Machine Learning for Analytical Transmission Electron Microscopy
Analytical scanning transmission electron microscopy

- Energy dispersive X-ray spectroscopy (EDXS)
  - Chemical composition
  - Heavy elements

- Electron energy-loss spectroscopy (EELS)
  - Chemical composition
  - Charge and coordination
  - Plasmons
  - Light elements

From 0.1 nm to a few μm → No homogeneity
The mixing problem

Pseudo HAADF

Transmission → Mixed

Scan → Noisy, redundant

→ How to retrieve true values?

Pure spectra

Abundances

1 pixel spectrum
Current unmixing algorithm for STEM-EDXS

Non-negative matrix factorization (Renormalized)

- Incomplete unmixing
- Remaining noise
- Missing physics

Angles:
- P0 : 7.8°
- P1 : 15.0°
- P2 : 9.1°
The MLATEM project

Initiated by Cécile Hébert and Guillaume Obozinski

Funded by the Swiss Data Science Center

Aim: Apply state-of-the-art machine learning tool for the analysis of electron spectro-microscopy data
Project outline

• Part I : EDXS
  → ML : On going
  → Exp : Hui Chen, Collab. with Robin Schaublin, Collab. with See Wee Chee Farhang Nabieei, Stéphane Poitel

• Part II : EELS
  → ML : Not started yet
  → Exp : To be discussed

• Part III : Simultaneous EELS/EDXS
  → ML : Not started yet
  → Exp : Future experiments in collaboration with Gerald Kothleitner
Simplex Non-negative Matrix Factorization

Similar approach as NMF
→ Loss function minimization + Non-negativity constraint

Additions in our algorithm

→ Loss function adapted to Poisson statistics

→ Constrain the solution:
  • Physical modelling
  • Sum-to-one

→ Penalize some solutions
  • Penalize minor phases
Physical modelling

- **Characteristic X-rays**
  - Sum of Gaussians
    - Energies: Atomistic theory
    - Intensity ratios: Atomistic theory
    - Widths: Detector characteristic

→ Fully calculated from $Z = 6$ to $Z = 100$

- **Continuum X-rays**
  - Self-absorption
    - Thin film approximation
  - Bremsstrahlung
    - Solid state theory
  - Detector
    - Without incomplete charge collection

→ Analytical expressions
Learning the model

• **Standard Initialization**
  - List of elements
  - Fitted continuum parameters
  - Tolerance
  - Continuum tolerance

• **Standard alternated gradient descent**
  Until the tolerance is reached, do:
  • Abundances step
    - Sum to one constraint
  • Gaussian weights step
  If the continuum tolerance is reached, do:
  • Continuum learning step
  • Calculate the loss function

• **Initialization with penalization**
  - penalization parameters
  - first phase spectrum

• **Alternated gradient descent with penalization**
  Until the tolerance is reached, do:
  • Abundances step
    - Sum to one constraint
    - **Penalization**
  • Gaussian weights step
  If the continuum tolerance is reached, do:
  • Continuum learning step
  • Calculate the loss function

Fix the sparse values of the abundances

Do a standard alternated gradient descent
Results on artificial data

\[ \mu = 0.0, \varepsilon = 0.0 \]

P0 : 7.7°
P1 : 7.2°
P2 : 7.2°

NMF Angles :
- P0 : 7.8°
- P1 : 15.0°
- P2 : 9.1°
Results on artificial data

$\mu = 0.1, \varepsilon = 1.0$

$P_0 : 7.7^\circ$
$P_1 : 8.8^\circ$
$P_2 : 4.2^\circ$
NMF on experimental data

Brigdmanite
Angle: 27.9°

Ferropericlase
Angle: 11.9°

Ca-Perovskite
Angle: 29.7°
SNMF on experimental data

μ = 3.0, ε = 1.0

Brigdmanite: 7.6°
Ferropericlase: 5.3°
Ca-Perovskite: 32.5°

No continuum learning
Conclusions

**Achieved so far:**
- Better hyperspectral unmixing
- Physical modelling
- Learning of the continuum X-rays
- Sparsity inducing regularization

**Possible improvements:**
- Spatial regularization
- Improve the learning of continuum X-rays
- Parallelization of the calculations

→ Scientific article?
SNMF on experimental data

μ = 0.0

Brigdmanite: 36.2°
Ferropericlase: 5.3°
Ca-Perovskite: 46.4°