Contemporary sampling techniques and compressed sensing
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Compressed sensing in electron microscopy

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Outline

1. Transmission electron microscopy & electron tomography
2. Case studies in CS-ET
3. CS in other aspects of electron microscopy
4. Summary
Exploiting sparsity at the nano/atomic-scale

Compressed sensing volume 1:

“Randomness is too important to be left to chance”

Compressed sensing volume 2:

“Structure is far too important not to be exploited”

Compressed sensing volume 3 (?):

“Sparsity is prevalent in everyday and novel applications…and a powerful tool to be harnessed…”

...sparsity is prevalent at the nano/atomic-scale
Transmission electron microscopy (TEM)

(a) Conventional TEM
- parallel acquisition
- broad beam illumination

(b) Scanning TEM (STEM)
- serial acquisition
- raster scanning
TEM – imaging, diffraction, spectroscopy...

Eggeman et al. Ultramicroscopy 2010, 110, 771-777


Phase identification via ADF-STEM: GaPd$_2$

Directly interpretable imaging + chemical sensitivity (atomic number ‘Z-contrast’)
Multi-dimensional electron microscopy
Why do we need 3D TEM?

3D characterisation

Electron tomography: a 3D structure is reconstructed from a series of 2D projections (images)

(Scanning) transmission electron microscope
Why do we need 3D TEM?

Electron tomography: a 3D structure is reconstructed from a series of 2D projections (images)

3D characterisation

(Scanning) transmission electron microscope
Electron tomography

**Tomography** = Visualisation of Slices

**Electron tomography**: a 3D structure is reconstructed from a series of 2D projections (images)

1917 - Principles of tomography go back to theoretical paper by **Radon**
1956 – Bracewell – Fourier slice theorem (see later)
1963 – proposal by **Cormack** for X-ray tomography for medical use
1971 – **Houndsfield** builds first ‘cat-scan’ – Nobel Prize with Cormack, 1979
1968 – First papers on electron tomography by:
   (i) David **De Rosier** and Aaron **Klug**
   (ii) Walter **Hoppe**
   (iii) Roger **Hart**
Why do we need 3D TEM?


Applications

Electron tomography in materials science

...and more
Electron tomography (ET)
Electron tomography (ET)
Electron tomography (ET): an established technique

A familiar diagram!

‘Structural’ electron tomography at the nanoscale is now well-established

...but a fully mature technique?

...some key aspects must be targeted to raise the scope and fidelity

...a number of aspects can be explored to open up new opportunities

Ever growing impact in materials science:
- heterogeneous catalysts, mesoporous solids, polymers, solar cells, semiconductors, dislocation structures...

See e.g. (amongst many others)

A typical example with established methods
- Resolution ~1 nm³
  Leary et al. JPCC 116 (2012) 13343-13352
Raising the fidelity of ET reconstruction

**Practical restrictions**

- Limited number of projections
  - Electron beam sensitivity
  - Carbonaceous contamination
- Limited tilt range (missing wedge)
  - Small pole-piece gap
  - Support grid/holder shadowing
- Limited ‘quality’ of projections
  - Low signal-to-noise ratio
  - Data inconsistencies
    - Inherent data deficiency
- Streaking artefacts, blurring, false elongation...

**Advanced reconstruction techniques**

- Discrete tomography
- Geometric tomography
- Dual-axis
- Equally sloped tomography
- Model based algorithms
  - Weighted SIRT
  - DIRECTT
  - ‘Compressed sensing’ *

**Hardware advances**

- New spectrometers, in-situ and time resolved capabilities, aberration-correction...

**Acquisition strategies**

- Low dose, link to reconstruction algorithm, harness prior knowledge...

**Sophisticated pre/post-reconstruction analysis**

- Data analysis techniques, e.g. PCA, ICA, NMF
- Image processing based segmentation
- Effective visualization
  May be limited by fidelity of the reconstruction!

**New opportunities in electron tomography**

*This presentation: compressed sensing electron tomography (CS-ET)*
A profusion of signal modes

