Model-free extraction of refractive index from measured optical data

A Tool for Refractive InDex Simulation

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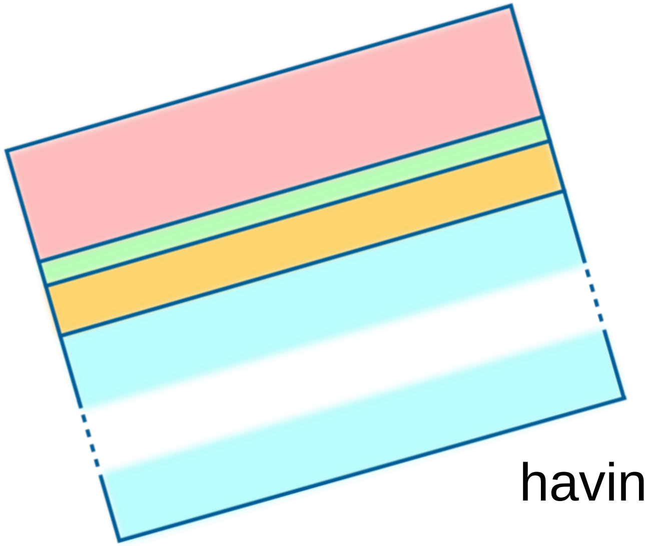
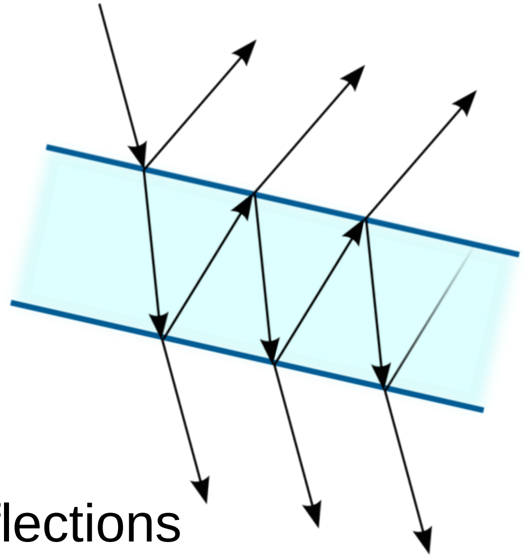
Motivation



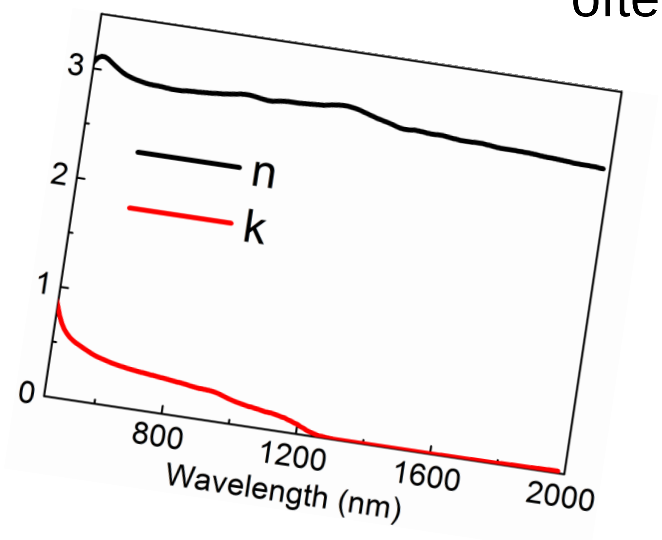


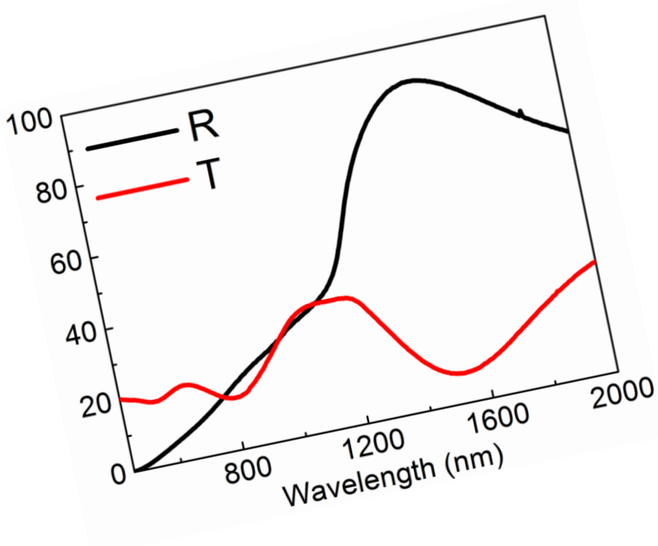
Thin film optics

having to deal with multiple reflections

and requiring refractive indices

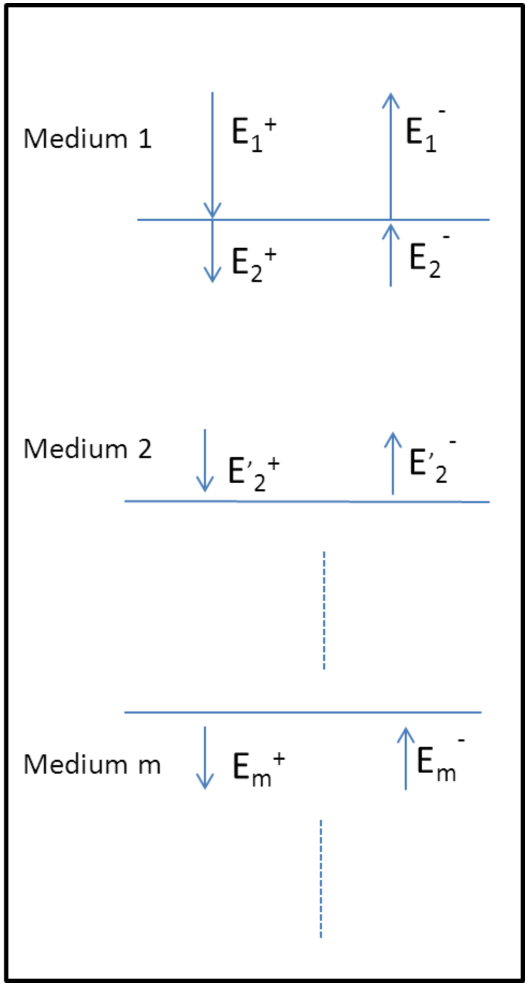
often only rely on optical measurements.





Contents

- Basic Principles
    - Transfer Matrix Method
    - Multilayer Stack
    - Comparison to Experiment
- Advanced Features
    - Surface Roughness
    - Inhomogeneous Layers
    - Effective Medium
- User Interface
- Outlook



Propagation through mediums at *normal incidence*

Superposition of electric field
- one wave with positive direction(E^+)
- one wave with negative direction(E^-)

Propagating through an interface:

$$\begin{bmatrix} E_i^+ \\ E_i^- \end{bmatrix} = \frac{1}{t_{i,j}} \begin{bmatrix} 1 & r_{i,j} \\ r_{i,j} & 1 \end{bmatrix} \begin{bmatrix} E_j^+ \\ E_j^- \end{bmatrix}$$

$$r_{i,j} = -r_{j,i} = \frac{N_i - N_j}{N_i + N_j}, \qquad t_{i,j} = \frac{2N_i}{N_i + N_j}, \quad t_{j,i} = \frac{2N_j}{N_i + N_j};$$

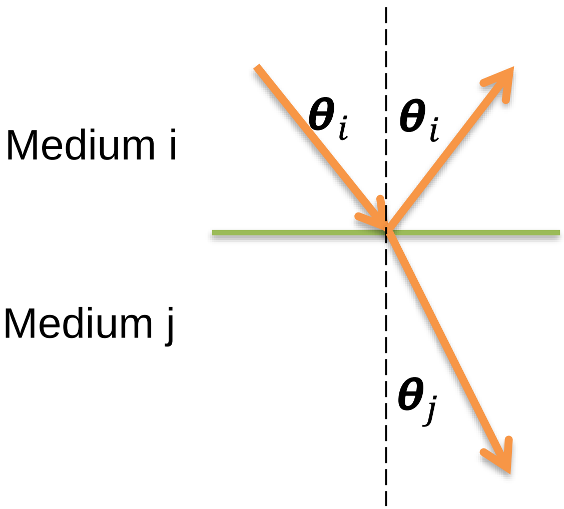
r, t are, respectively, the complex amplitude reflection and transmission Fresnel coefficients; N is the complex refractive index of the layer

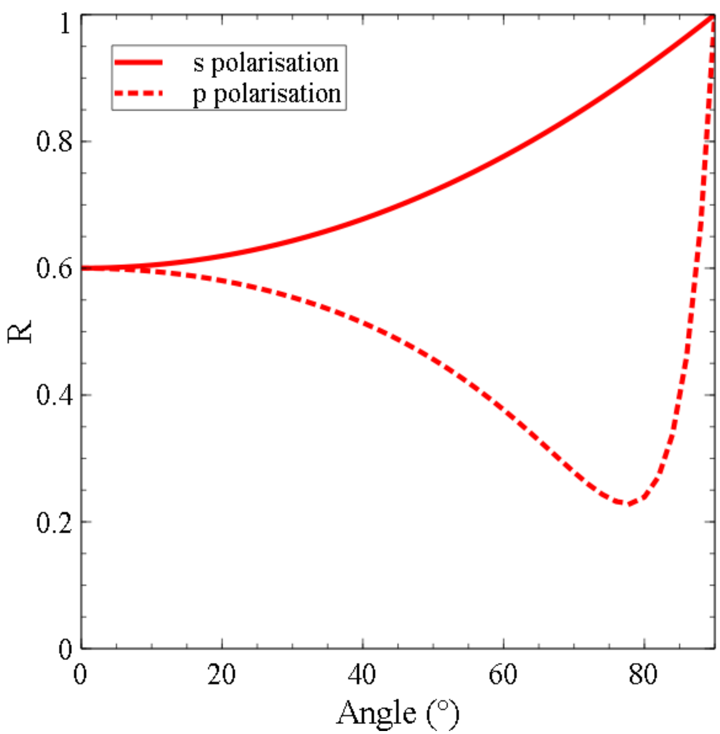
Propagating within a layer:

$$\begin{bmatrix} E_i^+ \\ E_i^- \end{bmatrix} = \begin{bmatrix} \Phi^{-1} & 0 \\ 0 & \Phi \end{bmatrix} \begin{bmatrix} E_i'^+ \\ E_i'^- \end{bmatrix} \qquad \Phi = e^{-i\frac{2\pi}{\lambda} N_i d}$$

Where *d* is the thickness of medium, ω is the frequency of the propagating light and c is the speed of light

Oblique incidence





At interface

P polarization:

$$r_{i,j} = \frac{N_j cos\boldsymbol{\theta}_i - N_i cos\boldsymbol{\theta}_j}{N_j cos\boldsymbol{\theta}_i + N_i cos\boldsymbol{\theta}_j}$$

$$t_{i,j} = \frac{2N_i cos\boldsymbol{\theta}_i}{N_j cos\boldsymbol{\theta}_i + N_i cos\boldsymbol{\theta}_j}$$

