# High-reflection optical thin films based on $SiO_2/TiO_2$ nanoparticles multilayers by dip coating

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The alternately stacking layers of $SiO_2$ and $TiO_2$ nanoparticles with different refractive indices were fabricated to form the distributed Bragg reflectors on the silicon wafer by dip coating method. By appropriate design of the thickness of the quarter-wave layers, the peak reflectance region can be tuned from the blue-green to the infrared portion of the electromagnetic spectrum. The peak reflectance of 65% at 800 nm and at 1600 nm has been achieved using seven periods, respectively; meanwhile, compared with the simulation, 80% at 800 nm and 70% at 1620 nm for seven periods, respectively. The scanning electron microscopy and the atomic force microscope studies confirm the thickness uniformity achieved along the fabrication direction, and a good quality of surfaces and interfaces.

**1. Introduction:** Distributed Bragg reflectors (DBRs) or multi-layered mirrors known as one-dimensional photonic crystals (1D-PCs) have been attracting increasing interest in recent years because of their potentially wide range of applications [1–6]. For the highly reflective coatings, the reflectivity and stop bandwidth are directly affected by the refractive index difference between the high-n and low-n materials and the thicknesses of the quarter-wave layers. As a result, the larger refractive index contrast provides higher reflectivity and wider stop bandwidth. The organic and polymeric 1D-PCs have been fabricated by use of self-assembly of block copolymers, co-extrusion of two polymers and spin coating [5–11]. The inorganic dielectric materials, the periodic structures can be fabricated with well-controlled thin film thickness, based on chemical vapour deposition and molecular-beam epitaxy. However, many layers were needed for high reflection and only low reflectivity was attained at the designed wavelengths in these studies, due to an inaccurate control of the periodic structure and the low contrast of refractive indices. Over the past years, some research works have been reported on the highly reflective coatings formed with two different alternating dielectric materials, such as $SiN_x/SiO_x$, $Ta_2O_5/SiO_2$, $ZrO_2/SiO_2$ and so forth [8–11].

In this Letter, we reported the fabrication and characterisation of the reflective coatings with $SiO_2$ and $TiO_2$ nanoparticles through a simple dip-coating method. Cross-section scanning electron microscopy (SEM) studies were carried out to study the growth behaviour of both nanoparticles on each other and to elucidate the origin of the discrepancy between experimental and simulated reflectance spectra.

## 2. Experimental section

2.1. Materials: A low refractive index silica and high refractive index titania colloidal nanoparticles with diameter of 10–15 nm were supplied by Nisan Chemical. The silicon wafers were supplied by Nova Electronic Materials Ltd. Silicon wafers were immersed in acetone for 10 min, and then rinsed by acetone and iso-propyl alcohol and dried using an $N_2$ stream.

2.2. Method: The reflective coatings were fabricated on a silica wafer by dip coating method, that is the alternating layers of silica and titania colloidal nanoparticles, which have different refractive indices. Two kinds of the reflective coatings with a quarter wavelength of 800 nm (sample 1) and 1610 nm (sample 2), respectively, were fabricated. Firstly, the silicon wafers were immersed in the silica colloidal nanoparticles with 10% (sample 1) and 7.5% (sample 2) concentration, respectively, for 1 min, then were dipped with 0.26 mm/s (sample 1) and 1.06 mm/s (sample 2), respectively, and dried at the room temperature for 15 min. Then the silicon wafers coated with the silica nanoparticles were immersed in the titania colloidal nanoparticles with 15% (samples 1 and 2) concentration for 1 min, then were dipped with 0.51 mm/s (sample 1) and 2.45 mm/s (sample 2), respectively, and dried at the room temperature for 15 min. Repeat the above process until we get satisfactory results. The measured effective index of silica and titania nanoparticles is 1.42 and 1.63, respectively.

Transmittance and reflectance of the coatings are measured under normal incidence using a JASCOV570UV/vis spectrophotometer in the spectral range of 400–2000 nm. The microstructure of the coatings is studied with a Zeiss SUPRA55VP scanning electron Microscope and Zeiss atomic force microscope (AFM).

