig. 4*) and operated at infrared wavelengths. Structures made by advanced silicon processing [3] and wafer fusion techniques operate at near-infrared wavelengths. It also was fabricated by direct laser writing to operate at infrared and near-infrared wavelengths. Inverse woodpile PCs made from silicon by CMOS-compatible deep reactive-ion etching were the first nanostructures to reveal spontaneous emission inhibition. In another approach, "3-cylinder" photonic crystals were fabricated by a deep X-ray lithography technique in polymers (see *Fig. 5*) and the transmission spectra show a 3D photonic bandgap centered at 125 µm (2.4 THz) in good agreement with theoretical calculations.

These photonic crystal designs (diamond lattice [1,2] and the woodpile structure [3], respectively) provided the largest completed 3D photonic bandgaps. Many experimental groups around the world still use the woodpile structure to fabricate photonic crystals at optical wavelengths. These designs have been instrumental in bringing forward the revolutionary fields of photonic crystals [1-2], negative diffraction, and metamaterials [4-5], extending the realm of electromagnetics while opening exciting new applications.

History of metamaterials: from microwaves to optics structures

Microwave metamaterials

Our group at Armes Lab demonstrated negative refraction properly designed PCs in the microwave regime and also demonstrated sub-wavelength resolution of $\lambda/3$. We fabricated a left-handed material (LHM) with the highest negative-index transmission peak corresponding to a loss of only 0.3 dB/cm at 4 GHz frequencies. We established that split-ring resonators (SRR) also have a resonant electric response, in addition to their magnetic response. The SRR electric response is cut-wire like and can be demonstrated by closing the gaps of the SRRs, thus destroying the magnetic response. We studied both theoretically and experimentally the transmission properties of a lattice of SRRs for different incident polarizations.

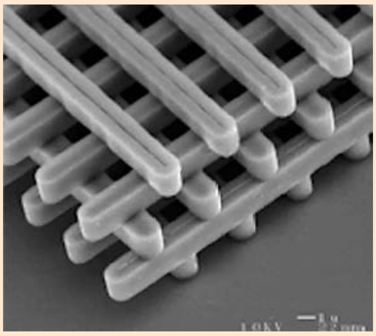

Figure 4. Electron-microscope image of a "woodpile" photonic crystal made by silicon with a complete band gap at 12 µm [1].

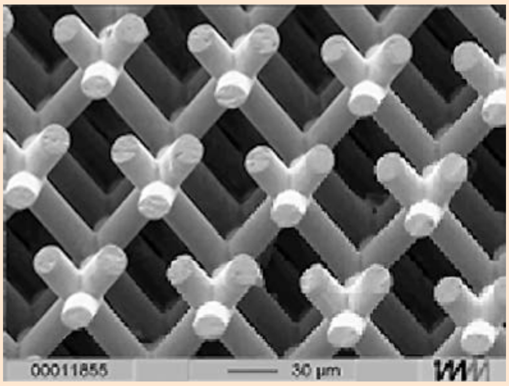

Figure 5. The "three-cylinder" photonic crystal made from negative tone resist [1].

We showed that as one decreases the size of the SRR, the scaling breaks down and the operation frequency saturates at some value. We will come back to this important point below. We suggested a new 3D left-handed material design that gives an isotropic negative index of refraction. This design has not been fabricated yet. We introduced new designs that were fabricated and tested at GHz frequencies. These new designs were very useful in fabricating negative-index materials at THz and optical wavelengths and can be easily measured for perpendicular propagation. A simple unifying circuit approach offered clear intuitive as well as quantitative guidance for the design and optimization of negative-index optical metamaterials

Optical metamaterials

In the optical transmission experiments on very small samples, we took advantage of the fact that, loosely speaking, one can couple to the magnetic-dipole resonance via the magnetic field of the light and via the electric field. The latter can be accomplished under normal incidence of light with respect to the substrate – which was much easier for these early experiments. We varied the lattice constant, opened and closed the slit and thereby unambiguously showed the presence of the predicted magnetic resonance at around 100 THz frequencies, equivalent to 3 µm wavelength [4].