<table>
<thead>
<tr>
<th>Signal mode</th>
<th>Early/selected studies</th>
<th>Status (Established/Advanced) + comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>ADF-STEM</td>
<td>Midgley &amp; Weyland (2001)</td>
<td>E Crystalline specimens, Z-contrast</td>
</tr>
<tr>
<td>EELS</td>
<td>Jarausch et al. (2009)</td>
<td>A Chemical segregation, optical properties, bonding variations; new fast spectrometers</td>
</tr>
<tr>
<td>EDXS</td>
<td>Möbus et al. (2003) Lepinay et al. (2013)</td>
<td>A Chemical segregation; new large area detectors</td>
</tr>
<tr>
<td>Cs-CTEM</td>
<td>Bar Sadan et al. (2008)</td>
<td>A Atomic-scale, WPOs</td>
</tr>
<tr>
<td>Cs-STEM</td>
<td>Van Aert et al. (2011) Goris et al. (2012)</td>
<td>A Atomic-scale; heavy metal nanoparticles</td>
</tr>
<tr>
<td>Holography</td>
<td>Twitchett-Harrison et al. (2007)</td>
<td>A Mean inner potential, electrostatic and magnetic fields</td>
</tr>
<tr>
<td>Lorentz TEM</td>
<td>Phatak et al. (2010)</td>
<td>A Magnetic fields</td>
</tr>
<tr>
<td>Diffraction</td>
<td>Kolb et al. (2007)</td>
<td>A Crystalline materials</td>
</tr>
<tr>
<td>Time resolved</td>
<td>Kwon &amp; Zewail (2010)</td>
<td>A New commercial TEMs</td>
</tr>
<tr>
<td>Cc-TEM</td>
<td>Baudoin et al. (2013)</td>
<td>A Thick biological specimens</td>
</tr>
<tr>
<td>Precession BF-CTEM</td>
<td>Rebled et al. (2011)</td>
<td>A Crystalline specimens</td>
</tr>
<tr>
<td>BF-STEM</td>
<td>Sousa et al. (2011)</td>
<td>A This specimens, polymers, biological sections</td>
</tr>
<tr>
<td>IBF-STEM</td>
<td>Ercius et al. (2006)</td>
<td>A Thick specimens</td>
</tr>
<tr>
<td>MAADF-STEM</td>
<td>Sharp et al. (2008)</td>
<td>A Dislocations</td>
</tr>
</tbody>
</table>

...and more!
A profusion of signal modes

Electron tomography
2014

...and more!

Signal mode
- BF-CTEM
- ADF-STEM
- EFTEM
- EELS
- EDXS
- Cs-CTEM
- Cs-STEM
- Holography
- Lorentz TEM
- Diffraction
- Time resolved
- Cc-TEM
- DF-TEM
- Precession BF-CTEM
- BF-STEM
- IBF-STEM
- MAADF-STEM

atomic-scale electron tomography

multi-dimensional electron microscopy
Composition, time, environment...
Adding dimensions to 3D TEM

Multi-dimensional electron tomography

Composition, time, environment...

STEM-EDX tomography
Genc et al. Ultramicroscopy
131(2013) 24-32

STEM-EELS tomography
Yedra et al. Ultramicroscopy
122 (2012) 12-18

Time resolved TEM tomography
Kwon & Zewail Science 328
(2010) 1668-1673

Lorentz TEM + tomography (vector potential)
253901

Rotation electron diffraction + tomography
Garcia-Martinez et al. ChemCatChem
DOI: 10.1002/cctc.201402499

Scanning precession electron diffraction + tomography
(‘6D’ data)

STEM-EELS tomography
(localized surface plasmon resonances)
Electron tomography reconstruction

• Fourier slice theorem

• Two ways to approach reconstruction…
Electron tomography reconstruction

• Fourier slice theorem

• Two ways to approach reconstruction…
Electron tomography reconstruction

- ET in terms of a system of linear equations

**Real-space formulation**

\[ \mathbf{b} = \Phi \mathbf{x} \]

Tilt-series projection images (sinogram)

Projection operator (electron microscope)

\[ \hat{\mathbf{x}} = 3D \text{ reconstruction} \]

**Fourier-space formulation**

\[ \mathbf{y} = \Phi_{F_u} \mathbf{x} \]

Radial Fourier data

Undersampled Fourier operator

‘missing wedge’
Electron tomography reconstruction

• ET in terms of a system of linear equations

Real-space formulation

\[ \mathbf{b} = \Phi \mathbf{x} \rightarrow \text{3D object} \]

Projection operator (electron microscope)

\[ \hat{\mathbf{x}} = \text{3D reconstruction} \]

Fourier-space formulation

\[ \mathbf{y} = \Phi F \mathbf{u} \rightarrow \text{3D object} \]

Undersampled Fourier operator

Limited tilt range (missing wedge)

• Small pole-piece gap
• Support grid/holder shadowing

Limited number of projections

• Electron beam sensitivity
• Carbonaceous contamination

Lead to: Streaking artefacts, blurring and false elongation in missing wedge direction
Electron tomography reconstruction

- Restricted tilt range

- Small pole-piece gap
- Support grid/holder shadowing

- Limited number of projections
- Electron beam sensitivity
- Carbonaceous contamination

