Differentiable Interference Modeling for Cost-Effective Growth Estimation of Thin Films

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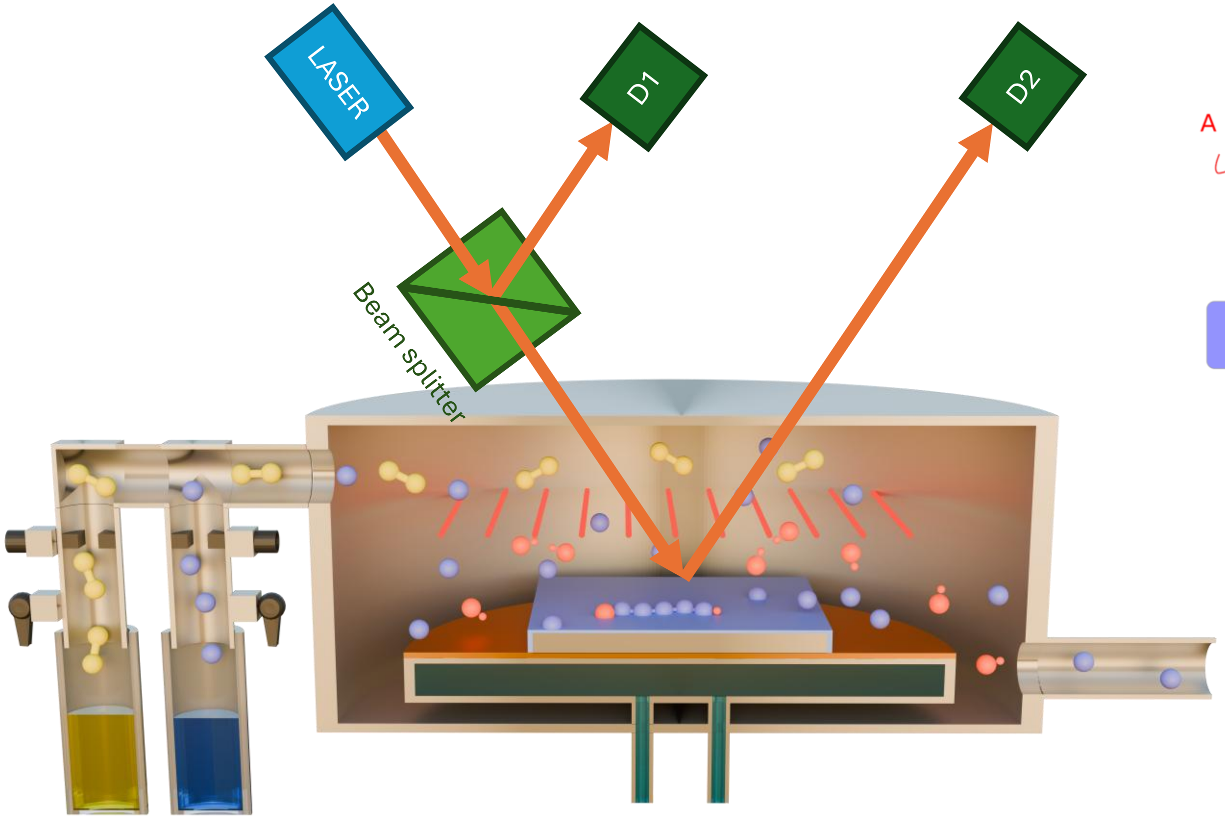
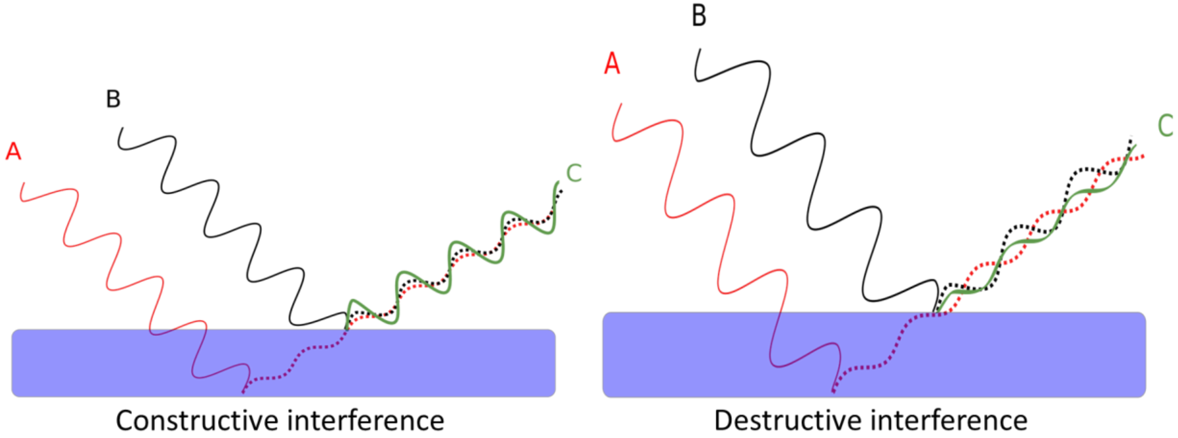
Contents

Introduction – iCVD and Interferometry





- Monomer & initiator evaporate and initiator decomposes at hot filament
- Free-radical polymerziation at the cooled substrate (adhesion through condensation)
- Corformal growth

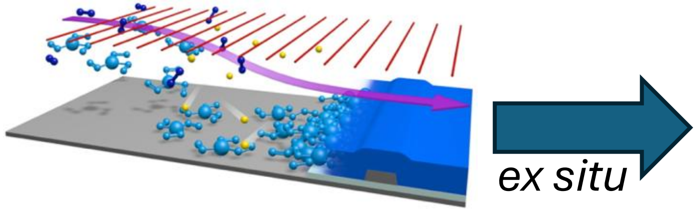
Schröder, S., Polonskyi, O., Strunskus, T., Faupel, F., Nanoscale gradient copolymer films via single-step deposition from the vapor phase, 2020, Materials Today, 37, 35-42, 10.1016/j.mattod.2020.02.004

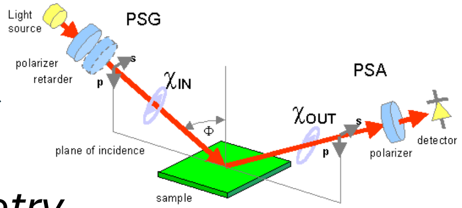
Initiator decomposition: I-I → 2 I•
Initiation: I• + M → I-M•
Propagation: I-M• + M → I-M-M•, I-M-M• + M → I-M-M-M•, ...
Termination: I-...-M• + •M-...-I → I-...-M-M-...-I or I-...-M• + •I → I-...-M-I

Motivation for Interferometry – a Cost-Effective Solution for Obtaining Growth Rates and Refractive Indices

Where? | **How?** | **Goal?** | **Problem?**

CAU Kiel
(right now)
worst



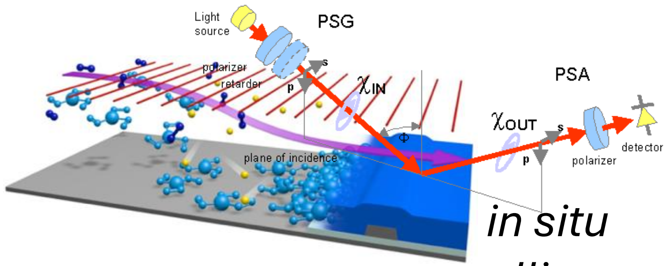


ex situ ellipsometry

Film thickness
d (*ex situ*)

- Only ex situ -> What happens during deposition?
- Refractive index assumed (or Cauchy)

Fancy, rich institute
best

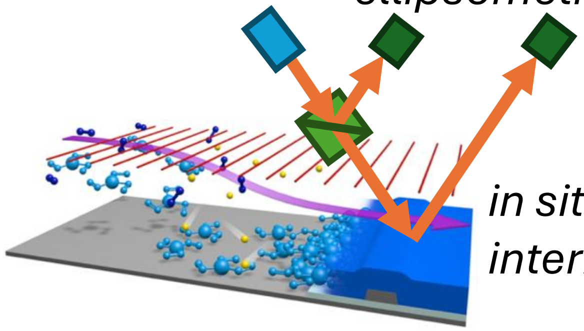


in situ ellipsometry

Film thickness
d & refractive index n (*in situ*)

- €
- Space in fume hood

CAU Kiel
(soon to be) **(hopefully)**
nearly best

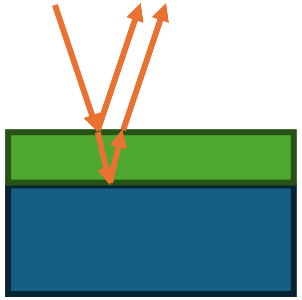


in situ interferometry

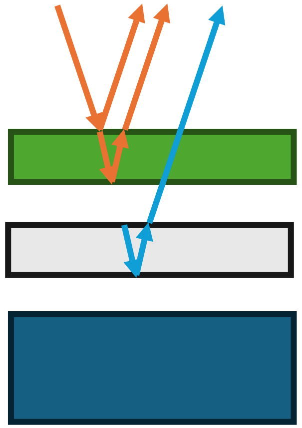
Film thickness
d & refractive index n (*in situ*)

- New model -> needs testing
- DIY

Model overview



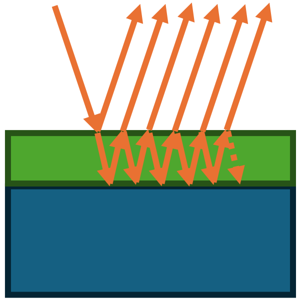
One layer,
no internal reflections



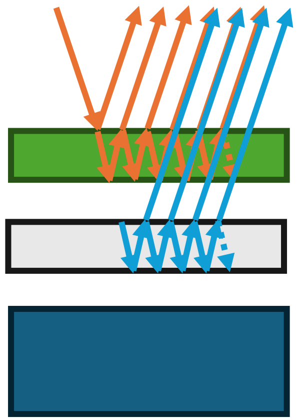
Multiple layers,
no internal reflections

