Novel metal-dielectric metameric optical filters for optical security devices*

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Optical security devices play an essential role in the fight against counterfeiting. In this paper, we study and design a pair of metal-dielectric optical filters based on metameric effect, which offer a hidden image effect by the color shift at a specific angle of observation. Compared with all-dielectric multilayer system, the metal-dielectric multilayer structure has larger color shift with varying incident angle, higher color saturation and fewer layers. Finally, the stacks with 5 layers and 7 layers are achieved, and the color difference index is only 0.71, which shows good metameric matching effect. Simultaneously, the sensitivity of filters to deposition errors is analyzed when the thickness deviation is ±2%, and the results show that the two filters have good manufacturability.

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The rapid development of optical anti-counterfeiting technology brought new high-tech means to the field of security. An innovative optical anti-counterfeiting concept based on metamerism was proposed by Bill Baloukas in 2006[1], and then the deep research work was proceeded for a combination of two different interference filters or a combination of an interference filter and a non-iridescent material (NIM)[2-4]. In 2008, a pair of metameric all-dielectric filters was fabricated by dual ion beam sputtering, which consist of 19 and 15 layers, respectively[5].

The optically variable devices can be based on metal-dielectric or all-dielectric multilayer system[6]. Compared with all-dielectric multilayer system, the metal-dielectric multilayer structure has larger color shift with varying incident angle, higher color saturation and fewer layers. In this paper, we investigate the design of metal-dielectric metameric filters.

To achieve the obvious color contrast effect for the pair of metameric filters, the clockwise color shift model is proposed, which makes the two metameric filters have the opposite color change trajectory. We design a pair of metal-dielectric filters of A and B, which consist of 5 layers and 7 layers, respectively. The metal materials of Al and Cr and the dielectric materials of SiO_2 and TiO_2 are used in design based on the optimization of multiple color targets. The color difference index is only 0.71. As the viewing angle is tilted, filter A shifts from green to blue-violet, while filter B goes from green to red.

When two objects have different reflection or transmission spectra but display the same color under a specific light source, for a specific observer, metamerism presents. A pair of metameric colors should meet the conditions as[7]

$$\begin{cases} X = k\int_\lambda \varphi_1(\lambda)\bar{x}(\lambda)\mathrm{d}\lambda = k\int_\lambda \varphi_2(\lambda)\bar{x}(\lambda)\mathrm{d}\lambda \\ Y = k\int_\lambda \varphi_1(\lambda)\bar{y}(\lambda)\mathrm{d}\lambda = k\int_\lambda \varphi_2(\lambda)\bar{y}(\lambda)\mathrm{d}\lambda \\ Z = k\int_\lambda \varphi_1(\lambda)\bar{z}(\lambda)\mathrm{d}\lambda = k\int_\lambda \varphi_2(\lambda)\bar{z}(\lambda)\mathrm{d}\lambda \end{cases}, \quad (1)$$

where X, Y and Z are the tristimulus values, $\varphi(\lambda)$ is the relative spectral energy distribution of the light source, $\bar{x}(\lambda)$, $\bar{y}(\lambda)$ and $\bar{z}(\lambda)$ are the relative quantities of the primary colors of the Commission International de l'Eclairage (CIE) 1931 color system, and k is a normalization factor.

Fig.1 shows the conceptual example of metameric filters. In this case, films A and B match with each other in color and display the same green, thus the pentagram pattern B is invisible at normal incidence. While changing the observation angle to 60°, film A begins to shift from green to blue, film B goes from green to red, and the hidden pentagram pattern appears.

Generally, there are two methods to evaluate the degree of metamerism, which are qualitative and quantitative methods[8]. Qualitative method is the reflectance spectra evaluation method. Intuitively, if two objects possess the same color while the spectral distributions differ

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widely, the degree of metamerism is high, and vice versa[9]. To quantify metamerism, we need to calculate the color difference ($\Delta E^*_{ab,\mathrm{I}}$) between two objects. The color difference between object 1 and object 2 is given as

$$\Delta E^*_{ab,\mathrm{I}}=\sqrt{(L^*_2-L^*_1)^2+(a^*_2-a^*_1)^2+(b^*_2-b^*_1)^2}\ , \qquad (2)$$

where L^*_i, a^*_i and b^*_i are the lightness and color-opponent dimensions of object *i*, respectively. I indicates the illuminant under which the color difference is calculated. When $\Delta E^*_{ab,\mathrm{I}}$ <2, it is considered to be acceptable, which means that very little color change can be observed by human eyes[10]. In this paper, the two methods are used to evaluate the degree of metamerism.

