Adjoint Electron Transport for Computational Modeling in EPMA

Tamme Claus[a,*], Gaurav Achuda[b], Silvia Richter[b] and Manuel Torrilhon[a]

[*] claus@acom.rwth-aachen.de

[a] RWTH Aachen University, Applied and Computational Mathematics (ACoM)

[b] RWTH Aachen University, Gemeinschaftslabor für Elektronenmikroskopie (GFE)

Electron Probe Microanalysis(EPMA): Quantification of solid material samples based on measured intensities of characteristic x-radiation induced by focussed beams of electrons.

Vision: Efficient Computational Model for EPMA

Modeling Approach:

- given a model $\mathcal{M}(\cdot)$ that maps a beam $\mathcal{B} \in \{\mathcal{B}\}$ to an x-ray intensity $\mathcal{M}(\mathcal{B})$
- loop the model $\mathcal{M}(\cdot)$ over all beam positions → becomes expensive

$$\begin{pmatrix} \\ \\ \\ \end{pmatrix} = \begin{pmatrix} \mathcal{M}(\mathcal{B}) & \mathcal{M}(\mathcal{B}) & \dots \\ \mathcal{M}(\mathcal{B}) & \dots \end{pmatrix}$$

($70 \times 70 \times 198\text{s} \approx 11\text{days}5\text{h}$)

If the model $\mathcal{M}(\cdot)$ is *linear (and bounded)* w.r.t. the beam $\mathcal{B}$, then
→ Riesz Representation: *there is a* $\mathcal{A}$ *such that* $\mathcal{M}(\mathcal{B}) = \langle \mathcal{A}, \mathcal{B} \rangle_{\{\mathcal{B}\}}$.

$$\begin{pmatrix} \\ \\ \\ \end{pmatrix} = \begin{pmatrix} \langle \mathcal{A}, \mathcal{B} \rangle & \langle \mathcal{A}, \mathcal{B} \rangle & \dots \\ \langle \mathcal{A}, \mathcal{B} \rangle & \dots \end{pmatrix}$$

($3 \times 198\text{s} \approx 10\text{min}$)

Adjoint Modeling Approach:

- compute representation $\mathcal{A} = \mathcal{M}^T(\blacksquare)$ [→ cost comparable to $\mathcal{M}(\mathcal{B})$]
- loop $\langle \mathcal{A}, \cdot \rangle_{\{\mathcal{B}\}}$ over all beam positions → much cheaper, same result

An Illustrative Example from Linear Kinetic Theory

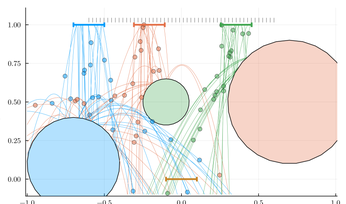
Independent particles accelerate under constant gravity F and undergo inelastic collisions with fixed background obstacles. Multiple sources $g^{(i)}$ are placed above the obstacles and a detector h below.

Non-Adjoint Model:

$v\partial_x f^{(i)} + F\partial_v f^{(i)} = g^{(i)}$

BC: no inflow + inel. refl.

$y^{(i)} = \int_{X\times V} f^{(i)} h \,\mathrm{d}x\,\mathrm{d}v$

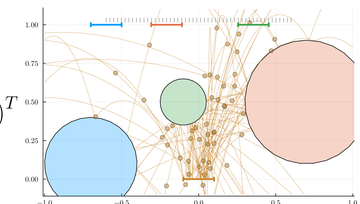


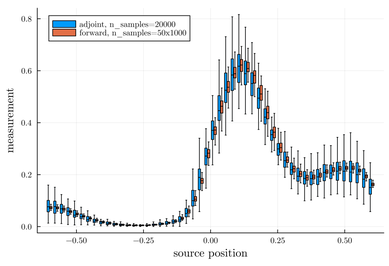
Adjoint Model:

$-v\partial_x \phi - F\partial_v \phi = h$

BC: no outflow + (inel. refl.)^T

$y^{(i)} = \int_{X\times V} \phi g^{(i)} \,\mathrm{d}x\,\mathrm{d}v$





- we use Monte Carlo to approximate the respective integrals
- sample sources $g^{(i)}$ → trace trajectory → integrate h over trajectory

↕ adjoint

- sample detector h → trace backwards → integrate $g^{(i)}$ over trajectory
- application requires uncertainty quantification (variance reduction)