**3. Results and discussion:** Fig. 1 shows the SEM and AFM images of single films of $SiO_2$ and $TiO_2$ nanoparticles. Seen from

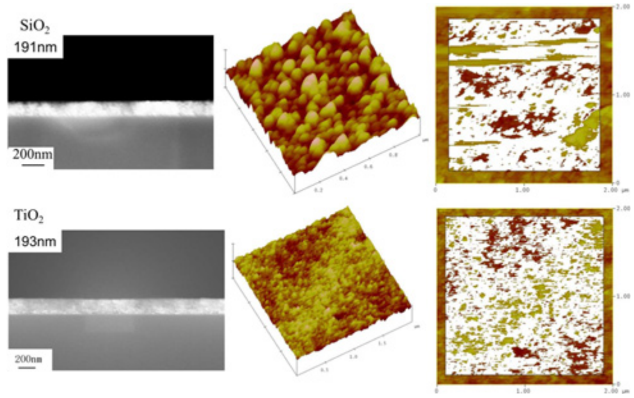


**Fig. 1** *SEM and AFM images of $SiO_2$ and $TiO_2$ nanoparticles films by dip coating*

Fig. 1, the root-mean-square roughness value of $SiO_2$ and $TiO_2$ nanoparticles is 2.315 and 1.461 nm, respectively. The results showed that $SiO_2$ and $TiO_2$ nanoparticles films prepared by dipping coating exhibited the best surface flatness.

We designed two kinds of the reflective coatings consisting of alternating layers of quarter-wave-thick $SiO_2/TiO_2$ nanoparticles on silicon substrates. The thickness of the layer is calculated according to the optical multilayers film theory. One consists of the alternating layers of 140 nm $SiO_2$ nanoparticles ($n_{SiO_2} = 1.42$) and 122 nm $TiO_2$ nanoparticles ($n_{TiO_2} = 1.63$), and the reflective wavelength is 800 nm. The other consists of the alternating layers of 280 nm $SiO_2$ nanoparticles and 240 nm $TiO_2$ nanoparticles, and the reflective wavelength is 1610 nm.

Figs. 2 and 3 show the reflectance spectra, SEM image and thickness versus number of layers of both of the reflective coatings, respectively. The inset is the photo of the sample. Seen from Fig. 2*a*, 65% reflectance is obtained in the wavelength ranges between 720 and 880 nm after seven periods of $SiO_2/TiO_2$ nanoparticles film with alternating layers. In Fig. 2*b*, the microstructure of a multilayer stack with the different refractive index is clearly seen. The dark bands are $TiO_2$ layers, and the bright bands are $SiO_2$ layers. In Figs. 2*b* and *c*, we can see that the experimental thickness of the layer did not completely match with a quarter wavelength of 800 nm; the thickness of $TiO_2$ nanoparticles film in the second and third periods is higher than that of the theoretical value. The thickness of layer has good repeated

