Supporting Information

Optical Contrast of Atomically-thin Films

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The reflectivity of a homogeneous thin film on a stratified planar substrate

For a stratified structure composed of M homogeneous planar layers (the M-th layer is a semi-infinite substrate) as depicted in the Figure 1 in the main text, the reflectivity of a TM polarization plane wave (with a time-dependent term of $e^{-i\omega t}$) is,[1]

$$r = \frac{K_0 - Z_1}{K_0 + Z_1} \quad \text{(A1)}$$

$$Z_1 = K_1 \frac{Z_2 + K_1 \tanh(-i\beta_1)}{K_1 + Z_2 \tanh(-i\beta_1)} \quad \text{(A2)}$$

$$Z_j = K_j \frac{Z_{j+1} + K_j \tanh(-i\beta_j)}{K_j + Z_{j+1} \tanh(-i\beta_j)} \quad \text{(A3)}$$

$$Z_{M-1} = K_{M-1} \frac{K_M + K_{M-1} \tanh(-i\beta_j)}{K_{M-1} + K_M \tanh(-i\beta_j)} \quad \text{(A4)}$$

$$K_j = \frac{\mu_j}{c} \frac{\cos\theta_j}{n_j} \quad \text{(A5)}$$

$$\beta_j = n_j\, k_0 h_j \cos\theta_j \quad \text{(A6)}$$

$\mu_j, n_j, \theta_j, h_j$ are the relative permeability, refractive index, refraction angle and thickness of the j-th layer film, respectively. c is the speed of light in free space. Z_1 is called the surface impedance, being the ratio of the tangential fields at the air-substrate interface, $Z_1 = \frac{E_t}{H_t}|_{z=0}$.[1]

With the addition of a thin film on top of the substrate, the reflectivity becomes

$$r' = \frac{K_0 - Z_f}{K_0 + Z_f} \tag{A7}$$

$$Z_f = K_f \frac{Z_1 + K_f \tanh(-i\beta)}{K_f + Z_1 \tanh(-i\beta)} \tag{A8}$$

The subscript f refers to the thin film layer, $\beta = nk_0 h \cos\theta_f$.

From eq A1, it is inferred

$$Z_1 = K_0 \frac{1-r}{1+r} \tag{A9}$$

Inputting the above formula of Z_1 into eq A8, eq A7 can be rewritten as

$$r' = \frac{K_0 - K_f \frac{K_0 \frac{1-r}{1+r} + K_f \tanh(-i\beta)}{K_f + K_0 \frac{1-r}{1+r} \tanh(-i\beta)}}{K_0 + K_f \frac{K_0 \frac{1-r}{1+r} + K_f \tanh(-i\beta)}{K_f + K_0 \frac{1-r}{1+r} \tanh(-i\beta)}} \tag{A10}$$

Let $\xi = \frac{K_f}{K_0} = \frac{n_0 \mu_f \cos\theta_f}{n_f \mu_0 \cos\theta_0} = \frac{n_0 \cos\theta_f}{n_f \cos\theta_0}$ (for non-magnetic materials, $\mu_f = 1$), we have

$$r' = \frac{\frac{2\xi r}{1+r} + [\frac{1-r}{1+r} - \xi^2] \tanh(-i\beta)}{\frac{2\xi}{1+r} + [\frac{1-r}{1+r} + \xi^2] \tanh(-i\beta)} \tag{A11}$$

and

$$\chi \equiv 1 - \frac{r'}{r} = \frac{\frac{1+r}{r}[\xi^2 - (\frac{1-r}{1+r})^2] \tanh(-i\beta)}{\frac{2\xi}{1+r} + [\frac{1-r}{1+r} + \xi^2] \tanh(-i\beta)} \tag{A12}$$

For TE-polarization, replacing ξ with $\zeta = \frac{n_f \cos\theta_f}{n_o \cos\theta_0}$.

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References

(1) Wait, J. R. *Electromagnetic waves in stratified media*; Pergamon Press Ltd.: Oxford, UK, 1970.