We published our experimental results on miniaturized gold SRR arrays, which showed a magnetic resonance at twice the frequency, *i.e.*, at around 200 THz, equivalent to 1.5 µm wavelength [4]. By experiments performed under oblique incidence of light, again loosely speaking, we not only coupled to the SRR via the electric field of light, but also via its magnetic field. Only this coupling can be mapped onto an effectively negative magnetic permeability.

We reached the limit of size scaling by further miniaturized SRRs (see *Fig. 6*). Thus, the LC frequency should also scale inversely proportional to size.

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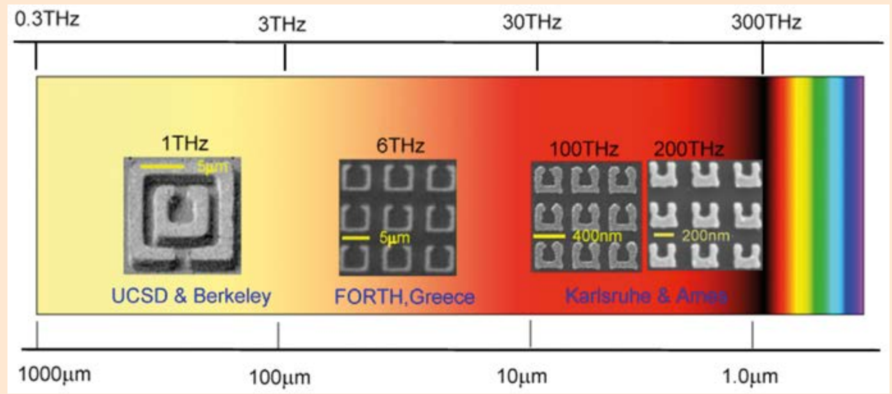

Figure 6. The operation frequency of magnetic metamaterials increased rapidly. The structures operating at 1, 6 and 100 THz frequency, respectively, were fabricated in 2004, and the one at 200 THz in 2005.

It does not though. At frequencies higher than the damping rate, the electrons in the metal develop a 90-degrees phase lag with respect to the driving electric field of the light. Our work showed that this limit is reached at around 900 nm eigenwavelength – unfortunately just outside of the visible regime.

Despite of this saturation, reducing the capacitance of the SRR can further increase the frequency. In this fashion, one gets a continuous transition between a traditional SRR and a pair of cut wires [4,5]. Of course, this frequency increase at fixed SRR side length comes with a price: It also means that the ratio of operation wavelength and size of the resonating object decreases significantly, bringing one close to the edge of validity of describing these structures by using effective material parameters. Nevertheless, our optical experiments and the retrieval of effective parameters suggested the possibility of obtaining a negative magnetic permeability – even under normal incidence of light with respect to the substrate (see *Fig. 7*).

We also investigated the optical properties of these double-fishnet optical metamaterials under oblique incidence of light and for different polarizations. We found an angular dispersion, which can be interpreted in terms of spatial dispersion due to interaction among the different unit cells (see *Fig. 8*).