S polarization:

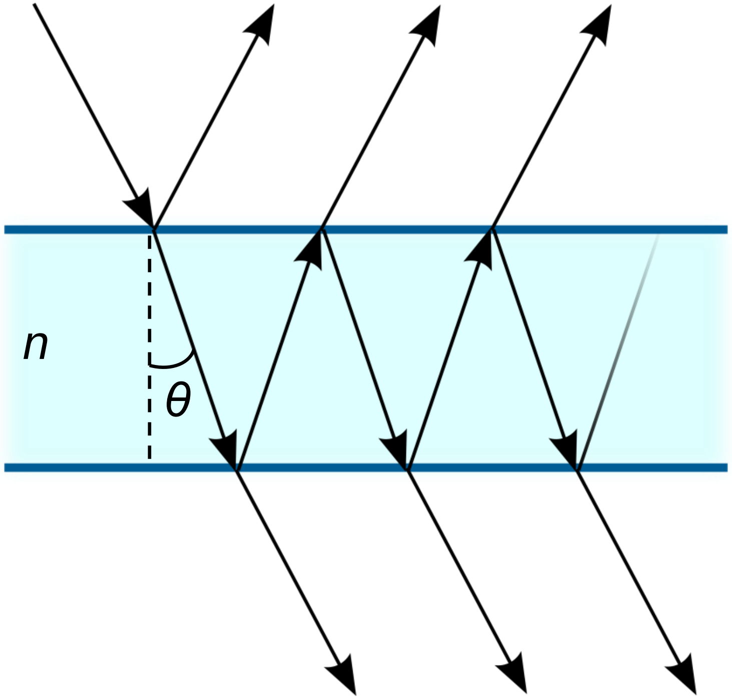
$$r_{i,j} = \frac{N_i cos\boldsymbol{\theta}_i - N_j cos\boldsymbol{\theta}_j}{N_i cos\boldsymbol{\theta}_i + N_j cos\boldsymbol{\theta}_j}$$

$$t_{i,j} = \frac{2N_i cos\boldsymbol{\theta}_i}{N_i cos\boldsymbol{\theta}_i + N_j cos\boldsymbol{\theta}_j}$$

Fresnel coefficiencts for *oblique incidence*

Within the layer

$$\Phi = e^{-i\frac{2\pi}{\lambda}N_m d/cos\boldsymbol{\theta}_j}$$



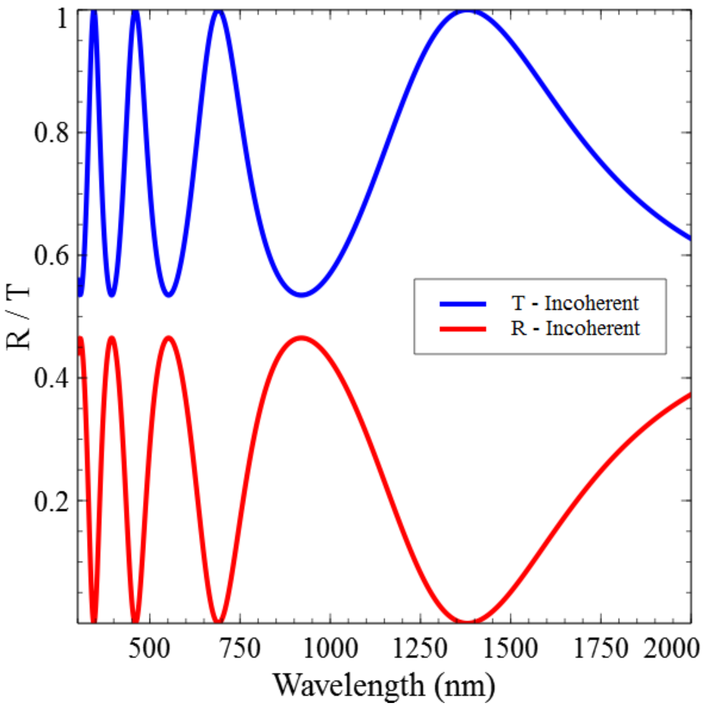
Validity condition:

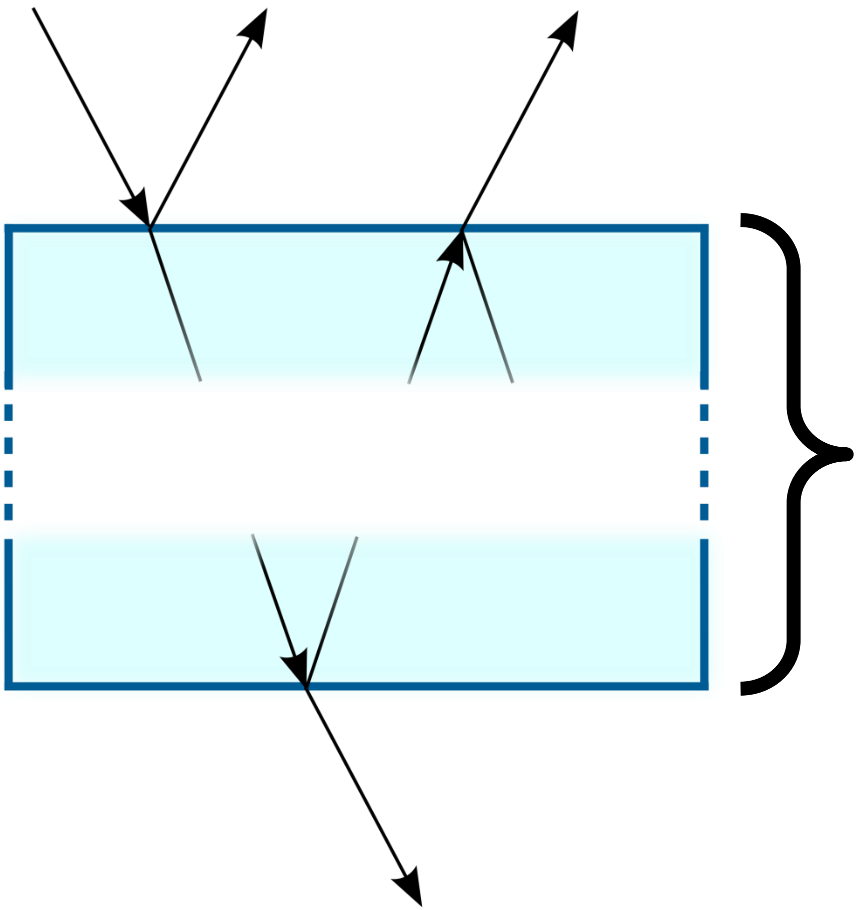
$$\frac{d}{\Delta\lambda_{Coherence}} << 1$$

Phase difference between transmission orders:

$$\delta = \left(\frac{2\pi}{\lambda}\right) 2nd\cos\theta$$

When $\delta = 2m\pi\ , m \in \mathbb{N}$ there will be *constructive interference* in T

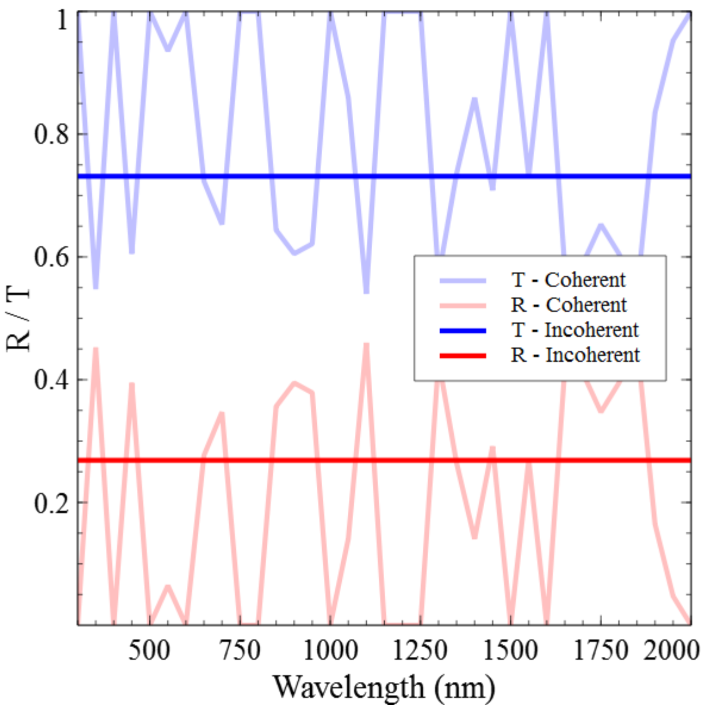




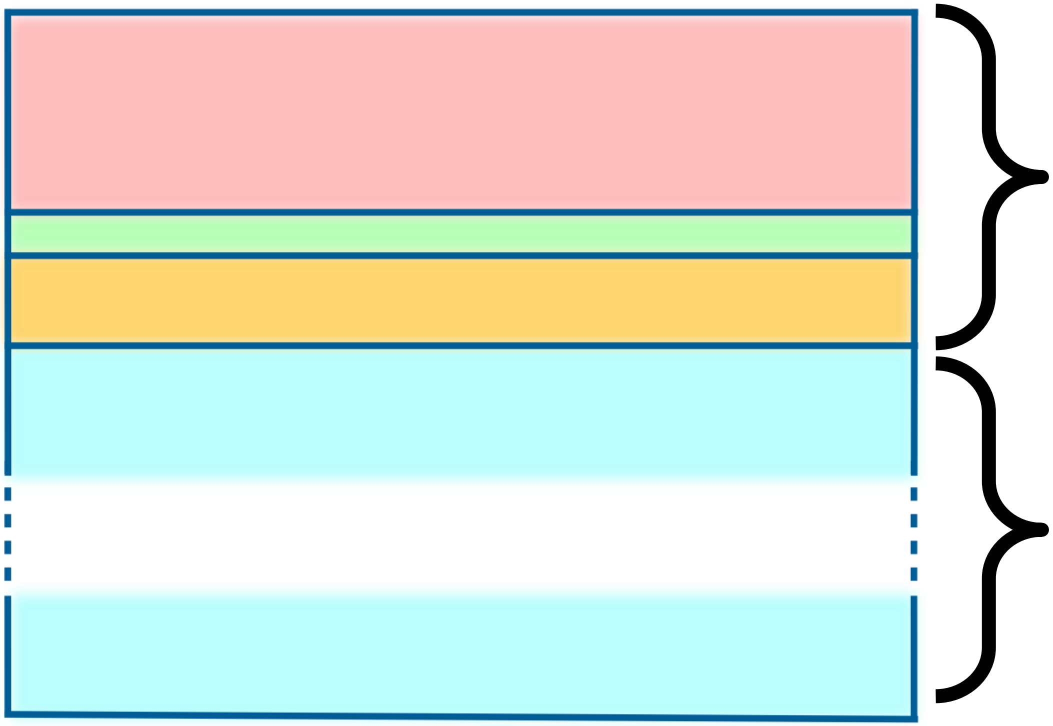
To removed coherency, calculate the *Intensity* instead of the *Electric Field*

$$I = E^*E = |E_0|^2 \underbrace{e^{ik\tilde{n}^*d} e^{-ik\tilde{n}d}}_{\text{phase information}} = |E_0|^2$$

$$\frac{t}{\Delta\lambda_{Coherence}} >> 1$$



Phase relationships between interior reflections is destroyed – therefore there is *no interference*