Ray propagation approaches

- Fresnel's equations for reflection and transmission at interfaces
- Phase gain per layer penetration described by $e^{i\delta}$
- Analyze each ray, then sum up all rays contributing to total reflectivity

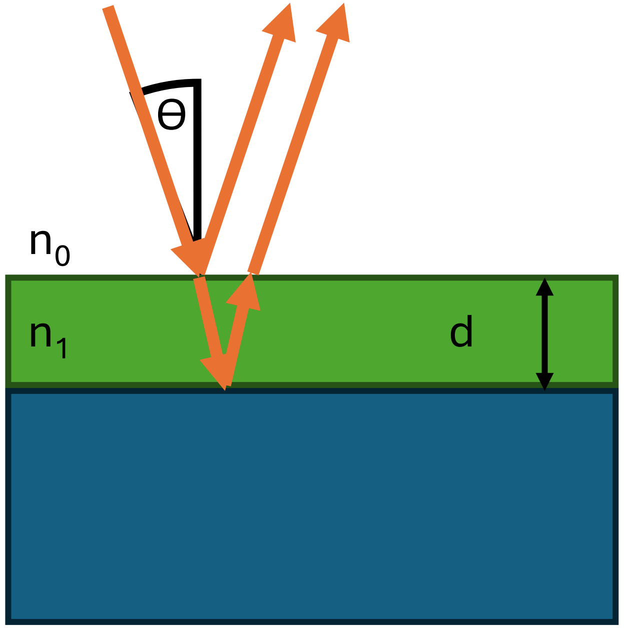


One layer,
internal reflections



Multiple layers,
internal reflections

Model 1: One Layer, no Internal Reflections



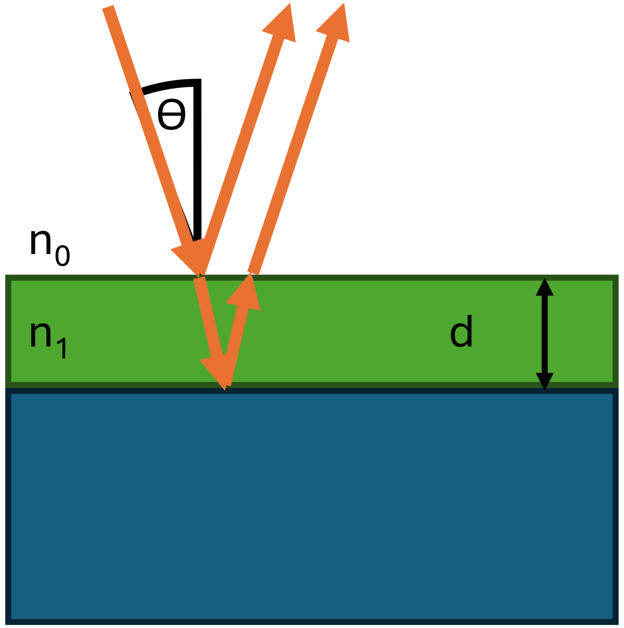
$$I(d) = 4{E_0}^2 \cos^2\left(\frac{2\pi d\sqrt{{n_1}^2 - {n_0}^2 \sin^2(\pi - \Theta)}}{\lambda_0}\right)$$

$$d = d_0 + \int_0^t v(t)dt \xrightarrow{v=\text{const.}} d = d_0 + vt$$

$$\boxed{I(t) = \underbrace{4{E_0}^2}_{I_{\text{inc}}} \cos^2\left(\frac{2\pi(d_0 + vt)\sqrt{{n_1}^2 - {n_0}^2 \sin^2(\pi - \theta)}}{\lambda_0}\right) + I_{\text{dark}}}$$

Kim, N., Real time thickness measurement of thin film for end-point detector (EPD) of 12-inch spin etcher using the white light interferometry, 2005, Microsyst. Technol., 11, 8-10, 958-964, 10.1007/s00542-005-0505-9

Model 1: One Layer, no Internal Reflections

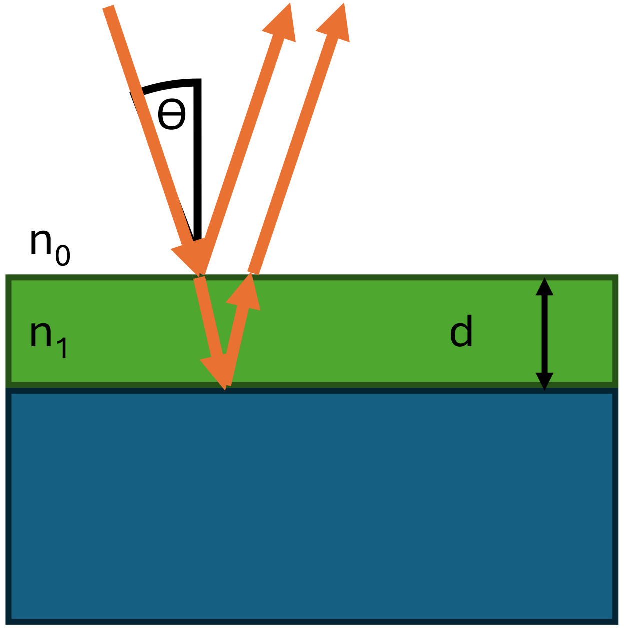


$$I(t) = I_{\mathrm{inc}}\sqrt{2\alpha\beta}\sqrt{\frac{\alpha^2+\beta^2}{2\alpha\beta} + \cos\left(2\pi\frac{d}{\Delta d_{\mathrm{opt}}}\right)}$$

$$\Delta d_{\mathrm{opt}} = \frac{\lambda_0}{2n_1\cos(\Phi)}, \frac{\sin(\Phi)}{\sin(\Theta)} = \frac{1}{n_1}, d = d_0 + vt$$

Cruden, B., Chu, K., Gleason, K., Sawin, H., Thermal Decomposition of Low Dielectric Constant Pulsed Plasma Fluorocarbon Films, 1999, J. Electrochem. Soc., 146, 12, 4590-4604, 10.1149/1.1392680

Model 1: One Layer, no Internal Reflections

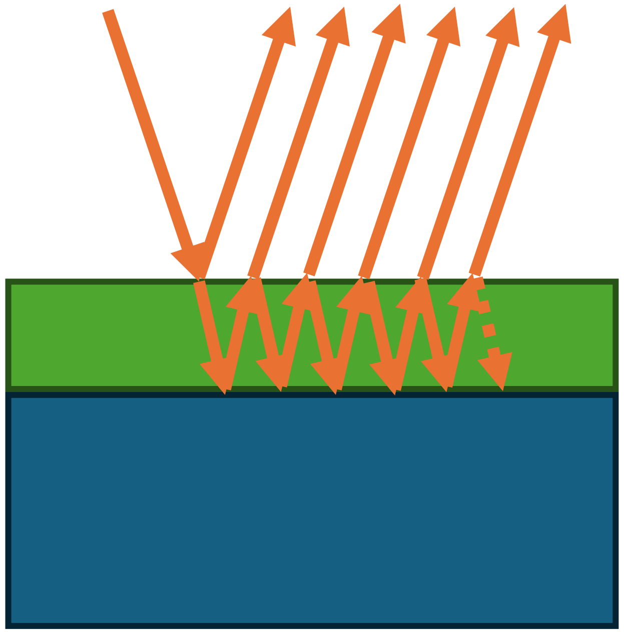


$$I(d) = I_{\text{inc}}R + I_{\text{dark}}, R = |r_{012}|^2$$

$$r_{012} = r_{01} + t_{01}e^{i\delta}r_{12}e^{i\delta}t_{10} = r_{01} + (1 - {r_{01}}^2)r_{12}e^{2i\delta}$$

$$\boxed{\begin{gathered} I(t) = I_{\text{inc}}\left|r_{01} + (1 - {r_{01}}^2)r_{12}e^{2i\delta}\right|^2 + I_{\text{dark}}, \\ \delta = \frac{2\pi(d_0 + vt)\sqrt{{n_1}^2 - {n_0}^2\sin^2(\pi - \theta)}}{\lambda_0} \end{gathered}}$$

Model 2: One Layer, Internal Reflections



$$I(d) = I_{\mathrm{inc}}R + I_{\mathrm{dark}}, R = |r_{012}|^2$$

$$r_{012} = r_{01} + t_{01}e^{i\delta}r_{12}e^{i\delta}t_{10} + t_{01}e^{i\delta}r_{12}e^{i\delta}r_{10}e^{i\delta}r_{12}e^{i\delta}t_{10} + \cdots$$

$$= r_{01} + t_{01}r_{12}t_{10}e^{i2\delta}(1 + r_{10}r_{12}e^{i2\delta} + \cdots)$$