A Model for Electron and Photon Transport

Linear Transport Equation: describes electron and photon transport in the sample $\mathcal{R}$

$$\Omega \cdot \nabla\psi + (\Sigma^{\text{tot}} + \Sigma^{\text{abs}})\psi = \int_{\mathbb{R}^+}\int_{S^2} \Sigma\psi \,\mathrm{d}\Omega'\,\mathrm{d}\epsilon' + q$$

(fluence $\psi(x,\epsilon,\Omega) = |v|f$, velocity $v(\epsilon)$, density $f(x,\epsilon,\Omega)$, direction $\Omega \in S^2$, energy $\epsilon \in \mathbb{R}^+$, position $x \in \mathcal{R} \subset \mathbb{R}^3$, macrosc. diff. scat. cross-section $\Sigma(x,\epsilon,\epsilon',\Omega\cdot\Omega')$, total macrosc. scat. cross-section $\Sigma^{\text{tot}} = \int_{\mathbb{R}^+}\int_{S^2}\Sigma(x,\epsilon',\epsilon,\Omega'\cdot\Omega)\,\mathrm{d}\Omega'\,\mathrm{d}\epsilon'$, source q)

Beam Model: source of e^--fluence on the sample surface $\mathcal{S}$

$q^{\text{e}^-} = \psi_{\text{beam}} \quad \forall x \in \mathcal{S}, n\cdot\Omega \le 0, \epsilon \in \mathbb{R}^+$ (sample surface $\mathcal{S} \subset \partial\mathcal{R}$, surface normal n)

X-Ray Source: source of x-ray-fluence in the sample $\mathcal{R}$ depends on e^--fluence

$$q^{\text{x-ray}} = \frac{1}{4\pi}\int_{\mathbb{R}^+}\int_{S^2} \Sigma^{\text{x-ray}}\psi^{\text{e}}\,\mathrm{d}\Omega\,\mathrm{d}\epsilon \quad \forall x \in \mathcal{R}, \Omega \in S^2$$ (macrosc. x-ray emission cross-section $\Sigma^{\text{x-ray}}(\epsilon)$)

Detector Intensity: extracts from the x-ray-fluence on the sample surface $\mathcal{S}$

$$y \propto \int_{\mathcal{S}} n\cdot\Omega\,\psi^{\text{p}}|_{\Omega=\Omega_{\text{takeoff}}}\,\mathrm{d}\Gamma$$ (Ω_{takeoff} detector takeoff direction)

Additivity Approximation: macrosc./microsc. cross-sections for compounds follow

$$\Sigma(x,\cdot) = \sum_{e=1}^{n_e} \mathcal{N}_e(x)\sigma_e(\cdot)$$ (# of compounds n_e, particle density $\mathcal{N}_e(\cdot)$, microscopic cross-section σ_e)

→ **spatial variability** of the background particle density $\mathcal{N}_e$
→ electron and x-ray transport/scattering/absorption happens at different **spatial scales**.
→ requires **coupling** of efficient numerical methods for RT.

P_N Moment Discretization and Numerical Solution

Approximation: *spherical harmonics*(Ω), *finite elements*(x) and *energy stepping*(ϵ):

$$\psi(x,\epsilon_m,\Omega) \approx \underbrace{\begin{pmatrix} \\ \\ \\ \\ \\ \end{pmatrix}^T \cdot \underbrace{\Psi_m^+}_{\in\mathbb{R}^{n_x^+\times n_\Omega^+}} \cdot \begin{pmatrix} \\ \\ \\ \\ \\ \end{pmatrix}}_{\text{even parity } \psi(-\Omega)=\psi(\Omega)} + \underbrace{\begin{pmatrix} \\ \\ \\ \\ \\ \end{pmatrix}^T \cdot \underbrace{\Psi_m^-}_{\in\mathbb{R}^{n_x^-\times n_\Omega^-}} \cdot \begin{pmatrix} \\ \\ \\ \\ \\ \end{pmatrix}}_{\text{odd parity } \psi(-\Omega)=-\psi(\Omega)}$$

