# Material Imaging in Electron Probe Microanalysis

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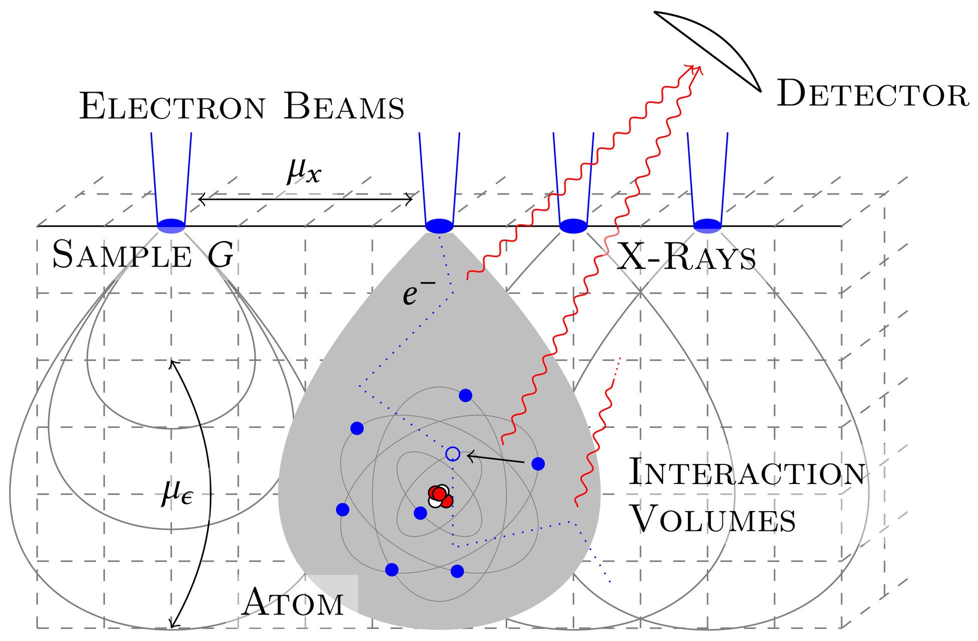
**Electron Probe Microanalysis (EPMA)[1]:** Quantification of solid materials based on **intensity** measurements of **characteristic x-radiation** induced by focussed beams of electrons.

**Inverse Problem of Material Reconstruction:**

$$\rho^*(x) = \underset{\rho(x)}{\arg\min}\, \mathrm{disc}(k^{\text{model}}[\rho(x)], k^{\text{exp.}})$$

*Find the material ($\rho(x)$: mass concentrations) such that a model ($k^{model}[\rho]$) reproduces the observations ($k^{exp.}$: k-ratios, normalized intensities).*

**Spatial Resolution[2]:** Currently limited by **k-ratio models, that assume homogeneity** inside the interaction volume.
Decrease of the interaction volume by physical means: → **less depth information**, → **decreased signal-to-noise ratio**
Employing availiable Monte Carlo models, that allow inhomogeneous materials: → hindered by their **statistical noise**.




**Our Goal:** Combine k-ratios from beams with **overlapping interation volumes**. Use **gradient based optimization** to solve the minimization problem. Employ a **deterministic k-ratio model**. Compute the gradient using **adjoint algorithmic differentiation**. Use different **material parametrizations** for regularization. → **Universal and efficient** reconstruction

## k-ratio Model based on $P_N$ Approximation of BCSD[3]

**X-Ray Generation and Attenuation:**

$$k_\alpha[\rho] = \frac{I_\alpha[\rho]}{I_\alpha[\rho_\alpha^{\text{std}}]} \quad \text{where} \quad I_\alpha[\rho] = \int_G \mathcal{A}_\alpha[\rho]\,\mathcal{N}_\alpha[\rho]\,\mathcal{I}_\alpha[\rho]\,\mathrm{d}x$$

$$\bar{\mathcal{A}}_\alpha = \frac{\mathcal{N}_\alpha \mathcal{I}_\alpha \bar{k}_\alpha}{I_\alpha^{\text{std}}} \quad \bar{\mathcal{N}}_\alpha = \frac{\mathcal{A}_\alpha \mathcal{I}_\alpha \bar{k}_\alpha}{I_\alpha^{\text{std}}} \quad \bar{\mathcal{I}}_\alpha = \frac{\mathcal{A}_\alpha \mathcal{N}_\alpha \bar{k}_\alpha}{I_\alpha^{\text{std}}} \quad \text{(adjoint/reverse operators)}$$