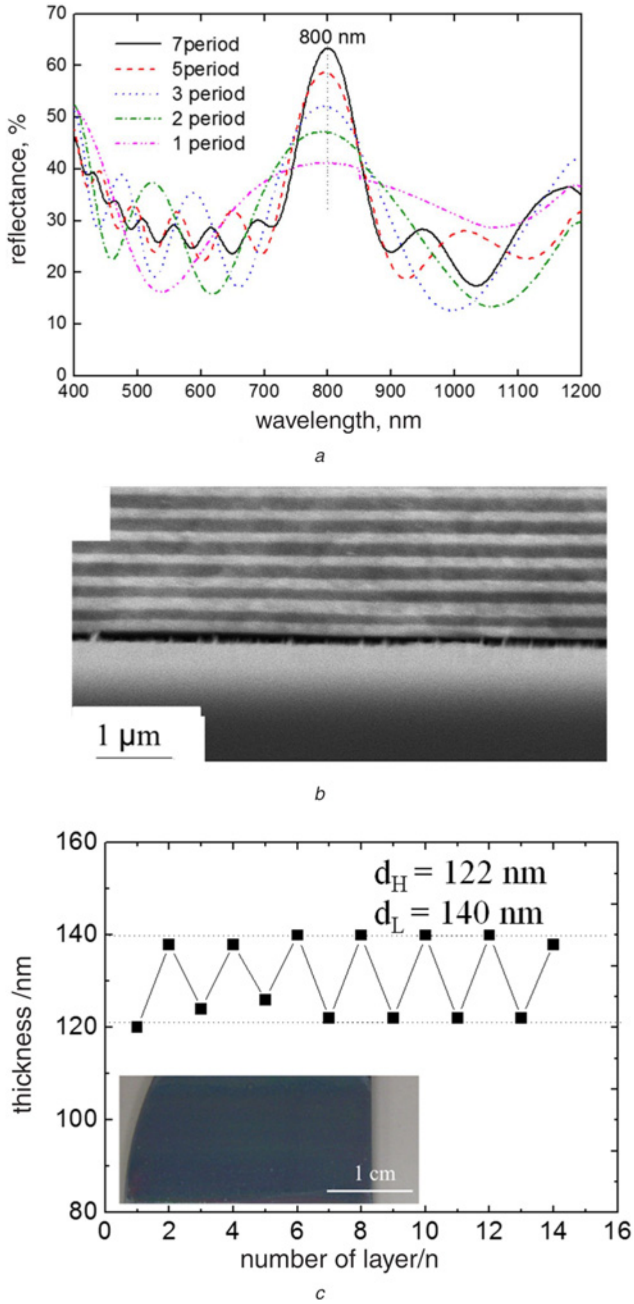


**Fig. 2** *Reflectance spectra, SEM image and thickness versus number of $SiO_2/TiO_2$ nanoparticles film with alternating layers. The inset is the photo of the sample*
*a* Reflectance spectra of the sample
*b* SEM image of the sample
*c* Thickness versus number of $SiO_2/TiO_2$ nanoparticles film with alternating layers. The inset is the photo of the sample

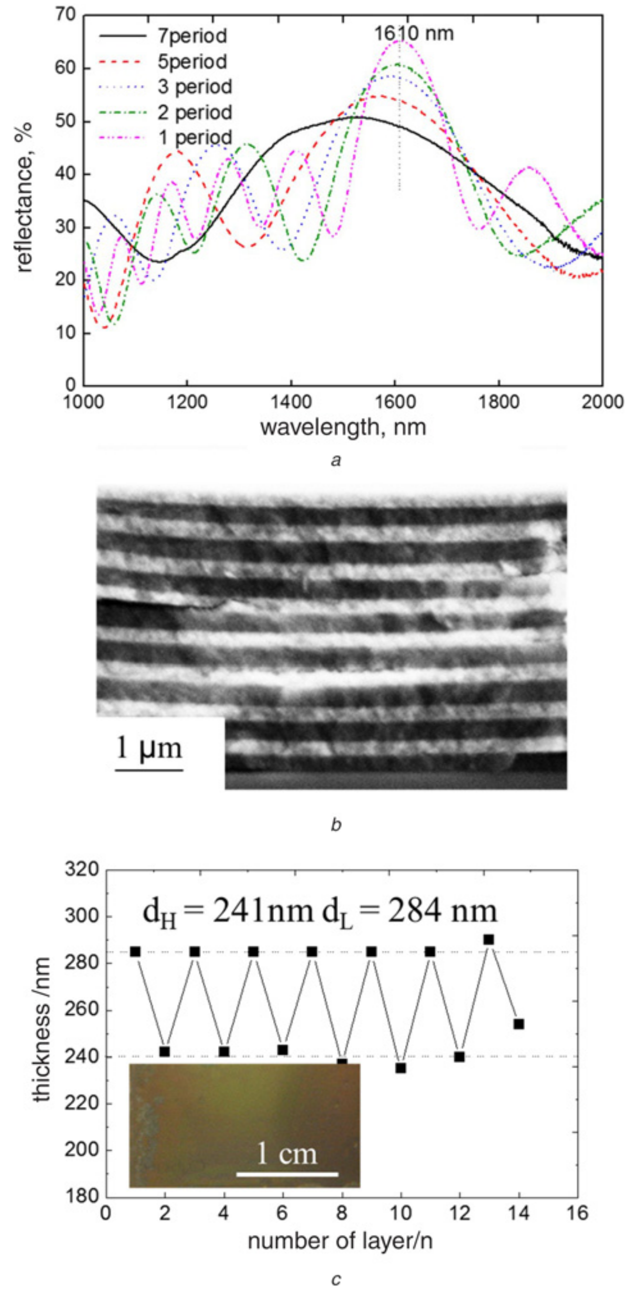


**Fig. 3** *Reflectance spectra, SEM image and thickness versus number of $SiO_2/TiO_2$ nanoparticles film with alternating layers. The inset is the photo of the sample.*
*a* Reflectance spectra of the sample
*b* SEM image of the sample
*c* Thickness versus number of $SiO_2/TiO_2$ nanoparticles film with alternating layers. The inset is the photo of the sample




