

1 Anti-reflection and polarising properties of a thin film

Consider the reflection due to the presence of a thin film on a substrate (example: TiO₂ thin film on a glass sheet) given by the expression

$$\mathcal{R} = \frac{r_{21} + r_{32}e^{2ik_zd}}{1 - r_{32}r_{12}e^{2ik_zd}}. \quad (1)$$

Here the various media would be Air (1), thin film medium (2) and the substrate material (3). The coefficients t_{ij} and r_{ij} are the Fresnel coefficients for transmission or reflection at the interface between the j^{th} and the i^{th} medium at the given angle and for a specified polarisation (p or s). Note that this is a periodic function of the phase $\delta = 2k_zd$ and unchanged on a change of the thickness of the thin film of $\Delta d = \lambda/(2n_2 \cos \theta_2)$. The maxima or minima of the reflectivity as a function of the thickness, obtained by setting $\partial|\mathcal{R}|^2/\partial d = 0$, occur when

$$d = m \frac{\lambda}{4n_2 \cos \theta_2} \quad \text{for } m = 0, 1, 2, 3, \dots$$

For normal incidence $\theta_2 = 0$, we have that the thickness should be an integral multiple of a quarter wavelength in the medium.

For odd m ($m = 1, 3, 5, \dots$), $e^{i\delta} = -1$, so that

$$|\mathcal{R}|^2 = \left(\frac{r_{21} - r_{32}}{1 - r_{21}r_{32}} \right)^2$$

At normal incidence, ($\theta_2 = 0$), we have

$$r_{21} = \frac{n_2 - n_1}{n_2 + n_1}, \quad r_{32} = \frac{n_3 - n_2}{n_3 + n_2} \quad \text{and} \quad |\mathcal{R}|^2 = \left(\frac{n_2^2 - n_1n_3}{n_2^2 + n_1n_3} \right)^2 \quad (2)$$

For even m , ($m = 2, 4, \dots$), $e^{i\delta} = 1$, in which case we have,

$$|\mathcal{R}|^2 = \left(\frac{r_{21} + r_{32}}{1 + r_{21}r_{32}} \right)^2$$

At normal incidence, ($\theta_2 = 0$), we will now have

$$|\mathcal{R}|^2 = \left(\frac{n_3 - n_1}{n_3 + n_1} \right)^2 \quad (3)$$

which is independent of the thin film. Thus, it is as if the thin film (medium-2) were absent. Note that the film thickness in these cases is half the wavelength in the medium in this case of even m .

Now we can decide whether we have a maximum or minimum reflection by analysing whether the second derivative $(\partial|\mathcal{R}|^2/\partial d)^2 < 0$ or > 0 . We obtain

$$(-1)^m r_{21} r_{32} [1 + r_{21}^2 r_{32}^2 - r_{21}^2 - r_{32}^2] > 0 \quad \text{or} \quad < 0$$

This leads to the conditions:

$$(-1)^m (n_1 - n_2)(n_2 - n_3) > 0 \quad \text{maximum}$$

$$(-1)^m (n_1 - n_2)(n_2 - n_3) < 0 \quad \text{minimum}$$

If the first medium is air ($n_1 = 1$), then we have a maximum if $n_2 > n_3$ and a minimum if $n_2 < n_3$ (note that m is always odd here and $(-1)^m = -1$).

The above tells us how to make an anti-reflection coating. For example, if we have a quarter wavelength thick film on top of a glass substrate, the reflectivity would be zero if $n_2^2 = n_1 n_3$. For glass ($n_3 = 1.5$), we have $n_2 = \sqrt{1.5} \simeq 1.225$ and there is no natural material that has this property. The closest is magnesium fluoride with $n_2 = 1.37$ or calcium fluoride with $n_2 = 1.43$, and some polymers with similar values ($n = 1.32$ to 1.42). There has been a continuous effort that has been concentrated to obtaining tailor made materials with low refractive index for such anti-reflection coatings. Examples are low density aerogels, porous thin films of silica or alumina, and more recently sculptured thin films with columnar structures. With the development of nano structured thin films ¹ and metamaterials, this requirement of materials with specific refractive indices at a specified frequency has essentially been solved.