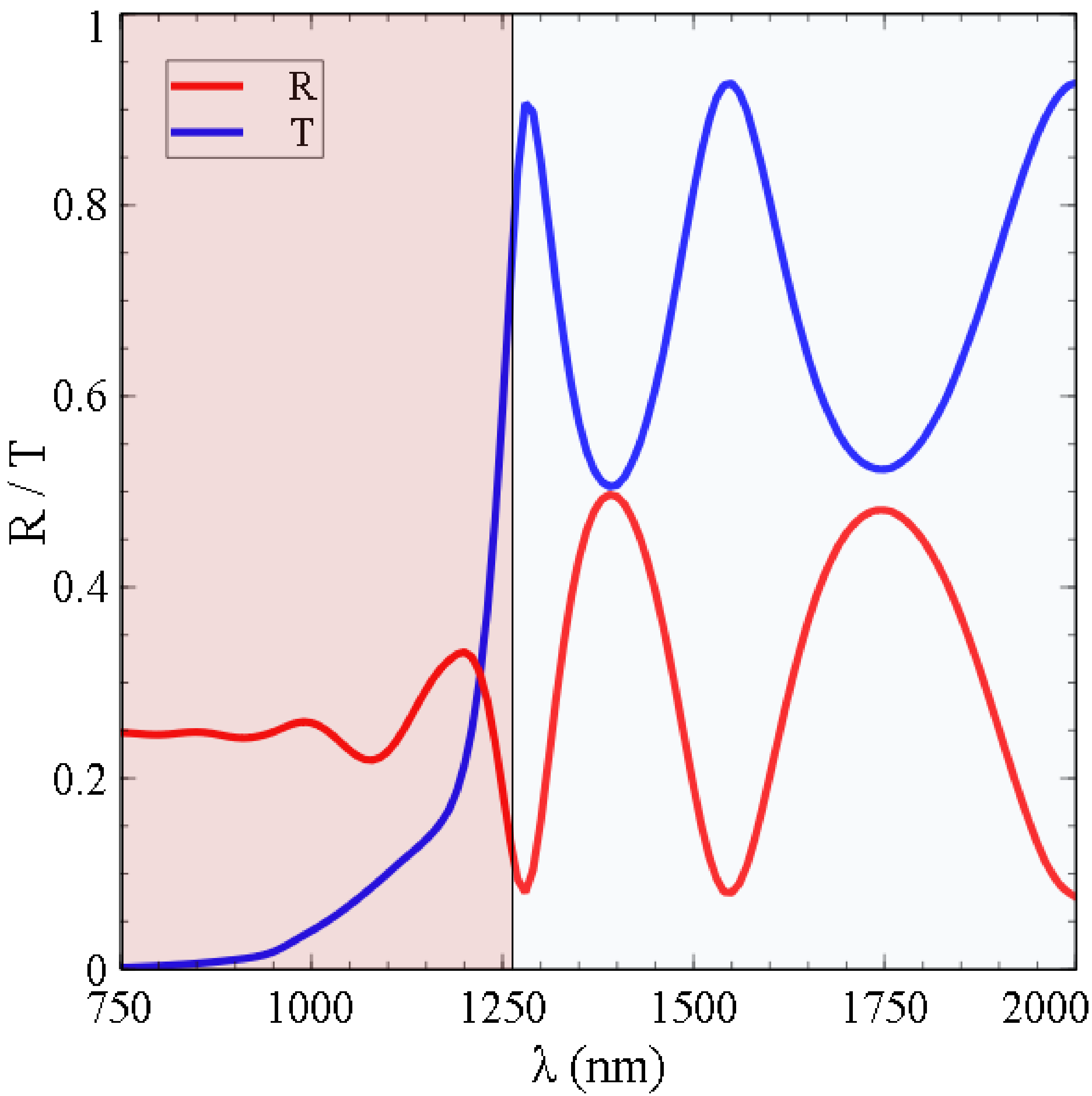
Coherent Stack

- Includes interference
- Typical thickness 0 ~ 2000 nm

Incoherent Layer

- Interference "turned off"
- Typical thickness 1 mm

- 9 Total layers implemented in RefDex
- Combine coherent and incoherent layers in any order
- For R,T calculation, d, n and k must be known for all layers

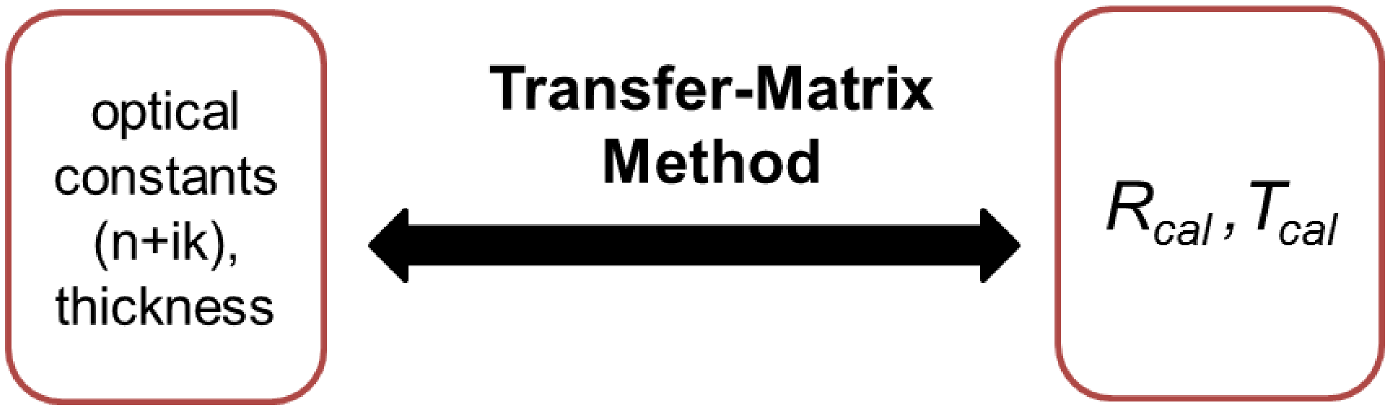


<u>Absorbing Region</u>

- Reflection loses coherency peaks
- Transmission drops to zero due to absorption

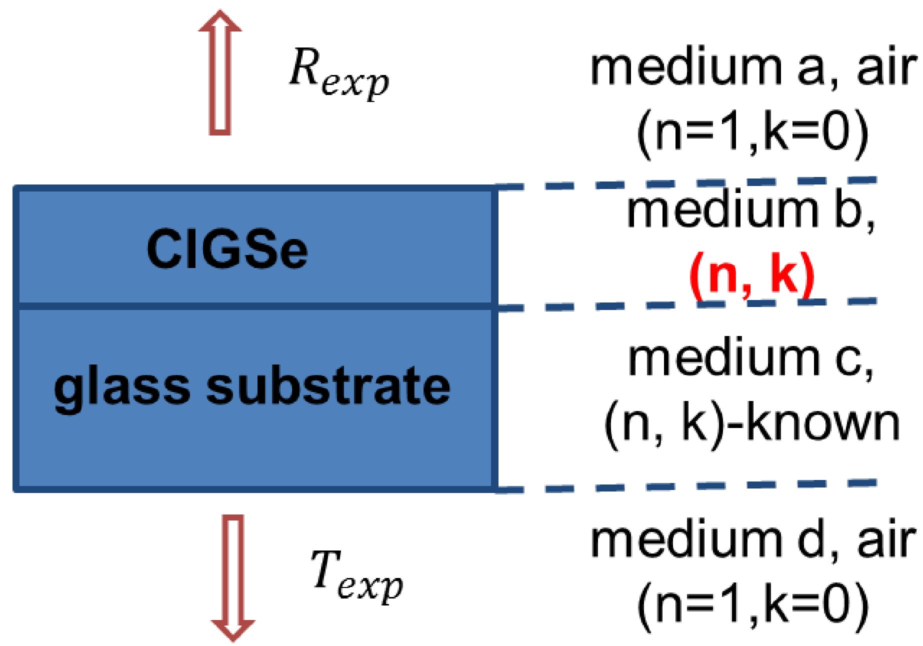
<u>Transparent Region</u>

- R and T both show coherency peaks
- R does not drop to 0 due to reflection from glass substrate



An example:

thin film
on substrate



Problem of Uniqueness

$$\left|R_{cal}\big(n(\lambda), k(\lambda)\big) - R_{exp}(\lambda)\right| = 0$$

$$\left|T_{cal}\big(n(\lambda), k(\lambda)\big) - T_{exp}(\lambda)\right| = 0$$

Choose the (n,k) values which minimise the difference between our model and experiment

$$F[n,k] = \left|R_{cal}\big(n(\lambda), k(\lambda)\big) - R_{exp}(\lambda)\right| + \left|T_{cal}\big(n(\lambda), k(\lambda)\big) - T_{exp}(\lambda)\right|$$

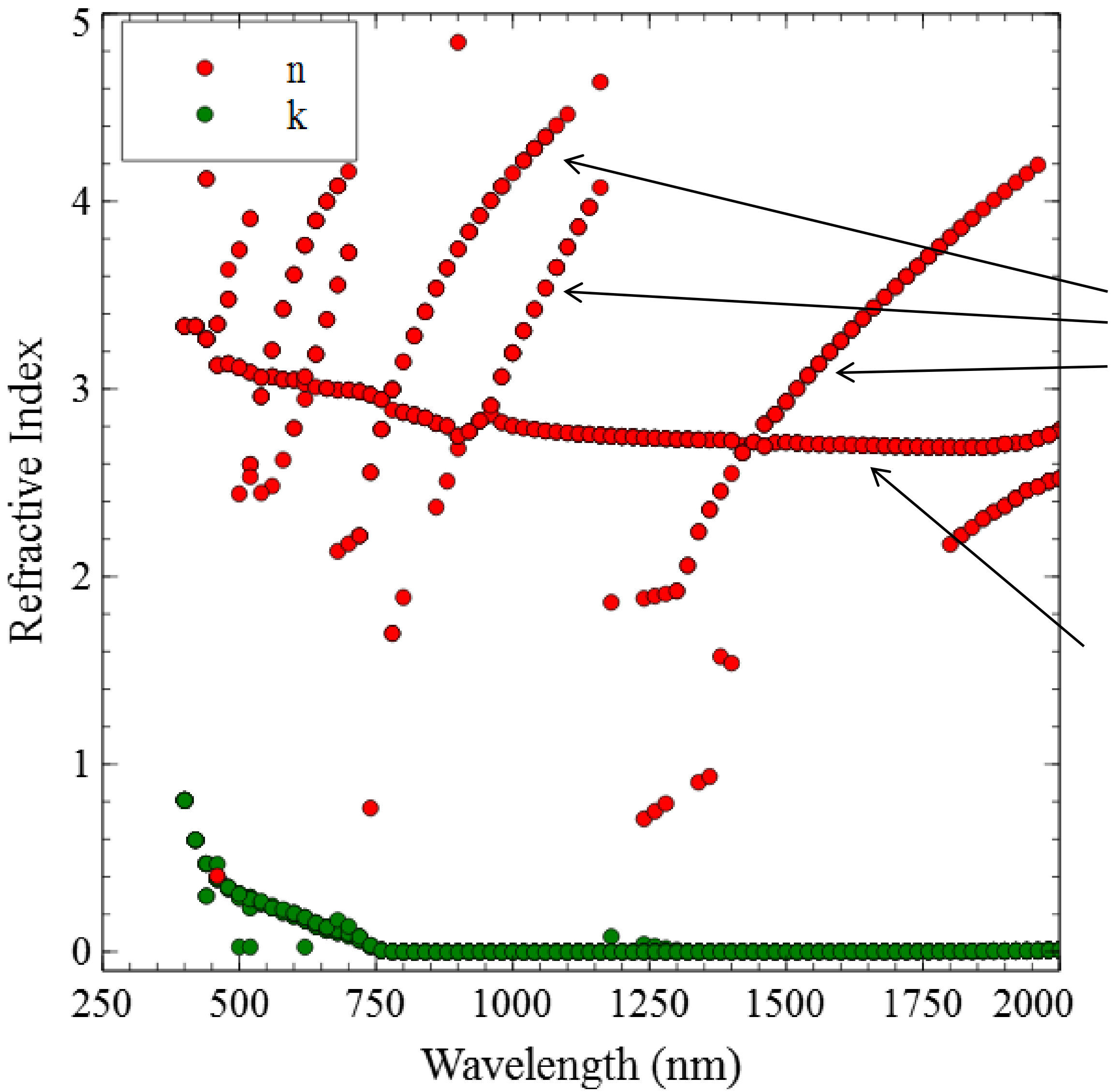
Adding these equations together we get a function F which takes n and k as input

