

## Chapter 4 Heterodyne Interferometry Kinetics

### 4.1 Film thickness measurement

A plane wave light propagating in a direction  $\tilde{s}$  can be expressed as electromagnetic waves<sup>1</sup> shown in equation 3.1.

$$\tilde{E} = \tilde{A} e^{i(\tilde{k} \cdot \tilde{r} + \omega t)}, \tilde{k} = \frac{\omega}{c} \tilde{n}(\omega, \tilde{s}) \tilde{s} \quad (4.1)$$

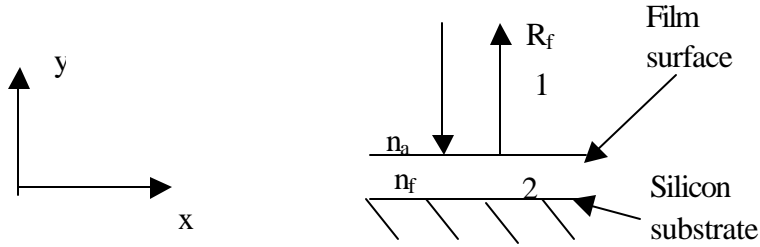
where  $\tilde{E}$  is the electric field vector.  $\tilde{A}$  is the complex vector (i.e. field amplitude), independent of coordinates and time,  $\omega$  is the angular frequency and the angular frequency  $\omega = 2\pi\nu$ , where  $\nu$  is light frequency and light frequency  $\nu = c/\lambda$ ,  $c$  is the speed of light and  $\lambda$  is the wavelength of the light.  $\tilde{k}$  is the wave vector,  $\tilde{n}$  is the complex index of refraction and  $\tilde{s}$  is a unit vector.

When electrical field  $\tilde{E}$  is parallel to the principle direction of refraction index as shown in figure 4.1a, 4.1b and 4.1c. The refractive index can be reduced to scalar. Therefore, the reference beam and the sample beam as shown in figure 3.1 can be reduced to scalar function. The reference beam at the photodetector is phase modulated by Bragg cell and can be expressed as,

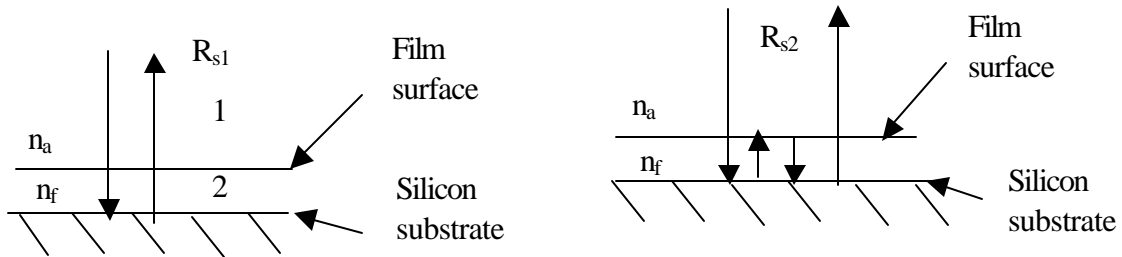
$$E_r = E_{r0} e^{i(\omega t + kx + \phi_r)} \quad (4.2)$$

Where  $E_r$  is the electric field of the reference beam.  $E_{r0}$  is the electric field amplitude of the reference beam.  $\omega$  is the laser angular frequency and the laser angular frequency  $\omega = 2\pi\nu$ , where  $\nu$  is laser frequency and laser frequency  $\nu = c/\lambda$ ,  $c$  is the speed of laser and  $\lambda$  is the wavelength of the laser.  $k$  is the wave vector  $k = 2\pi/\lambda$ .  $\phi_r$  is the phase of the reference beam and the phase of the reference beam  $\phi_r = \omega_b t$ .

The laser has multiple reflections at the film side shown in figure 4.1a, 4.1b, and 4.1c.



**Figure 4.1a** The first reflection at the film surface



**Figure 4.1b** The first reflection at the silicon

**Figure 4.1c** The second reflection at the silicon

\* The coordination is valid for all three figures 4.1a, 4.1b, and 4.1c. x axis is the principle direction which is parallel to the direction of applied electric field. y axis is the direction perpendicular to the film. z axis is the direction perpendicular to x and y axis and pointing out of the plane. Define  $R_f$  is the Fresnel reflectivity at the film.  $R_{s1}$  is the reflectivity of the silicon.  $R_{s2}$  are the reflectivity for the second reflection at the silicon.