The fields describe: Attenuation $\mathcal{A}$, Number of atoms $\mathcal{N}$, Ionization distribution $\mathcal{I}$.

$$\mathcal{A}_\alpha(x) = \exp\Big(-\int_{l(x,x_d)} \sum_{i=1}^{n_e} \tau_{\alpha,i}\rho_i(y)\,\mathrm{d}y\Big) \quad \mathcal{N}_\alpha(x) = \frac{\rho_i(x)|_{Z_i\in\alpha}}{A_\alpha} \quad \mathcal{I}_\alpha(x) = \int_0^\infty \sigma_\alpha^{\text{ion}}(\epsilon)u_\alpha^{(0,0)}\,\mathrm{d}\epsilon$$

$$\bar{\rho}_i(x) = \tau_{i,\alpha}\int_{l(x,x_d^*)} -\mathcal{A}_\alpha\bar{\mathcal{A}}_\alpha\,\mathrm{d}y \quad \bar{\rho}_i(x)|_{Z_i\in\alpha} = \frac{\bar{\mathcal{N}}_\alpha(x)}{A_\alpha} \quad \bar{u}_\alpha^{l,k}(x,\epsilon) = \begin{cases}\sigma_\alpha^{\text{emiss}}(\epsilon)\bar{\mathcal{I}}_\alpha(x) & l,k=0\\ 0 & \text{else}\end{cases} \quad \text{(adjoint/reverse operators)}$$

**Electron Transport:** Evolution of $u_\alpha^{(l,k)} = \int_{\mathbb{S}^2}|v(\epsilon)|f_\alpha Y_l^k\,\mathrm{d}\Omega$ is described by the **spherical harmonics** ($P_N$: $\{Y_l^k\}_{l\le N,|k|\le l}$) moment expansion of the **linear Boltzmann equation in continuous-slowing down (BCSD) approx.**

$$F_\alpha(u_\alpha,\rho) = -\partial_\epsilon(S[\rho]u_\alpha) + \sum_{d=1}^{3} A_{(d)}\partial_{x_d}u_\alpha - Q[\rho]u_\alpha = 0$$

$$S[\rho]\partial_\epsilon\bar{F}_\alpha - \sum_{d=1}^{3} A_{(d)}\partial_{x_d}\bar{F}_\alpha - Q[\rho]\bar{F}_\alpha = \bar{u}_\alpha \quad \text{and} \quad \bar{\rho}_i(x) = \int_E \bar{F}_\alpha^T(-\partial_\epsilon(S_i u_\alpha) - Q_i u_\alpha)\,\mathrm{d}\epsilon \quad \text{(adjoint equations)}$$

**Additivity approximation** for scattering-cross-sections → material coefficients $S, Q$ and transport matrices $A_{(d)}$ follow from CSD-scattering cross sections and the $P_N$-expansion

$$\{S,Q\}[\rho] = \sum_{i=1}^{n_e}\rho_i(x)\{S,Q\}_i(\epsilon) \quad A_{(d)}^{(l,k),(l',k')} = \int_{\mathbb{S}^2}\Omega_d Y_l^k(\Omega)Y_{l'}^{k'}(\Omega)\,\mathrm{d}\Omega$$

