8. Multilayer Films

Optical surfaces having virtually any desired reflectance and transmittance characteristics may be produced by means of thin film coatings. The purpose of this chapter is to find an orderly method for analyzing multi-layer thin films. We will first derive the characteristic matrix. From the elements of the characteristic matrix we will solve for the coefficients of reflection and transmission. Then we will look at examples of anti-reflection coatings and high-reflectance coatings.

8.1 Theory

Tangential components of $\vec{E}$ and $\vec{H}$ are continuous at an interface. (Figure 1 is from Optics by Hecht.)

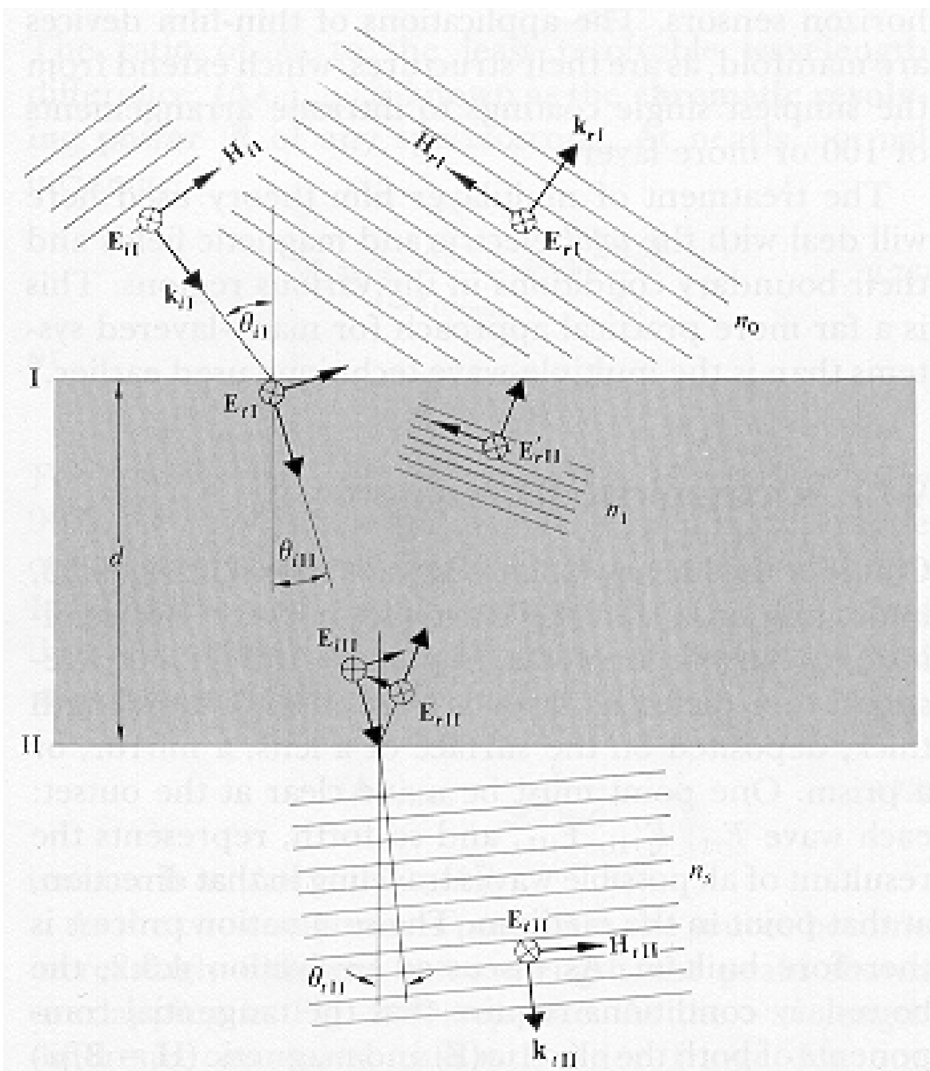


Figure 1

Boundary I

$$E_I = E_{iI} + E_{rI} = E_{tI} + E'_{rII} \qquad (1)$$

E_{iI}, E_{rI}, E_{tI}, and E'_{rII} represent the resultant of all waves traveling in a given direction.

$$\vec{H} = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n\,\vec{k} \times \vec{E} \quad ; \quad n \equiv \frac{c}{v} \equiv \sqrt{\frac{\mu\epsilon}{\mu_o\,\epsilon_o}}$$

