# Application of the Adjoint Method in gradient-based Optimization to the $\mathcal{M}1$-Model in EPMA

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## Forward Model: a k-ratio model based on the $\mathcal{M}1$-Model

The spatial resolution in electron probe microanalysis (EPMA) is currently limited to a pixel size approximately as large as the interaction volume of the electrons.
*Aim:* **increase the spatial resolution** of EPMA to a pixel size **smaller than the interaction volume**

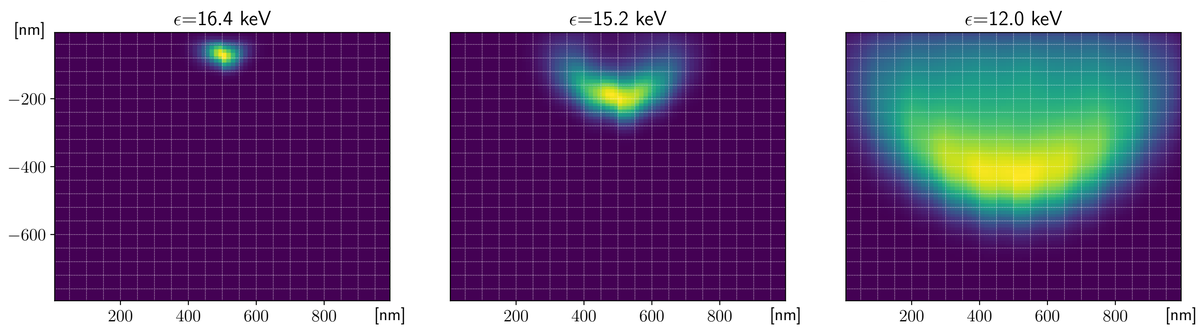
*How:* using **more sophisticated reconstruction methods** based on a model which precisely simulates electron transport and x-ray generation inside the material to resolve small concentration pixel

### $\mathcal{M}1$-Model to describe the electron fluence $\psi^0$

- deterministic transport/collision model of the electron fluence $\psi^0$ at energy $\epsilon$ and position $\bar{x}$ in a material domain $Q$ described by its mass concentration field $c(\bar{x})$
- first order moment approximation to the Boltzmann equation for electron transport in continuous slowing down approximation [1]

$$\underbrace{-\partial_\epsilon \left( S(c(\bar{x}),\epsilon) \begin{pmatrix} \psi^0(\bar{x},\epsilon) \\ \psi^1(\bar{x},\epsilon) \end{pmatrix} \right) + \nabla_x \begin{pmatrix} \psi^1(\bar{x},\epsilon) \\ \psi^2_{ME}(\psi^0,\psi^1) \end{pmatrix} + \begin{pmatrix} 0 & 0 \\ 0 & T(c(\bar{x}),\epsilon) I \end{pmatrix} \begin{pmatrix} \psi^0(\bar{x},\epsilon) \\ \psi^1(\bar{x},\epsilon) \end{pmatrix}}_{:=G(\psi(\bar{x},\epsilon),c(\bar{x}))} = 0$$

with: stopping power $S$, transport coefficient $T$, minimum entropy closure $\psi^2_{ME}$
**Solution** of the hyperbolic pde using the finite volume library CLAWPACK [2]



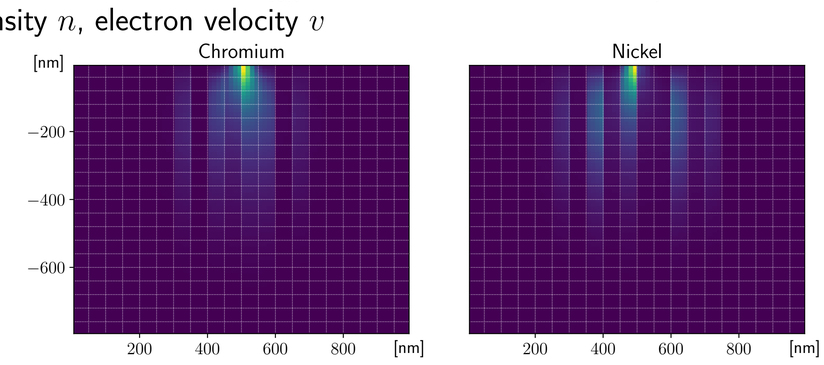

*Electron fluence $\psi^0$ for a material of chromium, nickel and iron (electron beam: 17keV at $x$=500nm)*