**Electron beam** described by energy-stable and characteristic **boundary conditions**

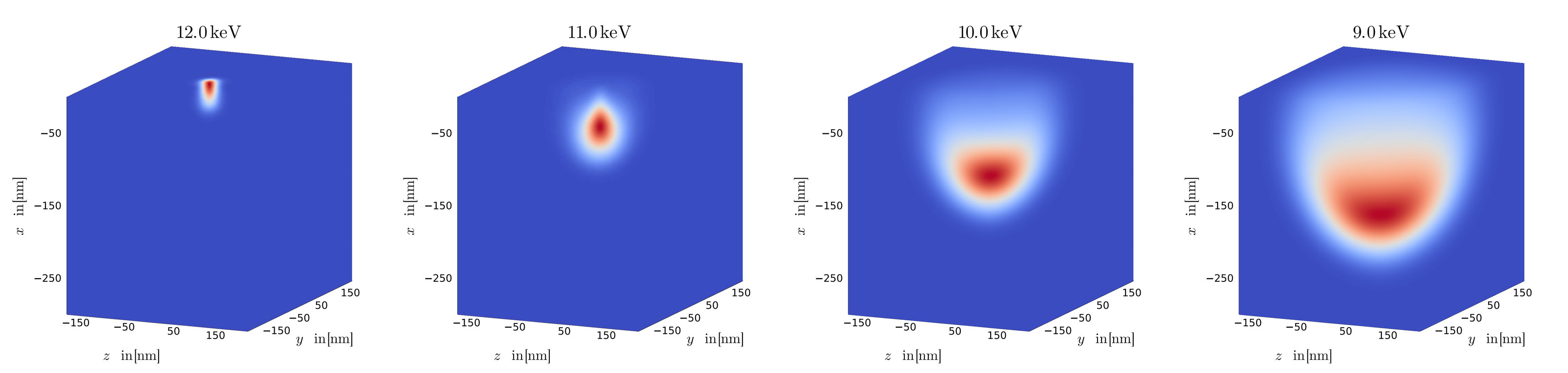
$$u_\alpha^{\{(l,k)\text{ odd}\}} = L_{(d)}\hat{A}_{(d)}u_\alpha^{\{(l,k)\text{ even}\}} + \int_{n\cdot\Omega<0}|v|f_\alpha^{\text{beam}}\,Y_l^k\big|_{(l,k)\text{ odd}}\,\mathrm{d}\Omega$$

$$\bar{F}_\alpha^{\{(l,k)\text{ odd}\}} = -L_{(d)}\hat{A}_{(d)}\bar{F}_\alpha^{\{(l,k)\text{ even}\}} \quad \text{(adjoint/reverse BC)}$$

**Structure/Sparsity** of $F_\alpha = 0$ motivates the solving using a **finite-difference staggered-grid method** (StaRMAP).

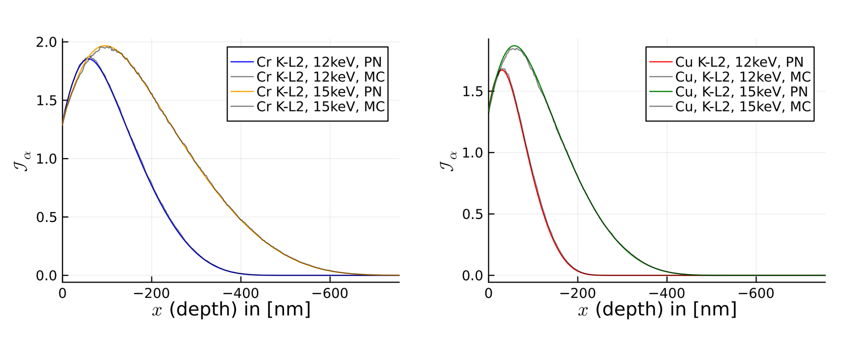
$\alpha$ multiindex (beam setup, x-ray line), $\tau_{\alpha,i}$ mass attenuation coefficient, $x_d$ detector position, $x_d^*$ reflection of $x_d$ about $x$, $A_\alpha$ atomic weight, $\sigma_\alpha^{\text{ion}}$ ionization cross section, $v(\epsilon)$ electron velocity, $f_\alpha$ electron number density, $f_\alpha^{\text{beam}}$ number density of beam electrons, $S_i$ stopping power, $Q_i$ transport coefficient

## 3D Electron Fluence in $Cu$ and $Ni$




The $u_\alpha^{(0,0)}$ moment of $|v|f_\alpha$ computed using $P_9$ in a material consisting of Copper and Nickel. 50x50x50 spatial discreatization. Beam energy $12 \pm 0.3keV$.

## Ionization Distribution $P_N$ vs $MC$ (Monte Carlo)




- **Good agreement** of ionization curves $\mathcal{I}_\alpha(x)$ computed using our method and the Monte Carlo code `NeXLCore.jl`.
- Both codes use the same physical parameters (stopping power/scattering cross section).
- Computed using $P_{21}$. 500 spatial discretization. (MC: number of electrons: 60000)
- MC results show typical **statistical noise**.

## Inverse Problem of Material Reconstruction

Reconstructing the infinite-dimensional $\rho(x)$ from the finite-dimensional $\{k_\alpha\}_\alpha$ is **ill-posed**. We focus on the **maximum likelihood/maximum posterior estimate** under the assumption of **gaussian noise** → squared error.
**Regularization** using a certain **parametrization** $\rho(x;p)$ ($p \in \mathbb{R}^n$: parameters) and **prior information** $\mathcal{R}(\rho(x;p))$.

