AI and ML in Microscopy: A Material Opportunity

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Multidimensional SPM modes

Physics perspective:
1. Local stimulus: spectroscopic (3D) SPM modes
2. Phase transitions are hysteretic: First Order Reversal Curves
3. Phase transitions can be rate-controlled

SPM Perspective:
4. SPM requires resonance enhancement (frequency domain)

Instrumental limit: photodetector bandwidth (~10 MHz) x DAQ performance (32 Bit)
- **Single frequency/heterodyne**: lock-in compression to ~ 1 kHz
- **Band excitation**: $10^2$ bins at ~ 1 kHz = 100 kHz
- **G-mode**: full streaming at ~10 MHz

We realized we are doing big data
We have the data... We need to do something with it!

M.P. Nikiforov, A.A. VerPEGel, V.V. Reukov, G.L. Thompson, S.V. Kalinin, and S. Jesse,
Opportunities in Materials Science

• “Improve”: Renewable energy, self-driving cars, transparent displays, new memory technologies
• “Discover”: Room temperature superconductivity, high mechanical stress materials
• “Engineer”: Quantum computing, single-atom catalysts, biomolecules

Functionality, manufacturability, cost
“New directions in science are launched by new tools much more often than by new concepts. The effect of a concept-driven revolution is to explain old things in new ways. The effect of a tool-driven revolution is to discover new things that have to be explained.”

Freeman Dyson
Materials in the Chemical Space

- Full atomic coordinate space: un-tractable
- Chemical space: minima corresponding to (meta)stable compounds

- Chemical space is non-differentiable
- The pathways between different regions are non-obvious
- “Useful” functionalities can be very complex and poorly understood
  - We can calculate bandgaps and ideal Young moduli
  - Biological activities and superconductivity, not so much

We typically need outliers!

But:
- There are underlying physical laws that determine what is possible

- Materials and molecules are points in chemical space
- Finding “the right” material is then a search/optimization problem.
- Machine learning is great at this. Right?
Molecular Systems:

- Chemical space is a graph: $10^{63}$ (only less than 30 atoms)
- Edges (reactions) can be optimized individually
- Amenable to the literature mining (*Chematica*)
- Area of known compounds can be expanded via retrosynthesis
- Amenable to laboratory robotics

**But:** functionalities at the nodes are defined within the context (CO$_2$ adsorption, water desalination, biological functionality)
Let’s think about it as a search problem:

- **Alloying**: need maintain composition ~1%
- **Doping**: need maintain composition ~ $10^{-6}$
- Grid search is out for $D > 3$ (experiment)
- MCMC type problem: how do we make it work?

Discovery of copper HTSC, MgB$_2$, iron pnictides. Serendipitous discovery followed by exploring large families. Could not predict – no theory
The Solid Problem

Chemical space is heavily degenerate: can use mean field descriptors

- Symmetry,
- Concentrations,
- Order parameters
- Thermodynamic potentials

- Really interesting materials are those when it is not the case (relaxors, Kitaev materials) – some correlations and disorder in ground state.
- Many physical properties cannot be predicted. Bandgaps ok, superconductivity not so much
- Large scale organization: defects, microstructures, etc.

How do we start describing structures of solids beyond symmetry-based methods, establish structure-property relationships, and use this information for prediction of materials and discovery of synthesis pathways?

C. Nelson

L.Q. Chen
More than atoms

Atomic positions can be determined to <10-pm precision

Bond length: Chemical reactivity, catalytic activity

Bond angles: Magnetism and transport

Configurations and repeating elements?

Nature 515, 487 (2014)

J.J. Guo et al., Nat. Comm. 5, 5389 (2014)
The Lab on a Beam

**Image**
- atomic columns

**Spectra from**
- single atoms
- Image light atoms and sensitive materials

**Diffraction from**
- subatomic volumes

**Beams with orbital momentum**

**Beam manipulation**

**0.61 Å resolution**

**2012**
- Segmented detectors

**2014**
- 4D STEM

**2016**
- Vortex beams

**2018**
- Physics extraction
- Atomic manipulation

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First AC (Nion) 1997

**2002**
- Prototype correctors

**2006**
- TEAM project

**2010**
- Broad adoption of AC STEM

**2012**
- Segmented detectors

**2014**
- 4D STEM

**2016**
- Vortex beams

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**Challenge 1:** Data Infrastructure