Lead to: Streaking artefacts, blurring and false elongation in missing wedge direction
Electron tomography reconstruction

- **Restricted tilt range**

  - +/- 90° tilt range
  - +/- 70° tilt range
  - +/- 50° tilt range
  - +/- 30° tilt range

  - Small pole-piece gap
  - Support grid/holder shadowing

- **Limited number of projections**

  - Electron beam sensitivity
  - Carbonaceous contamination

  Lead to: Streaking artefacts, blurring and false elongation in missing wedge direction
Example: densely-packed nanoparticles (pre CS-ET)

- Tilt series projections, ±76°, Δθ = 2°

- Segmented 3D data

- Slices through 3D reconstruction

- Quantitative analysis

Example: densely-packed nanoparticles (pre CS-ET)

Fidelity of the analysis is limited by artefacts in the reconstruction


(a) FR ≤ 2  
(b) 2 < FR ≤ 3  
(c) FR > 3

Missing wedge direction

50 nm
Exploiting sparsity (compressibility)

Signal compression

Sample

Sparsifying transform, $\Psi \rightarrow$

$n$ pixel image

$n >> s$

$(n - s)$ coefficients thrown away

$s$-sparse representation

Transmit/store

Compress

Receive

$s$-sparse representation

Decompress

$n$ pixel image

$\hat{x}$

$\Psi^*$

$\Psi$

$\hat{x}$

$\Psi$

$\Psi^*$

Non-linear

$n >> s$

Reconstruct

$m$ compressive measurements, $b$, against sensing basis, $\Phi$

$\hat{x}$

$m$ measurements

$s$-sparse representation

$n$ pixel image

$\Psi$

$\Psi^*$

$n$ pixel image

$\hat{x}$

$\Psi$

$\Psi^*$

Non-linear

$\hat{x}$

$n >> s$

Reconstruct

$\hat{x}$

$\Psi$

$\Psi^*$

$n$ pixel image

$\hat{x}$

$\Psi$

$\Psi^*$

Non-linear

$n >> s$

Reconstruct

$\hat{x}$

$\Psi$

$\Psi^*$

$n$ pixel image

$\hat{x}$

$\Psi$

$\Psi^*$

Non-linear

$n >> s$

Reconstruct

$\hat{x}$

$\Psi$

$\Psi^*$

$n$ pixel image

After Romberg et al., Lustig et al.
‘Sparsity’ & ‘Compressibility’

- A \textit{sparse} representation of a signal of length $n$ consists of $s \ll n$ non-zero coefficients.
- A \textit{compressible} signal is well approximated by $s \ll n$ non-zero coefficients.

<table>
<thead>
<tr>
<th>Signal characteristic(s)</th>
<th>Sparsifying transform, $\Psi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>\textit{Few non-zeros}</td>
<td>\textit{Identity (ID)}</td>
</tr>
<tr>
<td>\textit{Piecewise constant}</td>
<td>Finite differences (total variation, TV)</td>
</tr>
<tr>
<td>\textit{Local periodic}</td>
<td>\textit{Fourier}</td>
</tr>
<tr>
<td>\textit{Piecewise smooth}</td>
<td>\textit{Discrete cosine}</td>
</tr>
<tr>
<td>\textit{Curved edges}</td>
<td>\textit{Wavelets}</td>
</tr>
<tr>
<td>\ldots and more</td>
<td>\ldots Curvelets</td>
</tr>
</tbody>
</table>

...\textit{ridgelets, shearlets, bandlets} ...and more
...\textit{also ‘dictionary learning’ methods}. 
Incoherence & sparse recovery

**Incoherent sampling**

- **Φ** not easily represented in **Ψ**
- Low coherence between Sensing basis **Φ** and Sparse basis **Ψ**

- Global compressive measurements
- Each measurement: information about many sparse coefficients
- Undersampling (aliasing) artefacts add as noise-like interference

**Lustig et al.**:

- (Transform) point spread function to measure incoherence of aliasing artefacts in MRI acquisition schemes
- Radial sampling of Fourier space should be sufficiently incoherent to apply CS


- More recent mathematical treatment of sampling regimes:
  - see Hansen, Adock, Roman et al.
  - e.g. On asymptotic structure in compressed sensing, arXiv:1406.4178 [math.FA]

**Sparse recovery**

- Non-linear optimization that promotes sparsity
- Find the sparsest solution that is consistent with the measured data...to within a given tolerance

\[
\text{minimise } \|\Psi \hat{x}\|_{\ell_1} \text{ subject to } \|\Phi \hat{x} - b\|_{\ell_2} \leq \varepsilon
\]
CS-ET: a simple example

Leary et al. Ultramicroscopy 2013, 131, 70-91
CS-ET vs conventional reconstruction techniques

Our (current) implementation:

Leary et al. Ultramicroscopy 2013, 131, 70-91
**CS-ET simulation:** densely-packed nanoparticles

Unsupported Ga-Pd nanocatalysts: size, shape, spatial distribution, agglomeration state?

