1. Introduction

Color is one of the most important attributes of human visual perception. The pioneering experiments on visible light and colors can be traced back to the 17th century, when Sir Isaac Newton observed different transmitted colored bands when a narrow beam of sunlight struck the face of a glass prism at an angle. This early experiment implies that a desirable color can be obtained by spectrally filtering the white light. Indeed most of the modern colored displays rely on spectrum filters to produce primary colors (e.g., red, green and blue). Traditionally such spectrum filters are based on colorant pigments that absorb light in certain wavelength range, which finds dominant usage in many display systems such as liquid crystal displays (LCDs). On the other hand, color can also be created by light interaction with certain nanostructures that strongly influence the light propagation through the structures. In fact this is frequently observed in nature, for example, the wings of the Morpho butterfly (a special southern American butterfly family) shine a magnificent blue color, which was found to be due to surface-relief volume-diffractive nanostructures on its wings (Figure 1).\[1\]

The principle of these photonic nanostructures and the associated light management including tunable colors has been the subject of previous reviews\[2,3\] and will not be repeated here. Instead, this concept paper will focus on the color generation due to light interaction with conductive nanostructures. Specifically various patterned metallic nanostructures can perform spectrum filtering by the excitation of surface plasmons; and carbon nanotube-based structures can produce colors by tailoring the structures’ effective refractive index.

2. Color Generation via the Exploitation of Surface Plasmons in Metallic Nanostructures

A general overview is first provided on the most relevant concepts that are driving the development of structural color from plasmonic nanostructures. Several representative plasmonic nanostructures that are capable of filtering the color from the visible spectrum are presented. These metallic nanostructures could be advantageous for the optical applications such as spectral filtering, imaging and high-resolution color display.
2.1. Surface Plasmons and the Interaction of Light with Metallic Nanostructures

With the development of nanofabrication and characterization techniques, surface plasmons and related plasmonic nanostructures have generated considerable interest in recent years.[4–8] Surface plasmons are essentially surface-bound electromagnetic waves coupled with the collective oscillation of conduction electrons on the metal surface that are excited by an incident light. By exploiting plasmonic nanostructures, efficient conversion between free-space photons and confined plasmons can be controlled at a subwavelength scale. This means one can exploit the geometries of the plasmonic nanostructures to manipulate light properties, including aforementioned visible-light wavelength selection and thereby the generation of structural color. In contrast to the color produced by ordinary electronic absorption, which is determined by the material’s specific molecular spectrum, plasmon-based structural color is primarily determined by the geometry parameters of the plasmonic nanostructures. This characteristic brings in several advantages for the plasmon-based structural color: 1) different colors can be obtained by using the same material but only with different structural parameters, which could greatly simplify the manufacturing process for multiple colored units; 2) unlike traditional chemical pigment and dye that are affected by photobleaching, the plasmonic nanostructures are chemically stable, and therefore the structural color is attractive for applications requiring high light intensities or continuous illuminations; 3) comparing with other dielectric periodic structures such as photonic crystals[9–14] or semiconductor nanowires,[15,16] which also can generate structural colors, the highly confined surface plasmons in metallic nanostructures makes the device dimension more compact. Moreover, the metal in the plasmonic nanostructures can be used as electrodes and thereby offers better potential for the electro-optic applications.

2.2. Structural Color from Subwavelength Hole Arrays in Metal Films

In 1998, Ebbesen et al. experimentally demonstrated the extraordinary optical transmission (EOT) through perforated periodic subwavelength hole arrays in optically thick metallic films,[17] where the transmission is several orders higher than the value predicted by classic Bethe’s theory. The surface-plasmon modes excited by the subwavelength hole arrays are believed to play an essential role in the EOT phenomenon.[18–21]

The subwavelength hole arrays efficiently convert incident light into surface-plasmon modes by scattering, which provides the necessary momentum conservation. The excited surface plasmon waves tunneling through the subwavelength holes scatter again at the exit plane of the hole arrays, and the resulting propagation waves transmit away from the structure to the far field.