$$F[n', k'] = F[n'', k''] = 0$$

Problems arise because two different (n,k) input pairs can both equal zero!

One Physically Meaningful Solution

Many Unphysical Solutions

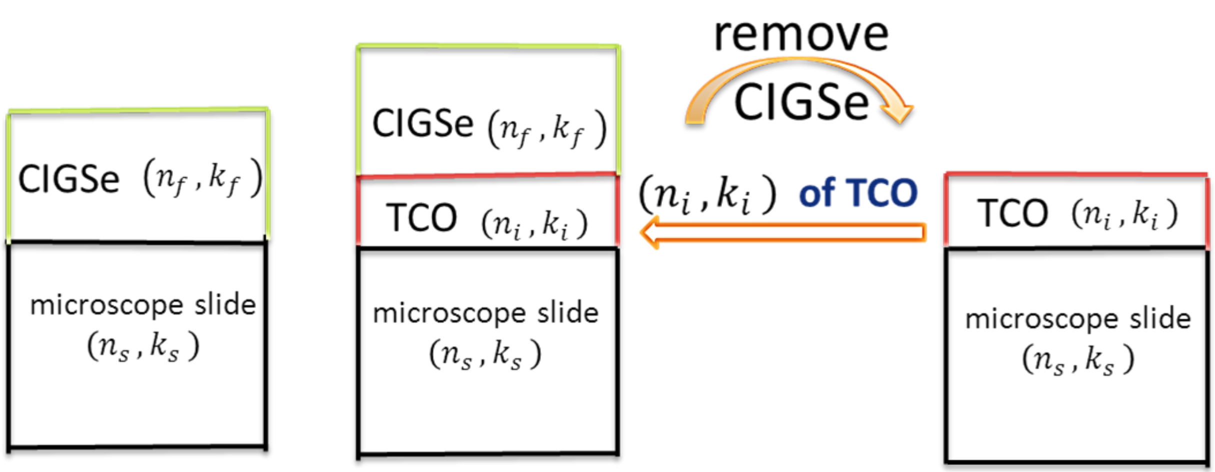


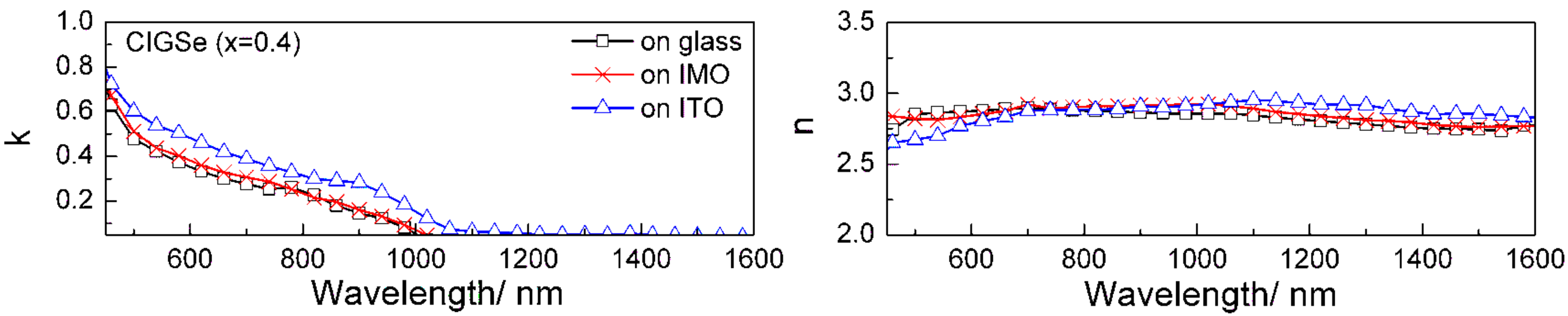
Spurious solution branches

Physically meaningful solution

Results need to be interpreted – More on this Later!

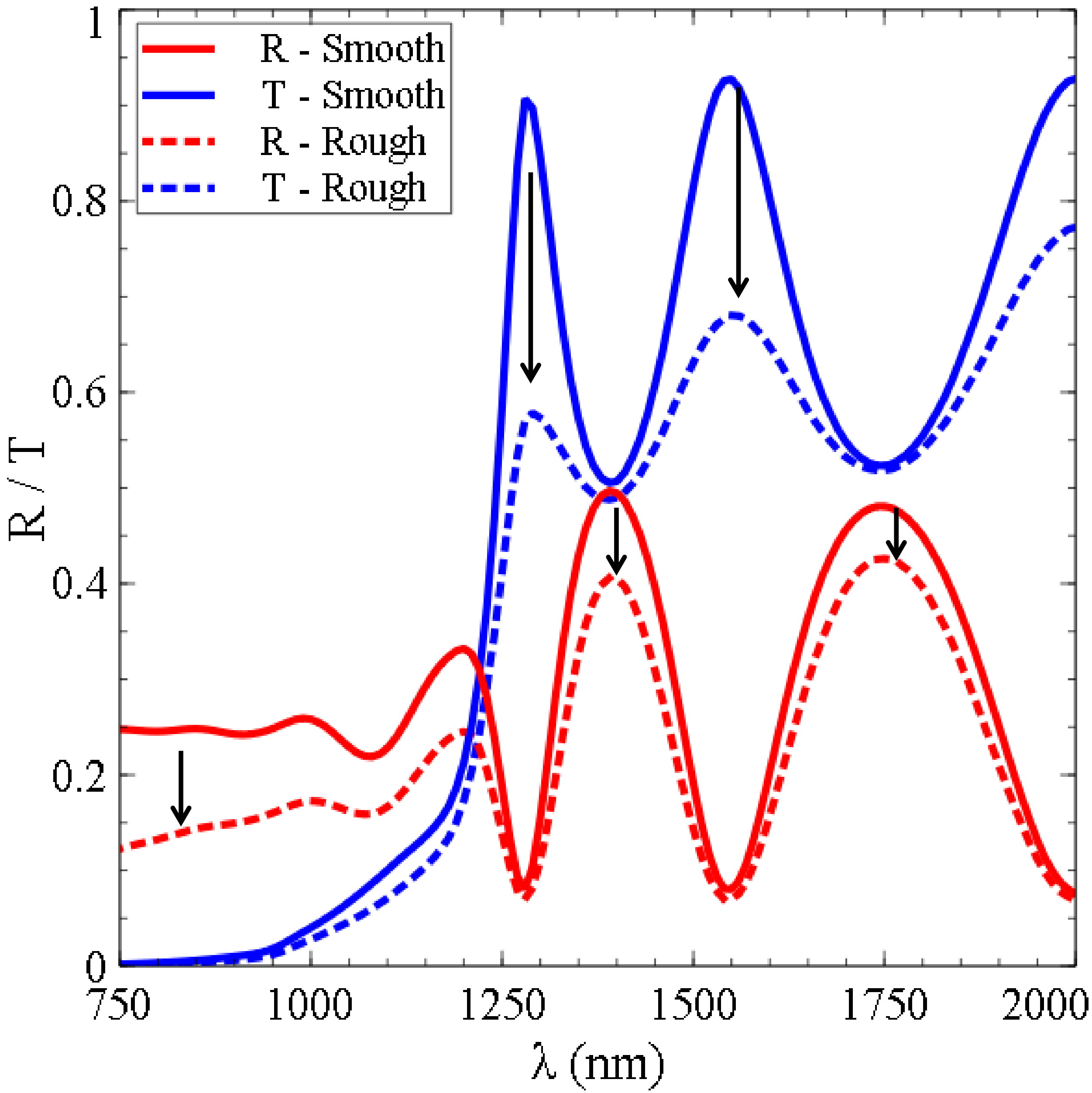
Take the configuration of CIGSe/TCO/glass substrate as an example:





G. Yin et al., *Influence of substrate and its temperature on the optical constants of $CuIn_{1-x}Ga_xSe_2$ thin films*, J. Phys. D: Appl. Phys., **47** 135101 (2014)

Surface Roughness – Effect on R and T



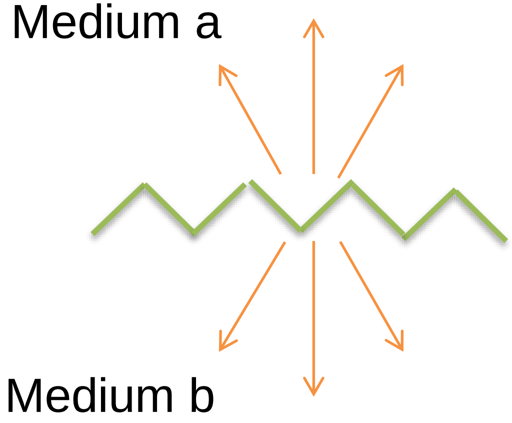
Absorbing Region

- Reflection Strongly Reduced
- Transmission Slightly Reduced

Transparent Region

- R and T reduced prefferentially at coherency peaks

Rough Interface



Modified → Fresnel coefficients

Scalar Scattering Theory

$$r'_{i,j} = r_{i,j} exp[-2(2\pi\sigma/\lambda)^2 {n_i}^2]$$

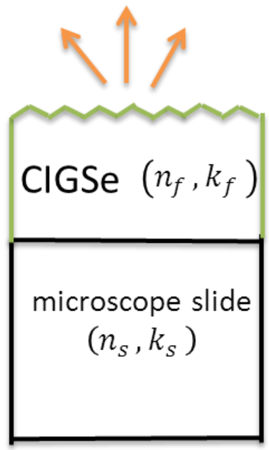
$$r'_{j,i} = r_{j,i} exp[-2(2\pi\sigma/\lambda)^2 {n_j}^2]$$

$$t'_{i,j} = t_{i,j} exp\left[-\left(\frac{2\pi\sigma}{\lambda}\right)^2 (n_i - n_j)^2/2\right]$$