**Standard image:** ~10 MB

**Dynamic data:** ~100 MB/S

**4D STEM:** ~TB data (limited by the availability of data infrastructure)

**Potentially:** Large Hadron Collider level data flows (from single microscope)
Physics from STEM and SPM data

• Can we get materials specific information (e.g. atomic coordinates from STEM, scattering potentials from 4D STEM, etc.) from microscopy data, at which level of confidence, and how this knowledge is affected and can be improved from knowledge of imaging system (e.g. classical beam parameters, resolution function, all the way to full imaging system modelling), and knowledge of material.

• Can we use the materials-specific information with uncertainties determined by incomplete knowledge of imaging system or intrinsic limitations to infer physics and chemistry, either via correlative models or recovery of generative physics (force fields, exchange integrals, etc.)

• Can we use thus determined materials information, either correlative or causative, to reconstruct materials behaviors (phase diagrams, etc) in the broader parameter space (e.g. for temperatures and concentrations different for specific sample studied), and determine how reliability of such prediction depends on position in parameter space.

• Can we harness the data stream from the microscope to engender real time feedback, e.g. for autonomous experimentation and atomic manipulation
Observations of atomic dynamics induced by beam (or temperature, field, etc.) gives information on multiple atomic configurations as they form and evolve. Can we learn:

• Effective interaction parameters (e-ph coupling)?
• Force fields?
• Phase transition dynamics
Deep learning for atomically resolved images

**Top 3 predictions**

Staffordshire bullterrier 43 %
American pit bull terrier 23 %
Basenji 11 %

*Very close*

**Meet Duffy:** Pitbull-Shepard-Collie mix

**Randomly selected feature maps**

**Top 3 predictions**

Wool 6.3 %
Velvet 5.5 %
Window screen 3.8 %

**STEM of WS2**
Deep learning works like a charm for:
• Drift correction
• Denoising
• Data processing/dimensionality reduction
• Feature finding (physics is in the training set)
Deep learning in AFM

Model trained on a single movie frame from the well-ordered phase and applied to the entire movie

Maxim Ziatdinov, Xin Li, Shuai Zhang, Harley Pyles, David Baker, James J. De Yoreo, Sergei V. Kalinin
Deep learning in AFM

Model trained on a single movie frame from the well-ordered phase and applied to the entire movie

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Defect Libraries

- Identify the “real” defects
- Theory to get energies, strains, etc

Analysis workflows

Open data

Open code: Jupyter papers
From Correlation to Causation:

- Problems can be intractable combinatorically, but have simple constitutive laws
- Low dimensional non-linear manifold in the very high dimensional linear spaces
- Observational data: astronomy
Problems:
- There are $10^{10-23}$ degrees of freedom
- Which (in most materials) correspond to very small number of collective variables (order parameters)
- Naturally, materials where this is likely not the case are really interesting
Physics and chemistry from structural STEM data?

On mesoscale, materials functionalities can often be described via order parameter fields:
- Mesoscopic order parameter is often known from macroscopic measurements
- What are the boundary conditions at surfaces and interfaces
- What are the roles of defects?
- Can we describe spatially inhomogeneous states (relaxors, charge ordered materials, MPB systems)?

\[
F = \int_V \left[ f_{\text{bulk}}(P_i) + f_{\text{grad}}(P_{i,j}) + f_{\text{elas}}(P_i, \varepsilon_{kl}) + f_{\text{elec}}(P_i, E_i) \right] dV
\]

LGD equation:
\[
\frac{dP_i}{dt} = -L \frac{\delta F}{\delta P_i}
\]

Conventional fixed \( \phi \) b.c.
\[
\phi \big|_{z=L+\lambda} = V_{\text{planar}}
\]
\[
\frac{\partial P_z}{\partial z} \bigg|_{z=0, L+\lambda} = 0
\]

Disordered states
Building the mesoscopic picture top down

Model System: Lanthanum-strontium cobaltite with topological defects and interfaces

Antiphase domain boundary

\[ \eta(x, z) \approx \eta_{SD}(z) \tanh \left( \frac{x - x_0}{L_C(z)} \right) \]