$n_a$  is the refractive index of air,  $n_a=1.0$ .  $n_f$  is the refractive index of the film,  $n_f=1.453$ . The reflectivity of the silicon  $R_{s1}=0.72$  for 633nm. The reflectivity of  $R_f$  can be calculated as,

$$R_f = \frac{(n_f - n_a)^2}{(n_f + n_a)^2} = 0.034 \quad (4.3)$$

The reflectivity of  $R_{s2}$  can be calculated as,

$$R_{s2} = R_{s1} \cdot R_f \cdot R_{s1} = 0.017 \quad (4.4)$$

From above calculation, we can see that the multiple reflections are small. Consider the absorption of the film. Define  $I_0$  is the intensity of incident sample beam.  $I_{Rf}$  is the intensity of light reflected at the film surface.  $I_{Rs1}$  is the intensity of light reflected first time from the silicon.  $I_{Rs2}$  is the intensity of light reflected second time on silicon.  $\alpha$  is the absorption coefficient of polymer.  $Z$  is the thickness of the film.

$$\begin{aligned} I_{Rf} &= I_0 R_f = 0.034 I_0 \\ I_{Rs1} &= I_0 R_{s1} e^{-\alpha 2Z} = 0.72 I_0 e^{-\alpha 2Z} \\ I_{Rs2} &= I_0 R_{s2} e^{-2\alpha Z} e^{-2\alpha Z} = 0.017 I_0 e^{-4\alpha Z} \end{aligned} \quad (4.5)$$

The absorption coefficient of polymer and the thickness of the film is very small,  $\alpha Z \approx 0$ , therefore, we can neglect the film absorption. Since the intensity of the reflected sample beam is different from the sample incident beam. We include a factor  $\gamma$  to modify the intensity of the sample reflected beam which interferes with the reference beam. The reflected sample beam is expressed as,

$$E_s = g E_{s0} e^{i(\omega t + kx + \mathbf{f}_s + \Delta \mathbf{f})} \quad (4.6)$$

Where  $E_s$  is the electric vector of the sample beam.  $\phi_s$  is the phase of the reflected sample beam.  $E_{s0}$  is the electric field amplitude of the incident sample beam. Define  $E_{s0}'$  as the amplitude of the reflected beam and set  $E_{s0}' = \gamma E_{s0}$  to simplify the derivation.  $\Delta \phi$  is the phase change of reflected beam due to the optical path of the sample which equals to the refractive index of the film times film thickness and can be expressed as,

$$\Delta \mathbf{f} = 2nkZ_0(1 - \cos \omega_g t) / 2 = nkZ_0 - nkZ_0 \cos(\omega_g t) \quad (4.7)$$

Where  $Z_0$  is the initial film thickness,  $\omega_g$  is the frequency of galvano mirror and  $k=2\pi/\lambda$ . Total power at the detector is given by,

$$\begin{aligned} P &= (E_r + E_s)(E_r + E_s)^* \\ P &= E_r^2 + E_s^2 + E_r E_s^* + E_r^* E_s \end{aligned} \quad (4.8)$$

By substituting equation 4.2 and 4.6 into equation 4.8, and rearrange, we obtain,

$$P = E_{r0}^2 + E_{s0}^2 + 2E_{r0}E_{s0} \cos(\mathbf{w}_b t - \mathbf{f}_s - \Delta \mathbf{f}) \quad (4.9)$$

Substituting equation 4.7 into equation 4.9, we obtain,

$$P = E_{r0}^2 + E_{s0}^2 + 2E_{r0}E_{s0} \cos(\mathbf{w}_b t - \mathbf{f}_s - nkZ_0 + nkZ_0 \cos(\mathbf{w}_g t)) \quad (4.10)$$

Next we set

$$\mathbf{f}_s - nkZ_0 = \mathbf{f}_1 \quad (4.11)$$

Simplify equation 4.10 to obtain,

$$P = E_{r0}^2 + E_{s0}^2 + 2E_{r0}E_{s0} \cos(\mathbf{w}_b t - \mathbf{f}_1 + nkZ_0 \cos(\mathbf{w}_g t)) \quad (4.12)$$

In equation 4.12,  $(E_{r0}^2 + E_{s0}^2)$  is constant power, invariant of time, (i.e. DC signal).