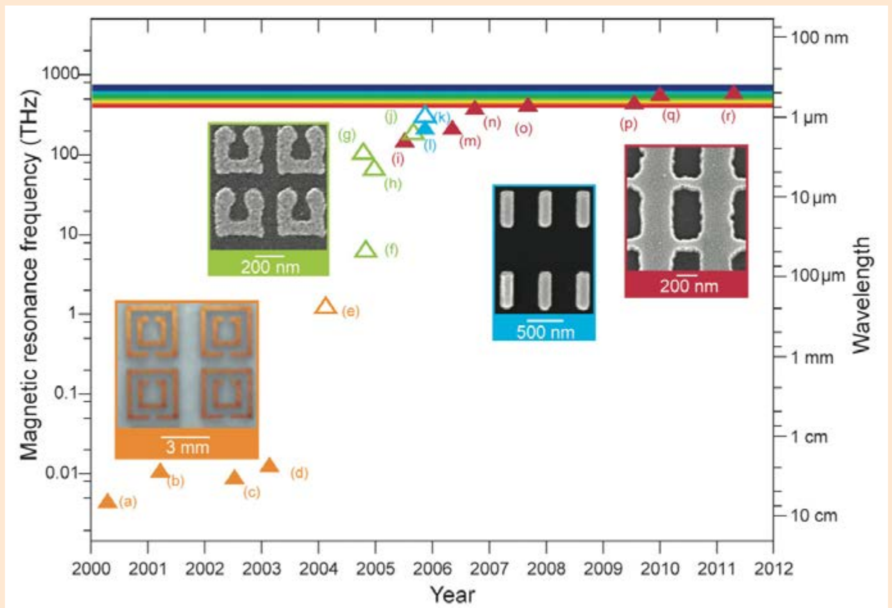

Figure 7. Fabrication progress of negative index materials vs year. This figure is taken from the article in *Nature Photonics* **5**, 523 (2011) written by Soukoulis and Wegener [4].

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These structures operating at around 1.5 µm wavelength were based on gold as metal. Soon thereafter, we could reproduce the same effects at comparable wavelength at much lower damping by using silver instead of gold.

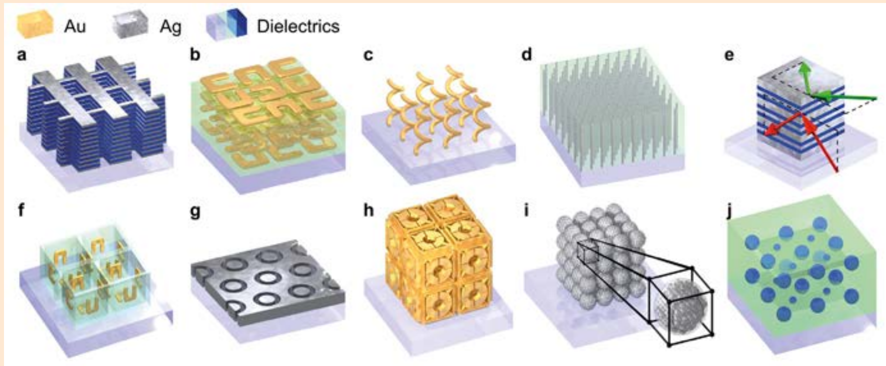

Figure 8. 3D Photonic-Metamaterials Structures. This figure is taken from the article in Nature Photonics 5, 523 (2011) written by Soukoulis and Wegener [4].

Optical metamaterials containing gain media

Our experimental efforts to bring gain media into close proximity of SRR arrays operating around 1.5 µm wavelength to compensate for these losses were motivated by our own early theoretical work suggesting this approach might work. However, our efforts over several years based on using thin semiconductor films as gain media in the near field of the SRR were not successful. The experiments did show some loss reduction, with line shapes in agreement with theory, but the beneficial effects were not large enough at the end of the day. Thus, we eventually gave up working in this direction.

Nevertheless, we studied loss compensation of SRR structures with a gain layer underneath in quite some detail theoretically. Numerical results showed that the gain layer could compensate the losses of the SRR, for light propagating parallel and perpendicular to the plane of the metamaterial layer. Three different gain pumping schemes were applied in the simulations and the efficiencies of their corresponding loss compensations were studied by investigating the line width of the resonant current. We have also studied the effect of the background dielectric of gain.

We have introduced an approach for pump-probe simulations of metallic metamaterials coupled to gain materials. We study the coupling between the U-shaped SRRs and the gain material described by a four-level gain model. Using pump-probe simulations, we find a distinct behavior for the differential transmittance $\Delta T/T$ of the probe pulse with and without SRRs in both magnitude and sign (negative, unexpected, and/or positive). Our approach verified that the coupling between the metamaterial resonance and the gain medium is dominated by near-field interactions. Our model can be used to design new pump-probe experiments to compensate for the losses of metamaterials.

Graphene for THz applications

Graphene – a one-atom-thick continuous sheet of carbon atoms – has special properties (e.g., electric and chemical tunability, high kinetic inductivity allowing for strongly confined plasmons, large THz optical conductivity) that make it a desirable material for manipulating terahertz waves.