### k-ratio model

The k-ratio $k^{i,j}(c)$ can be computed as ($i$ refers to an element, $j$ to one of its characteristic k-ratios)

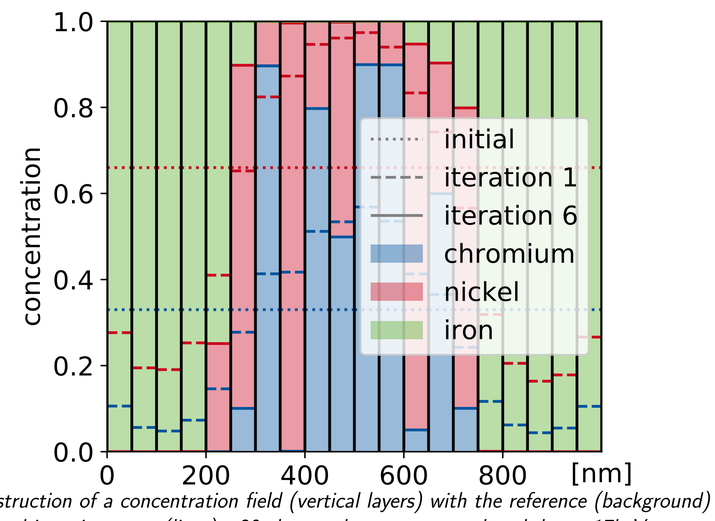
$$k^{i,j}(c) = \frac{1}{I^{i,j}_{std}} \int_Q e^{-\int_{d(\bar{x})} \mu^{i,j}(c(\bar{y}))\,\mathrm{d}\bar{y}} \int_{\epsilon_{cutoff}}^{\epsilon_{initial}} \omega^i_K \sigma^{i,j}_{ion}(\epsilon) N^i_V(c(\bar{x})) \underbrace{\int_{S^2} ||v(\epsilon)|| n(\bar{x}, v(\epsilon,\Omega))\,\mathrm{d}\Omega}_{=\psi^0(x,\epsilon)} \mathrm{d}\epsilon\,\mathrm{d}\bar{x}$$

with: standard intensity $I^{i,j}_{std}$, attenuation $e^{-\int \cdot\,\mathrm{d}\bar{y}}$, fluorescence yield $\omega^i_K$, ionization cross section $\sigma^{i,j}_{ion}$, number density of atoms $N^i_V$, electron number density $n$, electron velocity $v$




*Intensity of generated x-rays of chromium and nickel (including attenuation)*

## Inverse Problem: reconstruct concentrations from k-ratios




*Reconstruction of a concentration field (vertical layers) with the reference (background) and iteration steps (lines) - 20 electron beams: centered each layer, 17keV - Levenberg–Marquardt algorithm*

*Goal:* given **experimental** k-ratios $\tilde{k}^{i,j}$ and a **model** $k^{i,j}(c)$ we search for the concentration $c^*$ which **minimizes their squared error**

$$c^* = \arg\min_c \underbrace{\frac{1}{2} \sum (k^{i,j}(c) - \tilde{k}^{i,j})^2}_{:=J(c)}$$

### Gradient-based Optimization

$$c^{k+1} = c^k + f(c^k, \nabla_c J(c^k)) \quad \text{with initial } c^0$$

- iterative schemes based on the gradient of the objective function
- Levenberg–Marquardt algorithm (exploits least squares structure)

Gradient via **Finite Differences**

$$\frac{\partial J(c)}{\partial c_m} = \frac{J(c + \delta c_m e_m) - J(c)}{\delta c_m} \text{ (forward FD)}$$