The time dependent component ( $P'$ ) is given by:

$$P' = 2E_{r0}E_{s0} \cos(\mathbf{w}_b t - \mathbf{f}_1 + nkZ_0 \cos(\mathbf{w}_g t)) \quad (4.13)$$

Due to the reflectivity change and scattering at the edge of film, the amplitude modulation factor is considered. Wafer may absorb moisture on the surface. All these factors will introduce a phase lag of amplitude with the galvanometer rocking frequency. To correct this effect, a background factor  $\phi_g$  is introduced.  $S$  is the reflectivity of the sample surface and  $S_0$  is the initial reflectivity of the sample.  $S$  and  $S_0$  can be measured by the reflectivity measurement. Expand  $S/S_0$  at  $S_0$  and taking two terms for approximation.

$$s / s_0 = 1 + a \cos(\mathbf{w}_g t + \mathbf{f}_g) \quad (4.14)$$

where  $a = S/S_0$ .

By substituting equation 4.13 into 4.14, we obtain,

$$P' \approx 2E_{r0}E_{s0}'(1 + a \cos(\mathbf{w}_g t + \mathbf{f}_g))(\cos(\mathbf{w}_b t - \mathbf{f}_1) - nkZ_0 \cos(\mathbf{w}_g t) \sin(\mathbf{w}_b t - \mathbf{f}_1)) \quad (4.15)$$

By expansion and simplification of equation 4.15, we obtain,

$$P' \approx 2E_{r0}E_{s0}'(\cos(\mathbf{w}_b t - \mathbf{f}_1) + a \cos(\mathbf{w}_g t + \mathbf{f}_g) \cos(\sin(\mathbf{w}_b t - \mathbf{f}_1)) - nkZ_0 / 2 \sin((\mathbf{w}_b + \mathbf{w}_g)t - \mathbf{f}_1) - nkZ_0 / 2 \sin((\mathbf{w}_b - \mathbf{w}_g)t - \mathbf{f}_1)) \quad (4.16)$$

Further expand equation 4.16 and obtain,

$$P' \approx 2E_{r0}E_{s0}'(\cos(\mathbf{w}_b t - \mathbf{f}_1) - (nkZ_0 + a \sin \mathbf{f}_g) / 2 \sin((\mathbf{w}_b + \mathbf{w}_g)t - \mathbf{f}_1)) + a / 2 \cos \mathbf{f}_g \cos((\mathbf{w}_b + \mathbf{w}_g)t - \mathbf{f}_1) + (a \sin \mathbf{f}_g - nkZ_0) / 2 \sin((\mathbf{w}_b - \mathbf{w}_g)t - \mathbf{f}_1) + a / 2 \cos((\mathbf{w}_b - \mathbf{w}_g)t - \mathbf{f}_1) \cos \mathbf{f}_g) \quad (4.17)$$

The spectrum analyzer only measures the time-dependent components in the total power at the photodetector. Therefore, spectrum analyzer measures main signal (i.e. the carrier, the power portion at frequency  $\omega_b$ ), the first order left sideband (the power portion at  $(\omega_b - \omega_g)t$ ), the first order right sideband, (the power portion at  $(\omega_b + \omega_g)t$ ).

Let ratio of first order left sideband over carrier =  $R_1$

Let ratio of first order right sideband over carrier =  $R_2$

Using equation 4.16 and definition of  $R_1$ ,  $R_2$ , we obtain,

$$R_1 = 1/2\sqrt{((nkZ_0)^2 + a^2 - 2nkZ_0 a \sin \mathbf{f}_g)} \quad (4.18)$$

$$R_2 = 1/2\sqrt{((nkZ_0)^2 + a^2 + 2nkZ_0 a \sin \mathbf{f}_g)} \quad (4.19)$$

Manipulating the equation 4.18 and 4.19 and the values of  $Z_0$  and  $\phi_g$  are obtained.

$$Z_0 = 1/(kn)\sqrt{2(R_1^2 + R_2^2) - a^2 / 2} \quad (4.20)$$

$$f_g = \arcsin((R_2^2 - R_1^2) / nkZ_0 a) \quad (4.21)$$

Equation 4.19 indicates that if either the film thickness is known, the refractive index can be calculated and vice versa.