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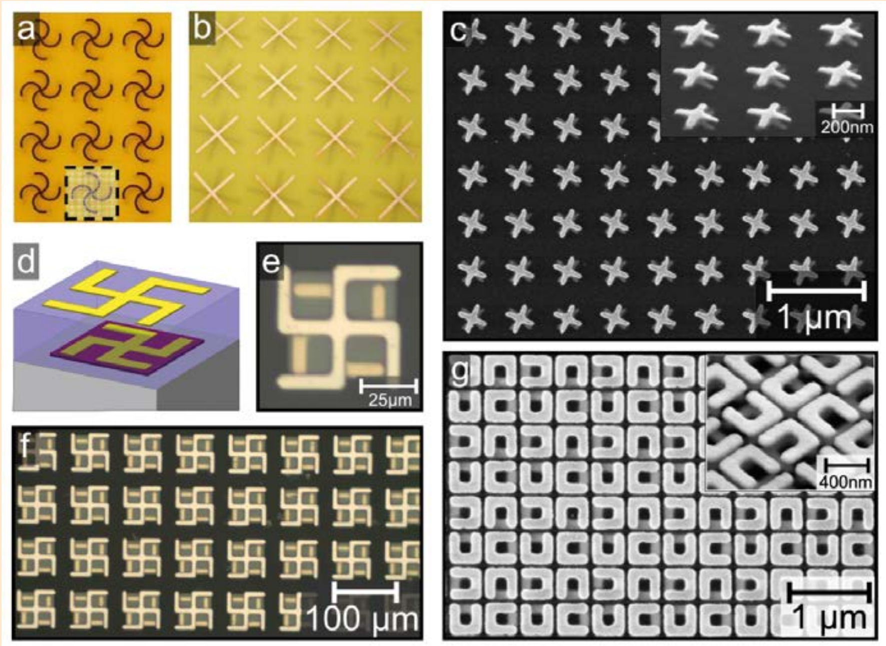

Figure 9. 3D Chiral Metamaterials. This figure is taken from the article in *Nature Photonics* **5**, 523 (2011) written by Soukoulis and Wegener [4].

THz applications operate at frequencies between microwave and far infrared. Some metamaterials could benefit by replacing the metals currently used in fabrication with graphene. Graphene also offers the advantage of a potential enhancement of terahertz wave confinement [6] and smaller, more sub-wavelength metamaterial resonators made from graphene rather than metals. However, as we pointed out in our analysis of published experimental measurements of the THz optical conductivity of graphene, the experimental data have shown significantly higher electrical losses at THz frequencies than have been estimated by theoretical work or have been assumed in mostly very optimistic numerical simulations, showing that more research needs to be done to make metamaterial devices from graphene. We also published an analysis and comparison of gold- and graphene-based resonator nanostructures for THz metamaterials and an ultrathin graphene-based modulator. Metamaterial resonators and plasmonic applications have different requirements for what can be considered a good conductor. We published a systematic comparison of graphene, superconductors, and metals as conducting elements in either metamaterials or plasmonics [6].

Chiral optical metamaterials

At this point we reminded ourselves that optical activity and circular dichroism can be explained by using magnetic-dipole resonances excited by the electric-field of the light and vice versa. This fact is related to bi-anisotropy (see *Fig. 9*).

Our early work in this direction used SRR or variations thereof arranged into single or different layers. These samples were fabricated by using electron-beam lithography of several aligned layers. This led to very large circular birefringence effects at negligible linear birefringence. Experiments and theory were in good agreement.

Conclusion

3D printing benefits from metamaterials and vice versa. By this combination, printing of thousands of different effective material properties may become accessible by using only a few ingredient material cartridges – in analogy to printing thousands of different colors from just three color cartridges in today's 2D graphical printers.

Today, many fundamental metamaterial effects have been demonstrated experimentally in microwaves, optics, thermodynamics, mechanics, and transport. Most scientifically fascinating to the authors are sign reversals of effective material parameters and the possibility of cloaking in all of these areas. Sign reversal has been shown for the magnetic permeability, refractive index, dynamic mass density, differential mechanical stiffness, thermal length-expansion coefficient, and the Hall coefficient.

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