Treatment of SPR background in Total internal reflection ellipsometry. Characterization of RNA polymerase II films formation.

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Supplementary information

The matrix formalism for light propagation in layered structures. The propagation of light in a layered structure with planar interfaces produces no diffraction, and can, hence, be described by impact (E_i), reflected (E_r) and transmitted (E_t) wave amplitudes of the electric field using simple matrix formalism:

$$\begin{bmatrix} E_\mathrm{i} \\ E_\mathrm{r} \end{bmatrix} = (\mathbf{V}_0)^{-1}(\mathbf{R}_1)^{-1}\dots(\mathbf{R}_m)^{-1}\mathbf{V}_\mathrm{f}\begin{bmatrix} E_\mathrm{t} \\ 0 \end{bmatrix} \equiv \mathbf{P}\begin{bmatrix} E_\mathrm{t} \\ 0 \end{bmatrix}, \tag{S1}$$

where matrices $\mathbf{R}_j$ belong to j-th layer of the structure and matrices $\mathbf{V}$ are connected with substrate (index $_0$) and ambient (index $_\mathrm{f}$). The expression

$$(\mathbf{R}_j)^{-1} = \begin{pmatrix} \cos\theta_j & -\dfrac{\mathrm{i}\sin\theta_j}{\tilde{n}_j} \\ -\mathrm{i}\tilde{n}_j\sin\theta_j & \cos\theta_j \end{pmatrix} \tag{S2}$$

is formally polarization independent (and $\det\mathbf{R}_j = 1$). Note, however, that the definition of $\tilde{n}_j$ is polarization dependent itself:

$$\mathrm{s}: \tilde{n}_j = n_j\cos\varphi_j \qquad \mathrm{p}: \tilde{n}_j = \frac{n_j}{\cos\varphi_j},$$

where n_j is the index of refraction of j-th layer and

$$\cos\varphi_j = \sqrt{1-\left(\frac{n_0\sin\varphi}{n_j}\right)^2},$$

with φ_j being an angle at which the j-th layer is propagated. The phase acquired in a j-th layer is given by

$$\theta_j = 2\pi\frac{d_j}{\lambda}n_j\cos\phi_j,$$

where d_j is the thickness of the j-th layer and λ is the vacuum wavelength of incident light.
For the $\mathbf{V}$ matrices we have

$$\mathrm{s}: \mathbf{V}_j = \begin{pmatrix} 1 & 1 \\ \tilde{n}_j & -\tilde{n}_j \end{pmatrix} \qquad \mathrm{p}: \mathbf{V}_j = \cos\varphi_j\begin{pmatrix} 1 & 1 \\ \tilde{n}_j & -\tilde{n}_j \end{pmatrix}$$

which brings

$$\mathrm{s}: (\mathbf{V}_0)^{-1} = -\frac{1}{2\tilde{n}_0}\begin{pmatrix} -\tilde{n}_0 & -1 \\ -\tilde{n}_0 & 1 \end{pmatrix} \qquad \mathrm{p}: (\mathbf{V}_0)^{-1} = -\frac{1}{2\tilde{n}_0\cos\varphi_0}\begin{pmatrix} -\tilde{n}_0 & -1 \\ -\tilde{n}_0 & 1 \end{pmatrix}.$$

The overall propagation matrix $\mathbf{P}$ from eq. (**??**) allows to compute the complex reflectivity through $r = P_{21}/P_{11}$ (distinguishing r_s and r_p by the appropriate entries of $\mathbf{P}$ for each of the polarizations) as well as the complex reflectivity ratio

$$\rho = r_\mathrm{p}/r_\mathrm{s}.$$

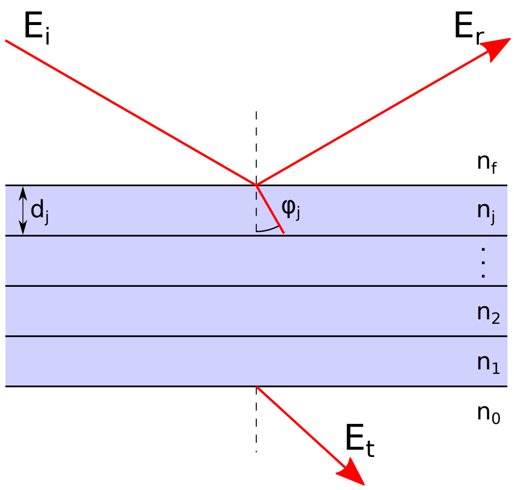

Figure 1. Notation for light propagation through a layered structure.

The difference ellipsometric spectra. The general propagation matrix from eq. (**??**) can be decomposed to $\mathbf{P} = \mathbf{B}\mathbf{V}_\mathrm{f}$ via

$$\mathbf{B} = (\mathbf{V}_0)^{-1}(\mathbf{R}_1)^{-1} \dots (\mathbf{R}_m)^{-1}.$$

Then, if another layer, described by matrix $\mathbf{S}$, is added on the top of the substrate just below the ambient, the overall propagation matrix gets $\mathbf{P} = \mathbf{B}\mathbf{S}\mathbf{V}_\mathrm{f}$. In the following, we will gather the (in principle unknown) properties of the substrate into $\mathbf{B}$ and study only the effect of adding a new layer over this structure.