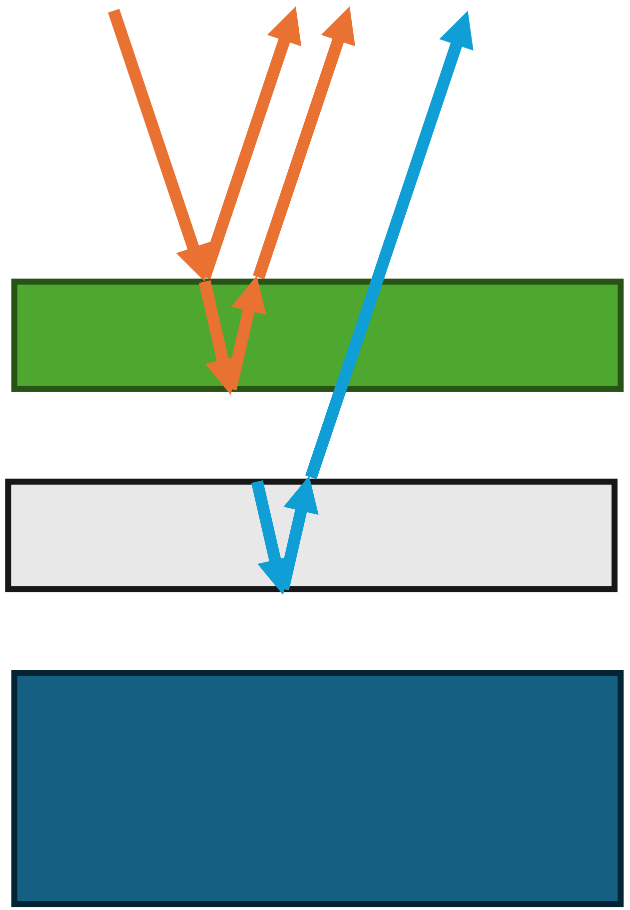
$$= r_{01} + \frac{t_{01}r_{12}t_{10}e^{2i\delta}}{1 - r_{10}r_{12}e^{2i\delta}}$$

$$= \frac{r_{01} + r_{12}e^{2i\delta}}{1 - r_{10}r_{12}e^{2i\delta}}$$

$$\boxed{I(t) = I_{\mathrm{inc}}\left|\frac{r_{01} + r_{12}e^{2i\delta}}{1 - r_{10}r_{12}e^{2i\delta}}\right|^2 + I_{\mathrm{dark}}, \delta = \frac{2\pi(d_0 + vt)\sqrt{{n_1}^2 - {n_0}^2\sin^2(\pi - \theta)}}{\lambda_0}}$$

Stenzel, O., The Physics of Thin Film Optical Spectra, 2016, Springer, 2^{nd}, 10.1007/978-3-319-21602-7

Model 3: Multiple Layers, no Internal Reflections

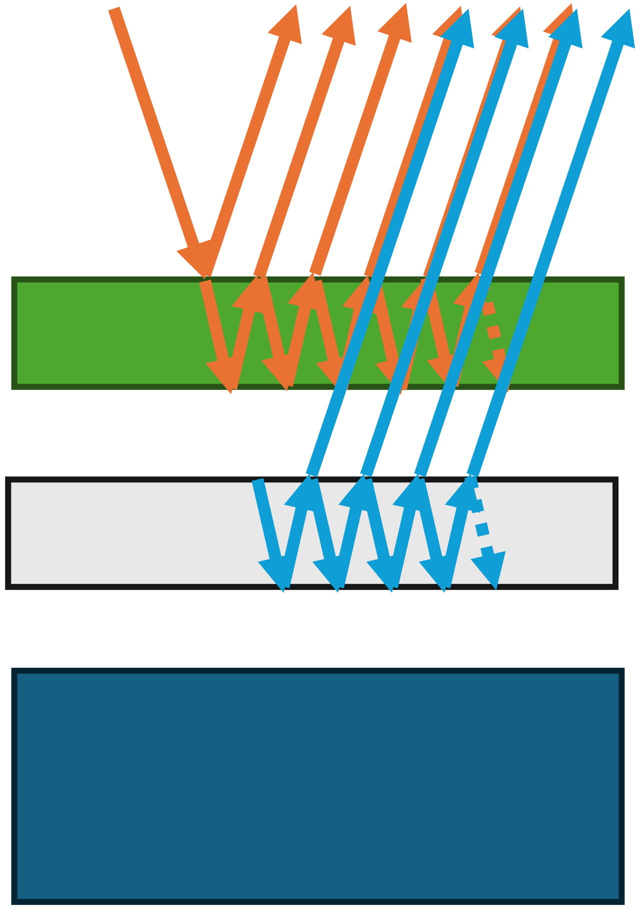


$$I(d) = I_{\text{inc}}R + I_{\text{dark}}, R = |r_{0123\ldots}|^2$$

$$r_{0123\ldots}$$
$$= r_{01} + t_{01}e^{i\delta_1}r_{12}e^{i\delta_1}t_{10} + t_{01}e^{i\delta_1}t_{12}e^{i\delta_2}r_{23}e^{i\delta_2}t_{21}e^{i\delta_1}t_{10}$$
$$+ t_{01}e^{i\delta_1}t_{12}e^{i\delta_2}t_{23}e^{i\delta_3}r_{34}e^{i\delta_3}t_{32}e^{i\delta_2}t_{21}e^{i\delta_1}t_{10} + \cdots$$
$$= r_{01} + \sum_{j=1}^{\text{NLAY}}\left(r_{j(j+1)}\prod_{k=1}^{j}\left(t_{(k-1)k}t_{k(k-1)}e^{i2\delta_k}\right)\right)$$
$$= r_{01} + \sum_{j=1}^{\text{NLAY}}\left(r_{j(j+1)}\prod_{k=1}^{j}\left(\left(1 - r_{(k-1)k}{}^2\right)e^{i2\delta_k}\right)\right)$$

$$\boxed{I(t) = I_{\text{inc}}\left|r_{01} + \sum_{j=1}^{\text{NLAY}}\left(r_{j(j+1)}\prod_{k=1}^{j}\left(\left(1 - r_{(k-1)k}{}^2\right)e^{i2\delta_k}\right)\right)\right|^2 + I_{\text{dark}},}$$
$$\boxed{\delta_k = \frac{2\pi d_k\sqrt{n_k{}^2 - n_{k-1}{}^2\sin^2(\pi - \theta_{k-1})}}{\lambda_0}, \frac{\sin(\theta_{k-1})}{\sin(\theta_{k-2})} = \frac{n_{k-2}}{n_{k-1}}, d_1 = d_{1,0} + v_1 t}$$

Model 4: Multiple Layers, Internal Reflections



$$I(d) = I_{\mathrm{inc}}R + I_{\mathrm{dark}}, R = |r_{0123\ldots}|^2$$

$$r_{0123\ldots} = r_{01} + \left(t_{01}e^{i\delta_1}r_{12}e^{i\delta_1}t_{10} + t_{01}e^{i\delta_1}r_{12}e^{i\delta_1}r_{10}e^{i\delta_1}r_{12}e^{i\delta_1}t_{10} + \cdots\right) +$$
$$\left(t_{01}e^{i\delta_1}t_{12}e^{i\delta_2}r_{23}e^{i\delta_2}t_{21}e^{i\delta_1}t_{10} + t_{01}e^{i\delta_1}t_{12}e^{i\delta_2}r_{23}e^{i\delta_2}r_{21}e^{i\delta_2}r_{23}e^{i\delta_2}t_{21}e^{i\delta_1}t_{10} + \cdots\right) + \cdots$$

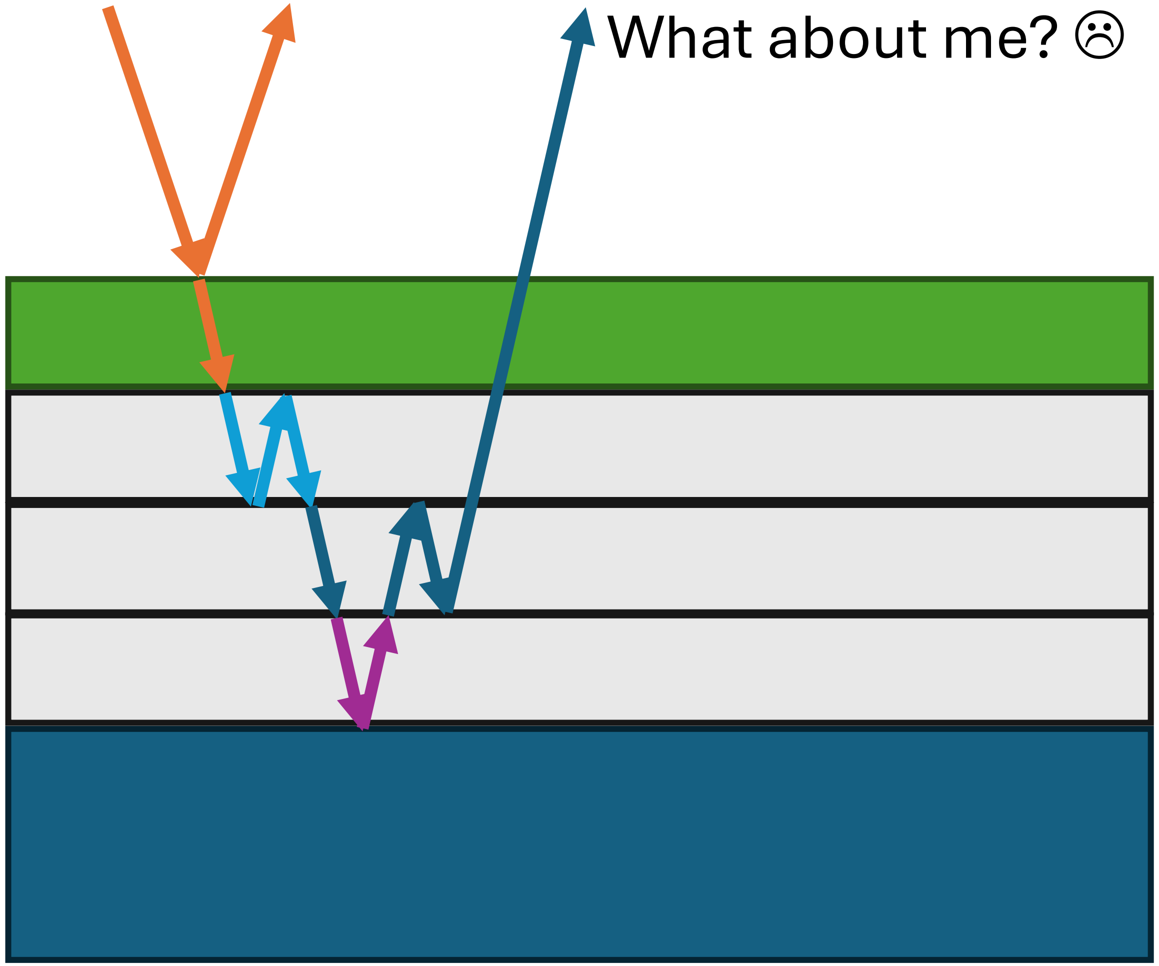
$$= r_{01} + \frac{t_{01}r_{12}t_{10}e^{2i\delta_1}}{1-r_{10}r_{12}e^{2i\delta_1}} + \frac{t_{01}t_{12}r_{23}t_{21}t_{10}e^{2i\delta_1}e^{2i\delta_2}}{1-r_{21}r_{23}e^{2i\delta_2}} + \frac{t_{01}t_{12}t_{23}r_{34}t_{32}t_{21}t_{10}e^{2i\delta_1}e^{2i\delta_2}e^{2i\delta_3}}{1-r_{32}r_{34}e^{2i\delta_3}} + \cdots$$