For the subwavelength hole arrays with square-lattice, the transmission peak wavelength in the normal incidence can be approximately calculated from the surface-plasmon dispersion relation as:[4]

$$\lambda_{\text{max}} \approx \frac{P}{\sqrt{i^2 + j^2}} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}}$$  \hspace{1cm} (1)$$

where $P$ is the square-lattice constant; the indices $i$ and $j$ are the scattering orders from the hole arrays; $\varepsilon_m$ and $\varepsilon_d$ are the permittivity for the metal and dielectric medium. This equation does not consider the presence of holes and the associated scattering loss, and it neglects the Fano-type interaction which results in a resonance red shift. Therefore the predicted transmission peak wavelength is slightly smaller than the real value.[22] From Equation 1, we can see that the transmission peak wavelength $\lambda_{\text{max}}$ is proportional to lattice constant of the hole arrays. Therefore the subwavelength hole arrays in metal film can act as optical filters for which the transmitted color can be selected by simply adjusting the lattice constant.[4,22] Figure 2a,b show the lowest order transmission peak ($i, j = 1, 0$ in Equation 1) of the perforated square-lattice hole arrays on a free-standing 300 nm-thick silver film. When the lattice constant of hole arrays increases from 300 to 550 nm, the transmission peak wavelength changes from 436 to 627 nm, covering primary RGB colors in the visible spectrum. In addition, by arranging the perforated and unperforated subwavelength hole arrays in the metal film, arbitrary colored patterns can be realized (Figure 2c).

In contrast to the square-lattice arrays, triangular-lattice hole arrays have improved color filtering performances.[21,24,25] Figure 2d,e show the microscope images of the subwavelength hole arrays with triangular-and square-lattice, respectively. In theory, the transmission peak wavelength $\lambda_{\text{max}}$ for the hole arrays with triangular-lattice can be approximated as:[21]

$$\lambda_{\text{max}} \approx \frac{P}{\sqrt{4(i^2 + j^2)}} \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}}$$  \hspace{1cm} (2)$$

where $P$ is the triangular-lattice constant. Comparing with Equation 1, we can conclude that the triangular-lattice subwavelength hole arrays will have a larger wavelength interval between the first two transmission peaks than the square-lattice hole arrays with the same lattice constant. This means triangular-lattice subwavelength hole arrays can reduce the color cross-talk and can produce purer color. Such improvement can be easily seen in the comparison between Figure 2d and e.

A further note on the color filters based on the subwavelength hole arrays is the symmetry of structure. A free-standing structure described above is difficult to fabricate.[19] However, if the filter is fabricated on a glass substrate but without an index-matching superstrate, the structure...
becomes asymmetric. Since both surfaces on either side of the hole arrays can support the excitation of surface-plasmon modes, the asymmetric structures will produce two sets of peaks in the transmission spectrum. The overlap of two sets of transmission peak will degrade the purity of the filtered color. Besides, asymmetric structure will reduce the coupling efficiency of the surface-plasmon modes on the two surfaces, which leads to a lower transmission peak. Therefore, in most cases an index-matching layer such as silica or oil always covers the whole structure when the device is fabricated on a glass substrate.

2.3. Structural Color from Metal–Insulator–Metal Nanoresonators

Metal–insulator–metal (MIM) geometry offers the ability to support both photonic and plasmonic modes at visible frequency and have been widely investigated for different applications, such as guiding waves at subwavelength scale concentrating light in the thin-film photovoltaics[27–28] and composing negative-index metamaterials[32–33]. Besides, the top and bottom metal layers of MIM structure can be integrated as electrodes in a straightforward manner: they are therefore of considerable interest for electro-optic applications.

The interference and resonance of the optical modes within the MIM resonators can be used to filter the white light into individual colors. For example, lateral Fabry-Perot resonators can be formed if input and output nanoslits are inserted in the MIM waveguide (Figure 3a). By proper design of the depth and location of the input and output slits, the resonator can preferentially couple to different waveguide modes and select any of the primary colors. Different separation lengths between the input and output slits produce different exhibited colors at the output slits. The typical length...
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of these MIM resonators is about several micrometers. As shown in Figure 3b, the resonators consist of a 500 nm-thick silicon nitride free-standing membrane sandwiched between a 400 nm-thick gold layer on top and a 400 nm-thick silver layer on the bottom. Since two metal claddings of the MIM resonators can be used as electrodes, the electro-optic material such as lithium niobate, liquid crystals, or electro-optic polymers, can be used as the middle layer to make active devices. An applied electric field can modulate the refractive index of the insulator layer and thereby change the output color, which yields the active tunable color filter.