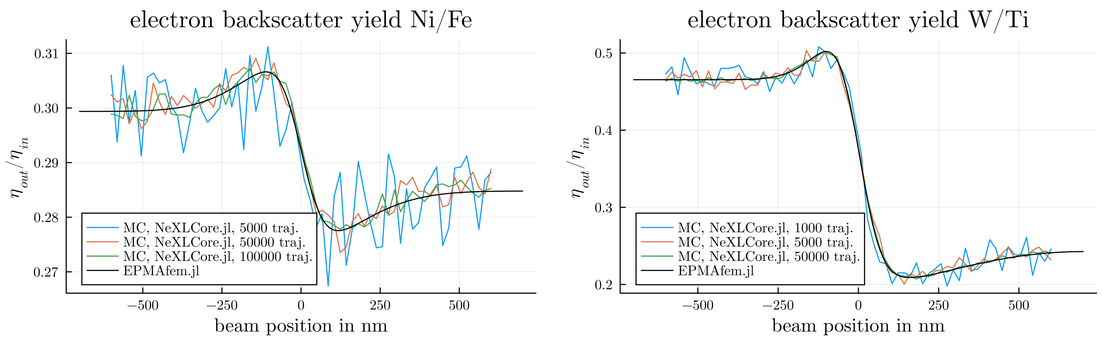
→ weak-form using a mixed variational framework based on e/o-parity splitting of ψ
→ large linear system $\mathcal{A}$ for $\Psi = \{\Psi_m^+, \Psi_m^-\}_{m=1,\dots}$ per beam $g^{(i)}$ or per k-ratio line $h^{(j)}$
→ $\mathcal{A}$ has tensor product structure $\mathcal{A}\cdot\Psi = \sum(\mathcal{X}\otimes\mathcal{W})\cdot\text{vec}(\Psi)$

$\mathcal{A}\cdot\Psi^{(i)} = g^{(i)};\ \mathcal{Y}^{(ji)} = h^{(j)}\cdot\psi^{(i)}$ **adjoint** $\mathcal{A}^T\cdot\Phi^{(j)} = h^{(j)};\ \mathcal{Y}^{(ji)} = \Phi^{(j)}\cdot g^{(i)}$

→ implementation: iterative linear solver, GPU acceleration, in Julia

Comparison P_N and Monte Carlo: Backscatter Coefficient

We compare the e^- backs. coeff. of two samples with a **vertical interface** at 0nm



- **excellent agreement** between P_N (EPMAfem.jl) and Monte Carlo (NeXLCore.jl)
- using identical physical parameters; Monte Carlo shows **statistical noise**
- non-adjoint Monte Carlo code *iterates over beam positions*
- adjoint P_N code *requires one linear solve for all beam positions*

Abstract Adjoint Method in a Computational Context

Abstract concept leveraged e.g., by algorithmic differentiation (backpropagation).

- Given vectors $g^{(i)} \in G, i = 1,\dots I$ and $h^{(j)} \in H, j = 1,\dots J$ in Hilbert spaces G, H
- and $A : G \to H$ a linear, bounded operator.

By *Riesz Representation Theorem* there is the equivalence

$$\mathcal{Y}^{(ji)} = \langle h^{(j)}, A(g^{(i)})\rangle_H = \langle A^*(h^{(j)}), g^{(i)}\rangle_G,$$

which enables **two** approaches to implement the computation of the matrix $\mathcal{Y}^{(ji)} \in \mathbb{R}^{J\times I}$:

Non-Adjoint Implementation:

for $i \leftarrow 1 : I$ **do**
  $v \leftarrow A(g^{(i)})$
  for $j \leftarrow 1 : J$ **do**
    $\mathcal{Y}^{(ji)} \leftarrow \langle h^{(j)}, v\rangle_H$
  end for
end for

Adjoint Implementation:

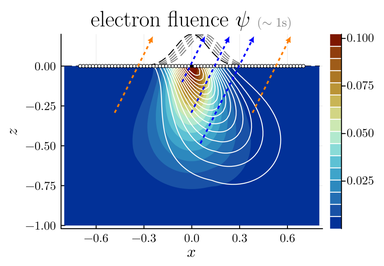
for $j \leftarrow 1 : J$ **do**
  $\lambda \leftarrow A^*(h^{(j)})$
  for $i \leftarrow 1 : I$ **do**
    $\mathcal{Y}^{(ji)} \leftarrow \langle \lambda, g^{(i)}\rangle_G$
  end for
end for

Both approaches have different computational cost:

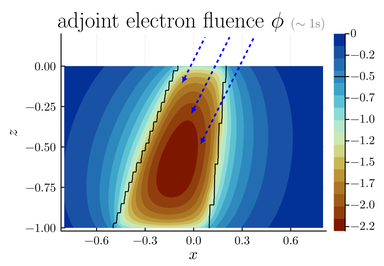
$I \times \mathcal{C}(A(\cdot)) + IJ \times \mathcal{C}(\langle\cdot,\cdot\rangle_H)$ $\qquad$ $J \times \mathcal{C}(A^*(\cdot)) + IJ \times \mathcal{C}(\langle\cdot,\cdot\rangle_G)$

Adjoint Electron Transport in EPMA

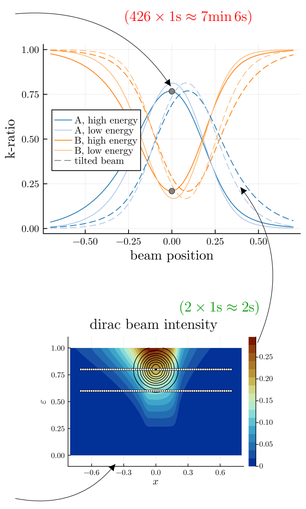
There are **two** different approaches to implement k-ratio line scans:



- source: e^--beam
- k-ratios: integration with x-ray emission and absorption
- a single solution $\psi^{(i)}$ yields **all** k-ratios for single e^--beam



- source: x-ray emission and absorption
- k-ratios: integration with the e^--beam
- a single solution $\phi^{(j)}$ yields k-ratios(A) for **all** e^--beams



Limitations: Effective adjoint transport requires **overlapping interaction volumes**. Only near the beam $\psi \neq 0$ allowing for reduction of the computational domain. However, $\phi \neq 0$ eventually throughout $\mathcal{R}$.

Outlook: Inverse Modeling using Numerical Optimization

Adjoint Derivatives: the computation of a Jacobian $Jf \in \mathbb{R}^{n\times m}$ can be interpreted as

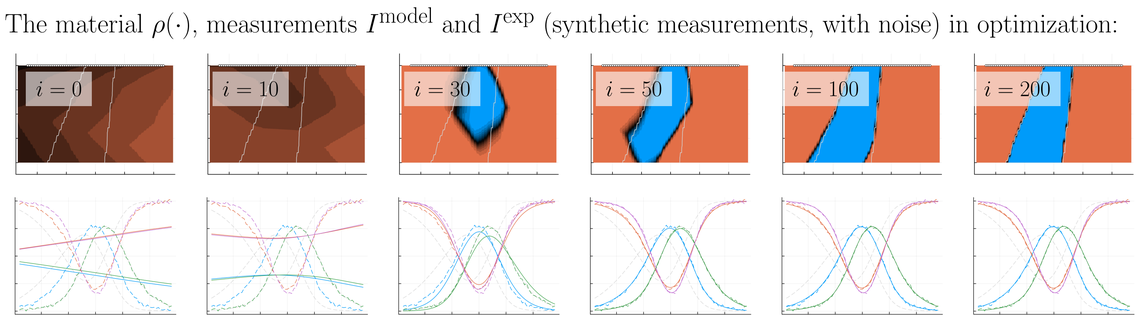
$$(Jf)_{ij} = \langle e^{(i)}, \frac{\partial f}{\partial x}[e^{(j)}]\rangle = \langle \frac{\partial f}{\partial x}^T[e^{(i)}], e^{(j)}\rangle$$

→ adjoint methods can be used for efficient implementations for:

Material Reconstruction: $\rho^*(\cdot) = \arg\min_{\rho(\cdot)} ||I^{\text{model}}[\rho(\cdot)] - I^{\text{exp}}||^2$

→ implement *gradient-based* optimization based on a computational model for EPMA

The material $\rho(\cdot)$, measurements I^{model} and I^{exp} (synthetic measurements, with noise) in optimization:



→ adjoint methods reduce the runtime of this example from multiple years (without adjoint approaches) to 12days (adjoint gradient) to 40min (adjoint gradient & transport).

References

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[3] Griewank, A., & Walther, A. (2008). Evaluating Derivatives: Principles and Techniques of Algorithmic Differentiation, Second Edition. In Other Titles in Applied Mathematics. Society for Industrial and Applied Mathematics.
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→ code repository for reproducibility: github.com/tam724/EPMAfem.jl