

# **Multi-functional antireflective surface-relief structures based on nanoscale porous germanium with graded refractive index profiles**

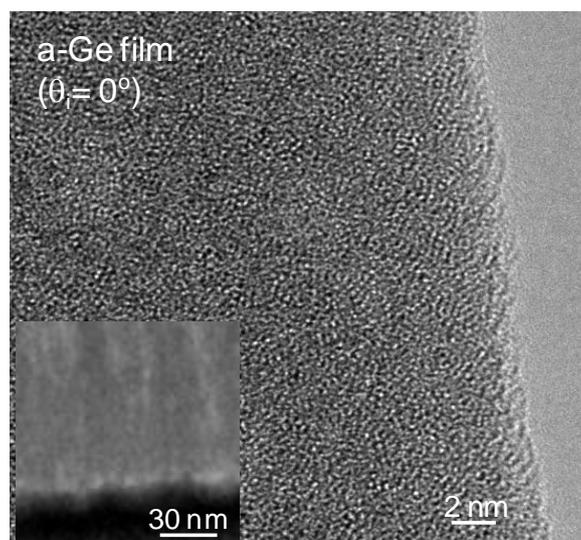
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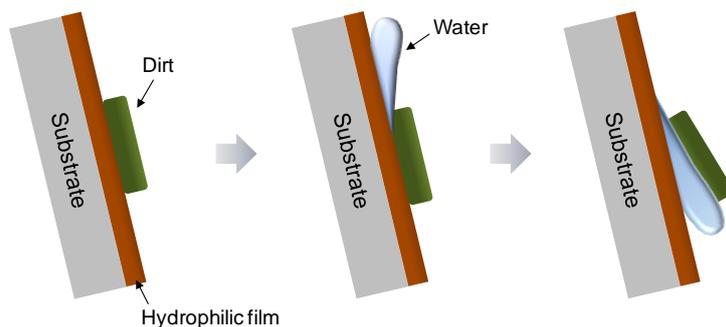
**Supplementary Information**

**S1. HRTEM image of the a-Ge film deposited by the GLAD at  $\theta_i = 0^\circ$ .**



**Fig. S1** HRTEM image of the a-Ge film deposited by the GLAD at  $\theta_i = 0^\circ$

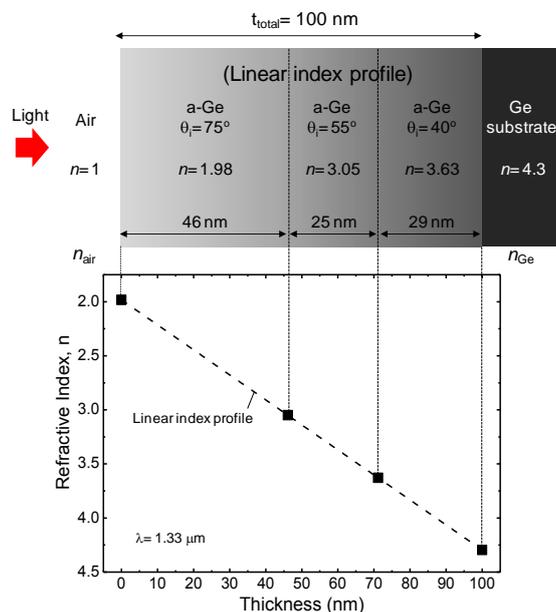
## S2. Mechanism of the self-cleaning on the hydrophilic surface.



**Fig. S2** Mechanism of the self-cleaning on the hydrophilic surface.

Fig. S2 shows the mechanism of the self-cleaning on the hydrophilic surface. The hydrophilic film attracts water and causes the droplets of water to spread out evenly like a thin film on the surface. This thin water film can squeeze into the space between the dusts and the hydrophilic film surface, and take the dusts away with the disappearing water due to the evaporation by the heat or the sliding down by the gravity. For the self-cleaning function on the hydrophilic surfaces, there have been some reports in previous studies.<sup>1,2</sup>

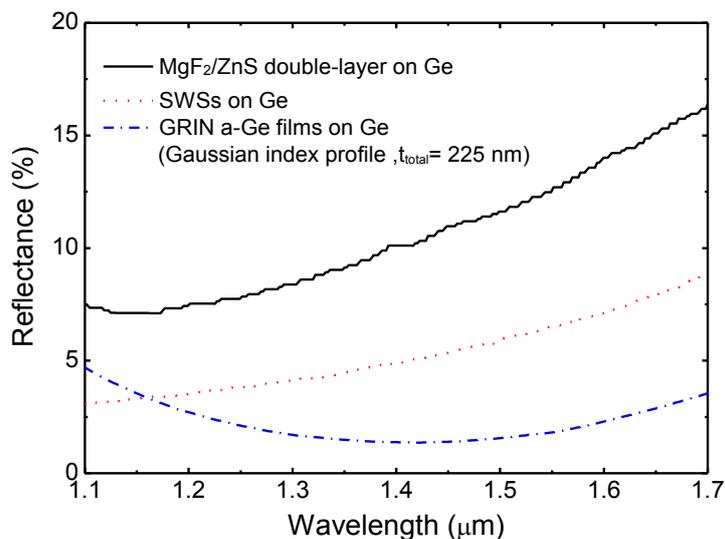
### S3. Calculation for each film thickness ( $t_i$ ) of the multi-stacked gradient-index (GRIN) a-Ge films.