2 Anti-reflection coatings and high reflection multi-layered coatings

Consider the reflection / transmission from multiple layers of materials (stratified material) with different refractive indices (or equivalently dielectric permittivities and magnetic permeabilities). We will first learn a technique by which we can calculate the reflectivity and transmission coefficients for a stack of layers. Since the layered geometry preserves the translational invariance along the layer, the parallel components of the wavevector in the different layers are all equal. Essentially each wave will create only one transmitted and one reflected wave at any interface. So

¹See for example, J.-Q. Xi et al, “Optical thin-film materials with refractive index for broadband elimination of Fresnel reflection” Nature Photon. Vol. 1, pp. 176 -179 (2007)

in each layer we will have two possible counter propagating waves ($e^{\pm ik_{j,z}d}$) where the Maxwell's relations within the homogenous layer medium require that

$$k_{j,z}^2 = \varepsilon_j \mu_j \frac{\omega^2}{c^2}.$$

The polarisation is also preserved in the scattered waves here - p-polarised light gives rise to only p-polarised light in the reflected or transmitted waves and similarly for the s-polarised light. Let

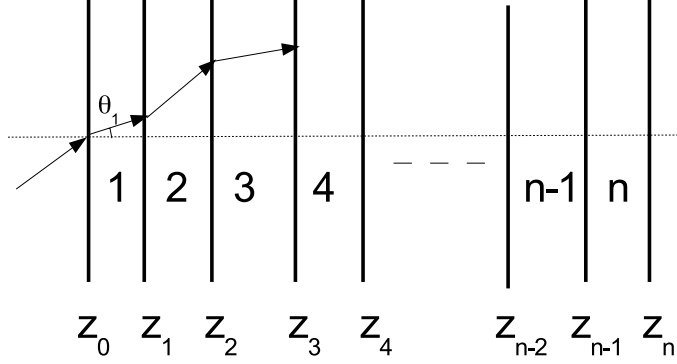


Figure 1: Schematic picture showing the transmission through a multi-layered stack of materials showing the coordinates of the individual layers.

us consider the p-polarised light with only one component of the magnetic field H_y as in Fig. 1. The magnetic field consists of the partial waves going towards the right and the left within the j^{th} layer and can be expressed as $H_{j,y}(z) = H_j^{(R)} \exp[ik_{j,z}z] + H_j^{(L)} \exp[-ik_{j,z}z]$. Equivalently, one can consider the field as a superposition of independent eigenmodes as

$$H_{j,y}(z) = A_j \cos(k_{j,z}z) + B_j \sin(k_{j,z}z). \quad (4)$$

The electric field can be obtained from Maxwell's equation as $\nabla \times \vec{H}_j = -i\omega\varepsilon_0\varepsilon_j E_j$, from which we obtain the parallel component of the electric field $E_{j,x}$ as

$$\begin{aligned} E_{j,x}(z) &= \frac{k_{j,z}}{i\omega\varepsilon_0\varepsilon_j} [-A_j \sin(k_{j,z}z) + B_j \cos(k_{j,z}z)] \\ &= -i\zeta_j [-A_j \sin(k_{j,z}z) + B_j \cos(k_{j,z}z)] \end{aligned} \quad (5)$$

where $\zeta_j = \sqrt{\mu_j/\varepsilon_j} \cos\theta_j$ is the impedance of the medium of the j^{th} layer multiplied by the cosine of the refraction angle. Now we will relate the tangential components of the magnetic

and electric fields between the left and the right edges of a given layer.

$$\begin{pmatrix} H_{j,y}(z_{j-1}) \\ E_{j,x}(z_{j-1}) \end{pmatrix} = \begin{pmatrix} P & Q \\ R & S \end{pmatrix} \begin{pmatrix} H_{j,y}(z_j) \\ E_{j,x}(z_j) \end{pmatrix}. \quad (6)$$

