Scale-up of the manufacturing of optical metamaterials

Yuan Wang and Xiang Zhang

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Recently, a new class of artificial materials known as metamaterials has emerged to manipulate light at the nanoscale. Contrary to natural materials, the physical properties of optical metamaterials are primarily dependent on the material structures rather than their chemical constituents. The structures forming the building blocks of the metamaterials are much smaller than the wavelength of light. By tailoring both effective electric permittivity and the magnetic permeability of metamaterials from a positive to a negative value, one can create negative refractive index metamaterials (NIMs).¹ Various 3D NIM structures have been previously reported,² but they are limited to small-scale samples of less than 1 mm. The unprecedented properties of metamaterials and their potential revolutionary applications, such as superlens imaging,3 remain a challenge to implement in practice due to the lack of innovative large-scale manufacturing methods. As a result, a scalable scheme that enables the large-area fabrication of 3D nanostructures must be developed.

In the work of Gao et al.,4 nanoimprint lithography with physical liftoff has been demonstrated as a successful route to produce 3D metamaterials with sizes of up to 8.5 cm and with fabrication throughputs several orders-of-magnitude larger than those based on focused ion beam milling and electron beam lithography. Multilayer fishnet geometries have been selected for use with nanoimprint lithography because of their unique optical properties.⁵ A soft nanoimprint lithography process forms a polymeric pattern to serve as a scaffold for the growth of multilayer metal-dielectric stacks. The scaffold defines the geometries with its interstitial space and protects the stacks during the physical liftoff process to remove the unwanted materials on top. A capping bilayer has been used to provide strong adhesion with a uniform elastomeric slab, leading to effective peeling of the top materials. This scheme enables high yields of multilayer nanostructures over large areas with low loss for optical bands (Figure 1).

Note that by avoiding typical lithography development and etching steps, this approach can effectively



Figure 1 (**a**–**c**) SEM images of nanoimprint lithography-formed fishnet structures (**a**) before and (**b**) after deposition of multilayers of Ag/MgF_2 , and (**c**) after removal of the polymer scaffold. (**d**, **e**) Macroscopic optical images of (**d**) telecom band and (**e**) visible band 3D NIM structures. Adapted with permission from Gao *et al.*⁴ Copyright 2014 American Chemical Society.

reduce sidewall roughness and prevent chemical contamination. In addition, the patterning step can be completed using commercially available nanoimprint lithography tools to achieve critical feature sizes deep into the nanometer range. One challenge to be addressed for applications with large-scale 3D NIM devices is that shadowing from the top layers of the stacks often leads to angular growth (typically 12–15°) in recessed regions, which limits the metamaterials' thickness that conventional thin-film deposition techniques can achieve. Nevertheless, the approach of Gao *et al.* offers the advantages of high throughput and low cost and is potentially applicable to roll-to-roll schemes for the large-scale manufacturing of metamaterials.

- 4 Gao, L., Shigeta, K., Vazquez-Guardado, A., Progler, C. J., Bogart, G. R., Rogers, J. A. & Chanda, D. Nanoimprinting techniques for large-area three-dimensional negative index metamaterials with operation in the visible and telecom bands. *ACS Nano* 8, 5535–5542 (2014).
- 5 Zhang, S., Fan, W., Malloy, K. J., Brueck, S. R. J., Panoiu, N. C. & Osgood, R. M. Near-infrared double negative metamaterials. *Optics Express* 13, 4922–4930 (2005).

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NSF Nanoscale Science and Engineering Center (NSEC), University of California at Berkeley, 3112 Etcheverry Hall, Berkeley, CA, USA E-mail: xiang@berkeley.edu

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³ Fang, N., Lee, H., Sun, C. & Zhang, X. Sub-diffractionlimited optical imaging with a silver superlens. *Science* **308**, 534–537 (2005).