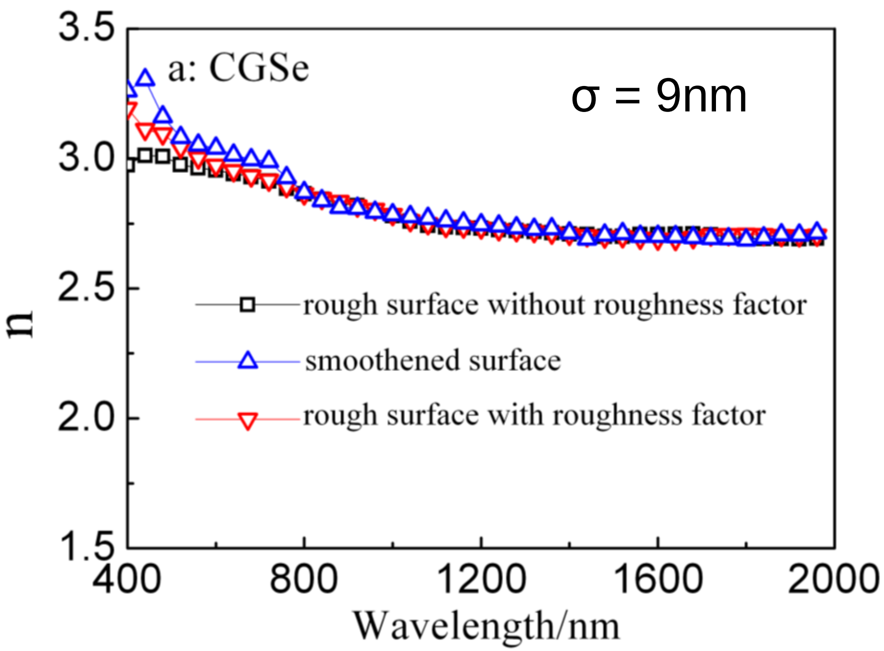
$$t'_{j,i} = t_{j,i} exp\left[-\left(\frac{2\pi\sigma}{\lambda}\right)^2 (n_j - n_i)^2/2\right]$$

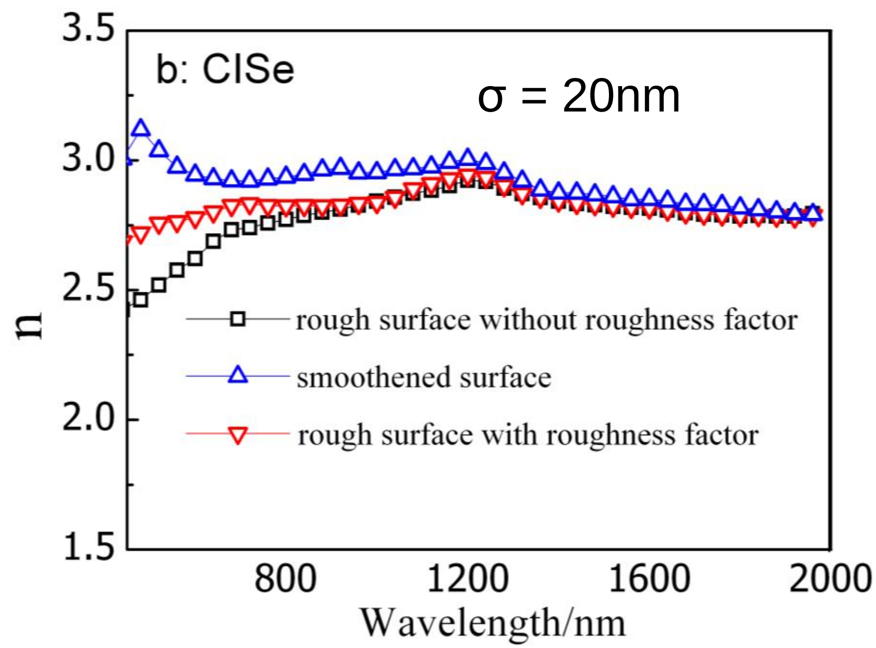
- σ is the interface roughness
- Gives us the loss of <u>*specular beam*</u> intensity due to interface roughness

Determination of optical constants

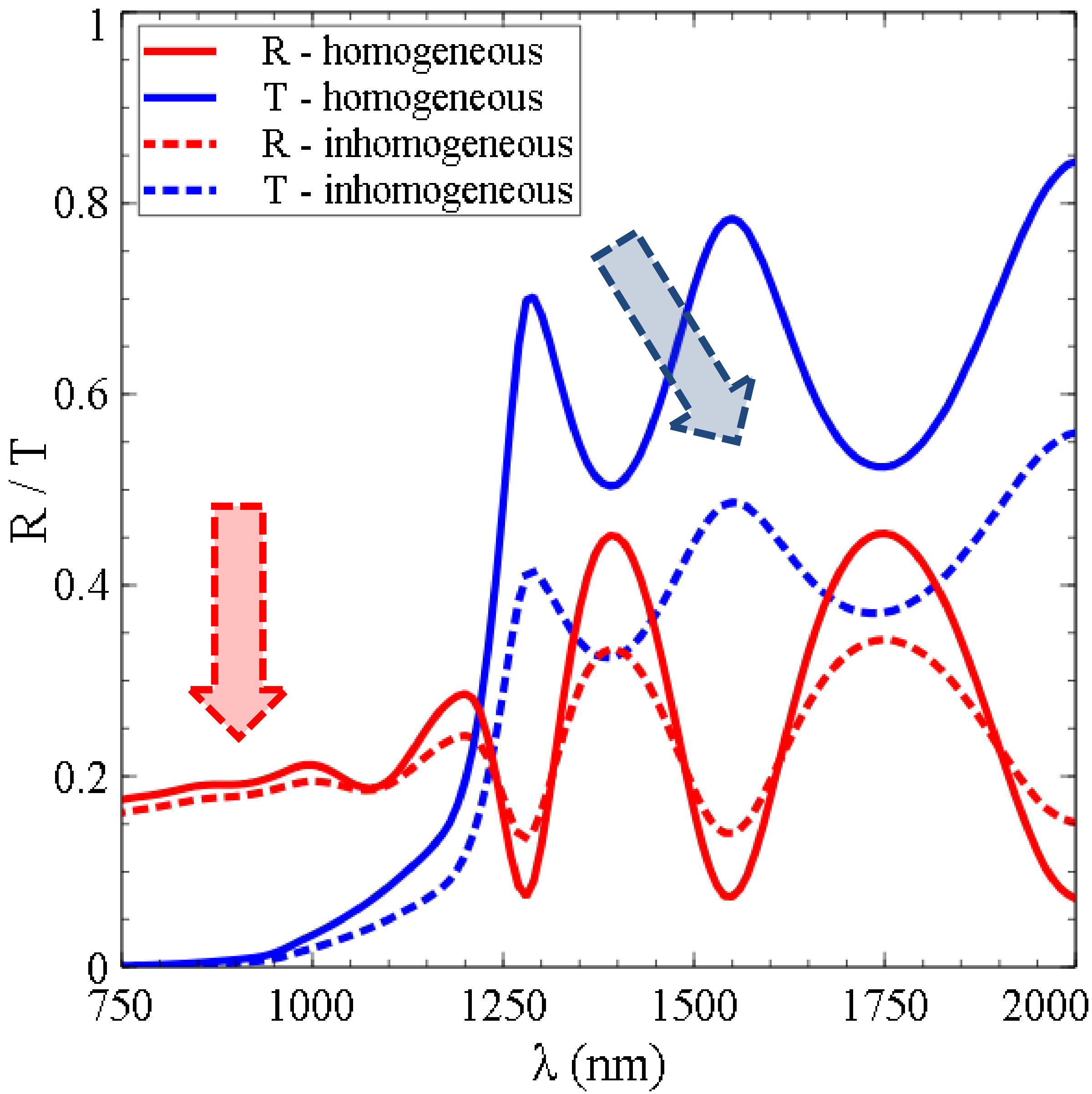


G. Yin et al., *The effect of surface roughness on the determination of optical constants of $CuInSe_2$ and $CuGaSe_2$ thin films*, J. Appl. Phys., **133**, 213510 (2013)





Inhomogeneous Layers – Effect on R and T



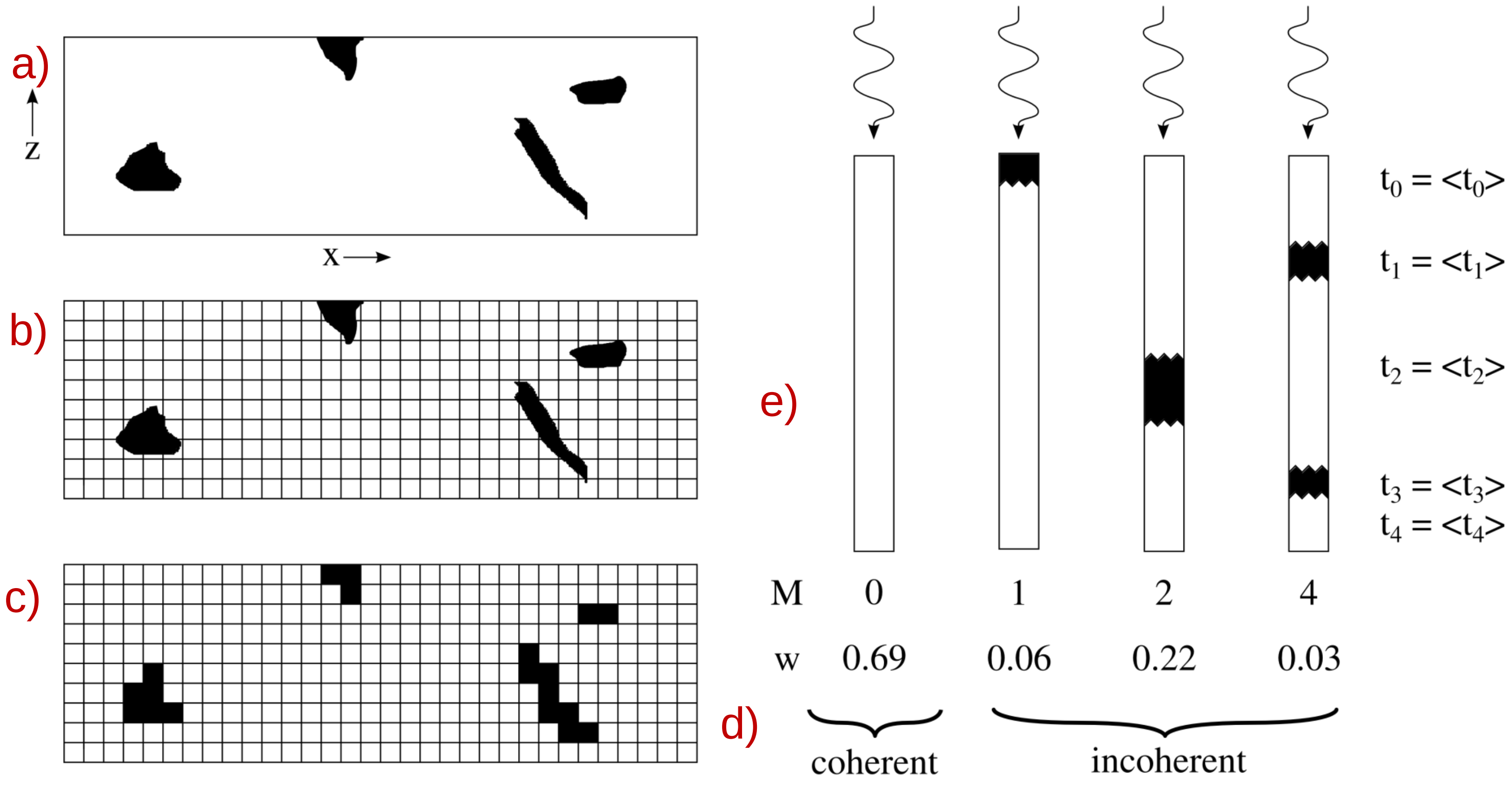
<u>Absorbing Region</u>

- Small reduction in R and T

<u>Transparent Region</u>

- Coherency reduced for both R and T
- Transmission strongly reduced

Inhomogeneous Layers – Coherent / Incoherent Decomposition



a) 2D slice through the 3D inhomogeneous film
b) Overlay a rectangular grid
c) The resulting discretised representation of the film
d) Layers containing voids can be modelled incoherently allowing the use of average layer thicknesses
e) This reduces the number of transfer matrix calculations to 4