Noting that the electric and magnetic fields differ in their units by a factor of the impedance, we can construct the matrix above as

$$\tilde{T}_j = \begin{pmatrix} P & Q \\ R & S \end{pmatrix} = \begin{pmatrix} \cos(k_{j,z}d_j) & -\frac{i}{\zeta_j} \sin(k_{j,z}d_j) \\ -i\zeta_j \sin(k_{j,z}d_j) & \cos(k_{j,z}d_j) \end{pmatrix}, \quad (7)$$

where $d_j = z_j - z_{j-1}$ is the thickness of the j^{th} layer. This matrix is characteristic of the particular layer and involves the material properties and dimensions only of that layer. It relates the fields across its thickness and is hence called the *transfer matrix* for that layer. The tangential components of the fields on the left side of the $(j+1)^{th}$ layer are seen to be equal to the tangential components of the fields at the right side of the j^{th} layer on the other side of the interface.

$$\begin{pmatrix} H_{j,y}(z_{j-1}) \\ E_{j,x}(z_{j-1}) \end{pmatrix} = T_j \begin{pmatrix} H_{j+1,y}(z_j) \\ E_{j+1,x}(z_j) \end{pmatrix}$$

Hence, if one had knowledge of the transfer matrices for the all layers, one may write that

$$\begin{pmatrix} H_{0,y}(z_0) \\ E_{0,x}(z_0) \end{pmatrix} = T_1 T_2 T_3 \cdots T_{n-1} T_n \begin{pmatrix} H_{n+1,y}(z_n) \\ E_{n+1,x}(z_n) \end{pmatrix} \quad (8)$$

Thus, the fields on either sides of the stack are related by the product of the transfer matrices of the individual layers. Now we seek to apply some conditions to reflect the incoming wave conditions from the left only. We will hence express the waves once again as plane waves moving to the left and to the right as

$$\begin{aligned} H_{0,y}(z) &= H_0^{(R)} + H_0^{(L)} \quad \forall z < z_0 \\ E_{0,y}(z) &= -i\zeta_j [H_0^{(R)} - H_0^{(L)}] \quad \forall z < z_0, \end{aligned}$$

where the phase of the incoming wave is conveniently set to be zero at $z = z_0$. Hence we have

$$\begin{pmatrix} H_{0,y}(z_0) \\ E_{0,x}(z_0) \end{pmatrix} = \begin{pmatrix} 1 & 1 \\ -i\zeta_0 & i\zeta_0 \end{pmatrix} \begin{pmatrix} H_0^{(R)} \\ H_0^{(L)} \end{pmatrix} = S_0 \begin{pmatrix} H_0^{(R)} \\ H_0^{(L)} \end{pmatrix}. \quad (9)$$

Similarly, at the right hand side, we have

$$\begin{pmatrix} H_{n+1,y}(z_n) \\ E_{n+1,x}(z_n) \end{pmatrix} = \begin{pmatrix} 1 & 1 \\ -i\zeta_{n+1} & i\zeta_{n+1} \end{pmatrix} \begin{pmatrix} H_{n+1}^{(R)} \\ H_{n+1}^{(L)} \end{pmatrix} = S_{n+1} \begin{pmatrix} H_{n+1}^{(R)} \\ H_{n+1}^{(L)} \end{pmatrix}. \quad (10)$$

Having only one incoming wave from the left hand side, we have $H_{inc} = H_0^{(R)} = 1$ and the reflectivity $r = H_0^{(L)}$. On the right hand side, we have the boundary conditions for only a outgoing wave as $H_{n+1}^{(L)} = 0$ and the transmission coefficient can be recognised as $t = H_{n+1}^{(R)}$. Hence we have

$$S_0 \begin{pmatrix} 1 \\ r \end{pmatrix} = \tilde{T} S_{n+1} \begin{pmatrix} t \\ 0 \end{pmatrix} \quad (11)$$

where $\tilde{T} = \prod_{j=1}^n \tilde{T}_j$. Defining the matrix $M = S_0^{-1} \tilde{T} S_{n+1}$, where

$$M = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix},$$

we can write the remittances as

$$t = \frac{1}{M_{22}}, \quad (12)$$

$$r = \frac{M_{21}}{M_{22}}. \quad (13)$$

This is known as the *transfer matrix method* and allows you to calculate the remittances for an arbitrary layered stack. note that the above remittances are the ratio of the magnetic fields

For s-polarised light, one can simply interchange H by E and μ by ε in these expressions and obtain the remittances as the ratios of the electric fields. The corresponding power fluxes in the z -direction are obtained by $I = 1/2c\zeta|H|^2 = 1/2c/\zeta|E|^2$ where $\zeta = \sqrt{\mu/\varepsilon} \cos \theta$ in the corresponding medium. The appropriate expression should be used to calculate the reflectance or transmittance.

