MATERION

Reflectance in Thin Films

Abstract

Reflectance (R) is the fraction of incident light reflected from a surface and is an intrinsic optical property of thin films. It is essential in determining color, transparency and polarization characteristics of the film. Total internal reflectance is also important in devices such as optical waveguides. Reflectance depends on the energy band structure and associated plasma frequency of charge carriers. As a result, high reflection spectral regions are different for metals, semiconductors and insulators. Basic relations that determine reflectance will be presented and related to refractive index, extinction coefficient, color and transparency of these three classes of thin film materials. Reflectance of thin films also depends on thickness and surface quality. In addition to spectral dependence, the color associated with reflectance can also be described by Tristimulus values and Chromaticity diagrams. Antireflection and high reflection multilayer thin film coatings will also be addressed.

Introduction

The reflectivity or reflectance (R), of a surface is an intrinsic optical property of a surface. In many optical, electrooptic, telecommunications, solar concentrator and architectural applications, reflectance must either be controlled (reduced or enhanced), or the color of the object changed (e.g., given a "gold" color). For example, heat mirrors are used to reflect infrared wavelengths to reduce heat loss or ingress through windows. Infrared reflectance must be maximized while keeping visible light transmission through the window high. Multilayer low-e and solar control coatings are used to achieve this performance but must be applied to low cost plastic films and glazings. Combined with absorption, reflectance determines color and intensity (or energy) of reflected light. Bare metal surfaces of jewelry and plumbing and hardware fixtures corrode, tarnish and scratch due to environmental exposure and every day use. Protective coatings that simulate the reflective color of gold, silver, bronze and copper can be applied to less expensive materials to reduce cost and extend their lifetime. New methods of reducing counterfeiting of currency and labels are constantly needed. Color shift coatings are applied to these items and hard to duplicate, and mitigate this problem to a large extent. Mirrors used in astronomical telescopes and solar concentrators are constantly exposed to the environment and must be highly reflective, abrasion resistant and not attackable by the environment. In these cases, refurbishing is very expensive. If left untreated, they will rapidly degrade. Additionally, areas as large as 80 m must be covered by mirror coatings. Laser mirrors are used extensively in every laser application. Metals (even highly polished metals) are generally unsuitable for laser mirrors because their reflectivity is not high enough and they have some optical absorption. Multilayer high reflector coatings can have almost perfect reflectivity and have wide applications as laser mirrors.

Reflectance is also important in telecommunication devices such as optical waveguides which function on total internal reflectance. In addition to waveguides, dense wavelength division multiplexing (DWDM) filters reflect over very narrow wavelength bands and are used to separate optical signals in telecommunication fiber optics. The list goes on….

In addition to improving performance and adding color, reflective coatings are also used to reduce the cost of a wide variety of materials. By applying reflective and color coatings, less expensive base materials can be used and lifetimes extended. Instead of using colored glass, architectural coatings can be applied to regular glass to obtain a range of colors not possible with the original substrates. Color shift coatings applied to currency and packaging have saved untold trillions of dollars in losses and theft. Obviously, reflective thin films are significantly less expensive than polishing metal surfaces, particularly on very large surfaces. By using DWDM filters, hundreds of optical signals can be propagated down a single fiber optics line instead of using separate fibers for each wavelength, thus saving cost of materials and installation.

To this end, thin reflective films can (1) improve the reflectance of virtually any surface, (2) turn a diffuse surface into a reflecting surface, (3) significantly reduce materials costs by using less expensive base materials, (4) change the color of the surface or window, (5) significantly decrease wear and corrosion of a surface, (6) increase the functionality of a surface or optical element, (7) replace highly toxic chromium in mirrors, plumbing fixtures and hardware and (8) increase security of currency and packaging.

<u>What is Reflectivity?</u>
Reflectance, as defined in physics and optics, is the ratio of the energy of a wave reflected from a surface to the energy possessed by the wave striking the surface. Some materials are used for mirrors because they reflect all spectral components of white light with little absorption or transmission. Other materials appear colored because they selectively absorb in certain wavelength ranges and reflect the unabsorbed wavelength bands. Because reflectivity is a directional property, it depends critically on surface quality, and can be either specular or diffuse. As shown in Figures 1a and 1b, for specular surfaces, such as glass or highly polished metal, reflectivity will be nearly zero at all angles except at the reflected angle; i.e., other than for radiation normal to the surface; incident radiation is reflected at an angle equal to the angle of incidence (angle of reflection (θ_R) = angle of incidence (θ_i)) (http://www.tulane.edu/~sanelson/geol211/proplight.htm).

Light can also be transmitted through the boundary and refracted. (Note: only reflection at metal surfaces) (http://qis.ucalgary.ca/quantech/471/labs/reflection_refraction.pdf). Diffuse scattering (Figure 1c) occurs when the surface is rough or matt in nature. In this case, reflectivity is uniform; radiation is reflected in all angles equally or near-equally. Such surfaces are said to be Lambertian. Thus, reflectance of a surface can be enhanced or reduced by controlling the roughness of the surface.
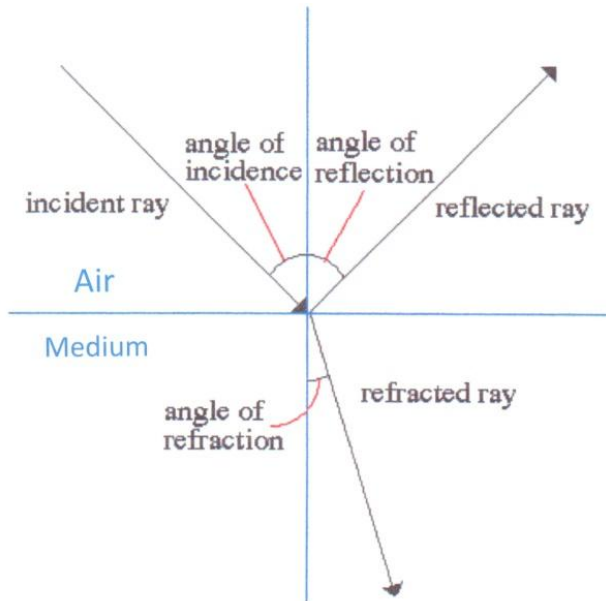
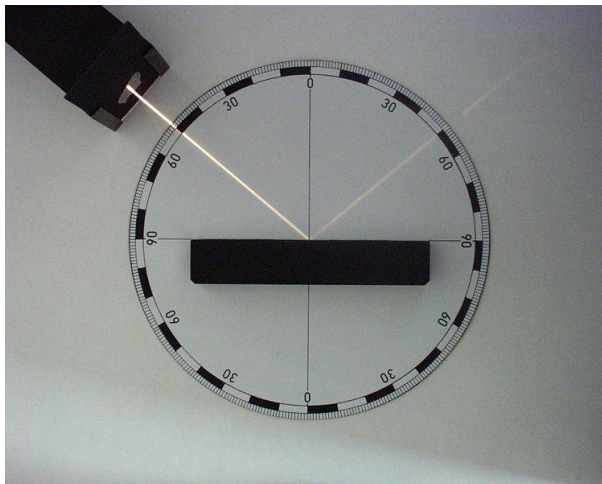Figure 1a. Reflection and refraction of light at a boundary between two media [1].