Ψ = TV + ID transform

Leary et al. Ultramicroscopy 2013, 131, 70-91
CS-ET case study: densely-packed Ga-Pd nanoparticles

→ Repeat for 512 different phantoms
→ Combine quantification results

Case studies in:

Compressed Sensing Electron Tomography (CS-ET)

(unpublished results removed from web version of presentation)
CS-ET: exploiting sparsity at the nanoscale

Nanoparticles – piecewise constant approximation

\[ \Psi = TV + \text{identity transform (reduces missing wedge artefacts)} \]

See also:
\[ \Psi = TV \text{ minimisation} \]

\[ \Psi = \text{anisotropic total variation (ATV)} \]
(limit edge detection from direction blurred by missing wedge)
CS-ET: exploiting sparsity at the nanoscale

Nanostructures – piecewise constant approximation

$$\Psi = \text{TV}$$

Elemental maps reconstructed using CS-ET, spectral reconstructions using SIRT

Haberfehlner et al. *Nanoscale* 2014, 6, 14563-14569
CS-ET: exploiting sparsity at the nanoscale

Smooth signals – state-of-the-art

- localised surface plasmon resonance

In general:
- magnetic fields
- electric fields
- compositional gradients

future of ET?

Ag nanocube localised surface plasmon resonances

STEM-EELS tomography

Ψ = DWT (Coiflet 8)

Nicoletti et al. Nature 2013, 502, 80-84

ET of electrostatic potential in pn junction

ET of magnetic fields


Phatak & Gursoy Ultramicroscopy 2015, 150, 54-64

Crystalline but electrically damaged silicon
- n-type silicon
- p-type silicon

Amorphous surface layers

Low          High

α   β   γ   δ   ε
CS-ET: exploiting sparsity at the atomic-scale

ET at atomic resolution – state-of-the-art

- Atomicity = sparsity
- Scope for development...

Simulated and experimental atomic resolution STEM images


Au/Ag nanorod

Gaussian peak fitting and reconstruction from the ‘model’ images

Ψ = ID transform


Au nanorod

Atomic resolution reconstruction from 4 images

Ψ = ID transform


Ag nanoparticle (simulated images)

Dictionary learning based reconstruction

**CS-ET:** exploiting sparsity at the nanoscale

**Biological CS-ET**  – low signal-to-noise-ratio
  – often line-like membrane structures
  – optimal sparsifying transform?

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**L²-gradient flows with TV**

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**Removal (inpainting) of fiducial markers**

Cryo-ET projections

CS-based in-painting

\[ \Psi = \text{DCT} \]


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Xu and Li *J Compt Math* 2011, 29(5), 501-525
CS and related methods in other electron microscopy techniques (emerging)

(unpublished results removed from web version of presentation)
2D imaging – lowering electron dose

Random subsampled scanning

Scanning EM (SEM): only scan randomly selected pixels

\[ \Psi = \text{DCT} + \text{TV} \quad \text{(inpainting)} \]

Anderson et al. Computational Imaging XI. Vol 8657, 86570C1-12

Aggregated measurements

Scanning TEM (STEM):
- Low resolution aggregated measurements
- Recover atom positions on a finer grid


Random subsampled scanning

Scanning TEM (STEM): only scan randomly selected pixels

Bayesian dictionary learning (inpainting)

Stevens et al. Microscopy 2014, 63(1), 41-51

(simulated sub-sampling)
Other/emerging applications

High resolution TEM 3D reconstruction

Scanning confocal electron microscopy

Ptychography (lensless diffractive imaging)

Van den Broek and Koch *Phys Rev B* 2013, 87, 184108

*Tom Furnival, Electron Microscopy Group; tjof2@cam.ac.uk*
Summary

Compressed sensing electron tomography (CS-ET)

• Only recently recognised in ET (compared to MRI, CT)

• ~10x fewer projections used

• Scope for optimisation of parameters

• New possibilities in ET
  (spectroscopic ET, atomic scale ET, beam sensitive specimens, time-resolved studies?)

Compressed sensing electron microscopy

• Lowering 2D imaging dose primary motivator, temporal imaging

• Methods/applications just beginning to be explored