Undersampling Raster Scans in Spectromicroscopy for reduced dose and faster measurements

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Joint work with Silvia Gazzola (Bath), Sergey Dolgov (Bath), and Paul Quinn (Diamond Light Source)

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Diamond Light Source

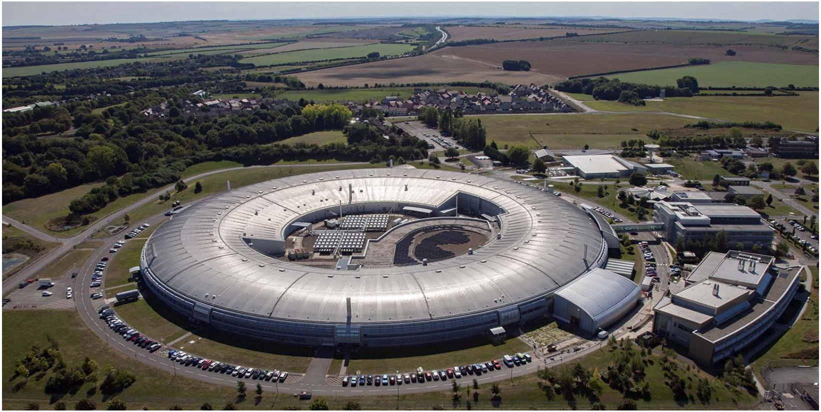

Figure 1: Aerial Picture of Diamond Light Source, Rutherford Appleton Laboratory

X-ray Spectromicroscopy

- Combination of Spectroscopy and Microscopy techniques.
- Beamline produced by 3^{rd} or 4^{th}generation synchrotron radiation sources.
- Scan over a 100×100 grid and at 150 energy levels - roughly 1.5 million measurements
- Experiments take several hours, creating a bottleneck.

Proposed Solution

Undersample the measurements taken of a specimen and use numerical techniques to recover the missing entries.

Experiment Schematics

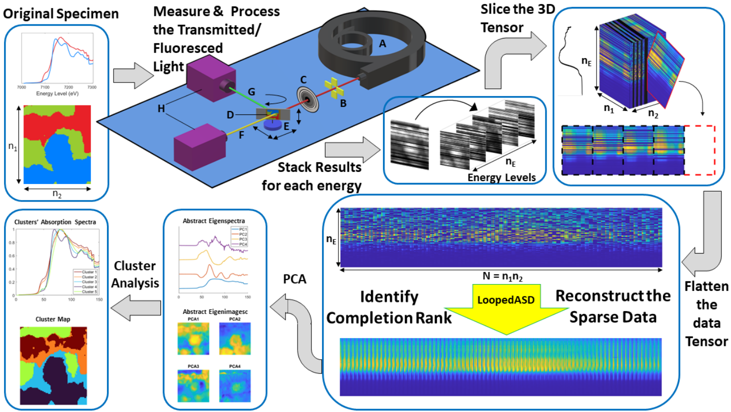

Figure 2: Schematics of Sparse Spectromicroscopy with real data. The original specimen contains a mixture of: FeO (blue region), Fe_2O_3 (red region), over a background (green region). Experimental setup: (A) - a x-ray light source, (B) - intensity of the incident beam is measured, (C) - the light is focused, (D) - specimen, (E) - specimen stage that can translate in 3D, (F) - transmitted flux, (G) - fluoresced flux, (H) - light intensity detectors.

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X-ray Spectromicroscopy

- Measure the intensity of the:
  incident x-rays: $I^0(E)$,
  fluoresced x-rays: $I^f(E)$.
  From these we infer the
  absorption coefficient, $\mu(E)$.
- Photon energies are tuned to the atoms' Absorption Edge.
- Scan all pixels in grid by moving scanner in a Raster Pattern.