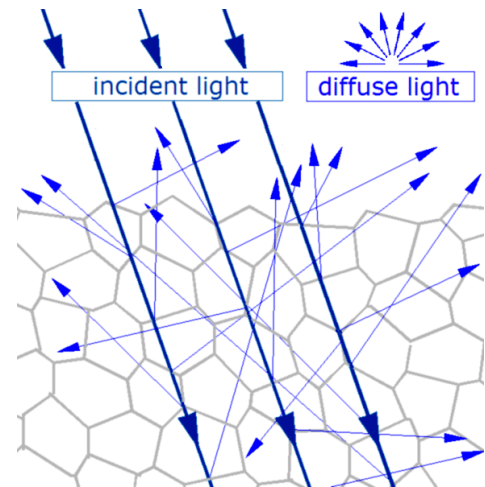Figure 1b. Specular reflectance of a laser beam.

Figure 1c. Diffuse reflectance of light.

Polarization of Light

Light specularly reflected from a surface also depends on the polarization of the incident light wave [1], and polarizers can be used to control reflectance. Light waves are composed of electric (E_i) and magnetic field (H_i or B_i) components, as shown in Figure 2 (note that $B = \mu H$, where μ is the permeability = 1 for air). Upon reflection, the electric field component (E_2) is flipped to the opposite direction (180 degrees) while the direction of the magnetic field component (B_2) is unchanged (see Figure 3). Wave vectors (k and k') indicate the direction of the incident and reflected waves. Electric and magnetic field directions

transmitted though the interface are unchanged (this will be important for thin films). A light wave is "*s*" polarized when the electric field is in the *x* direction (see Figure 2), which is also known as the transverse electric (TE) mode. The transverse magnetic (TM) mode is perpendicular to the TE mode and also known as "*p*" polarization.
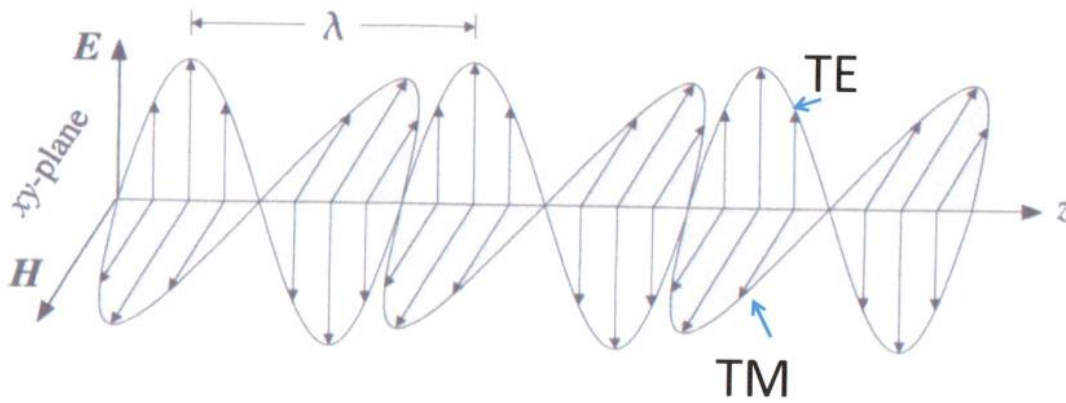

Figure 2. Electric and magnetic field components of light.
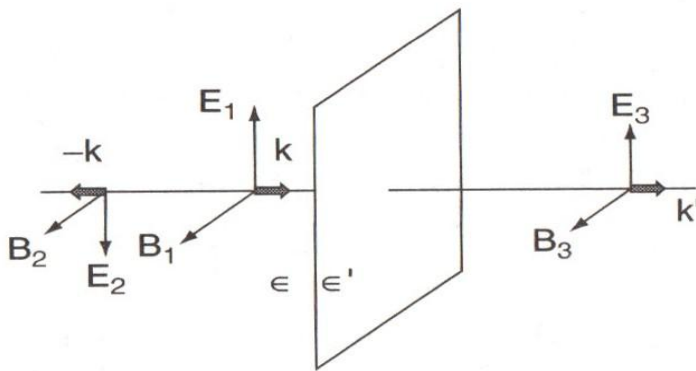

Figure 3. Reflection of electric and magnetic field components of light at an interface [2].

Calculation of Reflectivity and Reflectance

Reflectance of a surface can be described using the optical constants of the surface medium or by the free electron, or Drude, theory. The index of refraction *n* of a material is complex and can be written as

$$n = n + i\kappa$$

n = refractive index of the surface

κ = extinction coefficient of the surface

The extinction coefficient determines the attenuation of light intensity as it propagates in a material.