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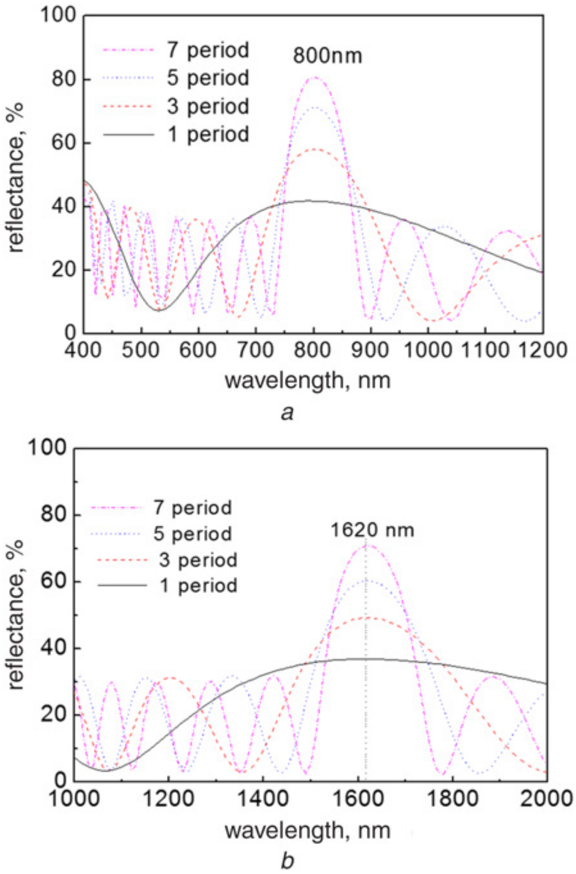


**Fig. 4** *Illustrates theoretical reflectance spectra of two structures of seven periods of $SiO_2/TiO_2$ nanoparticles film with alternating layers*

values in the last four periods. The colour of the sample is deep blue.

Seen from Fig. 3*a*, 65% reflectance is obtained in the wavelength ranges between 1470 and 1770 nm after seven periods of $SiO_2/TiO_2$ nanoparticles film with alternating layers, and the peak of reflectance spectrum red-shifts with the increase of period. We think that the spectral red-shifts may be related to the light scattering at interfaces and defects in the film. The microstructure shows that there are some cracks in the film. The experimental thickness of layer matches with a quarter wavelength of 1610 nm. The colour of the sample is brown-green.

Fig. 4 illustrates the theoretical reflectance spectra of two structures of seven periods of $SiO_2/TiO_2$ nanoparticles films with alternating layers. The reflectance for both of the structures is about 80 and 70%, respectively, is higher than experimental value. We think that the difference between the theoretical and experimental reflectance spectra may be attributed to thickness-deviation, light scattering at interfaces, and defects. Seen SEM images from Figs. 2 and 3, the experimental thickness of the layers is not in good accordance with theoretical quarter wavelength and there are some cracks in the film due to the stress and expansion parameter of thin films. With the increase of the number of layers the stress and expansion parameter of thin films keeps changing so that the thickness of the layer is not easy to be controlled and the defects gradually increases.

**4. Conclusion:** We have succeeded in fabricating the alternately stacking layers of $SiO_2$ and $TiO_2$ nanoparticles of different refractive indices to form the DBRs on the silicon wafer by dip coating method. By appropriate design of the thickness of the quarter-wave layers, the peak reflectance region can be tuned from the blue-green to the infrared portion of the electromagnetic spectrum. The peak reflectance of 65% at 800 nm and at 1610 nm has been achieved using seven periods, respectively, compared with the simulation, 80% at 800 nm and 70% at 1620 nm for seven periods, respectively. The SEM and the AFM studies confirm the thickness uniformity achieved along the fabrication direction, and a good quality of surfaces and interfaces. The reflection optical thin films using inorganic nanoparticles quarter-wave stacks provided a glimpse at the wide range of coating and photonic device applications where dip coating can be used.

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