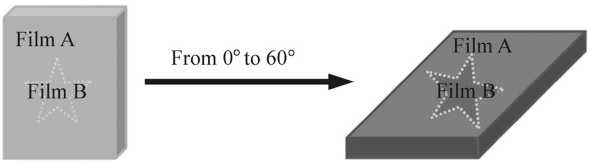


Fig.1 Conceptual example of metameric filters

In general, due to the change of optical thickness, the spectra of interference filters shift towards shorter wavelengths when the angle of incidence is increased. Therefore, the usual color change is from red to green then to blue. If the color shifts from green to red, the color variation path is completely on opposite side of the visible spectrum, which gives the conclusion that when we view at angle of a reverse shift, the anti-counterfeiting complexity and color varying effect are definitely increased. To obtain this effect, the double reflection peaks model is proposed. As can be seen in Fig.2, there must be a reflection peak in the green wavelength region, and the other reflection peak is in the near-infrared (NIR) wavelength region at normal incidence, so the color display is green. If the positions of the peaks are carefully chosen, when the filter is tilted, the green peak continues to shift to blue and disappears in the ultraviolet (UV) wavelength region, while the red peak moves from the near-infrared (NIR) into the visible wavelength region. Because this effect is more difficult to obtain, the difficulty of counterfeiting is increased.

Based on optical coating interference theory, the dielectric spacer layer can be used for inducing highly reflecting metal film to obtain the maximum reflectivity at the central wavelength, and the metallic film with high absorption can be used to achieve a wide-band cutoff. The classical structure of the metal-dielectric filter[11] is air/M1/D/M2 /glass, where M1 is an internal reflective metal acting as an absorber layer, D is a low-index dielectric material acting as a spacer layer, and M2 is an opaque and highly reflecting metal film.

The filters presented in this paper are designed based on multiple color targets optimization methods. The color optimization is performed in the *xyY* and $L^*a^*b^*$ color spaces, using standard illuminant D65 (average daylight with a correlated color temperature of 6 500 K).

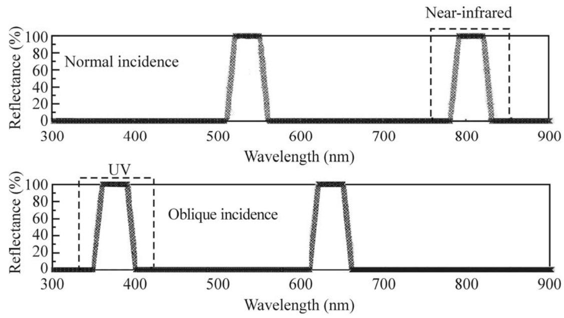


Fig.2 Clockwise color shift model

We concentrate on designing two metameric metal-dielectric filters for the use in reflection under illuminant D65. In our design, the central wavelength is 559 nm. The coating materials are as follows: metals are Cr (M1) and Al (M2), and dielectric materials consist of SiO_2 with refractive index *n* of 1.486 at 550 nm and TiO_2 with *n* of 2.401 at 550 nm. In order to achieve a dynamic color-display effect, as the viewing angle changing from 0° to 60°, we set the color of filter A from green to blue and that of filter B from green to red. The detailed color coordinates of filters A and B are given in Tab.1.

Tab.1 The designed reflection color targets of filters A and B with different viewing angles

Viewing angle (Color)	*x*	*y*	L^*	a^*	b^*
0°(A, B green)	0.364 2	0.477 6	47.717	–19.443	35.960
60°(Filter A, blue)	0.214 5	0.244 2	41.660	–2.690	–19.990
60°(Filter B, red)	0.514 8	0.335 5	52.125	58.772	33.712

Filters A and B have the identical initial stack as air/Cr/SiO_2/Al/glass. Firstly, for the filter A, the color change is from green to blue, this is a usual color realization by the typical film stack, so the color optimization target can be built only by *xyY* color space in 0° green and 60° blue. The finial film stack of filter A is with 5 layers as air/0.014Cr/0.701SiO_2/0.026Cr/0.251SiO_2/0.152Al/glass.

By $L^*a^*b^*$ color space, the $L^*a^*b^*$ coordinates of filter A with angle of incidence (AOI) of 0° are achieved. These can be used as the metameric color targets for filter B to assure the same green color display at normal incidence, while red *xyY* coordinates of filter B color target with AOI of 60° are set to assure the clockwise color shift. Therefore, for the filter B, the color optimization is based on the multiple color targets for the different color spaces, which leads to very little leeway during optimization. The synthesis optimization method is used to add the layers and TiO_2 material in the initial film stack, for avoiding the wrong convergence direction, the different tolerances are given for the color target, and the refinement method is used by fine-tuning the spectrum to minimize the final color difference. The final design of filter B consists of 7 layers as air/0.222SiO_2/0.02Cr/0.838TiO_2/0.471SiO_2/0.051Cr/0.561TiO_2/0.152Al/glass.

