## **Dielectric metalens by multilayer nanoimprint lithography and solution phase epitaxy**

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Metasurfaces have been rapidly developed in the past decades for the advanced characteristics of subwavelength thickness, large degree of freedom in optical functions design and integration, and superior optical properties, which are expected to be the potential alternatives of the refractive optical elements.<sup>1</sup> In recent years, metasurfaces are extensively investigated for the light manipulation, in magnitude, phase, frequency and polarization. N.Yu et al. proposed the generalized laws of reflection and refraction illustrating the working principle of metasurface design.<sup>2</sup> At the early stage plasmonic metasurfaces are studied but they suffer from low optical efficiency since the metallic nanoantenna absorbs light incidence.3-5 Dielectric materials are then adopted for metasurfaces design and fabrication with high optical efficiency across the spectrum of light, where  $HfO_2^6$  is used for UV;  $Si^7$ ,  $TiO_2^1$  and  $GaN^8$  are applied for visible, while  $\alpha$ -Si<sup>9-11</sup> and PbTe<sup>12</sup> are fabricated for infrared range. The large index dielectric nanoantenna strongly interact with light incidence to manipulate the optical properties.<sup>1, 8, 13</sup> Many applications such as metalens<sup>14,</sup>  $15$ , holography<sup>16-18</sup>, polarization beam splitter<sup>19, 20</sup> have been demonstrated. Moreover, compared with conventional single-function optical elements, metasurfaces can integrate multi-functions on one device, thus achieving several functions at the same time. $21-24$ 

At present the fabrication of metalens and other metasurface devices adopts two main routes: using electron beam lithography (EBL) to define the metasurface patterns into the resist layer and then depositing dielectric material, e.g., TiO<sub>2</sub>, by atomic layer deposition  $(ALD)^1$  or electron beam evaporation<sup>25, 26</sup>; while the second approach performs dry etching using either the patterned resist layer or lifted off metal layer as the mask to define the dielectric materials such as  $GaN<sup>15</sup>$  and  $TiO<sub>2</sub><sup>27</sup>$ . These methods typically suffer from high-cost and low-throughput and a limited aspect ratio restricted by the thin resist layer. To solve these problems, nanoimprint lithography (NIL) is investigated recently as a low-cost high throughput approach for the metasurfaces fabrication. G.Yoon et al proposed a single step nanoimprint fabrication of metasurfaces using UV-curable resin as a matrix with  $TiO<sub>2</sub>$  nanoparticles dopants.<sup>28</sup> C.A.Dirdal et al fabricated a Si metasurfaces using UV-nanoimprint and Bosch deep reactive ion etching.<sup>29</sup> However, although nanoimprint has proved able to successfully replicate sub-10 nm resolution nanostructures which is quite attractive for subwavelength devices, there are still some limitations for applying it in metasurface fabrication. As is shown in **Fig. 1.**, the schematic illustration of the metalens composed of

features with varying diameters, it's obvious a nonuniform structure. NIL is a process where features on the hard mold deform the polymer physically, which means that larger features need to displace more polymer over longer distance. Thus, when we try to fabricate nonuniform nanostructures like metasurfaces using NIL, the residual layer will be an important issue for consideration.



**Fig. 1.** Schematic diagram of the metalens and the basic unit nanopillar.



**Fig. 2.** Fabricated ZnO metalens SEM images

Here we demonstrated a multilayer-based nanoimprint lithography which can also solve the nonuniform residual layer issue. In our previous work we reported a metasurface subtractive color filter by solution phase epitaxy (SPE) growth of ZnO nanorods array.<sup>30</sup> ZnO is an ideal material for metasurface applications with a high refractive index and

low absorption in the visible wavelength. Combing the multilayer-based nanoimprint and SPE, we demonstrate a low-cost high-throughput fabrication of metalens, the focusing efficiency of the metalens is about 50% at 633 nm. **Fig. 2.** shows the SEM images of the metalens sample. The nanopillars at several positions are zoomed in. As can be seen from the images, the nanopillars heights are not uniform over the whole pattern area, which suffers from the loading effect during the epitaxy process, due to the variation of the local precursor concentration. This limits the focusing efficiency and needs to be optimized.

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