**Fig. S3** Schematic diagram (upper) of the multi-stacked GRIN a-Ge films with the linear index profile and calculated linear index profile (lower) as a function of the thickness of a-Ge layers deposited by the GLAD at  $\theta_i = 40, 55,$  and  $75^\circ$  for  $t_{total} = 100$  nm. The effective refractive indices of three a-Ge layers are chosen at  $\lambda = 1.33 \mu\text{m}$ .

Fig. S3 shows the schematic diagram (upper) of the multi-stacked GRIN a-Ge films with the linear index profile and calculated linear index profile (lower) as a function of the thickness of a-Ge layers deposited by the GLAD at  $\theta_i = 40, 55,$  and  $75^\circ$ . The total thickness ( $t_{total}$ ) of the three porous thin films was set to be 100 nm for simplicity in this calculation. The effective refractive indices of three layers at  $\theta_i = 40, 55,$  and  $75^\circ$  are chosen at a wavelength of  $1.33 \mu\text{m}$  in ellipsometry measurements. Different a-Ge layers have different degrees of porosity, leading to the different refractive indices, as shown in Fig. 2(a). The GRIN profile theories were referred in previous reports.<sup>3-5</sup> For the linear, quintic, and Gaussian index profiles, the corresponding equations are described in the main article. In this design, their own film thicknesses ( $t_i$ ) were determined by the matching points between the calculated GRIN profiles and their own measured refractive indices at a certain wavelength for the given  $t_{total}$  value. From this, for the linear index profile, the  $t_i$  values of the a-Ge films at  $\theta_i = 40, 55,$  and  $75^\circ$  were evaluated to be 29, 25, and 46 nm, respectively, for  $t_{total} = 100$  nm at  $\lambda = 1.33 \mu\text{m}$  using the  $n$  values of 3.63, 3.05, and 1.98 at  $\theta_i = 40, 55,$  and  $75^\circ$ , respectively, as shown in Fig. S3. For the other quintic and Gaussian index profiles, their own  $t_i$  values can be also obtained by the same method. Therefore, for all the GRIN profiles, each  $t_i$  value of the a-Ge films at  $\theta_i = 40, 55,$  and  $75^\circ$  is proportionally changed with the variation ratio of the given  $t_{total}$  value from the calculated one in Table 1 for  $t_{total} = 100$  nm. For example, for the linear index profile, at  $t_{total} = 200$  nm, the corresponding  $t_i$  values of the a-Ge films at  $\theta_i = 40, 55,$  and  $75^\circ$  are estimated to be approximately 58, 50, and 92 nm, respectively. These values are also twice compared to the  $t_i$  values at  $t_{total} = 100$  nm. Using this theory, the reflectance properties on the variation of the  $t_{total}$  value for the multilayer consisted of porous Si nanocolumnar structures in the visible wavelength range have been studied in the previous report.<sup>3</sup>

#### S4. Comparison with other reported structures for the antireflection properties



**Fig. S4** Reflectance spectra of the MgF<sub>2</sub>/ZnS double-layer, SWSs, and GRIN a-Ge films with the Gaussian index profile at  $t_{\text{total}} = 225$  nm on Ge surfaces.

Fig. S4 shows the reflectance spectra of the magnesium fluoride (MgF<sub>2</sub>)/zinc sulfide (ZnS) double-layer, subwavelength structures (SWSs), and GRIN a-Ge films with the Gaussian index profile at  $t_{\text{total}} = 225$  nm on Ge surfaces. Firstly, N. E. Posthuma et al. have investigated the reflectivity of MgF<sub>2</sub>/ZnS double-layer on the Ge surface for Ge thermophotovoltaic cell applications, exhibiting the average reflectance ( $R_{\text{avg}}$ ) value of  $\sim 10.1\%$  in the wavelength region of 1.1-1.7  $\mu\text{m}$ .<sup>6</sup> Secondly, the antireflection SWSs inspired from the moth eye effect on Ge substrates have been reported in our previous work.<sup>7</sup> Among the Ge SWSs, the lowest  $R_{\text{avg}}$  value was  $\sim 5.2\%$  at wavelengths of 1.1-1.7  $\mu\text{m}$ . For the MgF<sub>2</sub>/ZnS double-layer and SWSs on the Ge surfaces, the  $R_{\text{avg}}$  values are higher than that (i.e.,  $R_{\text{avg}} \sim 2.2\%$  in the wavelength range of 1.1-1.7  $\mu\text{m}$ ) of multi-stacked GRIN a-Ge films with a Gaussian index profile at  $t_{\text{total}} = 225$  nm on the Ge surface. In addition, for the conventional  $\lambda/4$  thickness stacked multilayer ARCs consisting of different materials with high and low refractive indices, there exist some disadvantages such as thermal expansion mismatch, diffusion of one material into another, and material selections. In the case of the SWSs, the relatively expensive, complex, or damage-induced fabrication technique including the patterning and etching processes is required, leading to the increase of fabrication costs and complexity. Compared to other reports, therefore, the multi-stacked GRIN a-Ge films by the GLAD can provide the relatively more efficient broadband antireflection in the near-infrared wavelength region as well as the low-cost and simple fabrication method for Ge-based optical and optoelectronic device applications.

### Supplementary References

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