Fig.3 shows the reflection spectra of filters A and B. For Fig.3, let us consider the luminous efficiency curve of the human eye, i.e., human eye is practically insensitive to wavelengths below 400 nm and above 700 nm. Thus, we can see from Fig.3 that as the viewing angle is rotated from 0° to 60°, the color shift of the filter A is from green to blue-violet, and that of filter B is from green to red.

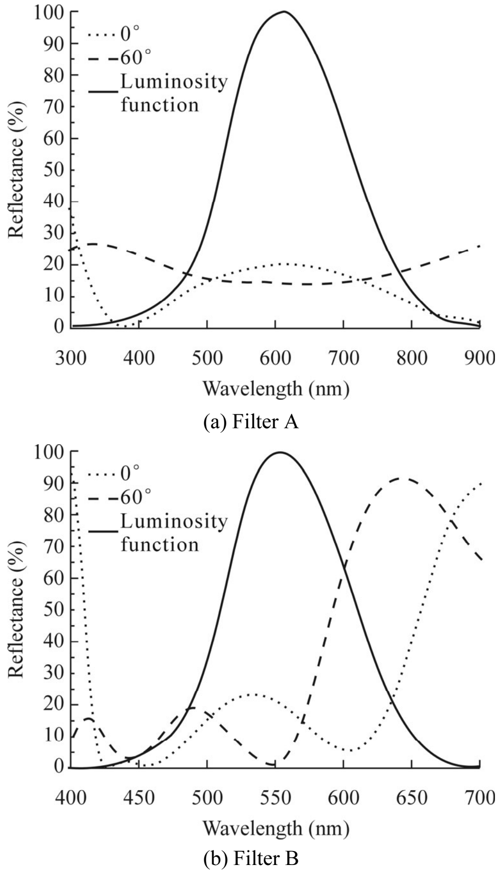


Fig.3 The reflection spectra of filters A and B at the viewing angles of 0° and 60°

Fig.4(a) shows the reflection spectra of the metameric filters A and B. It can be noted that the spectra of filters A and B are very different, and these filters have six intersections (as a rule of thumb, the minimum number of intersections for metamerism to be possible is three[12]). Fig.4(b) shows the color variation of filters A and B in *xyY* color space as a function of the viewing angle. As can be seen in Fig.4(b), at the beginning, they are almost at the same *xyY* coordinates. As the viewing angle is tilted, filter A varies from green to blue-violet following the simple color shift, while filter B goes from green to red following the clockwise color shift. This difference in color permits the creation of a hidden image which appears only at oblique incidence. To some extent, it increases the difficulty of anti-counterfeiting and enhances the intensity of the security.

Fig.5 presents the color variation paths with interval of 5° in reflection for metameric filters. Visibly, their colors at normal incidence match very well under illuminant D65. The color difference of $\Delta E^*_{ab,\mathrm{D65}}$ calculated by Eq.(2) is only 0.71, so no color difference can be perceived.

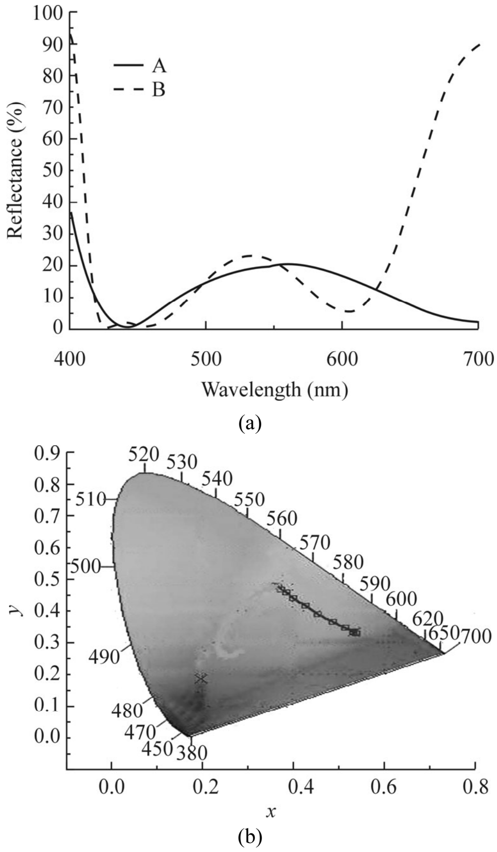


Fig.4 (a) The spectra of filters A and B; (b) Color variation in the *xy*Y color space for filters A and B