#### 4.2 Measurement of electric field effect

Under electrical field (E-field), the polymer film will orient in the E-field direction. The poling of polymer thin film will change the refractive index and the thickness of the polymer film along the E-field. Since polymers were poled uniformly along the field direction (i.e. principle direction x), therefore we can reduce light propagation as scalar equations 4.2 and 4.6 for electric properties measurements too. Since the film absorption and multiple reflections effect is very small, we can neglect the reflectivity change across the film. The phase change due to the refractive index and thickness change can be expressed similar to equation 4.7,

$$\Delta f = 2k(nZ - n_0Z_0) \quad (4.22)$$

Where  $Z_0, n_0$  are the initial polymer film thickness and refractive index.  $Z$  and  $n$  are the instantaneous film thickness and the refractive index along the applied field direction.  $Z$  and  $n$  are functions of time. The optical length of laser traveling in the film equals the film refractive index times film thickness and is expressed as,

$$nZ = 1/2(n_0Z_0 + n_1Z_1) - 1/2(n_1Z_1 - n_0Z_0) \cos(\omega_E t) \quad (4.23)$$

Where  $n_1$  and  $Z_1$  are the maximum refractive index and film thickness under certain frequency and amplitude of electric field.  $Z_0, n_0$  are the initial polymer film thickness and refractive index.  $\omega_E$  is the electric field frequency. By substituting equation 4.22 into 4.9, the total power is obtained,

$$P = E_{r0}^2 + E_{s0}^2 + 2E_{r0}E_{s0} \cos(\omega_b t - f_s - 2k(nZ - Z_0n_0)) \quad (4.24)$$

where  $E_{r_0}, E'_{s_0}$  are the magnitudes of the reference beam and the reflected sample beam;  
 $\omega_E$  is electric field frequency.

As we discussed before,  $(E_{r_0}^2 + E'_{s_0}{}^2)$  is DC component, invariant of time. The time dependent component  $P'$  is given by:

$$P' = 2E_{r_0}E'_{s_0} \cos(\mathbf{w}_b t - \mathbf{f}_s - 2k(nZ - n_0Z_0)) \quad (4.25)$$

Further expanding equation 4.24 after substituting equation 4.23 into 4.25,

$$P' = 2E_{r_0}E'_{s_0} \cos(\mathbf{w}_b t - \mathbf{f}_s + k(n_0Z_0 - n_1Z_1) - k(n_1Z_1 - n_0Z_0) \cos \mathbf{w}_E t) \quad (4.26)$$

Set

$$\mathbf{f}_s - k(Z_0n_0 - n_1Z_1) = \mathbf{f}' \quad (4.27)$$

Equation 4.26 is simplified as,

$$P' = 2E_{r_0}E'_{s_0} \cos(\mathbf{w}_b t - \mathbf{f}' + k(Z_1n_1 - n_0Z_0) \cos \mathbf{w}_E t) \quad (4.28)$$

From equation 4.28,

$$P' = 2E_{r_0}E'_{s_0} (\cos(\mathbf{w}_b t - \mathbf{f}') * \cos(k(Z_1n_1 - Z_0n_0) \cos(\mathbf{w}_E t)) - \sin(\mathbf{w}_b t - \mathbf{f}') \sin(k(Z_1n_1 - Z_0n_0) \cos(\mathbf{w}_E t))) \quad (4.29)$$

Using expansion series, the following terms are expressed as,

$$\sin(k(n_1Z_1 - Z_0n_0)) \cos(\mathbf{w}_E t) \approx k(n_1Z_1 - Z_0n_0) \cos(\mathbf{w}_E t) \quad (4.30)$$

$$\begin{aligned} \cos(k(n_1Z_1 - Z_0n_0) \cos(\mathbf{w}_E t)) &\approx 1 - k^2(n_1Z_1 - Z_0n_0)^2 \cos^2(\mathbf{w}_E t) / 2 \quad (4.31) \\ &= 1 - k^2(n_1Z_1 - Z_0n_0)^2 (1 + \cos(2\mathbf{w}_E t)) / 4 \end{aligned}$$

By substituting Equation 4.30 and 4.31 into equation 4.29 and simplifying, we obtained,

$$\begin{aligned}
P' = & 2 E_{s_0} E'_{s_0} ((1 - k^2 (n_1 Z_1 - n_0 Z_0)^2 / 4) \cos(\mathbf{w}_b t - \mathbf{f}')) \\
& - k (n_1 Z_1 - Z_0 n_0) / 2 \sin(\mathbf{w}_b t - \mathbf{f} + \mathbf{w}_E t) \\
& - k (n_1 Z_1 - Z_0 n_0) / 2 \sin(\mathbf{w}_b t - \mathbf{f} - \mathbf{w}_E t) \\
& - k^2 (n_1 Z_1 - Z_0 n_0)^2 / 8 \cos(\mathbf{w}_b t - \mathbf{f} - 2 \mathbf{w}_E t) \\
& - k^2 (n_1 Z_1 - Z_0 n_0)^2 / 8 \cos(\mathbf{w}_b t - \mathbf{f} + 2 \mathbf{w}_E t)
\end{aligned} \tag{4.32}$$

The spectrum analyzer measures only the time-dependent components of power as discussed earlier with film thickness measurements. This time-dependent power will include the carrier (i.e. main signal) at  $\omega_b t$ ; the first order left sideband at  $(\omega_b t - \omega_E t)$ ; the first order right sideband at  $(\omega_b t + \omega_E t)$ ; the second order left sideband at  $(\omega_b t - 2\omega_E t)$ ; the second order right sidebands at  $(\omega_b t + 2\omega_E t)$ .