The continuity of the tangential component of H gives us

$$H_I = \sqrt{\frac{\epsilon_o}{\mu_o}}\,(E_{iI} - E_{rI})\, n_o \,\mathrm{Cos}[\theta_{iI}] = \sqrt{\frac{\epsilon_o}{\mu_o}}\,(E_{tI} - E'_{rII})\, n_1 \,\mathrm{Cos}[\theta_{iII}] \qquad (2)$$

Boundary II

$$E_{II} = E_{iII} + E_{rII} = E_{tII} \qquad (3)$$

$$H_{II} = \sqrt{\frac{\epsilon_o}{\mu_o}}\,(E_{iII} - E_{rII})\, n_1 \,\mathrm{Cos}[\theta_{iII}] = \sqrt{\frac{\epsilon_o}{\mu_o}}\, E_{tII}\, n_s \,\mathrm{Cos}[\theta_{tII}] \qquad (4)$$

Let

$$k_o\,(n_1\, d\, \mathrm{Cos}[\theta_{iII}]) = k_o\, h$$

Therefore

$$E_{iII} = E_{tI}\, e^{i\, k_o\, h}; \qquad E_{rII} = E'_{rII}\, e^{-i\, k_o\, h}$$

Note that the sign of the exponent is different from some books because I write $e^{i\,(kz-\omega t)}$ instead of $e^{-i\,(kz-\omega t)}$. Thus the boundary conditions at boundary II can be written as

$$E_{II} = E_{tI}\, e^{i\, k_o\, h} + E'_{rII}\, e^{-i\, k_o\, h} \qquad (5)$$

$$H_{II} = (E_{tI}\, e^{i\, k_o\, h} - E'_{rII}\, e^{-i\, k_o\, h})\sqrt{\frac{\epsilon_o}{\mu_o}}\, n_1\,\mathrm{Cos}[\theta_{iII}] \qquad (6)$$

Solving the last two equations for E_{tI} and E'_{rII} and substituting into Equations 1 and 2 for boundary I yields

$$E_I = E_{II}\,\mathrm{Cos}[k_o\, h] - H_{II}\,(i\,\mathrm{Sin}[k_o\, h])\,/\,\gamma_1 \qquad (7)$$

and

$$H_I = -E_{II}\,\gamma_1\, i\,\mathrm{Sin}[k_o\, h] + H_{II}\,\mathrm{Cos}[k_o\, h] \qquad (8)$$

where

$$\gamma_1 = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_1 \,\text{Cos}[\theta_{\text{iII}}];$$

If we went through the same derivation for E in the plane of incidence we would obtain a similar equation except

$$\gamma_1 = \frac{\sqrt{\frac{\epsilon_o}{\mu_o}}\, n_1}{\text{Cos}[\theta_{\text{iII}}]};$$

In matrix notation

$$\begin{pmatrix} E_{\text{I}} \\ H_{\text{I}} \end{pmatrix} = \begin{pmatrix} \text{Cos}[k_o\, h] & -i\,\text{Sin}[k_o\, h]/\gamma_1 \\ -\gamma_1\, i\,\text{Sin}[k_o\, h] & \text{Cos}[k_o\, h] \end{pmatrix} . \begin{pmatrix} E_{\text{II}} \\ H_{\text{II}} \end{pmatrix} \tag{9}$$

or

$$\begin{pmatrix} E_{\text{I}} \\ H_{\text{I}} \end{pmatrix} = M_{\text{I}} . \begin{pmatrix} E_{\text{II}} \\ H_{\text{II}} \end{pmatrix} \tag{10}$$

The characteristic matrix M_I relates the field at two adjacent boundaries.

