

# All-dielectric metameric filters for optically variable devices

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In order to increase the anti-counterfeiting performance of optically variable devices, the innovative interference security image structures based on metamerism have been developed. In this letter, we show a pair of all-dielectric metameric filters offering a hidden image effect with the color shift at a specific angle of observation. These filters are designed by two materials TiO<sub>2</sub>/SiO<sub>2</sub> based on the different angle color target optimization. The 6-layer- and 9-layer stacks are achieved and the performance of prototype filters prepared by remote plasma sputtering is shown. The color difference index of the experiment is up to 1.19, which shows good metameric matching effect.

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With the development of science and technology, the counterfeiting of banknotes, valuable documents, common household items and something else is constantly increasing<sup>[1]</sup>, and there are different degrees of shoddy goods. As a result, it is not only harmful to the country's economy, but also has a negative impact on the public security because most counterfeit products fail to meet the safety standards. Developing advanced security technology becomes increasingly important<sup>[2]</sup>. The latest technology for anti-counterfeiting is optically variable devices, and they offer an interesting variation of color as a function of the observation angle; this character not only makes them simple to authenticate but also inhibits reproduction by most reprographic techniques, such as printing, scanning, etc.

The idea of using color shifting optical coatings to prevent counterfeiting of value documents such as bank notes, stock certificates, visas, passports, and the like, by color copiers and color printers was first suggested by Dobrowolski *et al.* in 1973<sup>[3]</sup>. In recent years, Bill Baloukas *et al.* have published a series of metamerism for security<sup>[4-8]</sup>. In 2008, they have fabricated two metameric all-dielectric filters *A* and *B*, which consist of 19 and 15 layers, respectively. Under illuminant *D*65, the color difference is 15.51.

In this letter, we design and fabricate a pair of metameric all-dielectric filters *A* and *B*, which have less layers and lower color difference index. Layers and color difference index at normal incidence under illuminant *D*65 of 6 layers, 9 layers and 1.1885, respectively. As the angle of incidence is increased, filter *A* varies from green to blue, while filter *B* goes from green to red. The detailed design process is given and the experimental samples are shown.

As defined by the International Commission on Illumination (CIE), two objects displaying the same color (identical tristimulus values) under a specific illuminant and for a specific observer will be termed metameric if their spectral distributions differ from each other in the visible spectrum<sup>[9]</sup>.

Stiles and Wyszecki found<sup>[10]</sup>: if a pair of different spectrally color stimulus want to match in color appearance under a certain view condition, the spectral reflection curves in the visible spectral region (400–700 nm), at least in three different wavelengths, must have the same value, that is, two reflectance curves have three intersections at least.

In order to evaluate the degree of metamerism more accurately, we use the  $L^*a^*b^*$  color space, and considered more perceptually linear than the  $xyY$  color space. The  $L^*a^*b^*$  coordinates are related to the  $XYZ$  coordinates by<sup>[11]</sup>

$$L^* = 116 \left( \frac{Y}{Y_0} \right)^{1/3} - 16 \quad Y/Y_0 > 0.008856, \quad (1)$$

$$a^* = 500 \left[ \left( \frac{X}{X_0} \right)^{1/3} - \left( \frac{Y}{Y_0} \right)^{1/3} \right] X/X_0 > 0.008856, \quad (2)$$

$$b^* = 200 \left[ \left( \frac{Y}{Y_0} \right)^{1/3} - \left( \frac{Z}{Z_0} \right)^{1/3} \right] Z/Z_0 > 0.008856, \quad (3)$$

where  $X_0$ ,  $Y_0$  and  $Z_0$  are the tristimulus values of the illuminant.

The color difference  $\Delta E_{ab,I}^*$  is obtained as

$$\Delta E_{ab,I}^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}. \quad (4)$$

The value of  $\Delta E_{ab,I}^*$  for a hardly perceptible color difference is close to 1.0<sup>[12]</sup>, a value which we will use as our metamerism limit.

In the initial stage of the work on metameric filters, the metameric filters' color change frame should be confirmed. As we know, this metameric color match strongly depends on the illumination source. The initial color design is green for the pair filters *A* and *B* under illuminant *D*65 at normal incidence for use in reflection.