$$= r_{01} + \sum_{j=1}^{\mathrm{NLAY}}\left(\frac{r_{j(j+1)}\prod_{k=1}^{j}\left(t_{(k-1)k}t_{k(k-1)}e^{i2\delta_k}\right)}{1-r_{j(j-1)}r_{j(j+1)}e^{2i\delta_j}}\right)$$

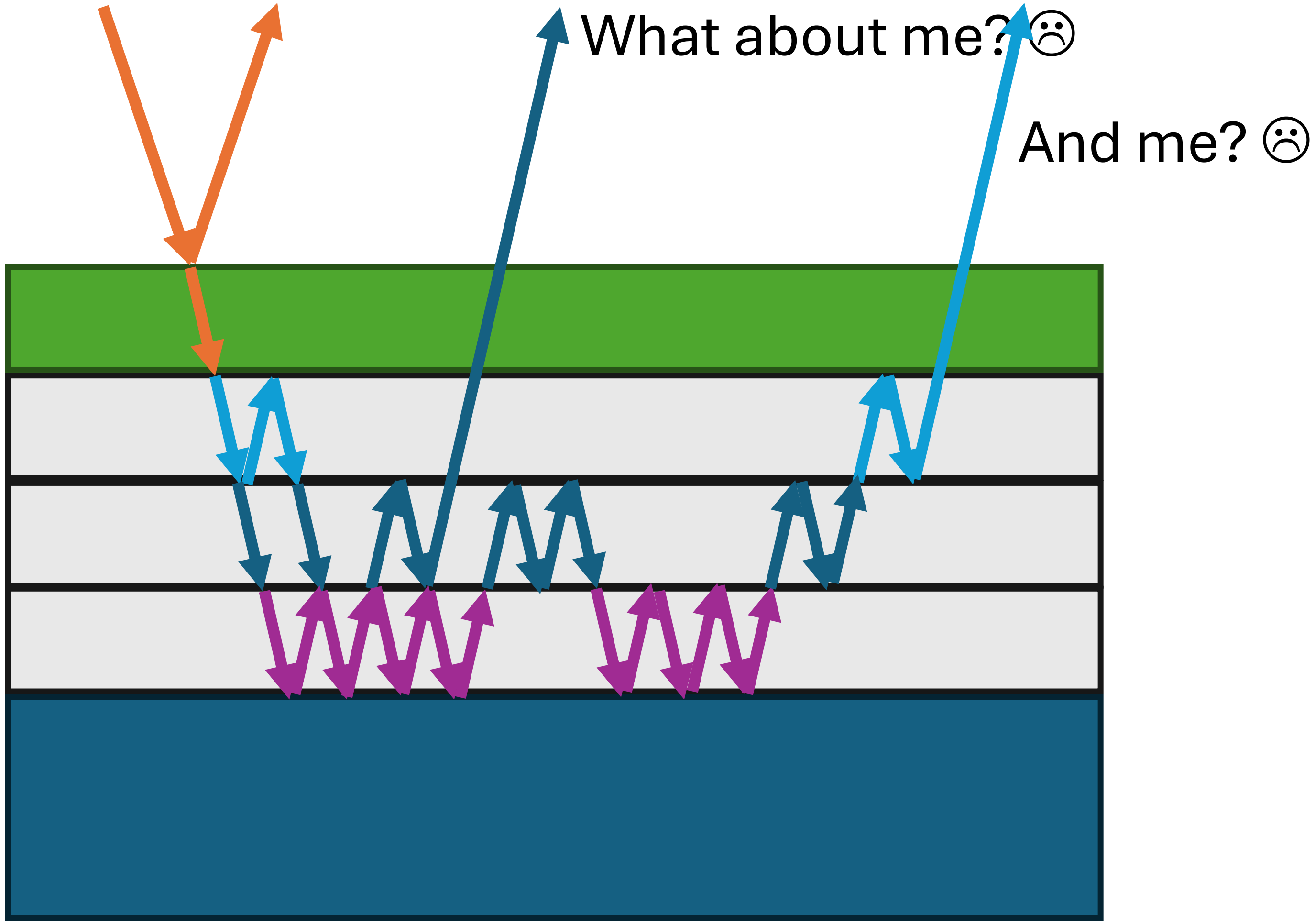
$$= r_{01} + \sum_{j=1}^{\mathrm{NLAY}}\left(\frac{r_{j(j+1)}\prod_{k=1}^{j}\left(\left(1-r_{k(k-1)}{}^2\right)e^{i2\delta_k}\right)}{1-r_{j(j-1)}r_{j(j+1)}e^{2i\delta_j}}\right)$$

$$\boxed{\begin{gathered}I(t) = I_{\mathrm{inc}}\left|r_{01} + \sum_{j=1}^{\mathrm{NLAY}}\left(\frac{r_{j(j+1)}\prod_{k=1}^{j}\left(\left(1-r_{k(k-1)}{}^2\right)e^{i2\delta_k}\right)}{1-r_{j(j-1)}r_{j(j+1)}e^{2i\delta_j}}\right)\right|^2 + I_{\mathrm{dark}},\\ \delta_k = \frac{2\pi d_k\sqrt{n_k{}^2 - n_{k-1}{}^2\sin^2(\pi-\theta_{k-1})}}{\lambda_0}, \frac{\sin(\theta_{k-1})}{\sin(\theta_{k-2})} = \frac{n_{k-2}}{n_{k-1}}, d_1 = d_{1,0} + v_1 t\end{gathered}}$$

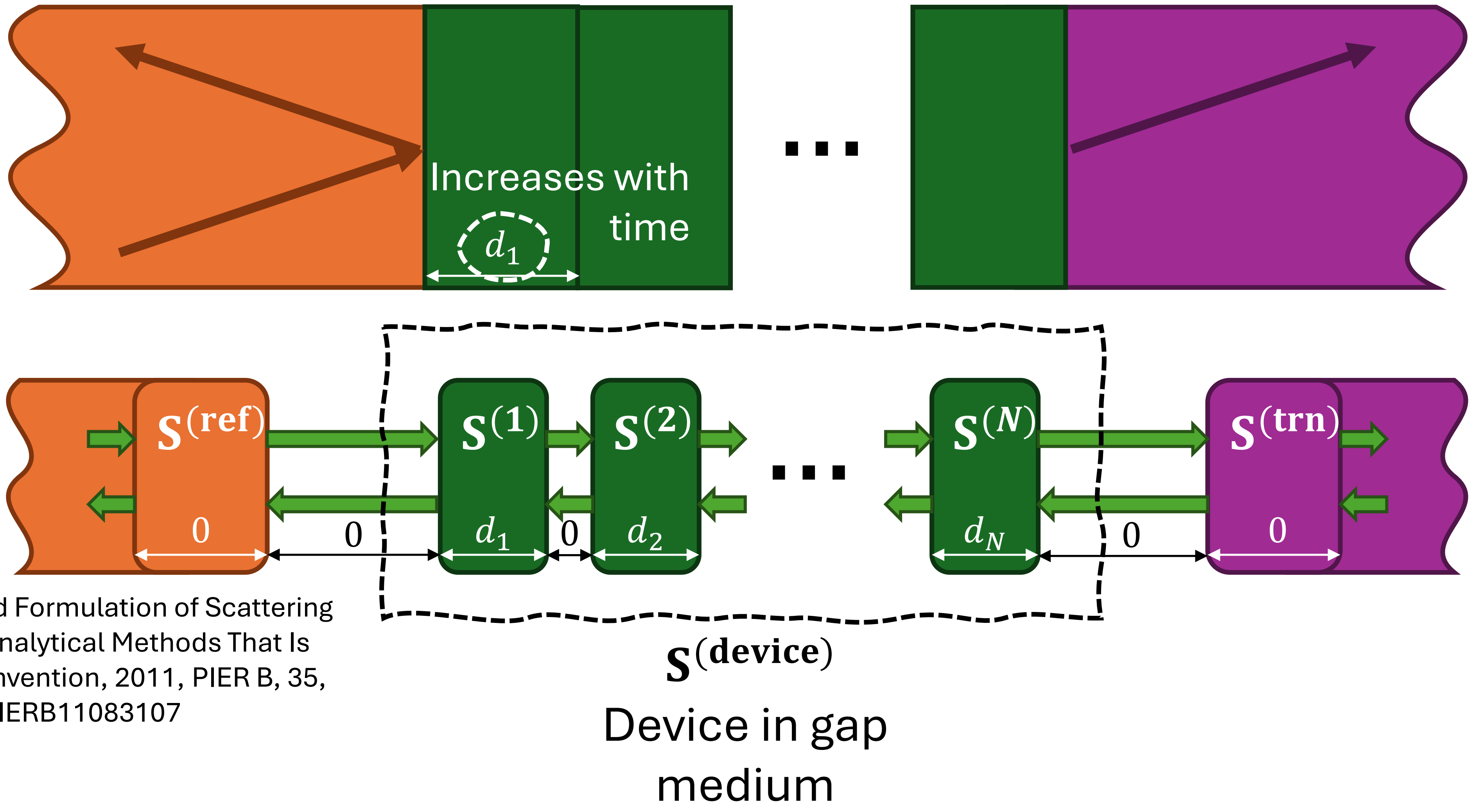
Problems with Ray Propagation Approaches