$$R_{calc}(n,k) = w_c R_C + w_I R_I$$

(Same equations for T not shown here)

Standard Calculation ← R_C

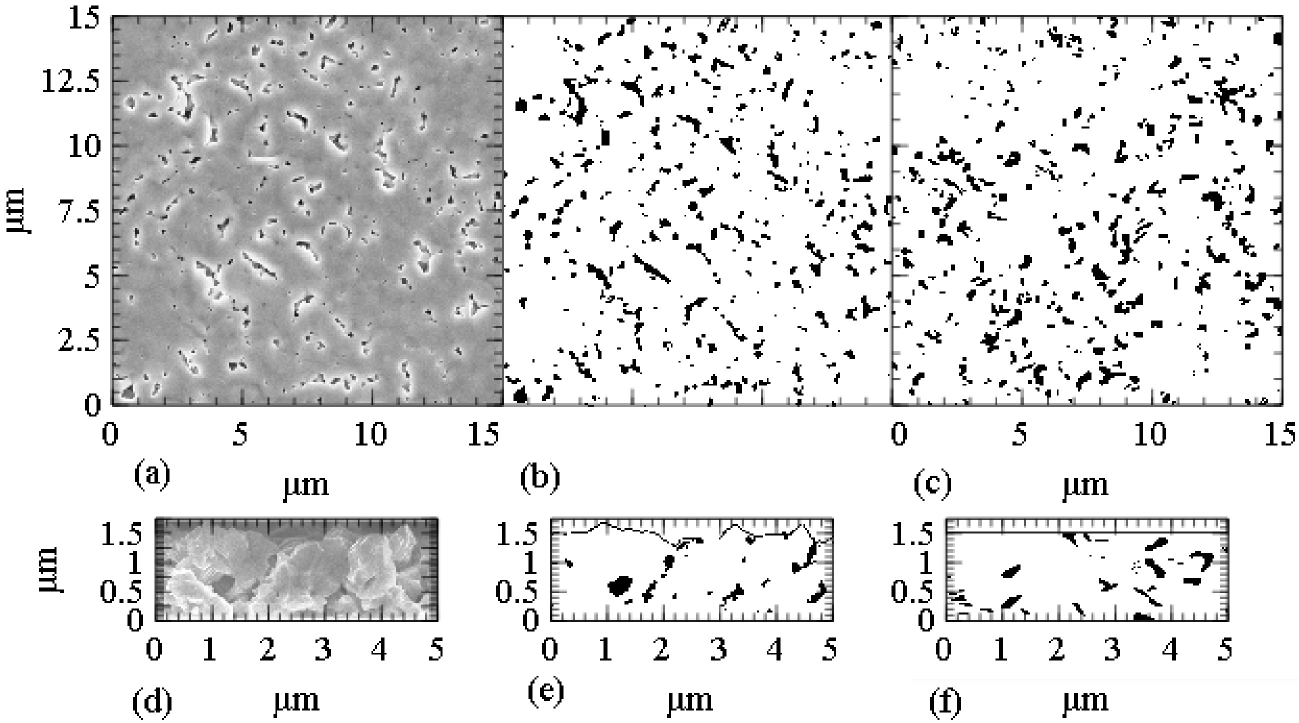
R_I ↓

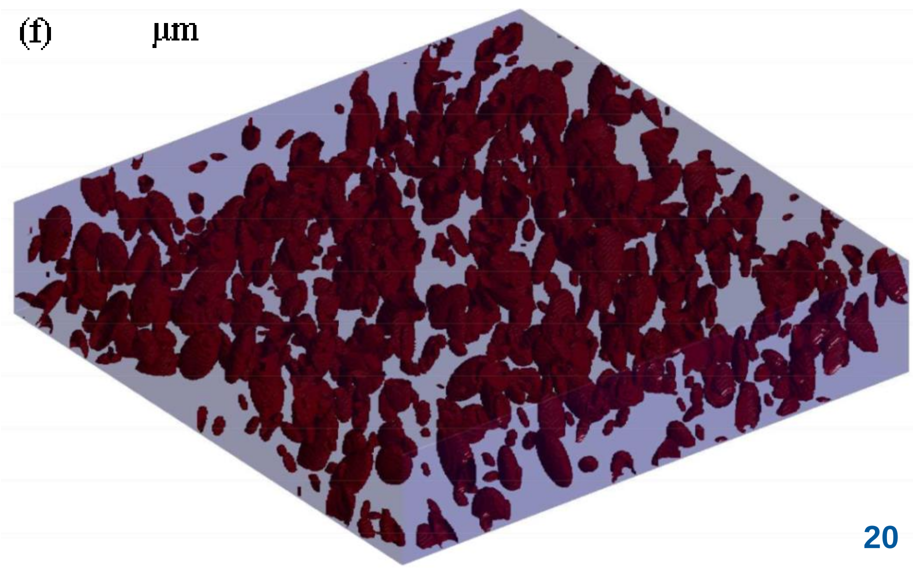
Replace propagation operator inside inhomogeneous layer with:

$$\widehat{\boldsymbol{P}}_i = \left\{\prod_{m=1}^{M} \widehat{\boldsymbol{P}}_m^{n(m)} \widehat{\boldsymbol{D}}_{m,m-1}^{n(m),n(m-1)}\right\} \widehat{\boldsymbol{P}}_0^{n(0)}, \quad n(m) = \begin{cases} n(0), m = even, m \neq 0 \\ \neg n(0), m = odd \end{cases}$$

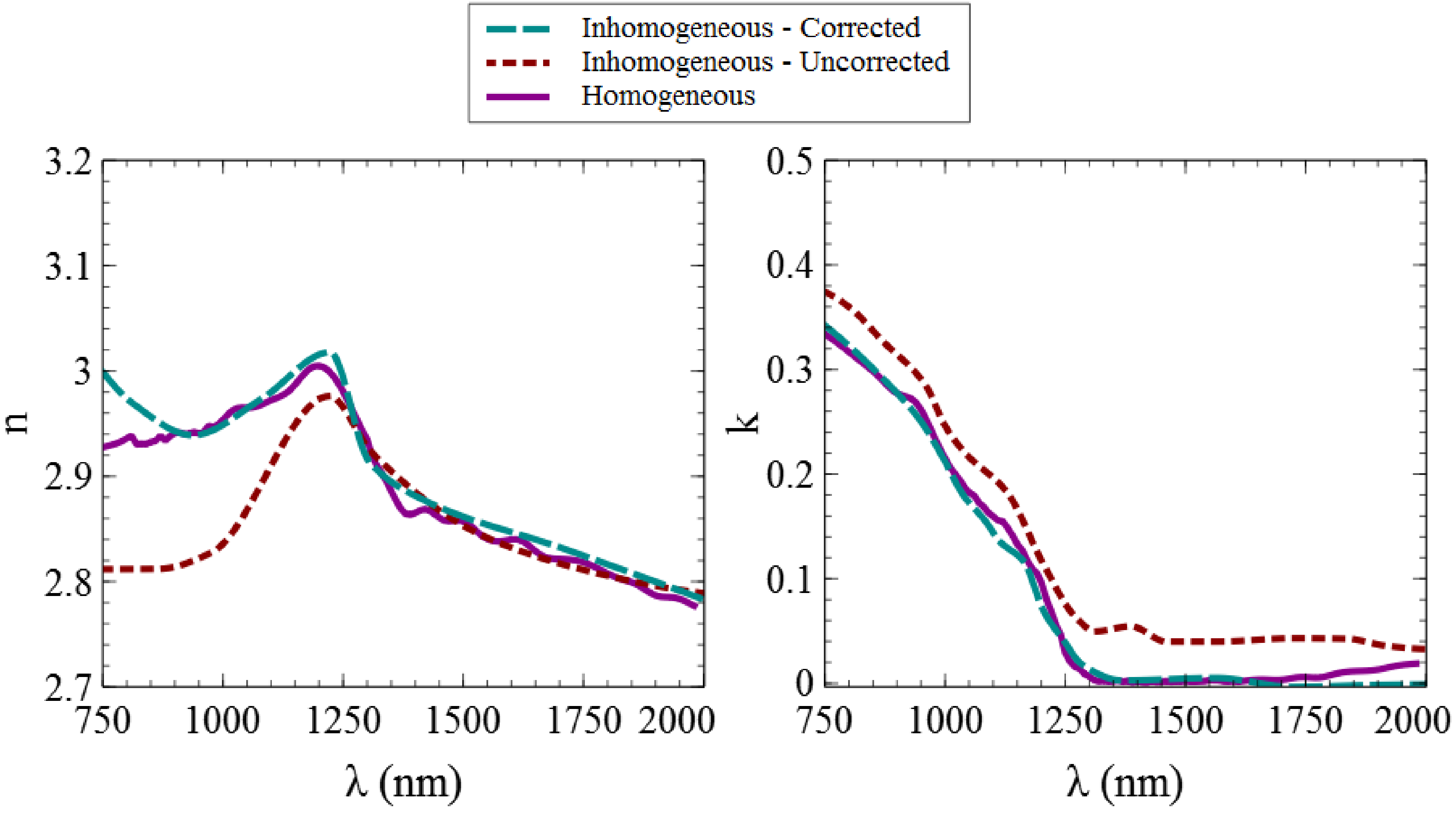
$$n(0) = n_i^* \quad \text{or} \quad n(0) = n_v^*$$

- Void scattering as from a rough surface. (Slide 13)
- Requires statistical knowledge of 3D void distribution as input





- Measurement of real 2D surface used to generate 3D distribution
- From 3D distribution we obtain inputs for the RefDex calculation



(n,k) data from an inhomogeneous $CISe_2$ film is in good agreement to the (n,k) data from a homogeneous film using the inhomogeneous layer feature.