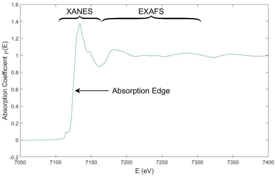


Figure 3: Absorption Coefficient of Hematite (Fe_2O_3, Fe3+)

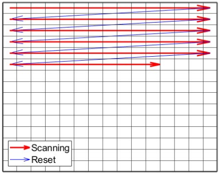


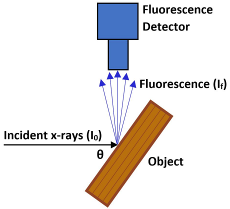
Figure 4: Scanner path for a Raster Scan

Storing the Data

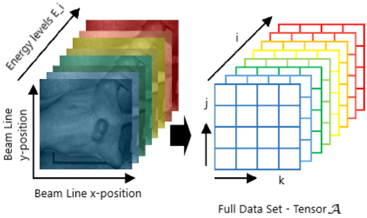
Fluorescence Model: $$I^f(E) \propto I^0(E)\mu(E)t.$$

We store the Optical Density in tensor $\mathcal{A} \in \mathbb{R}^{n_E \times n_1 \times n_2}$. Each layer is a full spatial raster scan at a different energy level:

$$\mathcal{A}_{i,j,k} = \frac{I^f_{j,k}(E_i)}{I^0} = \mu(E_i)t_{j,k}.$$



(a) Fluorescence Geometry



(b) Storing each 2D spatial scan of data.

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Flattening the Data

- Let the specimen consist of S different materials.
- $\mu \in \mathbb{R}^{n_E \times S}$, $t \in \mathbb{R}^{S \times N}$; columns of μ are the absorption spectra, rows of t are the corresponding thickness maps.
- Then

$$A_{ij} = \sum_{s=1}^{S} \mu_s(E_i) t_{sj} + \eta_{ij}, \qquad \eta \in \mathcal{N}(0, \delta^2 I).$$

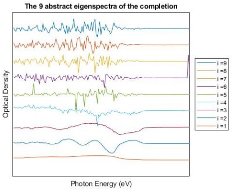
- Apply PCA: $\qquad A = C \cdot R, \qquad C \in \mathbb{R}^{n_E \times n_E}, R \in \mathbb{R}^{n_E \times N}$.
- Columns of C are the abstract eigenspectra, rows of R are the corresponding eigenimages.

Choice of Low Rank

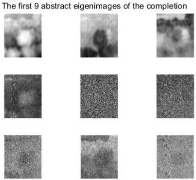
Compute **low rank** approximation $A' = C'R'$, where C', R' are the first L columns, rows of C, R respectively.

Set L at the elbow point (point of max curvature of the SVs of A).

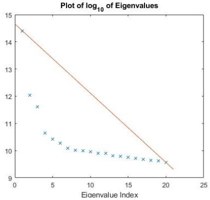
Use KNEEDLE algorithm - supports visual results from PCA.



(a) Columns of C



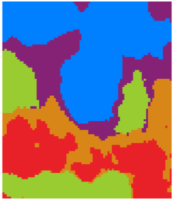
(b) Reshaped rows of R



(c) Illustration of Kneedle on DS1

Cluster Analysis

- Use kmeans to cluster the columns of R' in L-dimensional space.
- The centroid of each cluster approximates the absorption spectra of that material.



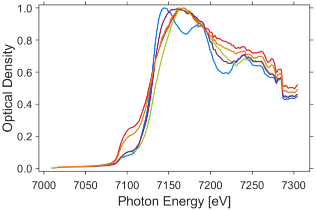


Figure 7: Cluster results of DS2 with $L = 5$. Images produced using Mantis X-Ray.

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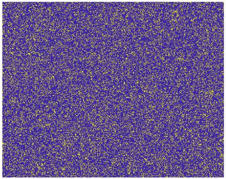
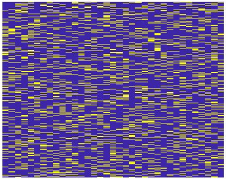
Sampling Methods

- Sampling pattern, $\Omega \subset [n_E] \times [n_1 n_2]$. Undersampling ratio, $p = |\Omega| / n_E n_1 n_2$.
- **Bernoulli Method**: Each entry sampled i.i.d. with probability p.
- **Raster Sampling**: block entries in their (physical) rows for greater experimental efficiency.
- **Robust Raster Sampling**: Slightly reduce randomness to ensure no zero-rows or zero-columns can occur.