The limitation of the MIM resonator color filters described above is their low transmission efficiencies, because only a very small portion of the incident light can couple out through the nanoslits. Even tiling several resonators together, the overall transmission efficiency is lower than 0.2%, not suitable for practical applications. Another type of MIM resonator color filter capable of filtering white light into individual colors across the entire visible spectrum with high resolution and high transmission efficiency simultaneously is introduced. For the TM-polarized waves (electric-field is perpendicular to the Al grating direction), the MIM stack resonators support the surface-plasmon modes whose mode momentum is larger than the free space photon momentum. Therefore the stack array should have a subwavelength period so that scattering from bottom Al grating can provide momentum matching for incident light coupling into surface-plasmon modes. The top Al grating, having the same period as the bottom one, serves to scatter the confined plasmon modes back to propagation waves and transmit to far-field. The relation between the resonant wavelength and stack array period can be obtained by solving the surface-plasmon dispersion equation. Arbitrary color in the visible spectrum can be filtered with this structure by selecting the proper stack array period. Figure 3d shows the optical microscopy images of the seven square-shaped color filters illuminated with the TM-polarized white light. The filters have the stack period ranging from 200 to 360 nm, corresponding to violet to red colors. The stack arrays show the expected filtering behavior with absolute transmission efficiency about 60% around the resonant wavelengths, which are several orders of magnitude higher than those of MIM resonator filters in reference. This transmission efficiency is comparable with the prevailing colorant-based filter used in an LCD panel, but the thickness of this MIM resonator device is 1–2 orders of magnitude thinner than that of the colorant one.
On the other hand, because the TE-polarized waves (the E-field is parallel to the Al grating direction) do not support the excitation of surface-plasmon modes, there is no obvious light conversion process. As a result, the TE-polarized incident light is strongly reflected. This indicates that the filtered light by such MIM resonator is naturally polarized, making it attractive for direct integration in LCD without a separate polarizer layer.\[36\]

### 2.4. Structural Color from Other Plasmonic Nanostructures

Besides the subwavelength hole arrays and MIM nano-resonators, there are several other relevant plasmonic nanostructures capable of color filtering in the visible spectrum. For example, the wavelength-filtering principle of the subwavelength hole arrays can be easily developed to the structures where single aperture is surrounded by the periodic grooves, i.e., a bull’s eye structure, as shown in Figure 4a. Here the whole structure acts as an antenna to efficiently couple the incident light into surface-plasmon modes at a resonant wavelength, which is also determined by the groove period as in Equation 1 for the normal-incidence illumination. This make the electromagnetic fields at the surface become intense above the aperture, resulting in extremely high transmission efficiency for the hole at this resonant wavelength.\[37,38\] In other words, arbitrary color in the visible spectrum can be obtained only by tuning the groove period, as shown in Figure 4b.\[39\] Although the transmission spectrum also can be modulated by the other structural parameters, such as groove depth, width, and hole shape,\[40\] controlling groove period is the simplest and most efficient way.

Another relevant structure is the metallic resonant waveguide grating (MRWG)\[41–44\] (Figure 4c), which is
3. Structural Color from Carbon Nanotubes and Generating Perfect Black

Although most of the previous plasmonic nanostructures are capable of producing various colors at visible frequency, they work at resonance condition and thus provide relatively narrow bandwidth. Carbon nanotubes (CNTs), allotropes of carbon with a cylindrical nanostructure, are an alternative type of conductive optical material for structural color applications. Actually, the optical properties of CNTs have been extensively studied in recent years, specifically on absorption, photoluminescence, and related topics for broadband applications. In this section, we will review the recent progress of enhanced color saturation and the ‘perfect black’ generated by CNTs.

Since the 1960s, it was well known that anodic aluminum oxide (AAO) film can produce a bright color in the visible-light range due to the interference of light. The color is bright but its saturation is very low. To make the color pure and highly saturated, colored substances have to be infiltrated into the AAO film nanochannels, but they suffer from fading. Recently, it was demonstrated that when a thin AAO film is uniformly coated with carbon by chemical vapor deposition (CVD), the CNT-coated film exhibits a vivid and tunable interference color with much higher saturation than the pristine AAO film. Figure 6a shows the schematic of the pristine AAO film and the CNT-coated one, and Figure 6b shows their corresponding photographs with pattern consists of three partly overlapped circles. Results show that the reflectance of the carbon-coated AAO films is lowered in such a way that the saturation of interference color is much enhanced, namely, the interference color becomes purer with the carbon coating. As colors of the CNTs and AAO composite thin films are mainly determined by the interference band with the maximum reflectance in the visible region, color tuning of the CNTs and AAO composite thin films can be attained by varying the AAO film thickness in the anodization process. In addition, by applying wet chemical etching to different areas of the AAO film for different durations, different colors can be produced from the patterned CNTs and AAO composite thin film. Such beautiful CNT-coated AAO films supported on an Al substrate are also useful for decorative purposes because they are dye-free and there is no problem of fading. The high-throughput fabrication of AAO films and improved mechanical stability should be addressed in future research for practical applications.