**PDE-constrained minimization problem**

$$p^* = \underset{p\in\mathcal{P}}{\arg\min}\sum_\alpha (k^{\text{model}}[\rho(x;p),u] - k^{\text{exp}})^2 + \mathcal{R}[\rho(x;p)]$$
$$s.t.\ F_\alpha(u_\alpha,\rho(x;p)) = 0 \quad \forall\alpha$$

## Differentiation Framework based on AD[4]

**Algorithmic Differentiation (AD):** rules for **modular** and **efficient** derivative computation

- *adjoint/reverse mode AD* facilitates **efficient** $O(1)$ gradient computation
- *chain rule for function composition* enables **modularity** and **encapsulation** of code

**Fundamental Modes of AD:**

- *tangent/forward mode* derived from the **directional derivative**

$$\dot{y} = \lim_{h\to 0}\frac{f(x+h\dot{x}) - f(x)}{h} = \frac{\partial f}{\partial x}[\dot{x}]$$

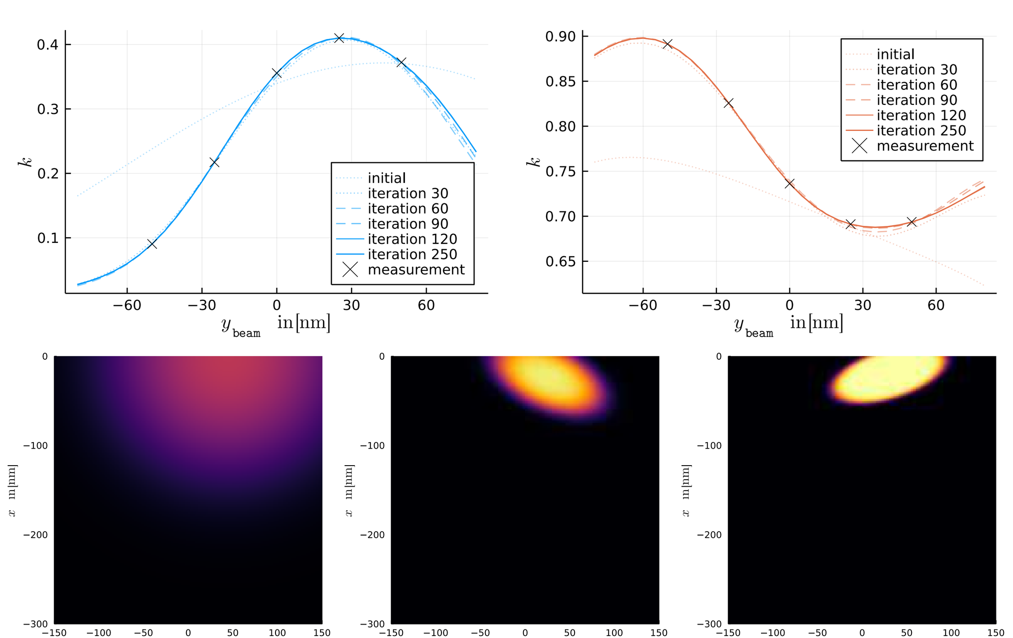
- *adjoint/reverse mode* derived from a scalar product identity

$$\langle\bar{y},\dot{y}\rangle = \langle\bar{y},\frac{\partial f}{\partial x}[\dot{x}]\rangle = \langle\frac{\partial f^*}{\partial x}[\bar{y}],\dot{x}\rangle = \langle\bar{x},\dot{x}\rangle \quad \forall\dot{x} \quad \text{(with proper definition of } \langle\cdot,\cdot\rangle)$$