Reflectance at normal incidence is given by

$$R = [(n-1)^2 + \kappa^2]/[(n+1)^2 + \kappa^2]$$

At nonnormal incidence, polarization becomes important and the amount of light reflected at an interface depends on the polarization of the incident wave, which is usually a combination of s and p components. If θ_1 is the angle of incidence (AOI) and θ_2 is the angle of refraction, reflectance of s polarized light is give by [1]:

$$R_S = (n_1\cos\theta_1 - n_2\cos\theta_2)^2/(n_1\cos\theta_1 + n_2\cos\theta_2)^2$$

While the reflection coefficient of light polarized in the plane (p-polarization) is given by:

$$R_P = (n_1\cos\theta_1 - n_2\cos\theta_2)^2/(n_1\cos\theta_1 + n_2\cos\theta_2)^2$$

Figure 4 shows s and p reflection for the cases of $n_1 < n_2$ and $n_1 > n_2$ against angle of incidence (AOI). For the first case, s polarization reflectance continues to increase at all angles of incidence but R_P approaches 0 at Brewster's angle ($\theta_B = \tan^{-1}n_2/n_1$) and then increases rapidly with increased AOI. As demonstrated in the figure, total internal reflection is only possible when $n_1 > n_2$., and occurs at the critical angle $\theta_C = \sin^{-1}(n_2/n_1)$.
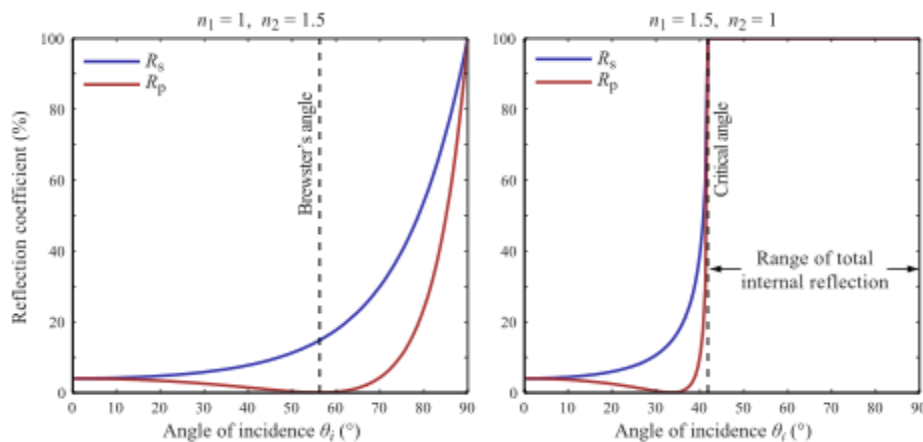

Figure 4. s and p reflection for $n_1 < n_2$ and $n_1 > n_2$.

Total internal reflection is critical for the operation of optical waveguides, including fiber optics. Figure 5 demonstrates total internal reflection in a block of PMMA. Here $n_1 = 1$ and $n_2 = 1.5$. If $n_1 < n_2$, some light would be transmitted out of the material and some reflected back into it according to the Fresnel equations. These concepts are critical for waveguide operation.
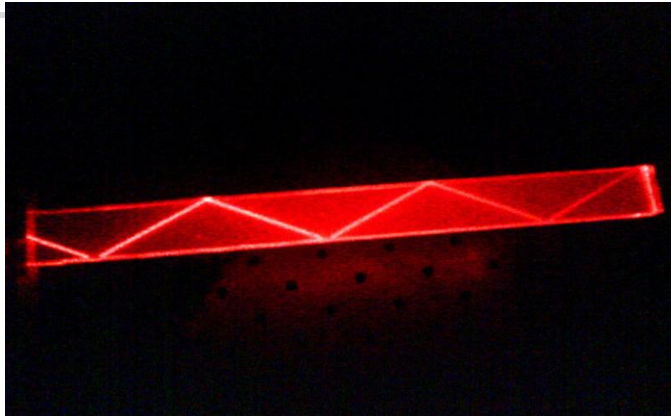
Figure 5. Total internal reflection in a block of PMMA.

Factors That Determine Reflectivity

In general, metals have very high extinction coefficients, making them good reflectors, while k for insulators are generally small, making them poor reflectors. Figure 6 shows the reflectance of selected metals at normal incidence (0° AOI). The plasma frequency (ω_P) is based on the frequency dependent electrical conductivity of metals and is important when describing the reflectance of metals [3]. It is the natural frequency of the electron gas in the conduction band of metals and generally occurs at ultraviolet and short visible wavelengths. As shown in Figure 7, the plasma frequency separates the frequency (or wavelength) domains at which the metal is absorbing ($\omega \sim \omega_P$), reflecting ($\omega < \omega_P$) or transparent ($\omega \gg \omega_P$) [4]. The Figure shows reflectance for several values of $\omega_P \tau$ (τ = electron relaxation time). Reflectance is very high and approaches 1 (or 100%) for very conductive metals (e.g., silver, gold, aluminum, copper). As expected, reflectance is very low for insulators with no electrons in the
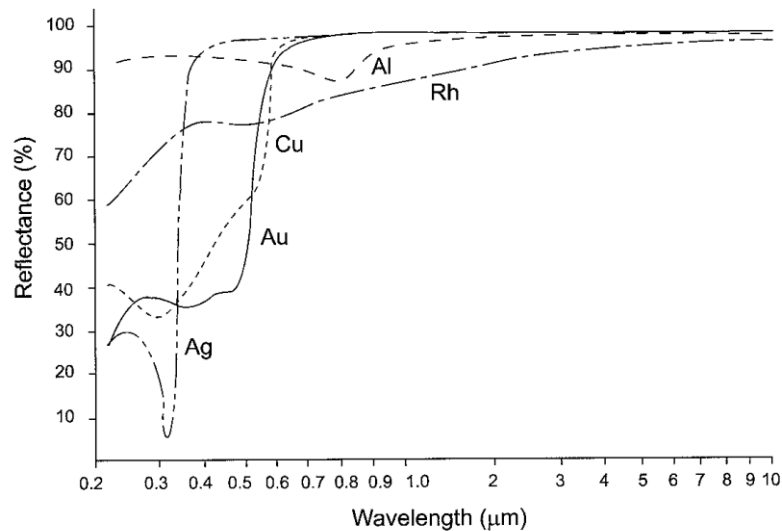
conduction band.