P. Manley et al., *A method for calculating the complex refractive index of inhomogeneous thin films*, J. Phys. D: Appl. Phys., **47** 205301 (2014)

$$n_{eff} = w_h n_h + w_i n_i$$

$$k_{eff} = w_h k_h + w_i k_i$$

Volume Fraction Approximation

- Direct mixing of the two materials via the volume fraction
- Does not consider polarisation effects arrising due to mixing

$$\left(\frac{\varepsilon_{eff} - \varepsilon_h}{\varepsilon_{eff} + 2\varepsilon_h}\right) = w_i \left(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h}\right)$$

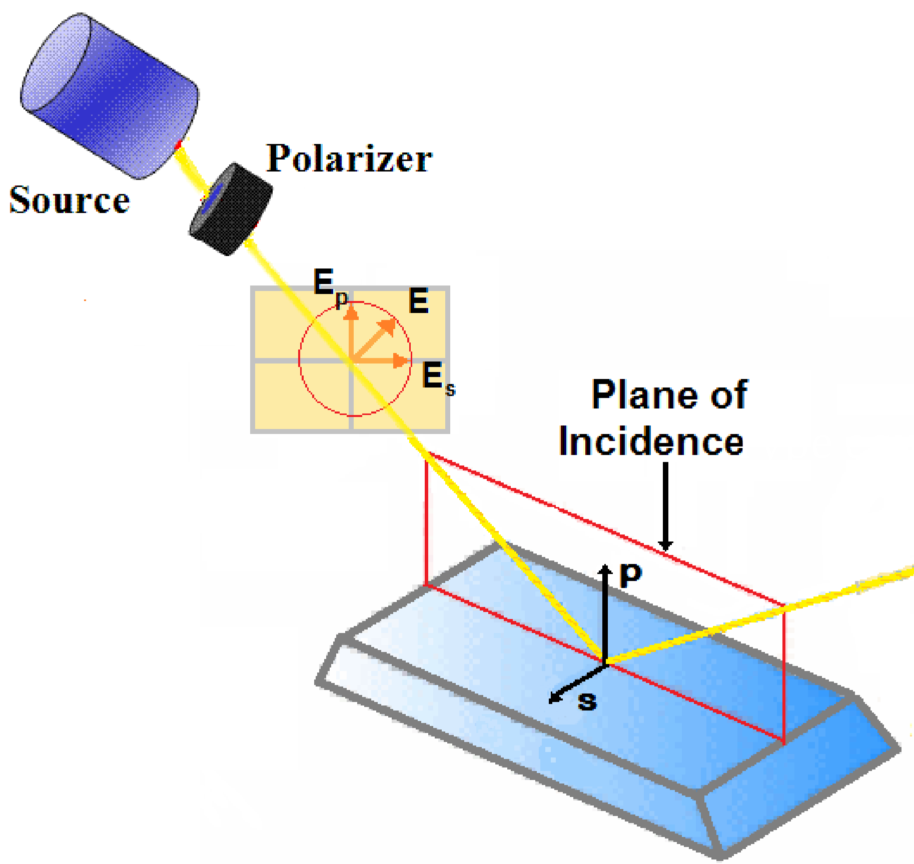
Maxwell Garnett Approximation

- Based on elementary electrostatics
- Assumes spatially separated polarisable particles

$$w_h \left(\frac{\varepsilon_h - \varepsilon_{eff}}{\varepsilon_h + 2\varepsilon_{eff}}\right) = -w_i \left(\frac{\varepsilon_i - \varepsilon_{eff}}{\varepsilon_i + 2\varepsilon_{eff}}\right)$$

Bruggeman Approximation

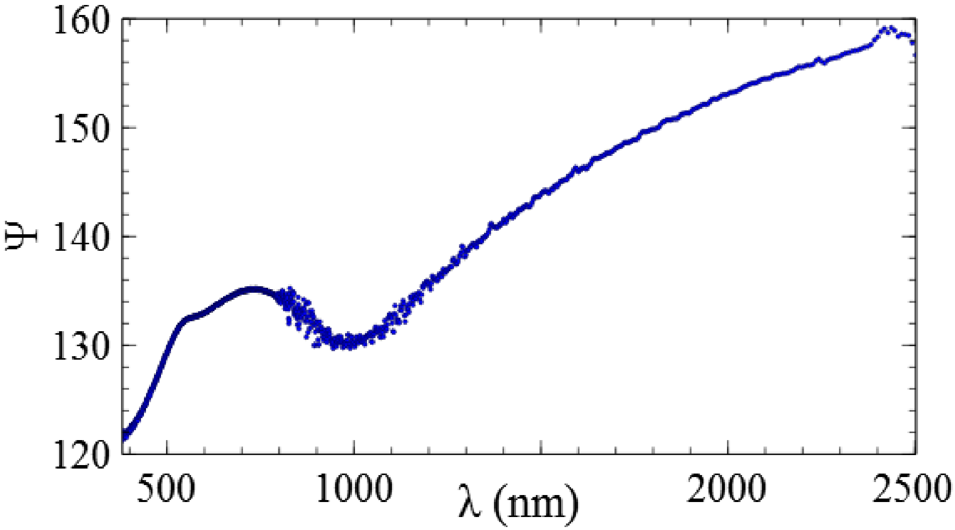
- Assumes two kinds of spherical particles randomly arranged.
- Spatial separation between particles should be small (i.e. w_i is large)

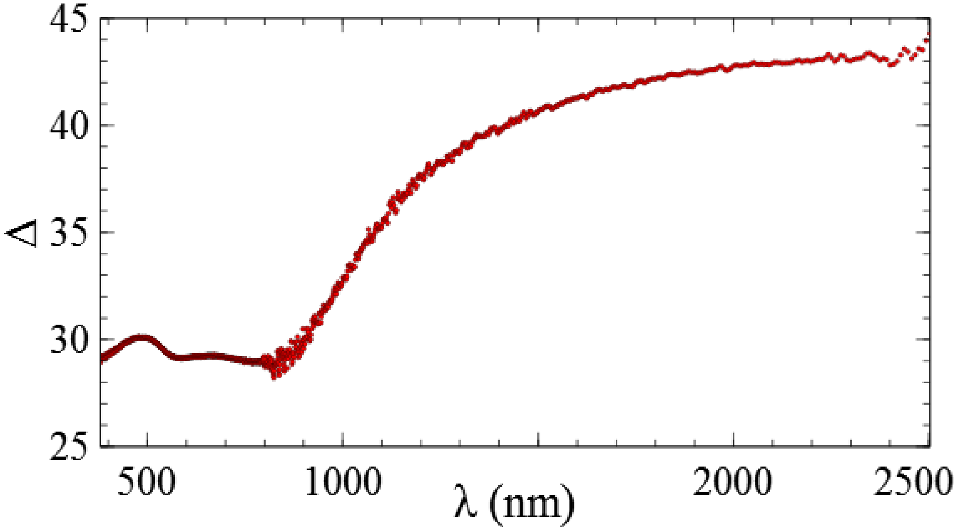


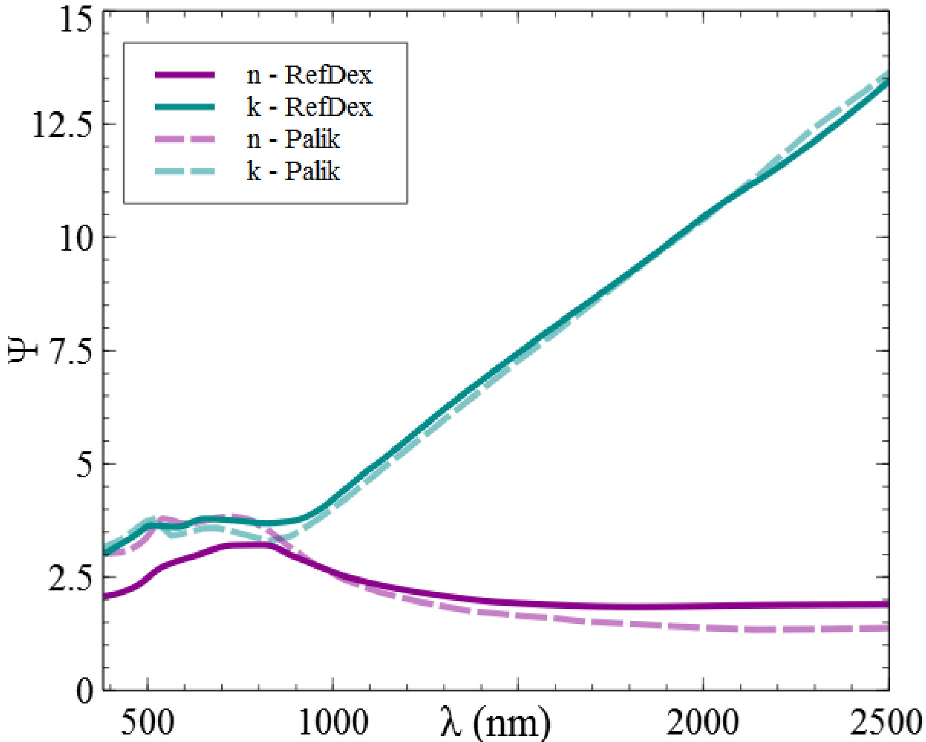
$$\frac{r_p}{r_s} = \tan\Psi\, e^{i\Delta}$$

- Ellipsometric parameters Ψ and Δ simulated by RefDex
- Useful for highly absorbing substrates
- Currently incompatable with roughness and inhomogeneity advanced features