**Explicit vs. Implicit AD:**

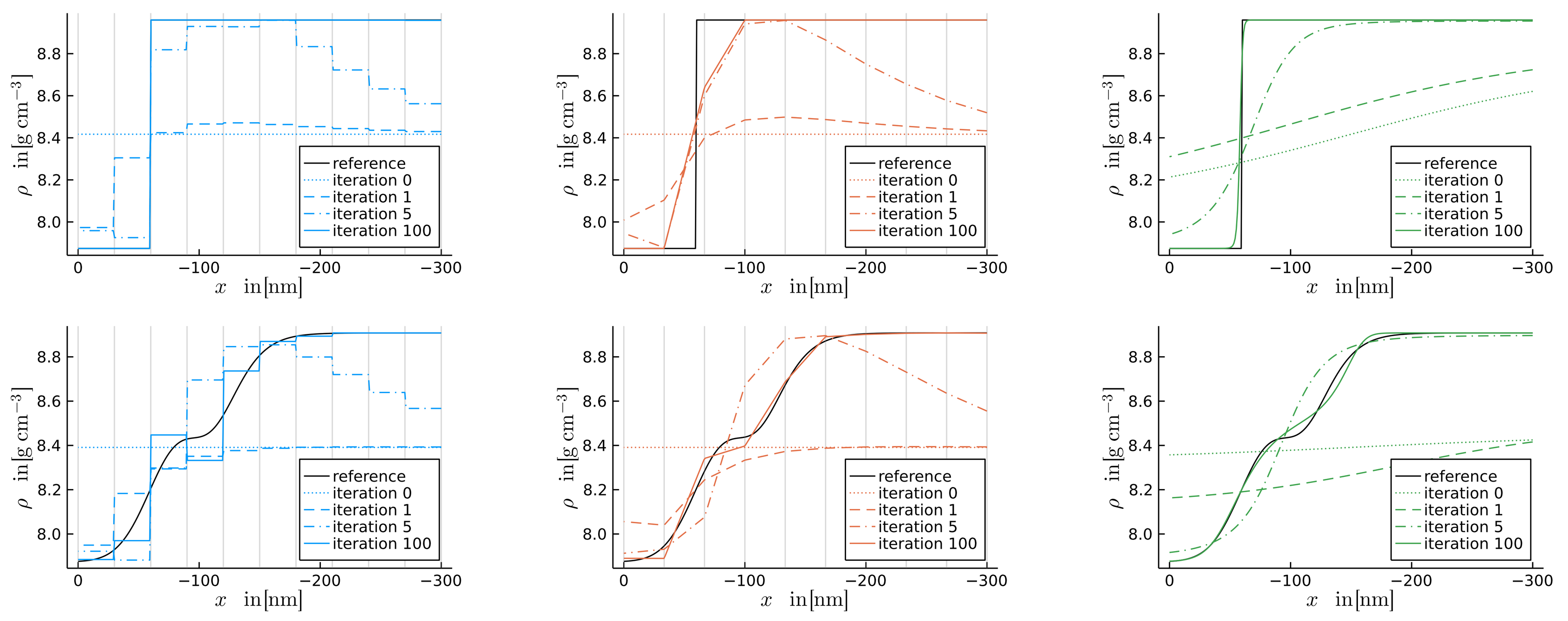
| Primal | Tangent mode | Adjoint mode |
|---|---|---|
| $y = f(x)$ | $\dot{y} = \frac{\partial f}{\partial x}[\dot{x}]$ | $\bar{x} = \frac{\partial f^*}{\partial x}[\bar{y}]$ |
| $F(y,x) = 0$ | $\dot{F} = \frac{\partial F}{\partial x}[\dot{x}]$ and $\dot{y} = -\frac{\partial F}{\partial y}[\dot{F}]$ | $\frac{\partial F^*}{\partial y}[\bar{F}] = \bar{y}$ and $\bar{x} = -\frac{\partial F^*}{\partial x}[\bar{F}]$ |

## Reconstruction of an Ellipsoidal Inclusion in 2D




- k-ratio profile of $(Cu, K-L_2)$ and $(Fe, K-L2)$ during L-BFGS iterations.
- Black crosses: measurements considered in the objective
- k-ratio profiles almost agree **quickly**
- Parameters: $(x, y \in \mathbb{R})$ position of ellipse, $(a, b \in \mathbb{R})$ principal axis, $(r \in \mathbb{R})$ angle
- Initial guess (left), iteration 60 (middle) and 250 (right) of the total density.
- With proper parametrization, **reconstruction is possible** given limited data

## Layer Reconstruction Using Different Parametrizations




Comparison of a *piecewise-constant* (left), a *bilinear*(middle) and a *non-linear*(right) parametrization for the 1D reconstruction of a *sharp*(upper) and a *diffusive*(lower) material interface between an $Fe$-layer on $Ni$-substrate ($x$ is depth). Beam energies: 9, 10.5, 12, 13.5, $15keV$. $P_9$.

**References**

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[4] Hiser J., Naumann U. and Herty M. (2022) *Discrete tangent and adjoint sensitivity analysis for discontinuous solutions of hyperbolic conservation laws*. PhD-Thesis (RWTH Aachen University).

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