As the newly added we choose a thin layer of thickness t and index of refraction n (the light will propagate through the layer at angle ϕ) with $(\mathbf{R}^{-1})_\mathrm{thin} \equiv \mathbf{S}$, where

$$\mathbf{S} \approx \begin{pmatrix} 1 & -\dfrac{\mathrm{i}}{\tilde{n}}\dfrac{2\pi}{\lambda} nt\cos\phi \\ -\mathrm{i}\tilde{n}\dfrac{2\pi}{\lambda} nt\cos\phi & 1 \end{pmatrix}$$

can be obtained using Taylor expansion with respect to t/λ of eq. (**??**).

We will now consider two measurements in terms of the complex reflectance ratio $\rho = \tan\Psi\exp(\mathrm{i}\Delta)$ produced: the background measurement ρ_0 without the additional layer and the sample measurement ρ with the additional layer. If we denote the components of $\mathbf{B}$ according to

$$\mathrm{s}: \mathbf{B} \equiv \begin{pmatrix} a_\mathrm{s} & b_\mathrm{s} \\ c_\mathrm{s} & d_\mathrm{s} \end{pmatrix} \qquad \mathrm{p}: \mathbf{B} \equiv \begin{pmatrix} a_\mathrm{p} & b_\mathrm{p} \\ c_\mathrm{p} & d_\mathrm{p} \end{pmatrix},$$

then one obtains

$$\rho_0 = \frac{(d_\mathrm{p} n_\mathrm{f} + c_\mathrm{p}\cos\phi_\mathrm{f})(a_\mathrm{s} + b_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}{(b_\mathrm{p} n_\mathrm{f} + a_\mathrm{p}\cos\phi_\mathrm{f})(c_\mathrm{s} + d_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}$$

with

$$\Psi_0 = \arctan\left|\frac{(d_\mathrm{p} n_\mathrm{f} + c_\mathrm{p}\cos\phi_\mathrm{f})(a_\mathrm{s} + b_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}{(b_\mathrm{p} n_\mathrm{f} + a_\mathrm{p}\cos\phi_\mathrm{f})(c_\mathrm{s} + d_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}\right| \qquad \Delta_0 = \mathrm{Arg}\left(\frac{(d_\mathrm{p} n_\mathrm{f} + c_\mathrm{p}\cos\phi_\mathrm{f})(a_\mathrm{s} + b_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}{(b_\mathrm{p} n_\mathrm{f} + a_\mathrm{p}\cos\phi_\mathrm{f})(c_\mathrm{s} + d_\mathrm{s} n_\mathrm{f}\cos\phi_\mathrm{f})}\right)$$

and

$$\rho = \frac{\left(a_s + n_f b_s\cos\phi_f - 2\pi\mathrm{i}n^2\frac{t}{\lambda}b_s\cos^2\phi - 2\pi\mathrm{i}n_f\frac{t}{\lambda}a_s\cos\phi_f\right)\left(d_p n_f + c_p\cos\phi_f - 2\pi\mathrm{i}n^2\frac{t}{\lambda}d_p\cos\phi_f - 2\pi\mathrm{i}n_f\frac{t}{\lambda}c_p\cos^2\phi\right)}{\left(c_s + n_f d_s\cos\phi_f - 2\pi\mathrm{i}n^2\frac{t}{\lambda}d_s\cos^2\phi - 2\pi\mathrm{i}n_f\frac{t}{\lambda}c_s\cos\phi_f\right)\left(b_p n_f + a_p\cos\phi_f - 2\pi\mathrm{i}\frac{t}{\lambda}n^2 b_p\cos\phi_f - 2\pi\mathrm{i}n_f\frac{t}{\lambda}a_p\cos^2\phi\right)}.$$

As before, $\Psi = \arctan|\rho|$ and $\Delta = \mathrm{Arg}\rho$ and one can perform the Taylor expansion with respect to t/λ. Of course, $\rho|_{t/\lambda\to 0} = \rho_0$, and we will limit ourselves to linear expansion.

Consequently, in case of Ψ to treat the derivative with complex numbers right, we use $|\rho| = \sqrt{\rho\rho^*}$, where $*$ denotes complex conjugation, and write

$$[\arctan\sqrt{\rho\rho*}]' = \frac{1}{1+|\rho|^2}\frac{\rho'\rho^* + \rho\rho^{*\prime}}{2|\rho|} = \frac{1}{1+|\rho|^2}\frac{\mathrm{Re}(\rho'\rho^*)}{|\rho|}.$$