To achieve the dynamic color display effect, we hope the pair metameric filters will have the obviously different color contrast at the oblique viewing angle. Here, the filter *A* will have the green-to-blue color with viewing angle  $60^\circ$ , and the filter *B* will have the green-to-red color with viewing angle  $60^\circ$ . To perform the color optimization design, the different viewing angle color coordinates of green, blue and red color should be given. Here, the calculation on metameric color of the green is performed in the  $L^*a^*b^*$  color space. The other color targets such as blue and red are performed in the  $xyY$  color space. The detailed color coordinates are showed for filter *A*, *B* in the Table 1.

The designs of filters *A* and *B* are all used in the all-dielectric materials, and they consist of a low-index material,  $\text{SiO}_2$  (index of refraction  $n$ : 1.486 at 550 nm) and a high-index material,  $\text{TiO}_2$  ( $n$ : 2.401 at 550 nm), both with negligible absorption in the visible region (380–780 nm). Based on the color theory, the filters' transmission color will be completely complementary color with the reflection color. The synthesis optimization method needle and refinement method simplex are used from the initial stack HL. The final designs of filters *A* and *B*, which consist of 6 and 9 layers respectively, are given as follows: filter *A* (*Sub/0.548H0.293L0.397H0.334L0.985H0.809L/Air*) and filter *B* (*Sub/0.339H0.391L0.268H0.255L0.284H0.367L0.246H0.35L0.05H/Air*).

Fig. 1(a) shows the all dielectric filter *A* design spectra shift with viewing angle in reflection and Fig. 1(b) shows the filter *A* color variation track in  $xy$  color space as a function of the viewing angle (green-to-blue).

Fig. 2(a) shows the metameric filter *B* design spectra shift with viewing angle in reflection, and Fig. 2(b) shows the filter *B* color variation track in  $xy$  color space as a function of the viewing angle (green-to-red).

Visibly, the two filters start off at the same color coordinates, but as the angle of incidence is increased,

their color paths diverge. This frame was designed specially in order to maximize the color difference at oblique incidence. Filter *A* varies from green to blue, while filter *B* goes from green to pink. This difference in color is what permits the creation of a hidden image that appears only at oblique incidence. The color difference ( $\Delta E_{ab,D65}^*$ ) calculated based on Eq. (4) is 0.56. For  $\Delta E^*$  values lower than 1, no color difference can be observed, and, evidently, for  $\Delta E^*$  higher than 1 the color difference increases. Usually, the color pairs possessing  $\Delta E^* \leq 2$  will be considered metameric, which is still a very good compromise in order to add more flexibility to the design process and to lower the number of layers.

Fig. 3 shows the reflection spectra intersection of the metameric filters *A*, *B*.

It can be seen there are six intersection points for the filter *A*, *B* in the visible spectrum range. Therefore, the design result shows a super metameric match performance. Fig. 4 shows the metameric filters color variation path as a function of the viewing angle ( $0^\circ$ -to- $60^\circ$ ) in reflection (a) and transmission (b).

The filters were fabricated using the remote plasma sputtering technique<sup>[13]</sup>. The technique relies on the generation of plasma remotely from the targets, and the internal magnetic elements behind the target are eliminated. The argon ions are accelerated into the target resulting in a high-energy plasma over the full surface area of the target. Hence, fully uniform erosion over the surface of the target is realized and results in a significant reduction in target poisoning. This allows for a uniform reaction in the plasma phase when performing reactive sputtering, leading to the formation and deposition of material with a uniform stoichiometry. The systems use cryo-pump with a base pressure of  $6 \times 10^{-6}$  Torr. And the oxygen is fed into the chamber through another diffusion ring placed as close as possible to the substrate. Silicon (99.999% purity) and titanium

**Table 1. The filter *A*, *B* design reflection color target with the different viewing angle**

Viewing/Color	$x$	$y$	$L^*$	$a^*$	$B^*$
$0^\circ$ ( <i>A</i> , <i>B</i> green)	0.3149	0.5508	66.932	-55.7026	56.1422
$60^\circ$ (Filter <i>A</i> , blue)	0.2563	0.2510	55.1526	7.4029	-26.6303
$60^\circ$ (Filter <i>B</i> , red)	0.3845	0.3170	69.6846	31.3020	6.9623