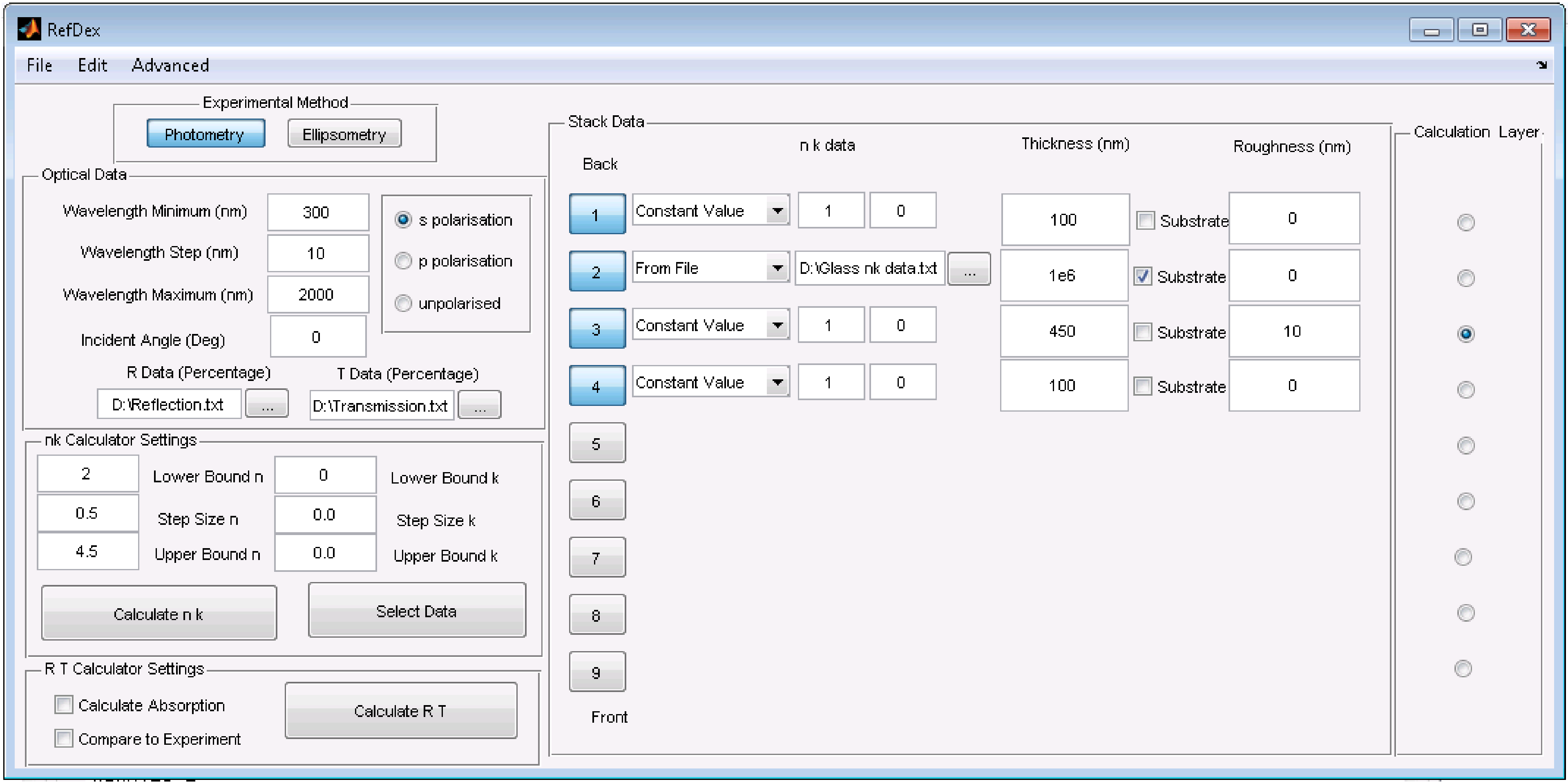
n k Data from Ellipsometry – Example of Mo film

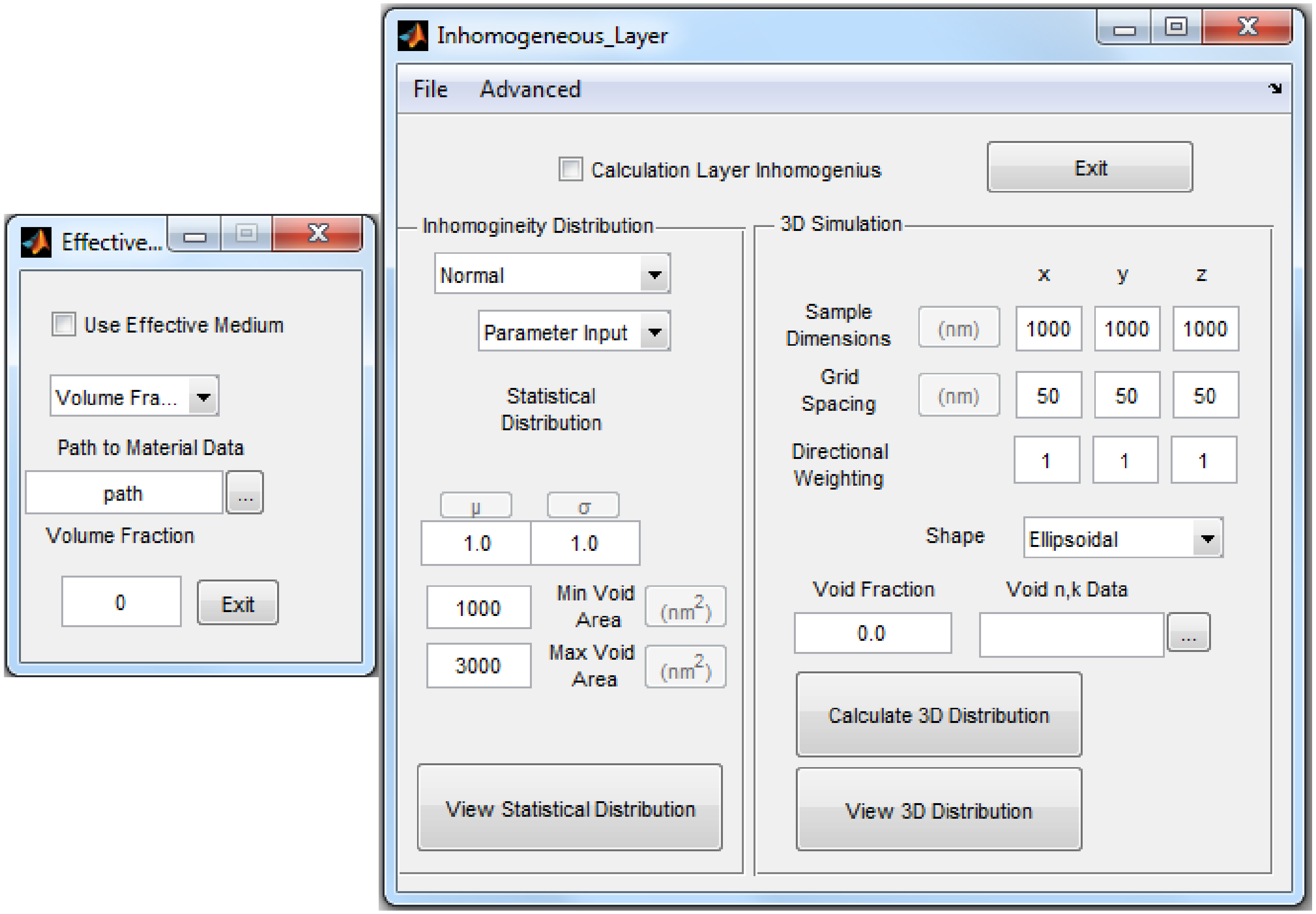




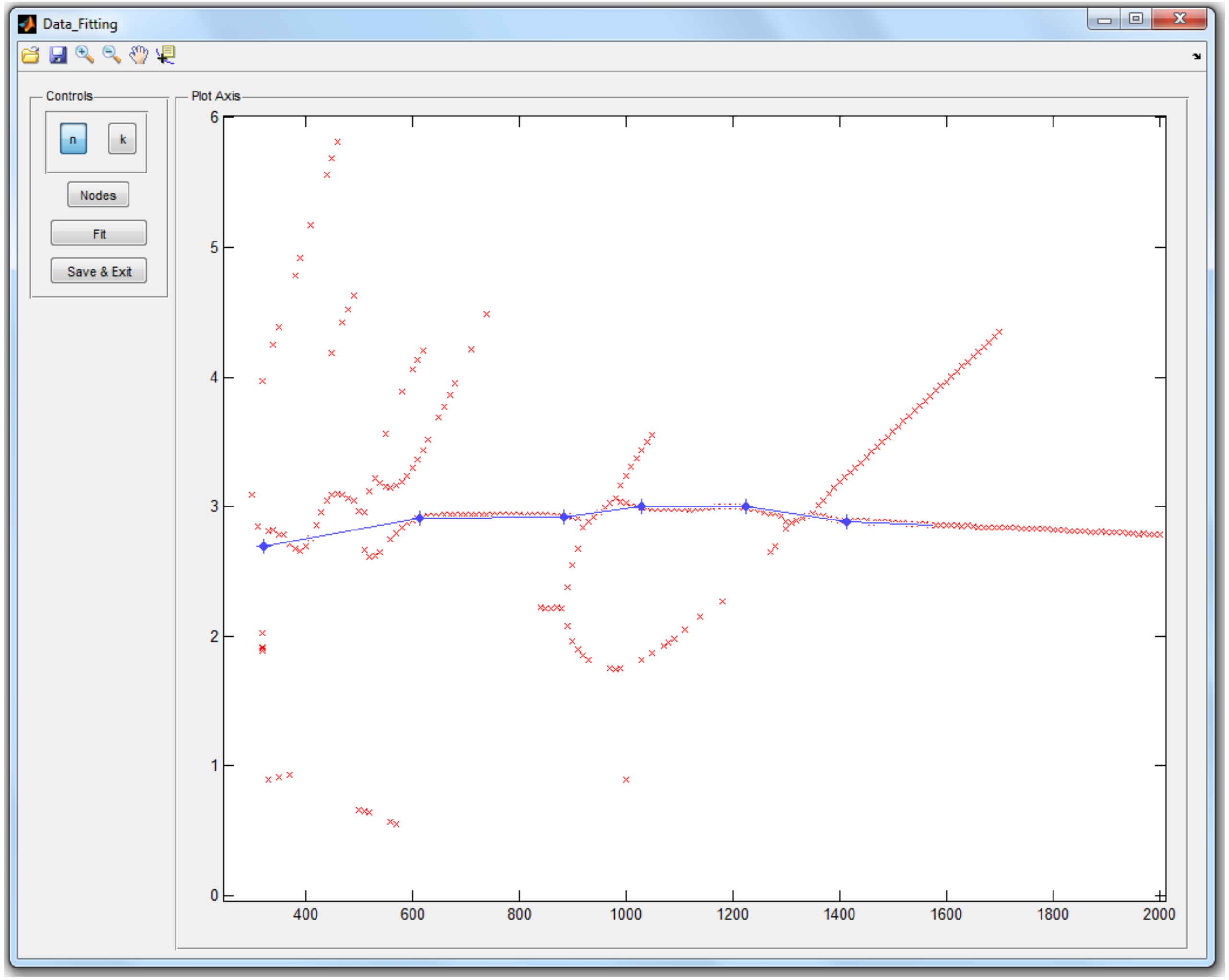


Main Interface





Effective...
Use Effective Medium
Volume Fra...
Path to Material Data
path
...
Volume Fraction
0
Exit
Inhomogeneous_Layer
File
Advanced
Calculation Layer Inhomogenius
Exit
Inhomogineity Distribution
Normal
Parameter Input
Statistical Distribution
μ
σ
1.0
1.0
1000
Min Void Area
(nm²)
3000
Max Void Area
(nm²)
View Statistical Distribution
3D Simulation
x
y
z
Sample Dimensions
(nm)
1000
1000
1000
Grid Spacing
(nm)
50
50
50
Directional Weighting
1
1
1
Shape
Ellipsoidal
Void Fraction
Void n,k Data
0.0
...
Calculate 3D Distribution
View 3D Distribution



- Interactive fitting process
- Place nodes which are automatically connected by a smooth function
- User selects physically meaningful solutions from multiply degenerate solution space

Summary and Outlook

RefDex

- calculates T, R (n,k) for a multilayer stack

  → extracts n, k from (T, R)
- considers surface roughness
- applies to inhomogeneous layers
- has also basic features for ellipsometry

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- is freely available from

  https://www.uni-due.de/ag-schmid/refdex.php

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