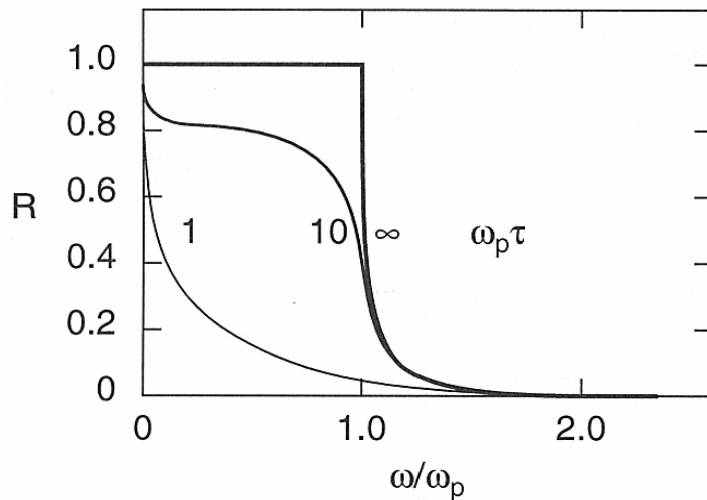Figure 6. Reflectance of selected metals at 0° AOI .

Figure 7. Reflectance as a function of frequency (in units of plasma freq.) for several values of $\omega_P\tau$ [4].

Thin Film Applications

Strictly speaking, reflectivity is the limiting value of reflectance when the surface becomes thick, and can only be modified by polishing or roughening. For very thin layers (thin films for example) internal reflection effects can cause the reflectance to vary with layer thickness. Thin films are applied to a surface to change color, enhance reflectance, reduce reflectance or change polarization. The most widely used optical surface treatment is the metal mirror with the multilayer thin film optical coating as second. However, even a humble single layer coating can significantly change the optical properties of a surface or substrate. A simple metal thin film can convert the optical properties of any non-scattering surface into a mirror or high reflector. Consider first the metallic reflector, which is essentially a broad band mirror. Figure 5 shows the reflectance of several metal coatings at 0° AOI. The color of each material depends on the wavelength range of high reflectance. Al and Ag have high reflectivity over the visible wavelength range and, essentially, no color and appear "silvery." The reflectance of Au and Cu films only becomes high above 500 nm wavelength and thus contains some yellow and red components.

High reflection thin film coatings have numerous applications, including telescope mirrors, laser mirrors, display optics, satellite optics, architectural glass, automobile mirrors and headlamps, defense optical systems, projectors, and much more. This family of coatings includes metal reflectors, protected metal reflectors, dielectric-enhanced metal reflectors, all dielectric multilayer reflectors, and rugate filters. Cousins to this group of coatings are dichroic coatings, high pass and low pass filters, beam splitters, dense wavelength division multiplexing (DWDM) filters, low-e coatings, and Fabry-Perot filters. Depending on the design and materials used, the high reflector can be broad band, covering a wide range of wavelengths, or selective, covering a specific wavelength band. An excellent summary of high reflector coatings can be found in *Optical Coating Technology* by Phil Baumeister [5].

There are six basic types of reflector coatings:

- First surface mirror (no overcoat)
- Second surface mirror (light incident on glass substrate)
- Overcoated metal mirror (thin film + metal)
- Enhanced metal high reflector (metal enhanced using interference effects)
- Multilayer all-dielectric reflector (high reflector, low and high pass filter, dichroic filter, beam splitter, DWDM filter, etc.)
- Photonic band gap structure

A highly technical application for high reflectance thin film coatings is for primary mirrors for large astronomical telescopes [6,7,8]. Protected and unprotected aluminum (Al) and silver (Ag) coatings are deposited onto primary telescope mirrors as large as 10 m in diameter [8]. A picture of the 8 m Ag-coated Gemini telescope (www.gemini.edu) primary mirror is shown in Figure 8 and Figure 9 shows the reflectance spectrum of a high reflector coating proposed for the Giant Segmented Telescope (Thirty Meter Telescope: www.tmt.org) [7].

To prevent environmental degradation, increase abrasion resistance and enhance reflectance, Al and Ag layers are protected by a single layer dielectric material such as silicon nitride (Si_3N_4) films or by multilayer coatings. Additionally, reflectance > 0.99999 can be achieved with multilayer optical coatings for use in high quality optics and laser mirrors.

Figure 8. Picture of 8 m Ag coated Gemini primary telescope mirror [www.gemini.edu].
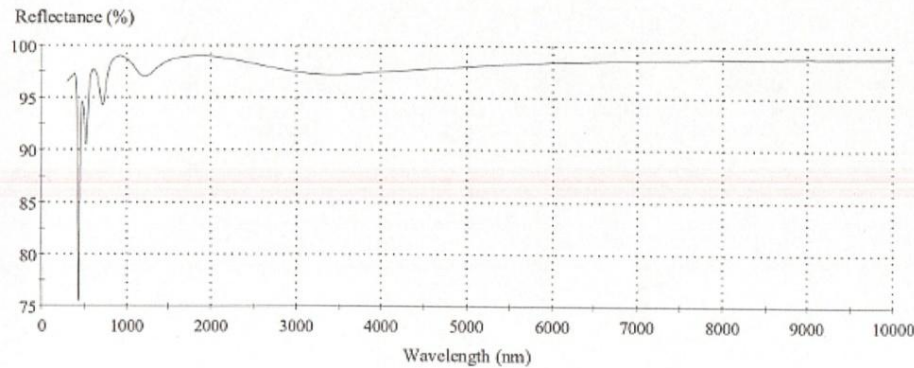
Figure 9. Reflectance spectrum of proposed high reflection coating for Giant Segmented Telescope [7].