(a) Bernoulli Sampling ($p = 0.15$)



(b) Raster Sampling ($p = 0.15$)

Fixing the rank

- Wish to solve:

$$\min_{X,Y} f(X,Y) \qquad f(X,Y) = \frac{1}{2}||\mathcal{P}_\Omega(A) - \mathcal{P}_\Omega(XY)||_F^2.$$

- Remain on the manifold of rank r matrices:

$$A = XY, \quad A \in \mathbb{R}^{n_E \times N}, \; X \in \mathbb{R}^{n_E \times r}, \; Y \in \mathbb{R}^{r \times N}$$

- Drawback: r must be set as an input.

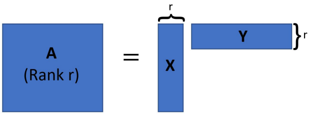


Figure 9: Illustration of X,Y decomposition

Alternating Steepest Descent (ASD)[1]

- Use low-rank matrix completion algorithm ASD (Alternating Steepest Descent).
- For each iteration, fix one component and minimise f(X,Y) using steepest descent with exact line search.
- Alternate the fixed component between X and Y.

ASD - Algorithm

$$\begin{cases} \text{Fix } Y_i, \text{ compute } \nabla f_{Y_i}(X_i),\ \delta_{X_{i+1}} \\ X_{i+1} = X_i - \delta_{X_{i+1}} \nabla f_{Y_i}(X_i) \\ \text{Fix } X_{i+1}, \text{ compute } \nabla f_{X_i}(Y_i),\ \delta_{Y_i} \\ Y_{i+1} = Y_i - \delta_{Y_i} \nabla f_{X_i}(Y_i) \end{cases}$$

$f_X(Y) = f(X,Y)$ for fixed X, $f_Y(X) = f(X,Y)$ for fixed Y, δ is the exact step size.

[1] J. Tanner and K. Wei. "Low rank matrix completion by alternating steepest descent methods". In: (2016).

LoopedASD

Our variation of ASD developed to improve the rate of successful completions for low undersampling ratios on noisy data:

- Select the completion rank r for the sampled data $\mathcal{P}_\Omega(A)$
- Begin by making a rank 1 completion with random initial matrices
- For $i = 2, ..., r$, we use the output of the $(i-1)^{\text{th}}$ completion as an initial guess for the i^{th} completion.
- For each Loop, we must concatenate the initial guesses with a random row/column.

If X^*_{i-1}, Y^*_{i-1} are the outputs of the $(i-1)^{\text{th}}$ ASD completion, and $\mu_X \in \mathbb{R}^{n_E \times 1}$, $\mu_Y \in \mathbb{R}^{1 \times N}$ are random matrices. Then

$$X_i^0 = \left[X^*_{i-1} | \mu_X\right] \qquad Y_i^0 = \left[\frac{Y^*_{i-1}}{\mu_Y}\right]$$

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Selecting the Completion Rank

Need a method to estimate the optimal completion rank r.

- Want the simplest approximation describing most of the data.
- Plot the completion errors, $e_c = ||A - A^*||_F^2 / ||A||_F^2$, for different ranks:

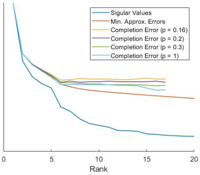
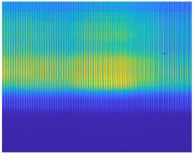


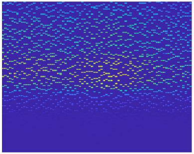
Figure 10: Scaled completion errors

- Cross-validation approach: run short completions on a training set and evaluate using the validation set.
- Set r at the point of max curvature of the validation errors.