Another important color produced by the CNT forest is ‘perfect black’, which is difficult to obtain through other methods. In 2008, it was discovered that a thin coating comprised of low-density arrays of loosely vertically-aligned carbon nanotubes, absorbs more than 99.9% percent of light over the entire visible frequency. The key to this perfect black was to...
concepts

![Diagram of nanochannel and CNT forest](image)

Figure 6. Brilliant and tunable color from carbon-coated thin anodic aluminum oxide films. a) Schematic of the pristine AAO film and the carbon-coated one. b) A photograph of the pristine patterned AAO film (left) and its carbon-coated one (right). The pattern consists of three partly overlapped circles. Reproduced with permission. Copyright 2007, AIP.

create a long and extremely porous vertically aligned carbon nanotube array with certain surface randomness, therefore minimizing light reflection and maximizing absorption simultaneously. It’s interesting to note that the reflectance of such nanotube arrays are two orders of magnitude lower than that of the glassy carbon, which is remarkable because both samples are made up of the same element. As a result, the reflectance at air and CNT forest interface is less than 0.1% for a sufficiently thick CNT forest. Meanwhile, the absorption inside the CNT forest is very effective. For 30 μm thick CNT carpet, the absorption is more than 99.5%; it is also easy to grow a CNT forest with a thickness of hundreds of micrometers.

Finally we point out an interesting application by exploiting the perfect absorption characteristics of a low-density CNT forest. By coating such a perfect black material on an arbitrary shaped object, the 3D object will appear as a 2D black sheet and all the geometric information disappears. In this case the CNT forest acts as a perfect magic black cloth that can completely conceal the 3D structure of the object. As a demonstration, an arbitrarily shaped object was fabricated on a 500 μm thick silicon substrate by focused ion beam (FIB) milling. In this case a tank pattern of 65 μm × 22.5 μm in size (SEM image in Figure 7c) was made and its reflection image was taken under an optical microscope illuminated by unpolarized broadband visible light (Figure 7f). To cover the object with the perfect black CNT forest, a 60 μm-thick CNT forest was grown on top of the whole silicon sample and therefore follows the profile of the original tank object (Figure 7d). To get the CNT carpet, first a 300 nm-thick SiO2 layer is deposited on the silicon sample by plasma-enhanced chemical vapor deposition, and then 1 nm-thick Fe catalyst layer is deposited by electron beam evaporation. The sample is loaded in a single-zone tube furnace, which is heated to 775 °C under the gas mixture of C2H4/H2/He. An optical image of the object covered with the CNT carpet was taken again. Figure 7g shows that the tank completely disappears and the surface looks exactly the same as a flat CNT sheet. As further proof, a control experiment was performed where a rectangle mark around the tank was made by FIB milling that removed the CNT. The optical image now clearly shows the rectangle mark, but the tank pattern inside the mark remains perfect black, as shown in Figure 7e,h. Since the incident visible light is broadband and unpolarized, such perfect black from a CNT forest is promising for various kinds of applications such as efficient solar-energy conversion, photo-detectors and display-related devices.

4. Discussion and Outlook

High resolution, slim dimension, and low power consumption are always the invariable goals for the development of display technologies. Typically, human eyes have a resolution limit of about 80 μm at 35 mm.[76] These plasmonic and carbon nanostructures can be used to build colored ‘super-pixels’ that are only several micrometers in the lateral dimension so that much smaller than the resolution limit of the human eyes for the application of ultrahigh resolution displays. Besides, they have a longitudinal thickness that is about 1–2 orders of magnitude thinner than that of chemical pigment ones, which can further reduce the device dimension and thereby increase the integration density. Also, color filter consisted by the 1D...
metallic grating structures can act as the polarizer simultaneously, which could not only benefit the applications in LCD by eliminating the need of a separate polarizer layer, but also can be used for extracting polarimetric information in spectral imaging.\cite{35,39} Furthermore, the metal and carbon in nanostructures can be directly used as electrodes and thereby offers better electro-optic modulation\cite{34} and photovoltaic self-powering\cite{31,71–73} in the system.

Although the plasmonic and carbon nanostructures have these intrinsic advantages which can potentially fulfill the requirements for the development of display technology, there is still a long way to go before final commercialization. The next step for the research on the nanostructured color devices will mainly focus on two aspects: first, the further improvement of their optical performances, including the transmission and reflection efficiencies, the purity of filtered color, and the incident angle-independency. Second, the development of a more efficient, high-throughput nano-fabrication method to realize mass production with low costs.

We believe these nanostructured color devices will open up a colored future for the next generation high-resolution display, spectral imaging, and relevant applications.

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