Reflectance of a diffuse reflecting (matte) surface can be significantly increased by application (deposition) of a polymer smoothing layer [9].The polymer layer creates an ultra-smooth, defect free surface for deposition of highly reflective metal layer or reflective optical coating. Figure 10 shows SEM pictures of a virgin polyester web substrate, a virgin polyester substrate with a sputtered Ta coating and a virgin polyester substrate with a sputtered Ta film deposited onto a polymer smoothing layer [9]. Note the significant improvement in surface smoothness of the bottom picture.
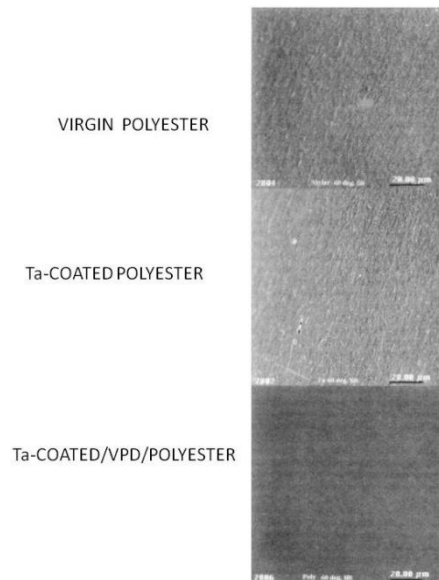

Figure 10. SEM picture of virgin polyester, sputter Ta-coated polyester and sputter Ta-coated polyester with a polymer smoothing layer.

Reflectivity and Color of a Surface

One of the primary applications of thin films is to change the color or reduce the color of a surface, window, optic, or object. The color of a thin film, whether in reflection or transmission, is critical in many applications, including low-e windows, antireflection coatings, hardware, plumbing fixtures, high reflector coatings, jewelry, automotive parts (including paints), and architectural glass. The "color" of a thin film results from its optical properties: transmittance, reflectance, and absorption. The optical properties of the substrate are often integral to the color of the thin film. When we look through a window we see both transmitted and reflected light. However, reflected and absorbed light are most important for coatings on structural components (plumbing fixtures, hardware, jewelry, etc.). Color can also be a function of angle of incidence (AOI) of the viewer.

Many thin film reflectance applications that require a specific color or lack of color for the film/substrate combination are based on the Tristimulus model for color [10]. This model is based on the fact that any color can be produced by the appropriate combination of primary colors:

C = B**B** + G**G** + R**R** (bold = unit value)

Here **B**, **G** and **R** are unit values for blue, green and red and B, G and R are relative intensities of these primary colors. In this case, white is characterized by "1" unit:

W = 1**B** + 1**G** + 1**R** (in appropriate intensity units)

The most widely used model used to describe color perception is the C.E.I Chromaticity diagram (Commission Internationale de l'Eclairage) [11,12]. A typical C.I.E chromaticity diagram with color regions is shown in Figure 11. Here "x" and "y" are chromaticity coordinates based on chromaticity matching functions and wavelengths (colors) are located on the perimeter of the color space. Thus, "x" and "y" can fully define the color of a surface [12]. Applying this diagram involves inserting chromaticity matching functions [13,14]. For example, white can also be achieved with many different mixtures of light, e.g., with complementary colors. If you have two illuminating sources which appear to be equally white, they could be obtained by adding two distinctly different combinations of colors. This implies that if you used them to illuminate a colored object which selectively absorbs and reflects certain colors, that object might look very different when viewed with the two different "white" lights. For instance, an object will have a different color outdoors in direct sunlight than under fluorescent or incandescent illumination. Note that thin film design software often has the capability of plotting C.I.E and other chromaticity diagrams (e.g., Essential Macleod) for a variety of light sources.
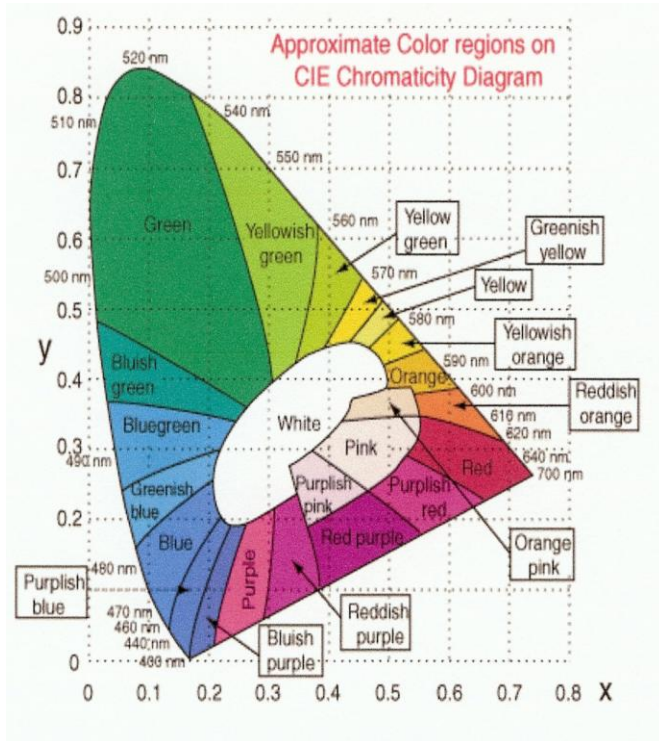
Figure 11. 1931 C. I. E chromaticity diagram [10,11].

Consider first the metallic reflector, which is essentially a broad band mirror. Recall that Figure 6 shows the reflectance of several metal coatings at 0° AOI. The color of each material depends on the wavelength range of high reflectance. Referring to the CIE 1931 Chromaticity diagram shown in Figure 11 for reflectance, we expect Ag and Al to fall close to the "white" zone and Au and Cu to have a considerable amount of yellow – red color. Gold is difficult to produce from CIE chromaticity diagrams, but has CIE coordinates (x,y) ~ (0.55,0.46) [15]. We see that Cu reflectance contains a bit more blue-green. Rh appears grayer due to absorption at lower visible wavelengths.