Problems with Ray Propagation Approaches

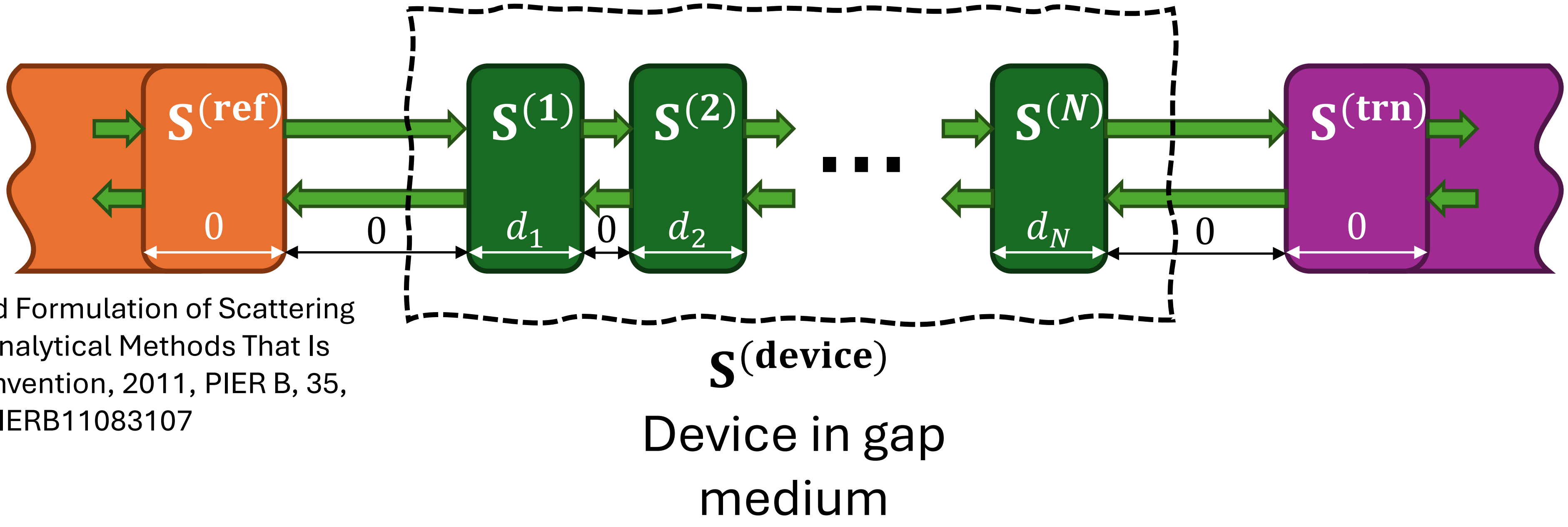


Model 5: Transfer Matrix Method



Rumpf, R., Improved Formulation of Scattering Matrices for Semi-Analytical Methods That Is Consistent with Convention, 2011, PIER B, 35, 241-261, 10.2528/PIERB11083107

Model 5: Transfer Matrix Method



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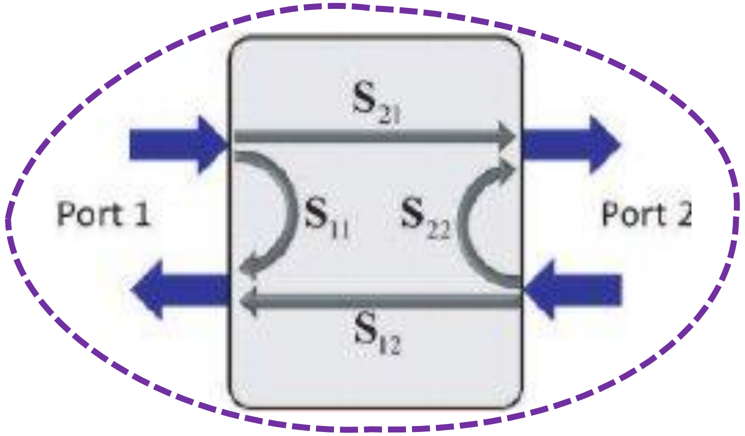
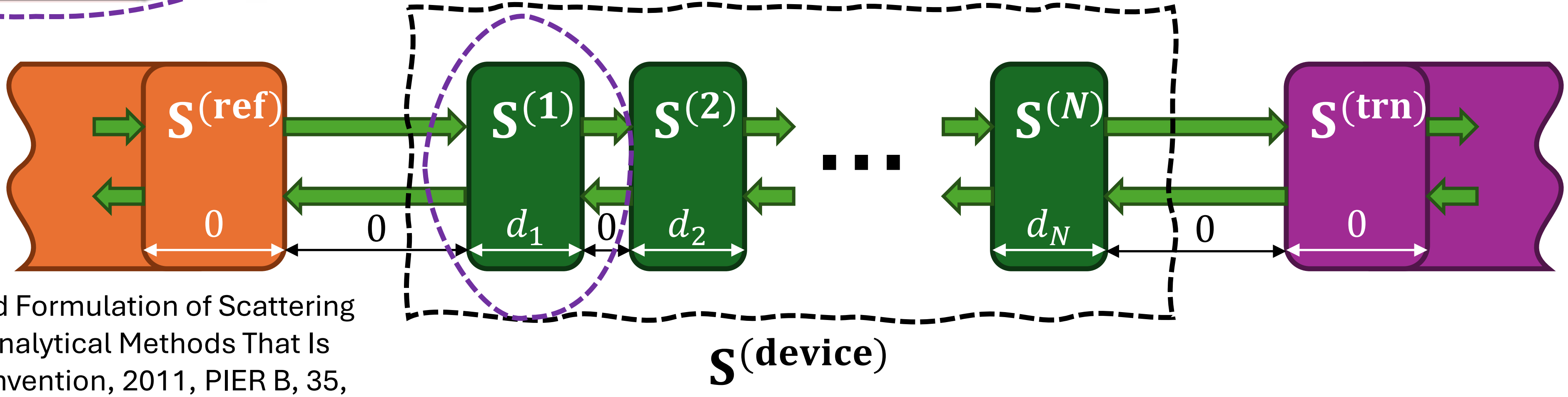
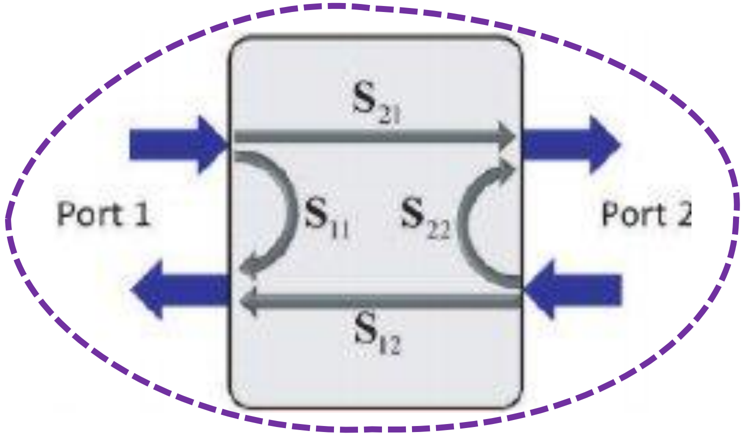




Rumpf, R., Improved Formulation of Scattering Matrices for Semi-Analytical Methods That Is Consistent with Convention, 2011, PIER B, 35, 241-261, 10.2528/PIERB11083107

Device in gap medium

Model 5: Transfer Matrix Method



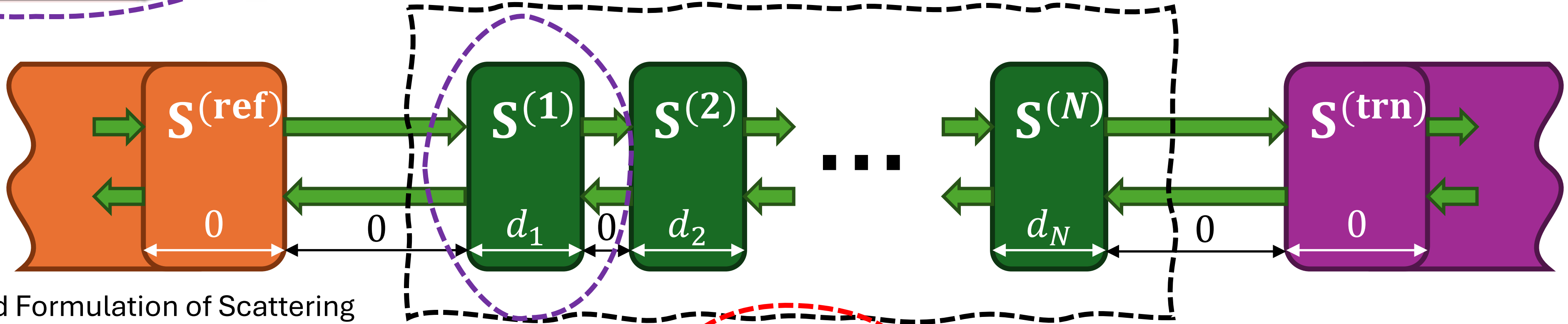
$$\boxed{\mathbf{S}^{(\text{global})} = \mathbf{S}^{(\text{ref})} \otimes \overbrace{\left[\mathbf{S}^{(1)} \otimes \mathbf{S}^{(2)} \otimes \cdots \otimes \mathbf{S}^{(N)}\right]}^{\mathbf{S}^{(\text{device})}} \otimes \mathbf{S}^{(\text{trn})}}$$