Fitting the experimentally observed atomic profiles to the functional form of order parameter

\[ \eta_{SD}(z) \approx \eta_b \left( 1 - \frac{1}{1 + \sqrt{2} \lambda/L_C(0)} \exp \left( - \frac{\sqrt{2}z}{L_C(0)} \right) \right) \]

Ordering behavior at the interfaces

\[ \eta(z) = 0.787 (1 - 0.5 \exp \left( - \frac{z-1}{1.1} \right)) \]

Allows to analyze the interplay between ordering, chemical composition, and mechanical effects at domain walls, interfaces and structural defects

A.Y. Borisevich et al., PRL 109, 065702 (2012)
Flexoelectricity by Computer vision

HAADF-STEM images

Theory-experiment matching

Model (best-fit flexo)

Effect of flexoelectricity

\[ F = \alpha_{ij} PP_{ij} + \alpha_{ijkl} PP_{ijkl} P_{ijkl} + \alpha_{ijklmm} PP_{ijklmm} P_{ijklmm} + \frac{1}{2} g_{ijkl} P_{ij} P_{kl} + \frac{1}{2} e_{ijkl} e_{kl} e_{kl} e_{kl} e_{kl} e_{kl} - q_{ijkl} e_{kl} P_{ijkl} - \frac{1}{2} \kappa_{ijkl} e_{ij} e_{ij} e_{ij} e_{ij} - E_{ij} P_{ij} + f_{ijkl}(P_{ijkl} e_{ij} - e_{ij} P_{ij}) \]

Longitudinal/transverse flexocoupling \( f_{ij}/f_{12} \) (\( f_{11} \sim -f_{12} \))

Similarity map

The flexocoupling in both PTO and STO layers is considered, revealing different modulation effects.

Li et al., Nat. Comm. 2017

- Polarization and tilt behavior at interfaces and topological defects: gradient terms and physical BCs
- Coupling with (electro)chemical boundary conditions
- Defect effects: transition from localized perturbation to collective responses
Physics from Microscopic Degrees of Freedom

**Microscopic models**
- Ising model
- Heisenberg model
- Kitaev model
- ...

**Macroscopic observables:**
- Property
- Average structure
- Fluctuations

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**On atomistic scale, we often use lattice Hamiltonian models:**
- Can we determine local interactions from STEM or SPM data
- What if some information is lost (1 << observables << degrees of freedom)

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**Can ML do it?**
(Melko 2017)

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**Macroscopic measurements**
Scattering data

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Real material

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A. Sefat
Can we study physics and chemistry locally?

Ising model

\[ H(H) = \sum_{i,j} J_{ij} S_i S_j + \sum_i (h_i + H)S_i \]

- Collection of two-state “spins” on a geometric lattice
- Interactions with nearest neighbors
- No long range depolarization fields
- Universal model that can represent physical (magnetism), chemical (alloys, surface adsorbates) systems
Minimize statistical distance between histograms

$$s = \arccos \left( \sum_{i=1}^{k} \sqrt{p_i} \sqrt{q_i} \right)$$

Utilize all available statistical information in the image

Using generative model, infer parameters from the experiment.
MoS$_2$ – ReS$_2$ Solid solutions by STEM

5% ReS$_2$  55% ReS$_2$  78% ReS$_2$  95% ReS$_2$

Data by Shize Yang and Matt Chisholm
Thermodynamics of solid solution

1. Pair-additive Model
   \[ u_i = w_1 \sum_{\{NN\}} \delta_{MoRe} + w_2 \sum_{\{NNN\}} \delta_{MoRe} \]

2. Many-body Model
   \[ u_i = w_1 \sum_{\{S\}} \delta_{MoMoRe} + w_2 \sum_{\{S\}} \delta_{MoReRe} \]

Exploring physics: statistical normal modes

- Traditionally, the order parameter is defined based on symmetry and atomistic representation is established in the *ad hoc* manner.
- But what if we define order parameter from the bottom up – based on the statistics of atomic distortions?
- And further correlate it to local chemical composition?
Phase transition via statistical normal modes

Three dominant distortion modes

Mode distributions vs. global and local composition

Local symmetry breaking!