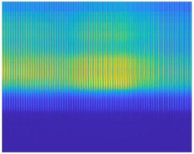
Completion of Real Data Sets:



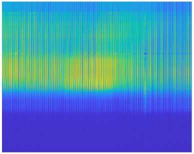
(a) Scaled Colour Image of Data Set 2 (flattened)



(b) Sampled DS2 (Robust Raster Sampling, $p = 0.2$)



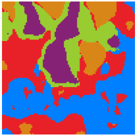
(c) LoopedASD completion of Data Set 2 ($r = 5$)



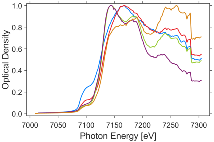
(d) Completion with intentional artefact sampling

Numerically Sampled Data

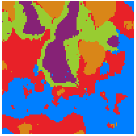
- Sample X-ray Spectromicroscopy data set numerically.
- Complete using LoopedASD, analyse to the completions.



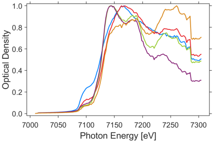
(a) Clusters for full dataset



(b) Absorption spectra for full dataset



(c) Clusters for completed data set

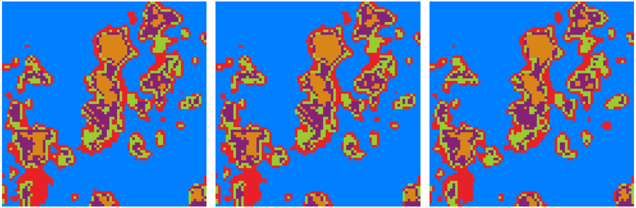


(d) Absorption spectra for completed dataset

Figure 12: Comparing Clustering Results of completion against full data set, $L = 5$, $p = 0.15$, $r = 5$.

Sparse Experiments

Visual comparison of full scans and sparse measurements. Unable to compare numerically due to material drift.



(a) $p = 0.15$ (b) $p = 0.25$ (c) $p = 1.00$

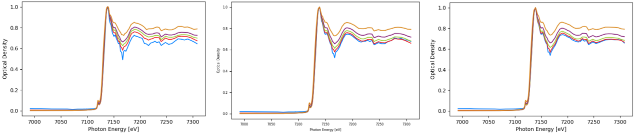


Figure 13: Imaging of chemical mixture of Fe_2O_3 and Fe_3O_4 with sparse measurements

Thank you for your attention!

[1] J. Tanner and K. Wei. "Low rank matrix completion by alternating steepest descent methods". In: (2016).

[2] O. Townsend et al. "Undersampling raster scans in spectromicroscopy for a reduced dose and faster measurements". In: (2022).

[3] Matthew Newville. "Fundamentals of XAFS". In: (2014).

[4] M. Ierotic et al. "Cluster Analysis of soft X-ray spectromicroscopy data". In: (2004).

[5] E. J. Candès and B. Recht. "Exact Matrix Completion via Convex Optimisation". In: (2009).

Core Electron Excitation[2][3][4]

What (roughly) happens when high-energy x-rays are incident on a material?

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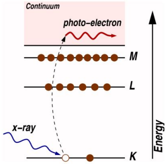

What (roughly) happens when high-energy x-rays are incident on a material?

- Electron's $E_{Binding} > E_{Photon}$ $\rightarrow$ **no absorption**.
- Electron's $E_{Binding} < E_{Photon}$ $\rightarrow$ **photon is absorbed** (photo-electron ejected from atom).

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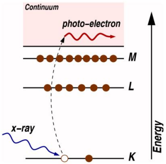

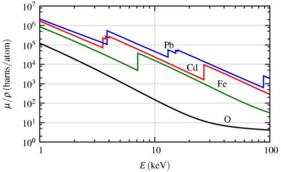

- For high E_{Photon}, we get **core electron excitation**.
- Causes sharp rise in **Absorption Coefficient** μ (absorption edge).

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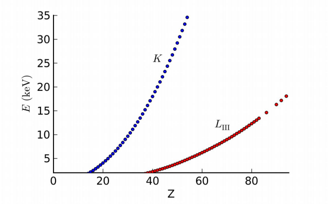

[2] Matthew Newville. "Fundamentals of XAFS". In: (2014)