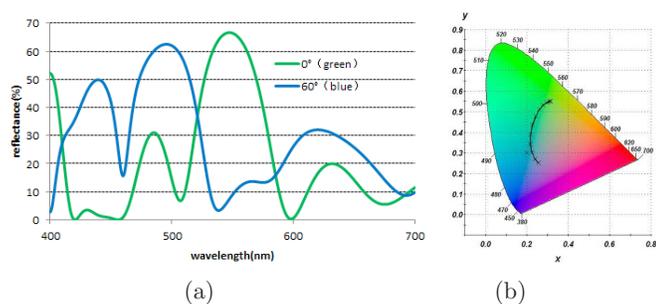


Fig. 1. (a) The reflection spectra of filter *A* at the  $0^\circ$  and  $60^\circ$  viewing angle; (b) Color variation in the CIE  $xy$  color space of filter *A* as a function of the viewing angle ( $0^\circ$ – $60^\circ$ ).

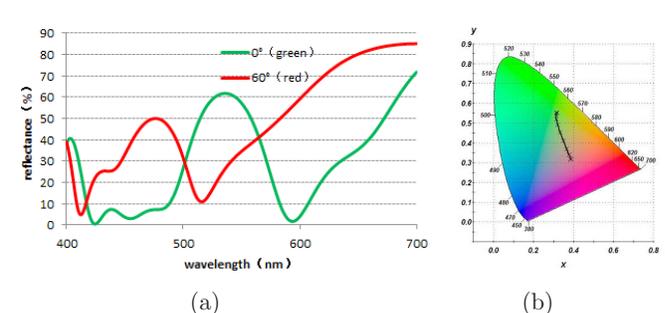


Fig. 2. (a) The reflection spectra of filter *B* at the  $0^\circ$  and  $60^\circ$  viewing angle; (b) Color variation in the CIE  $xy$  color space of filter *B* as a function of the viewing angle ( $0^\circ$ – $60^\circ$ ).

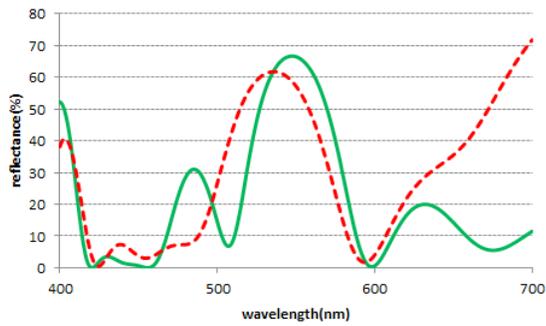


Fig. 3. The filter *A* (solid) and *B* (dash)spectra intersection.

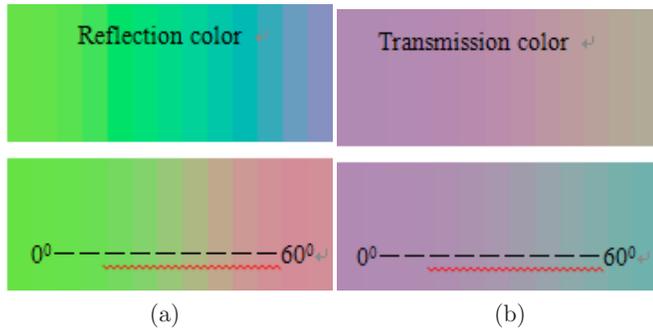
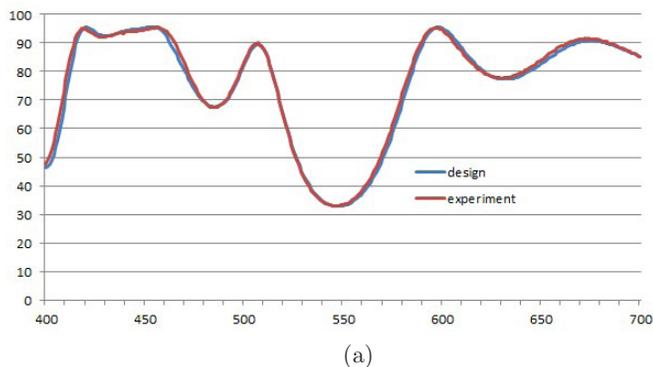