Let  $R_1$  and  $R_2$  be the ratio of first order left and right sidebands versus carrier.  $R_3$  and  $R_4$  are the ratio of second order left and right sidebands versus carrier.

The ratio of first order and second order sidebands versus the carrier can be calculated from equation 4.32 as,

$$R_1 = R_2 = -k(n_1 Z_1 - Z_0 n_0) / 2(1 - k^2 (n_1 Z_1 - n_0 Z_0)^2 / 4) \tag{4.33}$$

$$R_3 = R_4 = -k^2 (n_1 Z_1 - Z_0 n_0)^2 / 8(1 - k^2 (n_1 Z_1 - n_0 Z_0)^2 / 4) \tag{4.34}$$

$R_3$  and  $R_4$  can be further simplified as,

$$R_3 = R_4 = -1 / (8 / k^2 (n_1 Z_1 - n_0 Z_0)^2 - 2) \tag{4.35}$$

Equation 4.34 shows when  $(n_1 Z_1 - n_0 Z_0)$  increase,  $R_3$  and  $R_4$  also increase. Furthermore, the absolute change of  $(nZ)$  as defined earlier (the refractive index along the applied electric field direction times the instantaneous film thickness) can be calculated.

The changes in refractive index times thickness due to the first order sidebands is,

$$(n_1 Z_1 - n_0 Z_0) = (1 - \sqrt{(1 + 4R_1^2)}) / R_1 k = \mathbf{I} * (1 - \sqrt{(1 + 4R_1^2)}) / 2 * R_1 * \mathbf{p} \tag{4.36}$$



The changes in refractive index times film thickness in terms of second order sidebands is given by,

$$(n_1 Z_1 - n_0 Z_0) = -1/k * \sqrt{8R_3 / (1 + 2R_3)} = -\frac{I}{2 * p} \sqrt{8R_3 / (1 + 2R_3)} \quad (4.37)$$

From equation 4.36 and 4.37, if the initial film thickness and refractive index are known, the percent change of (nZ) of polymer film under electric field can be calculated.

$$\%change = (n_1 Z_1 - n_0 Z_0) / (n_0 Z_0) = n_1 Z_1 / n_0 Z_0 - 1 \quad (4.38)$$

This differential interferometer design also allows us directly measure the film thickness changes by removing the two polarizers as shown in figure 3.1. This special function of our interferometer is further discussed in chapter 5. Without polarizers, the laser beam is not polarized along the E-field, thus the interferometry cannot measure the change of polymer orientation along the E-field. The change of (nZ) is all contributed by film thickness change instead of both refractive index and thickness change. In this case, it is reasonable to assume  $n_1 = n_0 = n$ . By simplifying equations 4.36 and 4.37 with  $n_1 = n_0$ , the thickness change is calculated as,

$$R_1 = R_2 = -kn_0 (Z_1 - Z_0) / 2(1 - k^2 n_0^2 (Z_1 - Z_0)^2 / 4) \quad (4.39)$$

$$R_3 = R_4 = -1 / (8 / k^2 n_0^2 (Z_1 - Z_0)^2 - 2) \quad (4.40)$$

The above two equations can also be written as,

$$(Z_1 - Z_0) = (1 - \sqrt{(1 + 4R_1^2)}) / n_0 R_1 k = I * (1 - \sqrt{(1 + 4R_1^2)}) / 2 * R_1 * p * n_0 \quad (4.41)$$

$$(Z_1 - Z_0) = -1/n_0 k * \sqrt{8R_3 / (1 + 2R_3)} = -\frac{I}{2 * p * n_0} \sqrt{8R_3 / (1 + 2R_3)} \quad (4.42)$$

Thus from equation 4.41 and 4.42, if  $R_1, R_2, R_3, R_4$  are known, the change in thickness due to E-field (i.e. electrorestrictive effect) can be measured.

<sup>1</sup> E. Hecht, Optics, 2<sup>nd</sup> edition, Addison-Wesley publishing company, (1987).