Figure 11 is only part of the story. Reflectance has a strong dependence on AOI of the viewer. It is possible to shift color by variation of AOI as will be described below. The Fresnel equations shown above define the components of the reflected waves. Figure 12 shows s and p reflectance of Al for various AOI. The separation of reflectance of the two polarizations increases with increased AOI, yet note that the average reflectance is always the reflectance at 0° AOI. P polarization reflectance is always less than s polarization reflectance. Since our eyes see the average reflectance, the reflectance (therefore the color) of metals is virtually the same at all AOI and color never shifts in the CIE diagram.
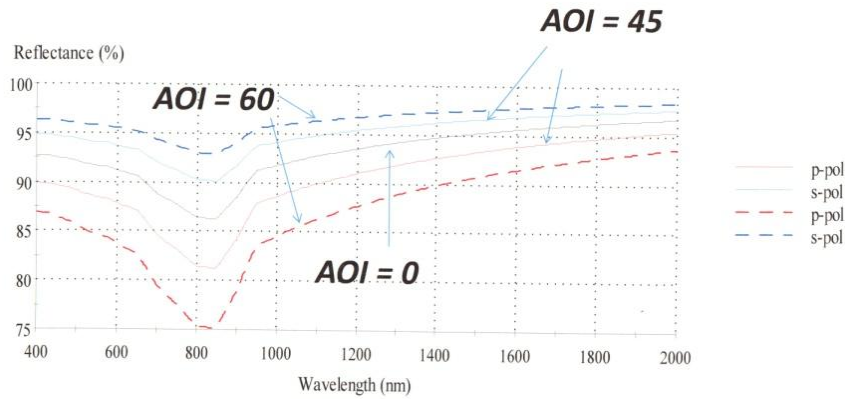
Figure 12. Reflectance of *s* and *p* polarizations for Al for AOI of 0°, 45° and 60°.

Color shift in the reflectance of multilayer optical coatings, however, is much more interesting. Employing interference effects, the layer structure of thin film coatings can be designed to provide high reflectance (or low reflectance) in specific narrow or broad spectral regions, thus transforming a transparent substrate into a mirror (broad band reflector) or giving it color (e.g., architectural glass). In addition to simple single band reflectors, multilayer designs include dichroic filters, laser mirrors, polarizers, high and low pass filters and Fabry-Perot filters [16]. Figure 13 shows various colors obtained for dichroic filters. Simple multilayer high reflector designs (used in laser mirrors for example) consist of alternating layers of high/low refractive index materials with designs expressed as $H(LH)^n$. Here H is the quarter wave thickness of the high index layer (= $n_H\lambda/4$) and L is the quarter wave thickness of the low index layer (= $n_L\lambda/4$). The design wavelength λ must be specified. Figure 14 shows the change in reflectance of a high reflector with design $H(LH)^{20}$ @ 510 nm with AOI. Here H = quarter wave optical thickness with n = 1.63 (Al_2O_3) and L = quarter wave optical thickness with n = 1.48 (SiO_2). The high reflectance peak shifts from 600 nm @ 0° AOI to 500 nm @ 60° AOI, essentially from red to blue-green. Figure 15 shows the shift in CIE Chromaticity coordinates (*x,y*) for reflectance and transmittance (yellow X's in the diagrams). In reflectance the CIE coordinates shift from (0.58,0.41) @ 0° AOI to (0.16,0.28) @ 60°, and from reddish orange to greenish blue. Transmitted chromaticity coordinates shift from (0.30,0.42) @ 0° to (0.42,0.42) @ 60°, or from blue green to yellow orange.
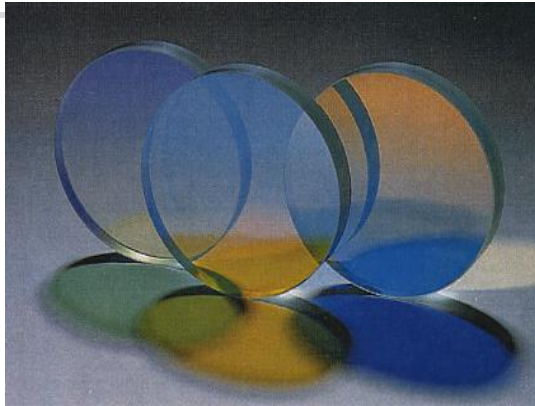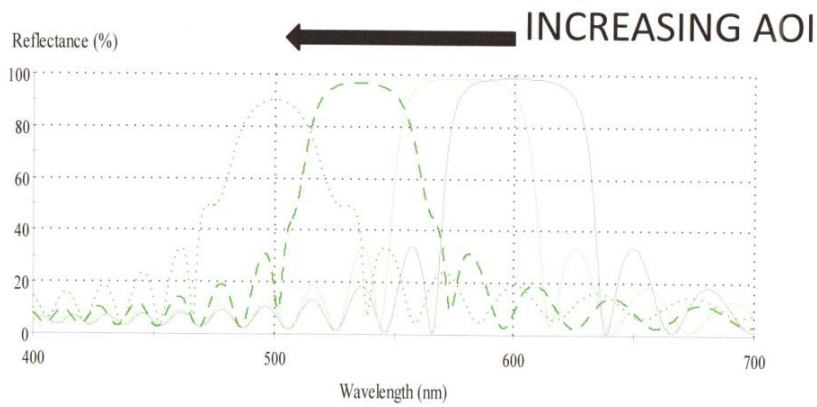
Figure 13. Colors reflected in dichroic filters.