As an exercise, compute the r and t for a layered stack of 10 alternating thin films of SiO_2 and TiO_2 of thicknesses 50 nm and 100 nm respectively by the transfer matrix method for both the polarisations as a function of the incident angle and plot it.

2.1 high reflectivity coatings from layered stacks

High reflection coatings require alternating layers of high and low refractive indices. Let each layer have a thickness of integral number of quarter wavelengths, implying that $k_{j,z}d_j = \pi/2$ – a condition that comes from our discussion of maximum reflection from a single layer. Since there is a phase shift of π in reflection for a wave incident from a low index to a high index layer, and the layers alternate, this occurs at every other boundary. The quarter wavelength spacing gives the maximum reflectivity as the reflected waves from each layer meets the wave from the previous layer in phase as shown in Fig. 2.

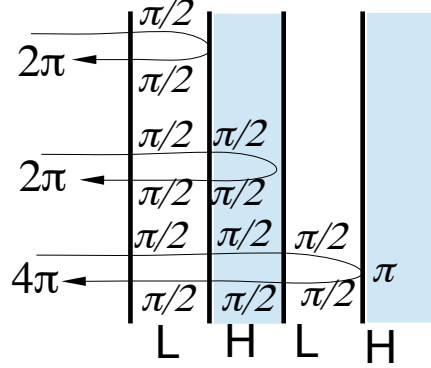


Figure 2: Schematic picture showing the partial waves for reflection constructively interfering in a multi-layered stack of high (H) and low (L) refractive index layers with quarter wavelength thicknesses. This causes a phase shift of $\pi/2$ upon traversal across the layer and a phase shift of π is induced whenever the wave that is incident from a low-index medium reflects at the interface between low index and a high index interface.

The transfer matrix in these cases (for p-polarised light) for the low index layer and the high index layer becomes

$$T_j^L = \begin{pmatrix} 0 & \frac{-i}{\zeta_j^L} \\ -i\zeta_j^L & 0 \end{pmatrix}, \quad \text{and} \quad T_j^H = \begin{pmatrix} 0 & \frac{-i}{\zeta_j^H} \\ -i\zeta_j^H & 0 \end{pmatrix}.$$

Thus, we have

$$\begin{pmatrix} 0 & -\frac{i}{\zeta_j^L} \\ -i\zeta_j^L & 0 \end{pmatrix} \begin{pmatrix} 0 & -\frac{i}{\zeta_j^H} \\ -i\zeta_j^H & 0 \end{pmatrix} = \begin{pmatrix} -\frac{\zeta_j^H}{\zeta_j^L} & 0 \\ 0 & -\frac{\zeta_j^L}{\zeta_j^H} \end{pmatrix}.$$

The produce of N such matrices is trivial to evaluate

$$\begin{pmatrix} -\frac{\zeta_j^H}{\zeta_j^L} & 0 \\ 0 & -\frac{\zeta_j^L}{\zeta_j^H} \end{pmatrix}^N = \begin{pmatrix} \left(-\frac{\zeta_j^H}{\zeta_j^L}\right)^N & 0 \\ 0 & \left(-\frac{\zeta_j^L}{\zeta_j^H}\right)^N \end{pmatrix}.$$

Noting that the impedance is inversely proportional to the refractive index, we note that $\left(-\frac{\zeta_j^H}{\zeta_j^L}\right)^N \rightarrow 0$ for $\zeta_j^H < \zeta_j^L$ and large N . Thus, we obtain the remittances as $t \rightarrow 0$ and

$r \rightarrow -1$. This is the essence of designing a high-reflectivity coating. Obviously the reflective property depends on the direction of the incident light, and the wavelength. As an exercise (Assignment), you are advised to design a high-reflectivity coating using 10 alternate layers of CaF_2 (low index) and ZnS (high index) at 1064 nm for 45° incidence. Using the known dispersions of these materials, plot the reflectivity as a function the wavelength across the wavelength range of 500 nm to 3000 nm.