$$\boxed{R = \left|\vec{E}_{\text{ref}}\right|^2}$$

$$\begin{bmatrix} E_x^{\text{ref}} \\ E_y^{\text{ref}} \end{bmatrix} = \mathbf{S}_{11}^{(\text{global})} \begin{bmatrix} E_x^{\text{inc}} \\ E_y^{\text{inc}} \end{bmatrix} \qquad \begin{bmatrix} E_x^{\text{trn}} \\ E_y^{\text{trn}} \end{bmatrix} = \mathbf{S}_{21}^{(\text{global})} \begin{bmatrix} E_x^{\text{inc}} \\ E_y^{\text{inc}} \end{bmatrix}$$

$$T = \left|\vec{E}_{\text{trn}}\right|^2 \frac{\text{Re}\left[\widetilde{k_z^{\text{trn}}}/\mu_{r,\text{trn}}\right]}{\text{Re}\left[\widetilde{k_z^{\text{inc}}}/\mu_{r,\text{inc}}\right]}$$

$$E_z^{\text{ref}} = -\frac{\widetilde{k_x} E_x^{\text{ref}} + \widetilde{k_y} E_y^{\text{ref}}}{\widetilde{k_z^{\text{ref}}}} \qquad E_z^{\text{trn}} = -\frac{\widetilde{k_x} E_x^{\text{trn}} + \widetilde{k_y} E_y^{\text{trn}}}{\widetilde{k_z^{\text{trn}}}}$$



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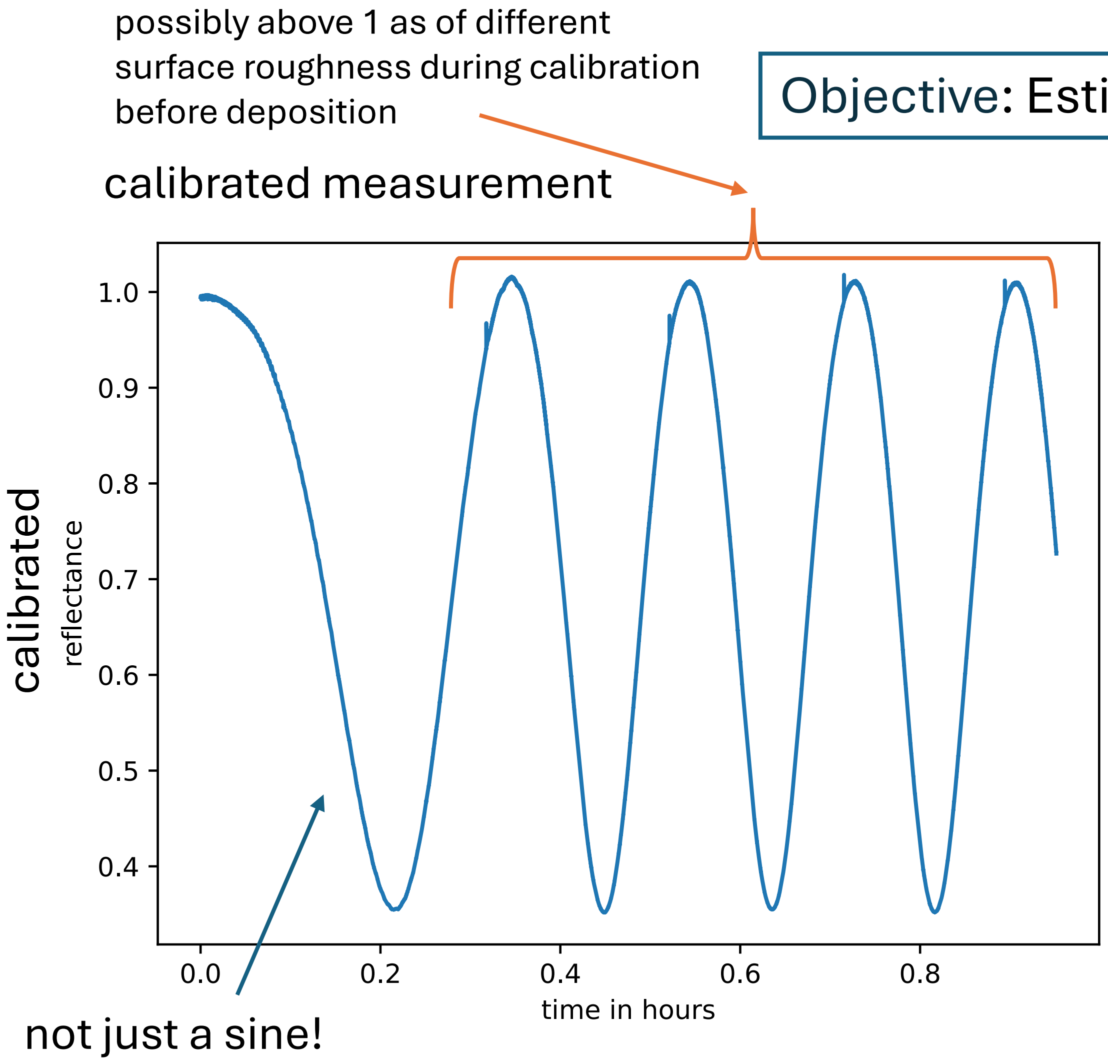
$\mathbf{S}^{(\text{device})}$

Device in gap medium

Our Modeling Problem and Challenges with Traditional Approaches to Instantaneous Frequency Estimation

Objective: Estimate the growth function of the thin layer.



Note: The phase of the interference signal is a linear function of the thickness of the thin layer.

Classical Idea: Obtain the instantaneous frequency / phase using Short Time Fourier Transform / Wavelet analysis / Superlets / Hilbert Transform / ChirpGP / ...

Problem: The Gabor-Limit is especially limiting in this low-frequency setting, classical approaches are **not aware of the underlying signal form**.

Interlude on Differentiable Simulators for Inverse Modeling – How to Smartly Go from Data to Input?

model parameters
e. g. refractive indices, measurement angles, …

Input
e. g. thicknesses
of a thin layer

simulator
(forward model)

Output
e. g. reflectance

Objective
e. g. reproduce
the correct
reflectance

Loss
e. g. L2 or SoftDTW

Aim: Adapt model parameters / input to minimize the loss.

in a **differentiable simulator** the simulation output can be Automatically Differentiated (AD) to its inputs → efficient optimization of e. g. thicknesses *parameterized by a neural network with lots of parameters*

But why?

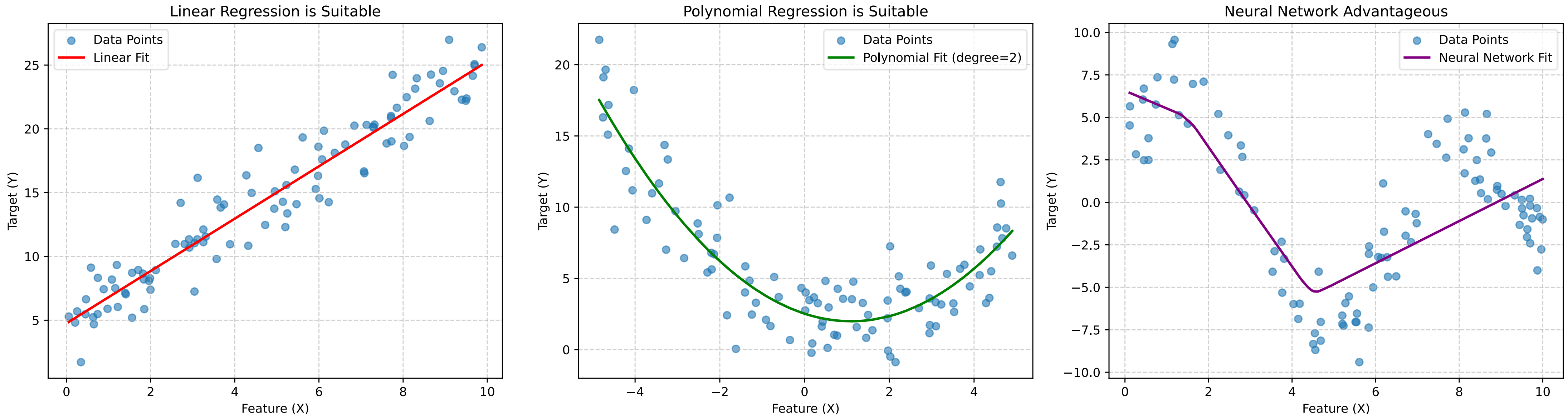
Problem without differentiability: Vast optimization space, might have hundreds to billions of parameters → brute force and smarter related approaches are not feasible, need gradient information but finite differencing is too costly (would need one model evaluation per parameter)

One approach for differentiability: Use a programming library like JAX where the primitive operations themselves are differentiable.

Interlude on Neural Networks (NN)

+ numerically advantageous
+ suited for higher dimensional in- and outputs
+ fast optimization based on backpropagation

Neural Networks are Universal Function Approximators



(just linear regression with additional X^2 feature)

Challenges: overfitting (also fit the noise) and underfitting (model not sufficiently complex), optimization into local minima neural network: flexible statistical model with fast way to adapt all parameters

input → input layer → hidden layer(s) → output layer → output

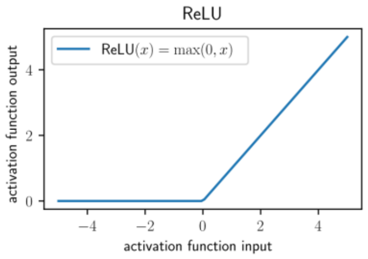
Elements of Statistical Learning

- model
  - e. g. fully connected
- target / loss
  - e. g. squared error to the data (L2 loss)
- optimizer
  - typically gradient descent variants, like ADAM

where the layers consist of neurons, with some neuron m

input_{1m} (weight_{1m}), …, input_{km} (weight_{km}) → $\boxed{\sum \mathrm{input}_i \mathrm{weight}_i + \mathrm{bias}_m}$ ← bias_m → activation function → output_m

activation function,
e. g. ReLu



Aim: Find the weights and biases so that the network learns the data.