If two overlaying films are deposited on one substrate there would be three boundaries and

$$\begin{pmatrix} E_{\text{II}} \\ H_{\text{II}} \end{pmatrix} = M_{\text{II}} . \begin{pmatrix} E_{\text{III}} \\ H_{\text{III}} \end{pmatrix} \tag{11}$$

For p layers

$$\begin{pmatrix} E_{\text{I}} \\ H_{\text{I}} \end{pmatrix} = M_{\text{I}} . M_{\text{II}} \cdots M_p \begin{pmatrix} E_{p+1} \\ H_{p+1} \end{pmatrix} \tag{12}$$

$$M = M_{\text{I}} . M_{\text{II}} \cdots M_p = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} \tag{13}$$

To see how this fits together we will derive the expression for the amplitude coefficient of reflection and transmission. Let

$$\gamma_o = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_o \,\text{Cos}[\theta_{\text{iI}}];$$

$$\gamma_s = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_s \,\text{Cos}[\theta_{\text{tII}}];$$

Combining Equations 1-4 and 10 yields

$$\begin{pmatrix} E_{\text{iI}} + E_{\text{rI}} \\ (E_{\text{iI}} - E_{\text{rI}})\, \gamma_o \end{pmatrix} = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} . \begin{pmatrix} E_{\text{tII}} \\ E_{\text{tII}}\, \gamma_s \end{pmatrix}$$

$$E_{\text{iI}} + E_{\text{rI}} = m_{11}\, E_{\text{tII}} + m_{12}\, E_{\text{tII}}\, \gamma_s$$

$$(E_{\text{iI}} - E_{\text{rI}})\, \gamma_o = m_{21}\, E_{\text{tII}} + m_{22}\, E_{\text{tII}}\, \gamma_s$$

Let

$$r = \frac{E_{rI}}{E_{iI}}; \qquad t = \frac{E_{tII}}{E_{iI}};$$

Then

$$1 + r = m_{11}\, t + m_{12}\, \gamma_s\, t$$

$$(1 - r)\, \gamma_o = m_{21}\, t + m_{22}\, \gamma_s\, t$$

Solving for r and t yields

```
Solve[{1 + r == m11 t + m12 γs t, (1 - r) γo == m21 t + m22 γs t}, {r, t}]
```

$$\left\{\left\{r \to -1 - \frac{2\,\gamma_o\,(-m_{11} - m_{12}\,\gamma_s)}{m_{21} + m_{11}\,\gamma_o + m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s},\ t \to \frac{2\,\gamma_o}{m_{21} + m_{11}\,\gamma_o + m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s}\right\}\right\}$$

Consequently,

$$\mathbf{r = Together\left[-1 - \frac{2\,\gamma_o\,(-m_{11} - m_{12}\,\gamma_s)}{m_{21} + m_{11}\,\gamma_o + m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s}\right]}$$

$$\frac{-m_{21} + m_{11}\,\gamma_o - m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s}{m_{21} + m_{11}\,\gamma_o + m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s}$$

$$\mathbf{t = \frac{2\,\gamma_o}{m_{21} + m_{11}\,\gamma_o + m_{22}\,\gamma_s + m_{12}\,\gamma_o\,\gamma_s};}$$

Therefore for any combination of films we only need to compute the characteristic matrix and substitute the matrix elements into the above.

$$R = \mathrm{Abs}[r]^2; \quad T = \mathrm{Abs}[t]^2;$$

8.2 Anti-reflection coatings (AR)

We will look at the case of normal incidence.

$$\theta_{iI} = 0; \quad \theta_{iII} = 0; \quad \theta_{tII} = 0;$$

$$\gamma_o = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_o; \qquad \gamma_s = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_s; \qquad \gamma_1 = \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_1$$

$$M = \begin{pmatrix} \mathrm{Cos}[k_o\, h] & -i\,\mathrm{Sin}[k_o\, h]/\gamma_1 \\ -\gamma_1\, i\,\mathrm{Sin}[k_o\, h] & \mathrm{Cos}[k_o\, h] \end{pmatrix}$$

Since only 1 layer present we will write the amplitude reflectance as r_1.

```
r1 = r;
```

```
r1 = Simplify[r1 //.
   {m11 → Cos[ko h], m12 → -ⅈ Sin[ko h] / γ1, m21 → -γ1 ⅈ Sin[ko h], m22 → Cos[ko h]}]
```

$$\frac{-\mathrm{Sin}[h\,k_o]\,\gamma_1^2 + i\,\mathrm{Cos}[h\,k_o]\,\gamma_1\,(\gamma_o - \gamma_s) + \mathrm{Sin}[h\,k_o]\,\gamma_o\,\gamma_s}{\mathrm{Sin}[h\,k_o]\,\gamma_1^2 + \mathrm{Sin}[h\,k_o]\,\gamma_o\,\gamma_s + i\,\mathrm{Cos}[h\,k_o]\,\gamma_1\,(\gamma_o + \gamma_s)}$$