In order to show the advantages of metal-dielectric structure clearly, the comparison among three pairs of different metameric filters is shown in Tab.2. Samples 1 and 2 are all-dielectric metameric filters which have been fabricated by Bill Baloukas' team and our lab, respectively, and Sample 3 is the metal-dielectric filter proposed in this paper. An unnoticeable color difference value of metameric structure is below or equal to 1. We can see from Tab.2 that the color difference indexes are all lower than 1, and the metal-dielectric filters have less layers and higher color saturation.

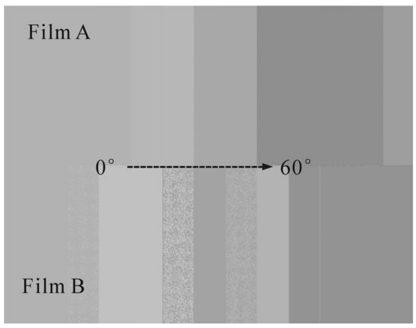
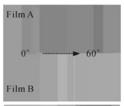
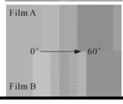


Fig.5 The color variation paths of filters A and B in reflection

Tab.2 The comparison among three pairs of metameric filters

Sample	Filter A layers	Filter B layers	Designed $\Delta E^*_{ab,\mathrm{D65}}$	Experimental $\Delta E^*_{ab,\mathrm{D65}}$	Color path in reflection
1	19	15	0.24	15.51	
2	6	9	0.56	1.19	
3	5	7	0.71		

If one wants to obtain a good color match at normal incidence, a more precise control of the layer thickness is needed. Because the SiO_2 layers are thick, just using traditional optical control method, a good result can be achieved. But for Cr layers, because the thickness is only a few nanometers, the deposition error is difficult to control and has a great effect on color. Therefore, we study the effect of thickness deviation of Cr layers. It can be seen from Fig.6 that when the thickness deviation of Cr layers is ±2%, both spectral properties of filters A and B suffer from a very slight shift. In order to accurately reflect the effect, we calculate the color difference ($\Delta E^*_{ab,\mathrm{D65}}$) between filters A and B. For thickness deviation of –2%, the color difference is 1.41, and for thickness deviation of 2%, the color difference is 1.57. The color differences are both lower than 2, which shows that filters A and B are not very sensitive to deposition errors.

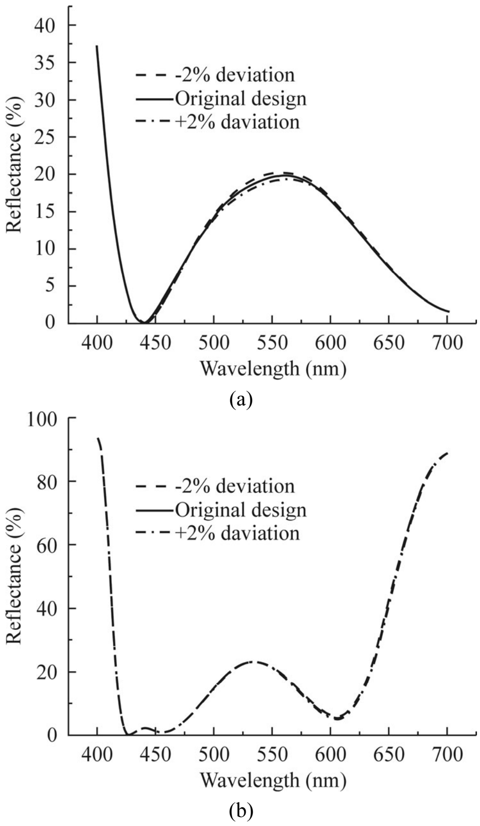


Fig.6 The effect of thickness deviation of Cr layers for (a) filter A and (b) filter B

We show that a pair of metal-dielectric filters based on metamerism can be used in reflection under illuminant D65. Based on the multiple color targets optimization approach, the stacks consisting of 5 layers and 7 layers are achieved, and their colors at normal incidence match with each other very well in the green region. The color difference is only 0.71, and no color difference can be perceived by naked eyes. When the viewing angle is tilted, the filters provide two distinct color shifts. The property offers augmented security protection due to the design complexity is increased and the authentication is easy. Our future work will focus on optimizing the multilayer structure and increasing the practicability.

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