→ one pde solve per parameter $c_m$

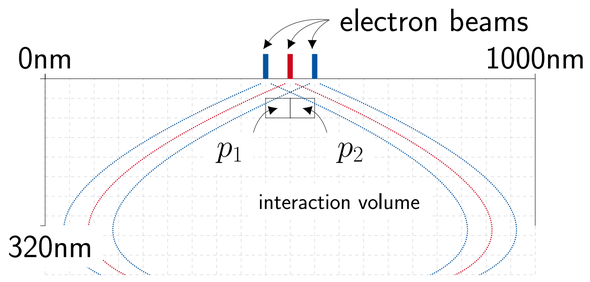
Gradient via **Adjoint State Method** [4]

$$G(\psi, c) = 0 \qquad \text{(forward model)}$$
$$\frac{\partial J}{\partial \psi} - \left(\frac{\partial G}{\partial \psi}\right)^* \lambda = 0 \qquad \text{(adjoint equation)}$$
$$\nabla_c J(c) = \nabla_c h(\psi, c) - \langle \lambda,\ \nabla_c G(\psi, c) \rangle$$

with: the adjoint operator $(\cdot)^*$ and $J(c) = h(\psi, c)$

→ two pde solves per gradient

## Investigation of the objective function (Example: variation of the concentration in two cells)




*Sketch of experimental setup (material grid, beam positions, variable cells $p_{\{1,2\}}$, interaction volume)*

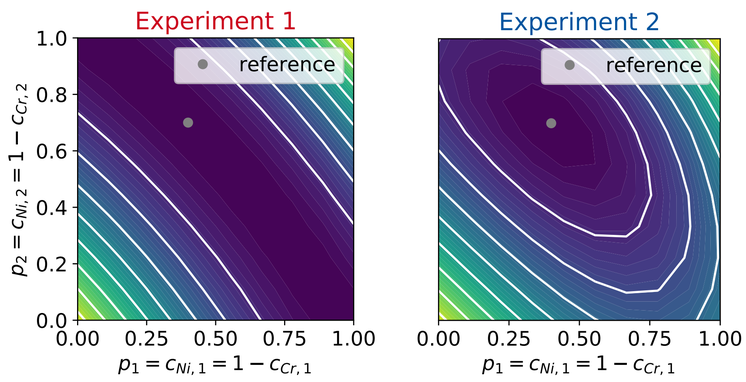
*Assume:* only $c_{Ni} = p_{\{1,2\}}$ and $c_{Cr} = 1 - p_{\{1,2\}}$ are variable

*Result:* Experiment 1 (one beam position): insufficient information to reconstruct $p_{\{1,2\}}$

Experiment 2 (two beam positions): unique minimum for $p_{\{1,2\}}$

*Also:* multiple beam energies for depth information [3]

*Further investigation:* Definiteness and eigenvalues/vectors of the hessian $\nabla^2_c J(c)$ at the minimum




*The objective function $J(p_1, p_2)$ for experiment 1 & 2 with variable $p_1$ and $p_2$*

## Next steps

- Investigation of the objective function and inverse problem with noisy measurements
- Uncertainty quantification for reconstructed parameters (e.g. confidence interval)

## References

[1] Bünger J, Richter S and Torrilhon M 2018 A deterministic model of electron transport for electron probe microanalysis. *IOP Conf. Ser.: Mater. Sci. Engng.* **304** 012004

[2] LeVeque R J 2002 *Finite volume methods for hyperbolic problems. Cambridge Texts in Applied Mathematics.* (Cambridge: Cambridge University Press)

[3] Claus T 2018 *Application of the adjoint method in gradient-based optimization to the M1-model in electron beam microanalysis.* BSc-thesis (Aachen, Germany: RWTH Aachen)

[4] Plessix R E 2006 A review of the adjoint-state method for computing the gradient of a functional with geophysical applications. *Geophys. J. Int.* **167** 495-503