$$\mathbf{r_1 = Simplify\left[r_1 \;//.\; \left\{\gamma_o \to \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_o,\ \gamma_s \to \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_s,\ \gamma_1 \to \sqrt{\frac{\epsilon_o}{\mu_o}}\, n_1\right\}\right]}$$

$$\frac{-\mathrm{Sin}[h\,k_o]\,n_1^2 + i\,\mathrm{Cos}[h\,k_o]\,n_1\,(n_o - n_s) + \mathrm{Sin}[h\,k_o]\,n_o\,n_s}{\mathrm{Sin}[h\,k_o]\,n_1^2 + \mathrm{Sin}[h\,k_o]\,n_o\,n_s + i\,\mathrm{Cos}[h\,k_o]\,n_1\,(n_o + n_s)}$$

$$R_1 = \frac{\text{Cos}[h\,k_o]^2\,(n_o - n_s)^2\,n_1^2 + \text{Sin}[h\,k_o]^2\,(n_o\,n_s - n_1^2)^2}{\text{Cos}[h\,k_o]^2\,(n_o + n_s)^2\,n_1^2 + \text{Sin}[h\,k_o]^2\,(n_o\,n_s + n_1^2)^2};$$

This becomes a simple formula if

$$k_o\,h = \frac{\pi}{2}$$

$$k_o\,h = \frac{2\,\pi}{\lambda_o}\,(n_1\,d); \qquad n_1\,d = \frac{\lambda_o}{4}$$

$$R_1\; //.\; k_o\,h \to \frac{\pi}{2}$$

$$\frac{(-n_1^2 + n_o\,n_s)^2}{(n_1^2 + n_o\,n_s)^2}$$

$$R_1 = 0 \quad \text{if} \quad n_1 = \sqrt{n_o\,n_s}$$

If $n_o = 1$ and $n_s = 1.5$, $n_1 = \sqrt{1.5} = 1.225$.

Commonly use $M_g\,F_2$ which has an index of 1.38 which is larger than desired. However, a single 1/4 λ layer reduces the reflectance from 4% to approximately 1% over the entire visible spectrum.

The reflectance can be reduced by using a double layer $\lambda/4$ AR coating. As an example, put index n_2 on the substrate and index n_1 on top of n_2.

$$M = M_1 . M_2$$

$$M = \begin{pmatrix} 0 & -i/\gamma_1 \\ -i\,\gamma_1 & 0 \end{pmatrix} . \begin{pmatrix} 0 & -i/\gamma_2 \\ -i\,\gamma_2 & 0 \end{pmatrix} \; // \; \text{MatrixForm}$$

$$\begin{pmatrix} -\frac{\gamma_2}{\gamma_1} & 0 \\ 0 & -\frac{\gamma_1}{\gamma_2} \end{pmatrix}$$

Since $\gamma_i = \sqrt{\frac{\epsilon_o}{\mu_o}}\;n_i$

$$M = \begin{pmatrix} -\frac{n_2}{n_1} & 0 \\ 0 & -\frac{n_1}{n_2} \end{pmatrix};$$

$$R_2 = \left(\frac{m_{11}\,\gamma_o - m_{22}\,\gamma_s}{m_{11}\,\gamma_o + m_{22}\,\gamma_s}\right)^2;$$

$$R_2 = \left(\frac{n_2^2\,n_o - n_s\,n_1^2}{n_2^2\,n_o + n_s\,n_1^2}\right)^2;$$

$$R_2 = 0 \quad \text{if} \quad \left(\frac{n_2}{n_1}\right)^2 = \frac{n_s}{n_o}$$

Thus, $n_2 > n_1$.

Common materials for the high refractive index are zirconium dioxide, n = 2.1; titanium dioxide, n= 2.4; zinc sulfide, n = 2.32. Common materials for the low refractive index are magnesium fluoride, n = 1.38 and cerium fluoride, n = 1.63.

By using two layers, one of high index and one of low index, zero reflectance can be obtained at one wavelength. Three layers can give zero reflectance at two wavelengths, etc.