Evaluating now the derivative near zero, one obtains

$$[\arctan\sqrt{\rho\rho*}]'|_{t/\lambda\to 0} = \frac{1}{1+|\rho_0|^2}\frac{1}{|\rho_0|}\mathrm{Re}(\rho'\rho^*)|_{t/\lambda\to 0},$$

where

$$(\rho'\rho^*)|_{t/\lambda\to 0} = -2\pi\mathrm{i}\left[\begin{array}{c} \dfrac{b_\mathrm{s}n^2\cos^2\phi + a_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}}{a_\mathrm{s}+b_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}} + \dfrac{d_\mathrm{p}n^2\cos\phi_\mathrm{f} + c_\mathrm{p}n_\mathrm{f}\cos^2\phi}{d_\mathrm{p}n_\mathrm{f}+c_\mathrm{p}\cos\phi_\mathrm{f}} - \\ -\dfrac{d_\mathrm{s}n^2\cos^2\phi + c_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}}{c_\mathrm{s}+d_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}} - \dfrac{b_\mathrm{p}n^2\cos\phi_\mathrm{f} + a_\mathrm{p}n_\mathrm{f}\cos^2\phi}{b_\mathrm{p}n_\mathrm{f}+a_\mathrm{p}\cos\phi_\mathrm{f}} \end{array}\right]|\rho_0|^2.$$

Denoting now

$$A = (d_\mathrm{p}n^2\cos\phi_\mathrm{f} + c_\mathrm{p}n_\mathrm{f}\cos^2\phi)(a_\mathrm{s}+b_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}) + (b_\mathrm{s}n^2\cos^2\phi + a_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f})(d_\mathrm{p}n_\mathrm{f}+c_\mathrm{p}\cos\phi_\mathrm{f})$$

$$E = (b_\mathrm{p}n^2\cos\phi_\mathrm{f} + a_\mathrm{p}n_\mathrm{f}\cos^2\phi)(c_\mathrm{s}+d_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}) + (d_\mathrm{s}n^2\cos^2\phi + c_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f})(b_\mathrm{p}n_\mathrm{f}+a_\mathrm{p}\cos\phi_\mathrm{f}).$$

$$A_0 = (d_\mathrm{p}n_\mathrm{f}+c_\mathrm{p}\cos\phi_\mathrm{f})(a_\mathrm{s}+b_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f})$$

$$E_0 = (b_\mathrm{p}n_\mathrm{f}+a_\mathrm{p}\cos\phi_\mathrm{f})(c_\mathrm{s}+d_\mathrm{s}n_\mathrm{f}\cos\phi_\mathrm{f}),$$

one can write $\rho_0 = A_0/E_0$ and

$$\mathrm{Re}(\rho'\rho^*)|_{t/\lambda\to 0} = 2\pi\mathrm{Re}\left(-\mathrm{i}\left[\frac{A}{A_0}-\frac{E}{E_0}\right]\right)|\rho_0|^2 = 2\pi\mathrm{Im}\left(\frac{A}{A_0}-\frac{E}{E_0}\right)|\rho_0|^2,$$

so that we arrive to

$$\Psi = \Psi_0 + 2\pi\mathrm{Im}\left(\frac{A}{A_0}-\frac{E}{E_0}\right)\frac{|\rho_0|}{1+|\rho_0|^2}\frac{t}{\lambda} + \dots$$

Please note that A_0 and E_0 are introduced for convenience only and have no physical interpretation; in particular, they do not coincide with r_p and r_s.

Concerning Δ, we use the definition

$$\mathrm{Arg}\rho = \arctan\frac{\mathrm{Im}\rho}{\mathrm{Re}\rho},$$

whence for the purposes of Taylor expansion

$$[\mathrm{Arg}z]' = \frac{1}{1+\left(\frac{\mathrm{Im}\rho}{\mathrm{Re}\rho}\right)^2}\frac{\mathrm{Re}\rho\,\mathrm{Im}'\rho - \mathrm{Re}'\rho\,\mathrm{Im}\rho}{\mathrm{Re}^2\rho} = \frac{\mathrm{Re}\rho\,\mathrm{Im}'\rho - \mathrm{Re}'\rho\,\mathrm{Im}\rho}{|\rho|^2}.$$

Realizing now that $\mathrm{Re}\rho\,\mathrm{Im}'\rho - \mathrm{Re}'\rho\,\mathrm{Im}\rho = \mathrm{Im}(\rho'\rho^*)$ one can make use of the derivation of Ψ and write

$$\mathrm{Im}(\rho'\rho^*)|_{t/\lambda\to 0} = 2\pi\mathrm{Im}\left(-\mathrm{i}\left[\frac{A}{A_0}-\frac{E}{E_0}\right]\right)|\rho_0|^2 = -2\pi\mathrm{Re}\left(\frac{A}{A_0}-\frac{E}{E_0}\right)|\rho_0|^2,$$

so that

$$[\mathrm{Arg}z]'_{t/\lambda\to 0} = -2\pi\mathrm{Re}\left(\frac{A}{A_0}-\frac{E}{E_0}\right),$$

and, finally,

$$\Delta = \Delta_0 - 2\pi\mathrm{Re}\left(\frac{A}{A_0}-\frac{E}{E_0}\right)\frac{t}{\lambda} + \dots$$