Figure 14. Shift in high reflectance peak of $H(LH)^{20}$ ($H = 1.63$ (Al_2O_3) and $L = 1.48$ (SiO_2)) design with increased AOI.
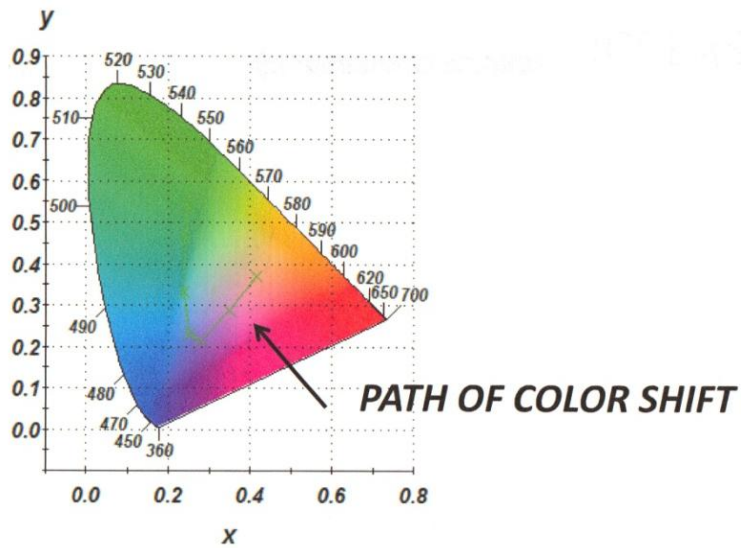

Figure 15. CIE chromaticity diagram for reflectance color shift of high reflector shown in Figure 13.

Color Shift Coatings

Color shift coatings, paints and inks are based on the change in reflectance of Fabry-Perot (FP) filters with angle of incidence. These and similar coatings are used in anticounterfeiting, including currency, labeling and packaging applications [17](http://ftgsoftware.com/fscolor1.htm). Figure 16 shows the basic concept of a color shift coating.

There are two basic types of FP filters, metal/dielectric (MD) and all-dielectric. We are concerned here with the MD type. The FP filter is a narrowband filter with basic design MDM, where M is a semitransparent metal layer, usually Al or Ag, and D is the dielectric cavity layer. While there are a number of more sophisticated designs, Figure 17 shows the reflectance of a Ag/SiO$_2$/Ag structure and Figure 18 shows a picture of a car with color shift paint (http://www.coatingsworld.com/contents/view_breaking-news/2008-10-15/ppg-says-global-vehicle-color-popularity-starting-/). Available color shifts are shown in the Figure. For example, color can shift from magenta to gold with a variation in AOI. In addition to the above applications, there are many applications for these pigments including football helmets, fabrics, golf clubs, pens, skis, calculators, and electric guitars.
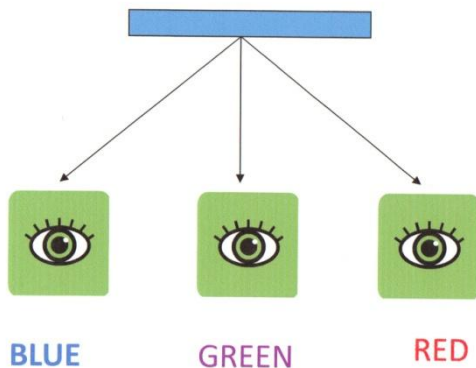

Figure 16. Color shift concept.

Figure 17. Shift in reflectance of Ag/SiO$_2$/Ag Fabry Perot filter.

Figure 18. Car with color shift paint.

Reflectance and Solar Control Coatings

As mentioned above, heat mirrors are used to reduce transmission of infrared light through a window to control temperature. As shown in Figure 19, heat mirrors are low-e coatings that transmit visible radiation and reflect infrared (heat) radiation. While a heat mirror can be as simple as a single layer of a transparent conductive oxide such as indium tin oxide (ITO), multilayer thin film dielectric/metal (DMD) structures provide better visible transmittance and infrared reflectance. Figure 20 shows the layer structure of low-e and solar control coatings with high infrared reflectance. The silver layer is ~ 80 Å thick and provides much of the infrared reflectance, which is enhanced by the dielectric layers. Heat mirrors are generally deposited onto flexible plastic films and sandwiched between a pane of glass. A low-e window can have as many as three heat mirror films sandwiched between either standard glass or low-e glass. Figure 21 shows transmittance and reflectance of the low-e heat mirror in Figure 20. Note the high infrared reflectance.
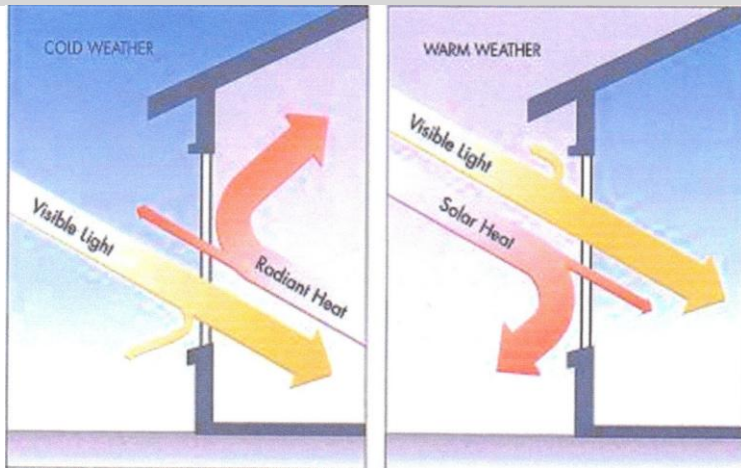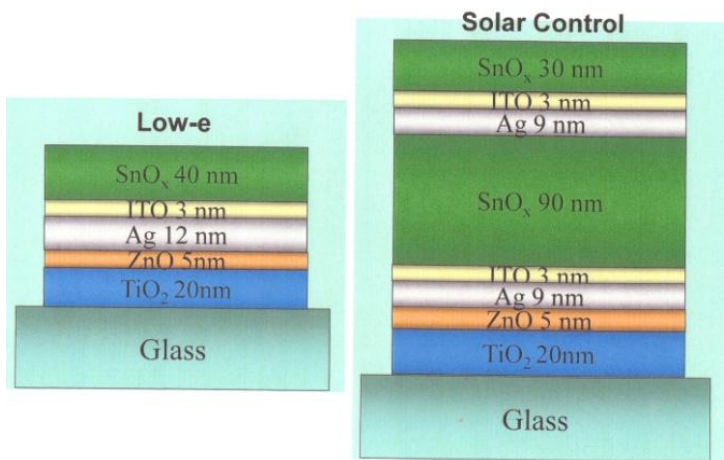
Figure 19. Heat mirror concept.