Differentiable Interference Modeling I: Model

Design challenge: Ensure monotonicity of the layer thickness.

find a thickness function reproducing the measurement

initialized to guess
of constant growth rate

optimizer: Adam with Learning Rate Decay and Early Stopping

Neural Network
Modeling the **Layer Growth**

feed-forward neural net
one hidden layer with 512 neurons,
dropout against overfitting

softplus
(ensuring growth positivity)

thicknesses by numerical integration

further parameters

differentiable interference models in JAX

simulated reflectance

measured reflectance

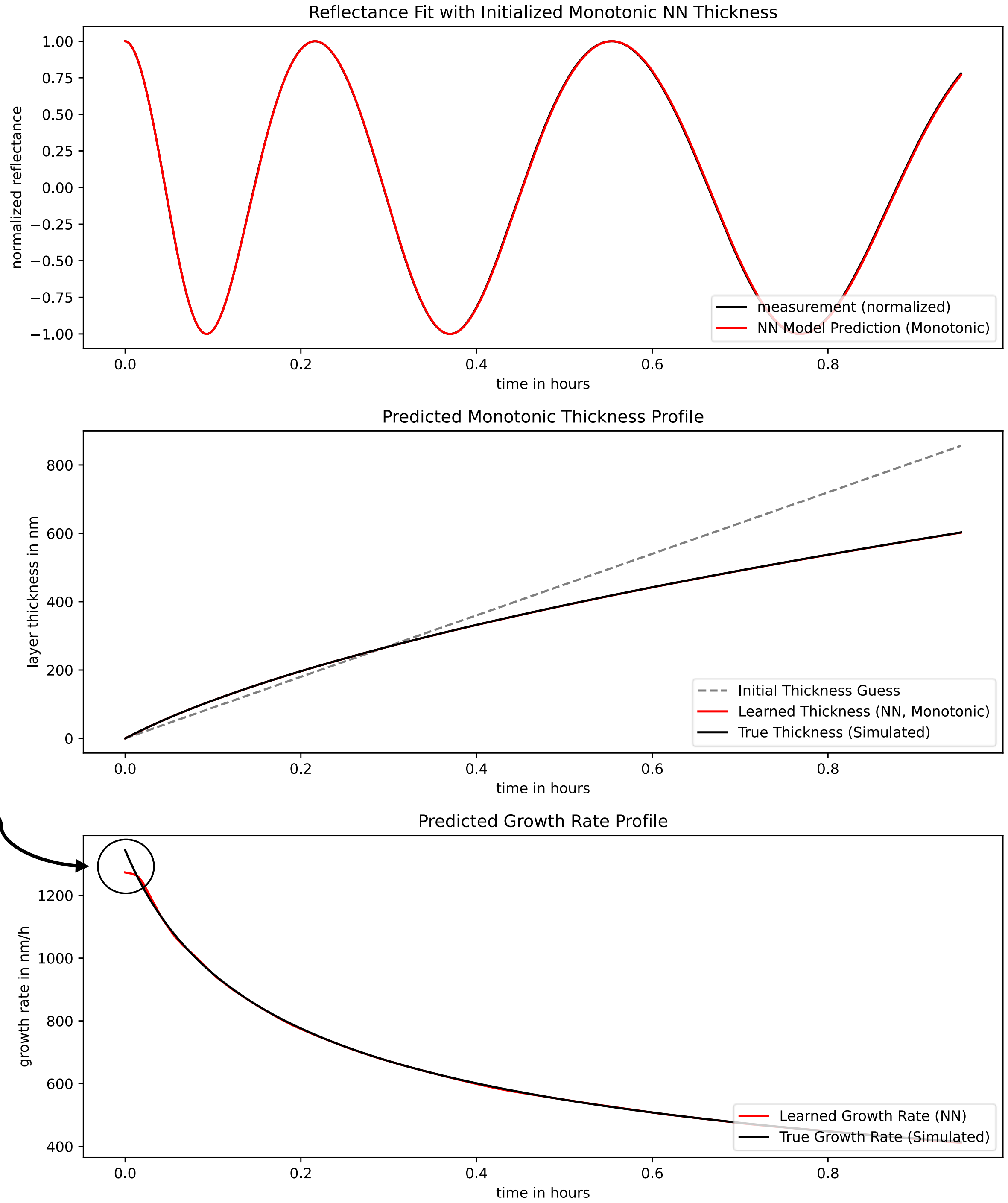
L2 loss

(as an alternative to the neural network,
one might also use a Gaussian Process here)

Differentiable Interference Modeling II: Validation

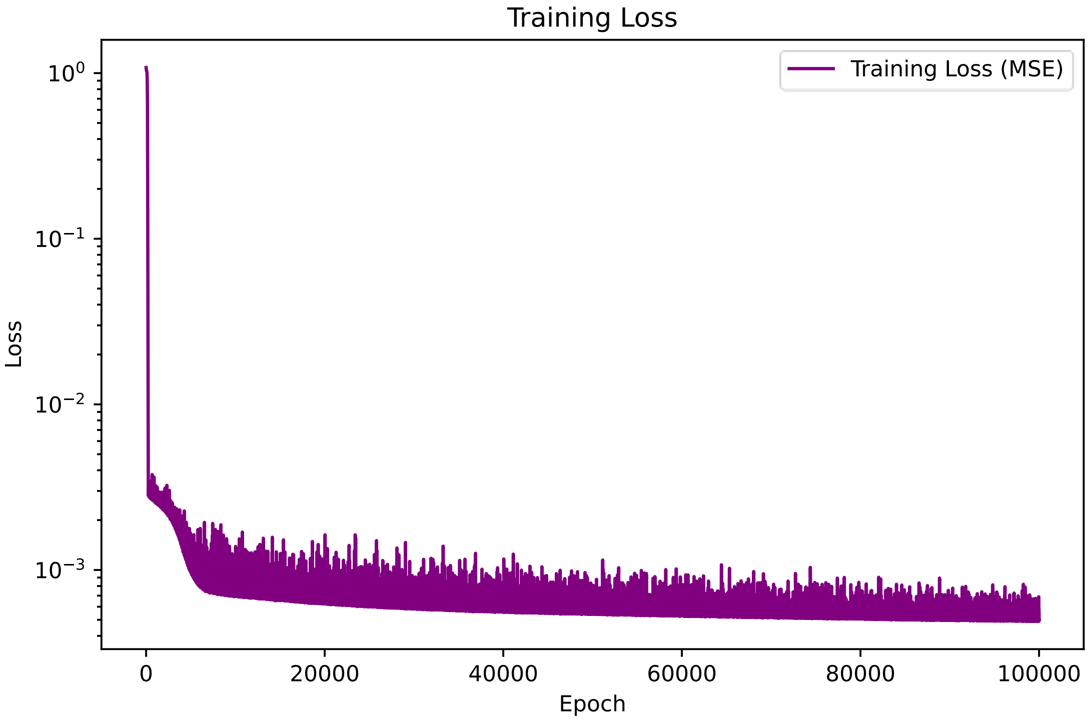
with Smoothed Square-Root thicknesses

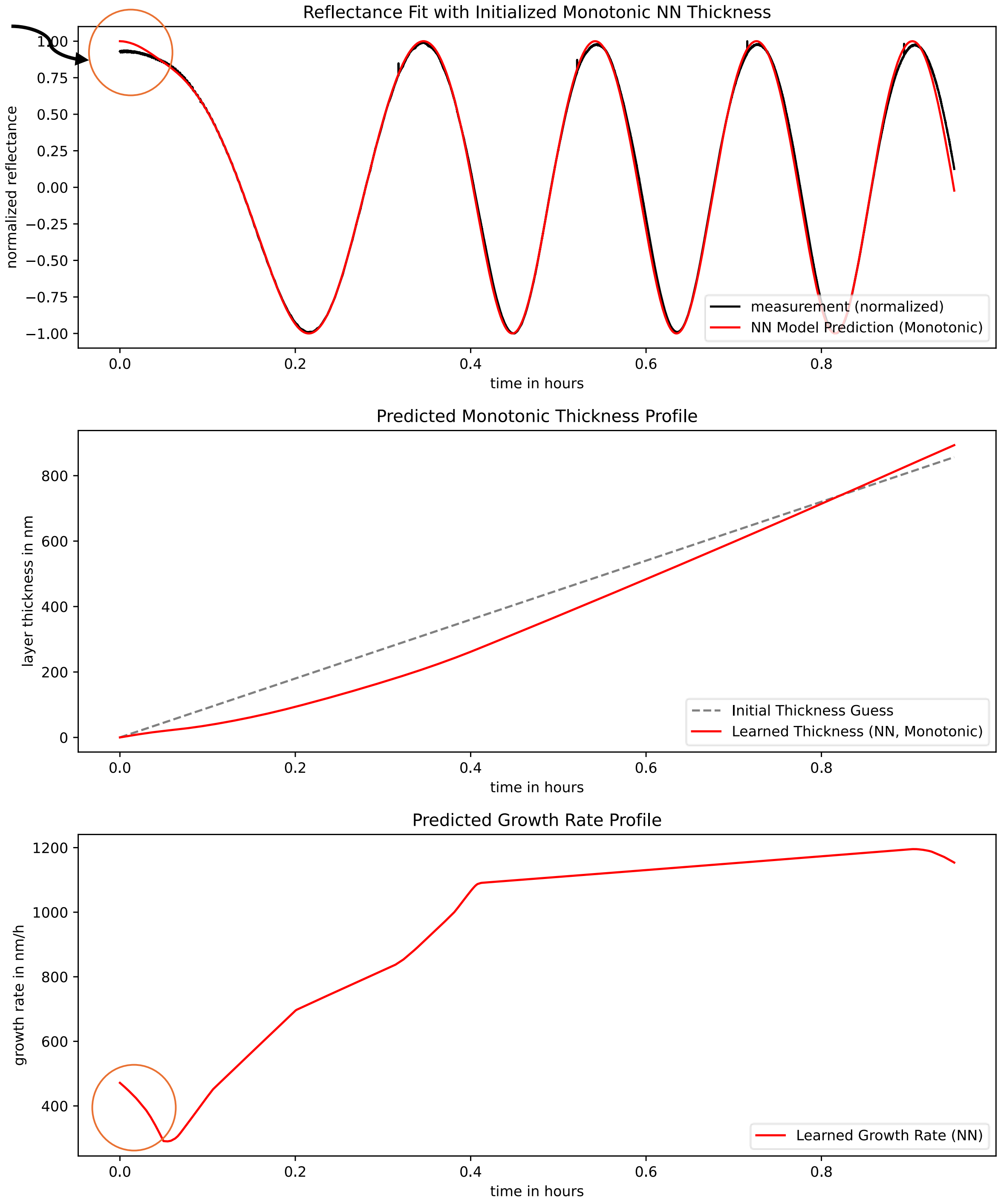
possibly a problem of how we train on the cumulative sum of the “growth rates”