8.3 High-reflectance coatings

The simplest periodic thin film system is a quarter-wave stack which is made up of a number of quarter-wave layers. For example, $s(\mathrm{HL})^3\, a$, where s is the substrate and a is air. As an example we will look at the reflectance of $s(n_2\, n_1)^p\, a$. From above it follows that

$$M = \begin{pmatrix} \left(-\frac{n_2}{n_1}\right)^p & 0 \\ 0 & \left(-\frac{n_1}{n_2}\right)^p \end{pmatrix};$$

$$R_{2\,p} = \left(\frac{n_o \left(-\frac{n_2}{n_1}\right)^p - n_s \left(-\frac{n_1}{n_2}\right)^p}{n_o \left(-\frac{n_2}{n_1}\right)^p + n_s \left(-\frac{n_1}{n_2}\right)^p}\right)^2;$$

$$\mathbf{R_{2\,p}} = \left(\frac{1 - \frac{n_s}{n_o}\left(\frac{n_1}{n_2}\right)^{2p}}{1 + \frac{n_s}{n_o}\left(\frac{n_1}{n_2}\right)^{2p}}\right)^2;$$

For zero reflectance

$$\left(\frac{n_2}{n_1}\right)^{2p} = \frac{n_s}{n_o}$$

Therefore, $n_2 > n_1$.

We previously looked at the case where p=1.

For high reflectance we want $n_2 >> n_1$ or $n_2 << n_1$. Since $n_s > n_o$, the result will converge a little faster if $n_1 > n_2$. The reflectance becomes higher as p increases. For a given p, the larger the ratio of refractive indices the better we are.

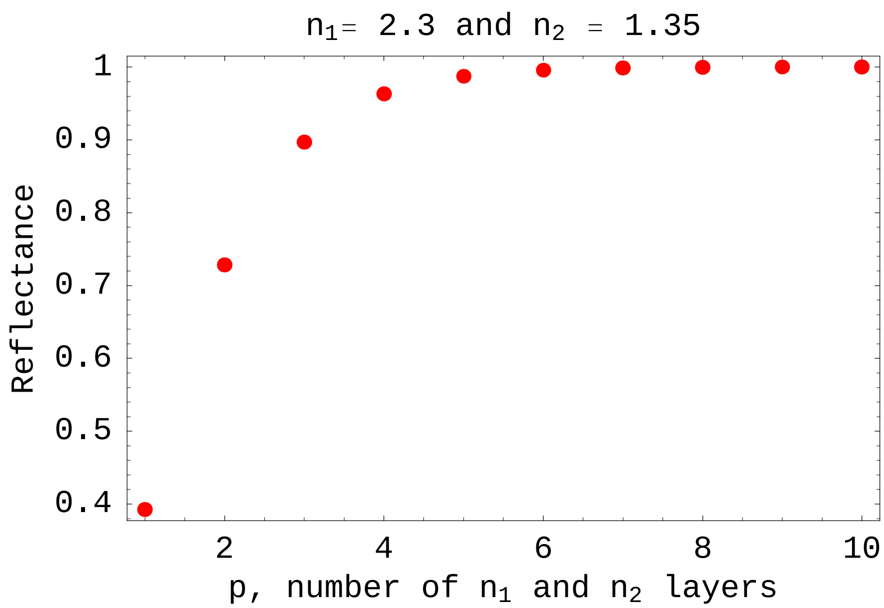
```
R2 p /. {n1 → 1.35, n2 → 2.3, p → 4, ns → 1.5,  no → 1}
0.918935
```

```
R2 p /. {n1 → 2.3, n2 → 1.35, p → 4, ns → 1.5,  no → 1}
0.963128
```

Having $n_1 > n_2$ helped increase the reflectance. If p becomes 8 (16 layers) the reflectivity is greater than 99.9%.

```
R2 p /. {n1 → 2.3, n2 → 1.35, p → 8, ns → 1.5,  no → 1}
0.999471
```

The following plot shows the reflectance as a function of p, the number of $n_1\, n_2$ layers.



By using various combinations of quarter-wave stacks it is possible to make band pass filters, high pass filters, low pass filters, etc. An excellent book on the topic is *Thin-Film Optical Filters* by H. A. Macleod.

8.4 Non-normal incidence

At non-normal incidence up to approximately 30^o there is generally little degradation in the response of thin film coatings. In general, the effect of increasing angle is a shift in the reflectance curve down to shorter wavelengths. Remember one of my favorite equations $2\ n\ d\ \mathrm{Cos}[\theta] = m\ \lambda$. If θ increases, $\mathrm{Cos}[\theta]$ decreases, and therefore we would expect λ to decrease.