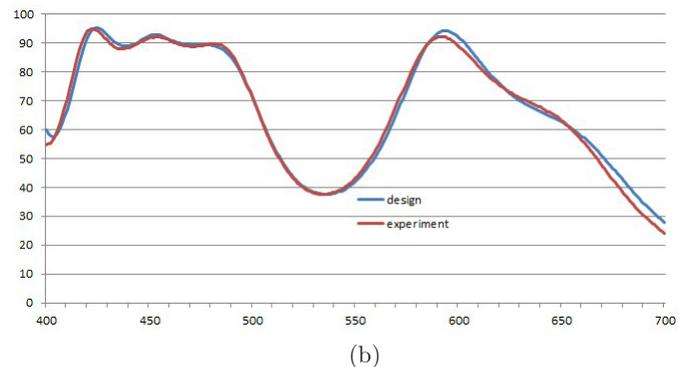
Fig. 4. (a) The color variation path of filter *A*, *B* in reflection; (b) the color variation path of filter *A*, *B* in transmission.

(99.99% purity) targets are used in direct current (DC) sputtering mode. By adding  $O_2$  as well as Ar to the chamber,  $SiO_2$  and  $TiO_2$  were deposited. The quartz glasses with high surface quality of 10/5 scratch-dig are used to deposit the films. The deposited film thickness is monitored by time-power. So, the deposition optimization conditions were well performed and the deposit rates of the  $SiO_2$  and  $TiO_2$  were accurately calibrated by the single-layer deposition. The rate of  $SiO_2$  is 0.35nm/s, and the rate of  $TiO_2$  is 0.55nm/s.

For the all-dielectric coating, the transmission color observation is easily more than reflection, and the transmission spectra measurements are more convenient and accurate. So, the optical transmittances of the samples were measured using Perkin-Elmer Lambda 750 spectrometer. As shown in Figs. 5(a) and (b), the spectra of filters *A* and *B* are compared with the design and experiment spectra. The spectra of filter *A* is



(a)



(b)

Fig. 5 (a) Transmission spectra of filter *A* with design and experiment; (b) Transmission spectra of filter *B* with design and experiment.

well matched, and the spectra of filter *B* has a little slight variation. The color coordinates for the  $xyY$  and Lab spaces were calculated by the measurements data, and the experimental final color difference ( $\Delta E_{ab,D65}^*$ ) is up to 1.19 in transmission. The data still show better metameric matching degree. The experimental samples for filter *A* and *B* are taken pictures to show the color practical change effect in Fig. 6. In transmission, the purple is observed of the metameric filters with viewing angle close to  $0^\circ$ ; when changing the viewing angle close to  $60^\circ$ , the filter *A* shows the fawn and filter *B* shows baby blue. The experimental results show the well matching with the design color path.

We show the innovative all-dielectric metameric filters for the next generation of optically variable devices due to an increased fabrication complexity and ease of authentication. Based on the color target optimization approaches, the perfect color matching filter structures has been realized by less layers and provide two distinct color shifts, as so for the best experimental result of the 1.19 ( $\Delta E_{ab,D65}^*$ ) color difference index is achieved for the dielectric metameric filters frame. On-going work has been focusing on developing devices containing metal-dielectric layers in order to reduce the cost and increase the practicability.

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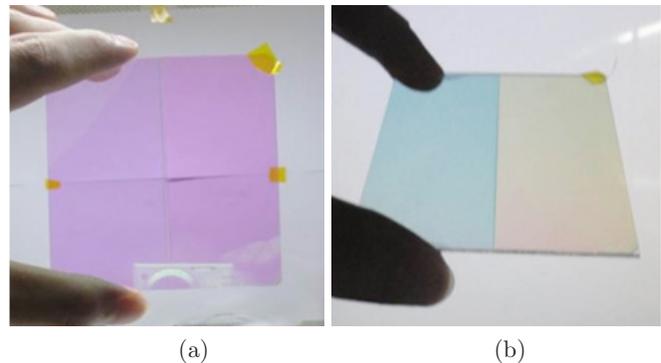


Fig. 6. The pictures of the metameric filters *A* and *B* with viewing angle ( $0^\circ$ – $60^\circ$ ).

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