Differentiable Interference Modeling II: Results

problem as discussed





Training takes ~ three minutes on an NVIDIA A100 with the internal reflections single layer model. We don't yet have data where the more advanced models would be necessary but training with TMM gets similar results (at higher training cost though).

Conclusion and Outlook

- we can cheaply find the thickness function over time, we do not need spectral information
- if we would also consider spectral information, we could fit both the refractive index as a function of wavelength as well as the thickness function over time

Literature

General References on Differentiable Physics

A Review of Differentiable Simulators

RHYS NEWBURY[1,2], JACK COLLINS[3], KERRY HE[1], JIAHE PAN[4], INGMAR POSNER[3], DAVID HOWARD[5], and AKANSEL COSGUN[6]

[1]Monash University, Australia
[2]Australian National University, Australia
[3]University of Oxford, United Kingdom
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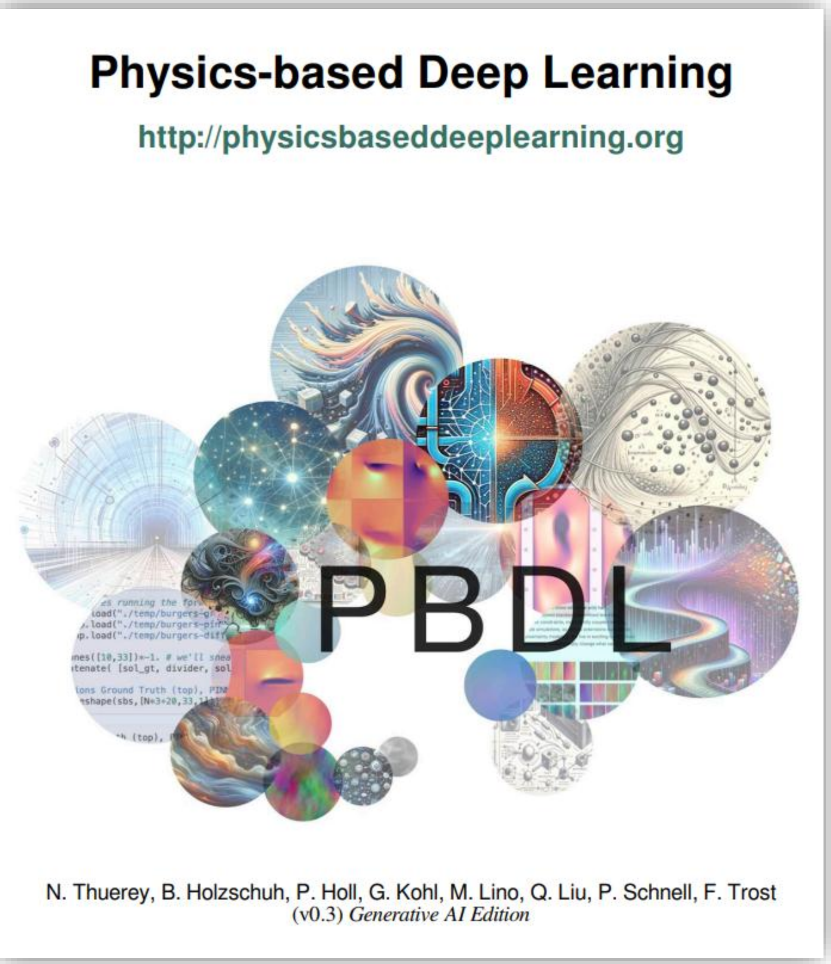
ABSTRACT Differentiable simulators continue to push the state of the art across a range of domains including computational physics, robotics, and machine learning. Their main value is the ability to compute gradients of physical processes, which allows differentiable simulators to be readily integrated into commonly employed gradient-based optimization schemes. To achieve this, a number of design decisions need to be considered representing trade-offs in versatility, computational speed, and accuracy of the gradients obtained. This paper presents an in-depth review of the evolving landscape of differentiable physics simulators. We introduce the foundations and core components of differentiable simulators alongside common design choices. This is followed by a practical guide and overview of open-source differentiable simulators that have been used across past research. Finally, we review and contextualize prominent applications of differentiable simulation. By offering a comprehensive review of the current state-of-the-art in differentiable simulation, this work aims to serve as a resource for researchers and practitioners looking to understand and integrate differentiable physics within their research. We conclude by highlighting current limitations as well as providing insights into future directions for the field.

INDEX TERMS Differentiable Simulator, Review, Differentiable Physics, Soft Body Simulation, System Identification, Trajectory Optimization, Morphology Optimization, Policy Optimization, Robotics



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https://physicsbaseddeeplearning.org/intro.html

Transfer Matrix Method and Interferometry References

Research Letter

Evaluation of the Thickness in Nanolayers Using the Transfer Matrix Method for Modeling the Spectral Reflectivity

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The reflectivity spectra have been traditionally used to determine the thicknesses in semiconductor films. However, thicknesses of nanofilms are not easy to evaluate because the interference fringes are not visible in the transparent region. In this paper, we present a computed method based on the transfer matrix (TM) which is used to match the calculated and experimental room temperature reflectivity spectra of the ZnTe/GaAs films and to determine its thickness film values afterwards. The TM method needs only to know refraction indices and absorption coefficients as a function of wavelength for the film and the substrate. The thickness nanofilms evaluated by our method are in agreement with the values measured by ellipsometry, Rutherford backscattering spectroscopy and transmission electron microscopy techniques. The present procedure extends the application of the standard spectral reflectance technique to determine semiconductor nanolayer thicknesses.

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Transfer-matrix formalism for the calculation of optical response in multilayer systems: from coherent to incoherent interference

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Abstract: We present a novel way to account for partially coherent interference in multilayer systems via the transfer-matrix method. The novel feature is that there is no need to use modified Fresnel coefficients or the square of their amplitudes to work in the incoherent limit. The transition from coherent to incoherent interference is achieved by introducing a random phase of increasing intensity in the propagating media. This random phase can simulate the effect of defects or impurities. This method provides a general way of dealing with optical multilayer systems, in which coherent and incoherent interference are treated on equal footing.

https://doi.org/10.1364/OE.18.024715

Transfer Matrix Method and iCVD References

IMPROVED FORMULATION OF SCATTERING MATRICES FOR SEMI-ANALYTICAL METHODS THAT IS CONSISTENT WITH CONVENTION

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Abstract—The literature describing scattering matrices for semi-analytical methods almost exclusively contains inefficient formulations and formulations that deviate from long-standing convention in terms of how the scattering parameters are defined. This paper presents a novel and highly improved formulation of scattering matrices that is consistent with convention, more efficient to implement, and more versatile than what has been otherwise presented in the literature. Semi-analytical methods represent a device as a stack of layers that are uniform in the longitudinal direction. Scattering matrices are calculated for each layer and are combined into a single overall scattering matrix that describes propagation through the entire device. Free space gaps with zero thickness are inserted between the layers and the scattering matrices are made to relate fields which exist outside of the layers, but directly on their boundaries. This framework produces symmetric scattering matrices so only two parameters need to be calculated and stored instead of four. It also enables the scattering matrices to be arbitrarily interchanged and reused to describe longitudinally periodic devices more efficiently. Numerical results are presented that show speed and efficiency can be increased by more than an order of magnitude using the improved formulation.

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Nanoscale gradient copolymer films via single-step deposition from the vapor phase

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Organic gradient materials are an inherent part of many functional structures in the natural world. Synthetic organic materials, like polymers, are thus the perfect choice to artificially recreate these structures for functional purposes. This work reports on new high-quality gradient copolymer films via large-area deposition from the vapor phase and circumvents thus problems related to current wet chemistry approaches. It enables furthermore for the first time the transfer of this gradient approach to the nanoscale, introducing a new class of organic gradient nanomaterials. This facilitates completely new organic gradient functionality on the nanoscale, not achievable with materials currently in use. Fully functional gradient films of 21 nm have been synthesized, which open up new pathways for many application fields. Their versatility is demonstrated by some application examples ranging from every day life (adhesion of PTFE in frypans) to advanced subwavelength devices on large-area substrates as well as complex geometries.

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