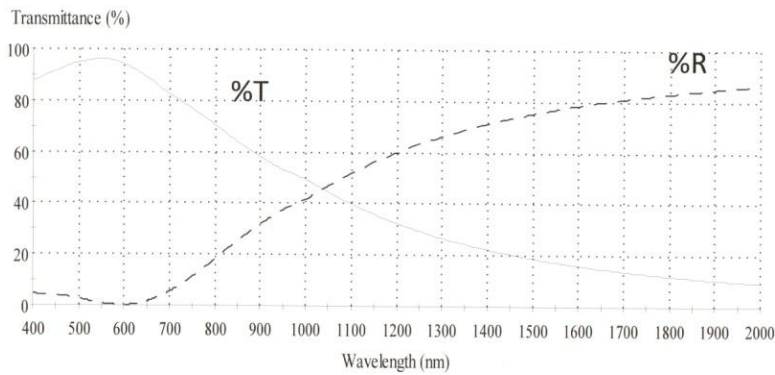Figure 20. Layer structures of low-e and solar control coatings (Courtesy Pilkington).

Figure 21. Reflectance and   transmittance of low-e coating showing high infrared reflectance.

Decorative and Wear Resistive Reflective Coatings

Finally, many thin film materials can change the color of a surface without interference effects. Thin film materials such as transition metal nitrides (TiN, HfN, ZrN, NbN, CrN and TaN) are applied to a wide range of objects for decorative and protective applications, and can have a gold, brass, or silvery color. There materials are extremely hard and wear resistant and can also reduce friction at the surface. Applications include plumbing fixtures, jewelry, hardware, cutting tools and medical implants. Transition metal nitrides are replacing highly toxic chromium in mirrors, plumbing fixtures and hardware. Figure 22a shows the reflectance spectrum of a TiN thin film and Figure 22b shows a picture of TiN coated dies. (http://books.google.com/?id=pbt-RWodmVAC&pg=PA193). Figure 23 shows the CIE chromaticity diagram for this film with an AOI shift from 0° to 60°. Note that reflectance of the film is virtually independent of AOI, behaving much like a metal. Similar to metals, TiN has a high electrical conductivity.
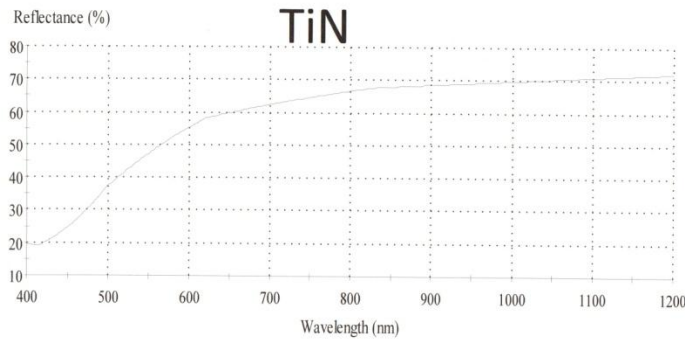

Figure 22a. Reflectance spectrum of TiN thin film.

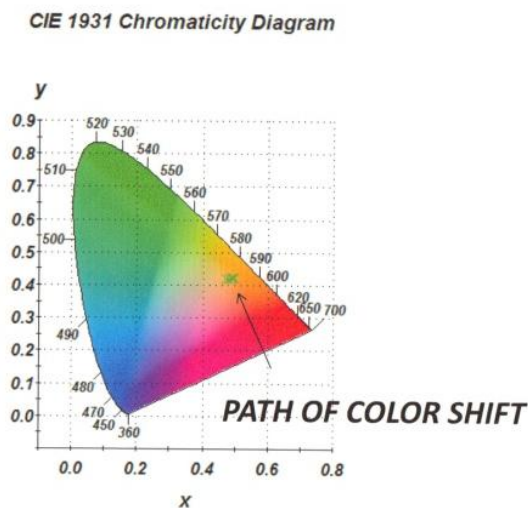Figure 22b. Picture of TiN coated dies.

Figure 23. CIE chromaticity diagram
For a TiN thin film for an AOI shift
from 0° to 60°.

Summary

This has been a brief review of the origins and applications of reflectance of surfaces and thin films. Reflectance is a ubiquitous property that is critical in a wide variety of applications, and results primarily from free electrons in the materials conduction band and interference effects. It is an intrinsic optical property of the material and surface quality, but can be enhanced or reduced by application of thin films. Reflective thin films can add functionality to virtually any surface or object. Reflectance of metals can easily be degraded by handling and environmental and surface damage. To this end, a transparent protective coating such as aluminum oxide should be applied over the metal layer.

Additionally, protective reflective coatings such as TiN are applied to less expensive materials for jewelry, plumbing and hardware fixtures to simulate gold and silver and improve lifetime. Brass can be simulated by NbN for application to plumbing and hardware fixtures, which makes them abrasion and corrosion resistant. These reflective thin film materials are replacing highly toxic chromium in mirrors, plumbing fixtures and hardware. Color shift coatings applied to currency, labels and packaging reduce the likelihood of counterfeiting and thus saving the consumer and taxpayer trillions of dollars. Reflectance of a diffuse surface can also be improved application of a polymer smoothing layer under a metal film, thus converting a poor reflector into a high reflector.

Multilayer coatings can be designed for high reflectance at virtually any wavelength or spectral region, and can change simple glass or plastic windows into a highly reflective mirror. Applications include low-e and solar control coatings, astronomical telescope mirrors, laser mirrors, satellite optics, DWDM filters, optical cavities, dichroic beam splitters, polarizers and electromagnetic shielding. Unlike polished metals, multilayer high reflector coatings can have almost perfect reflectivity and are used as laser mirrors.

Suggested Reading:

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