Component #0

Component #1

Component #2

5% ReS$_2$

55% ReS$_2$

78% ReS$_2$

95% ReS$_2$
Phase transition from the bottom up

Global symmetry breaking

Local symmetry breaking!
Transition to mesoscopic free energy

Free energy functional for 2D materials

\[ F[\eta] = \int d^2r \left( f_L[\eta(r)] + f_{KL}[\eta(r)] + g_{ijkl} \frac{\partial \eta_i}{\partial x_j} \frac{\partial \eta_k}{\partial x_l} + \frac{f_{KL}}{2} \left( \frac{\partial \eta_i}{\partial x_j} - \eta_i \frac{\partial \eta_i}{\partial x_j} \right) \right) \]

Local free energy term:

\[ f_L[\eta(r)] = \alpha_{11} \eta_1^2 + \alpha_{33} \eta_3^2 + \beta_{11} \eta_1^4 + \beta_{33} \eta_3^4 + \beta_{13} \eta_1 \eta_3 (\eta_1^2 - 3\eta_3^2) + \gamma_{111} \eta_1^6 (\eta_1^2 - 3\eta_3^2)^2 + \gamma_{222} \eta_2^6 (3\eta_1^2 - \eta_3^2)^2 + \gamma_{113} \eta_1^2 \eta_3^4 + \gamma_{133} \eta_1^4 \eta_3^2 + \gamma_{333} \eta_3^6 \]

Opportunity:
- Phase diagrams
- Electronic phenomena induced by curvature via flexoelectric coupling
- Field- and doping induced transitions
A Bit of (Ancient and Modern) Wisdom

...there are known knowns; there are things we know we know. We also know there are known unknowns; that is to say we know there are some things we do not know. But there are also unknown unknowns—the ones we don't know we don't know.

D. Rumsfeld

Har kas ke bedanad va bedanad ke bedanad
Asb-e kherad az gombad-e gardun bejahanad
Har kas ke nadanad va bedanad ke nadanad
Langan kharak-e khish be manzel beresanad
Har kas ke nadanad va nadanad ke nadanad
Dar jahl-e morakkab'abad od-dar bemanad

Anyone who knows, and knows that he knows
Makes the steed of intelligence leap over the vault of heaven
Anyone who does not know, but knows that he does not know
Can bring his lame little donkey to the destination nonetheless
Anyone who does not know, and does not know that he does not know
Is stuck forever in the double ignorance

Naser od-Din Tusi (1201-1274)
The World is Bayesian: Physics from Observations

...there are known knowns; there are things we know we know. We also know there are known unknowns; that is to say we know there are some things we do not know. But there are also unknown unknowns—the ones we don't know we don't know.

D. Rumsfeld

$P(\text{Theory}|\text{Data}) = \frac{P(\text{Data}|\text{Theory}) P(\text{Theory})}{P(\text{Data})}$

• Experimentalists know the priors. Albeit they do not know that they know it, or how to convert them to algorithmic form

• How can we add Bayesian priors to:
  – Reinforcement learning (functionality optimization in experiment)
  – GANs (inverse problems, image reconstruction)
  – VAEs (physical constraints on latent variables)

Hypothesis driven science:
What we want to learn

Forward model:
Theory

Domain expertise:

High Performance Computing
The chemistry challenge

R. Ishikawa

Only one atom moves in well-defined potential

One or few atoms move in defined potential

One or few atoms move in weakly changing potential

Point defect dynamics

Extended defect dynamics

Everything changes: defect formation, nucleation, mechanical deformation

What about chemistry?

- Markov state descriptions
- Potential energy landscape reconstructions
- Force fields (and excitation) from observed dynamics
Learning the defect evolution

Experimental

Sample: WS2
E-beam energy: 60 kV

Data collected by Ondrej Dyck (CNMS/ORNL)
Spatio-temporal trajectories

Diffusion parameters for selected defect types

Diffusion coefficient: $3 \times 10^{-4}$ nm$^2$/s - $6 \times 10^{-4}$ nm$^2$/s (within 2D random walk approximation)

- Identification of dominant point defects and their characteristic statistical behaviors
- Analysis of diffusion parameters for the selected defect species
- Study of transformation pathways and transition probabilities for composite defects

Maksov et al., npj Computational Materials 5, 12 (2019)
Spatio-temporal trajectories

Evolution of defects as Markov process

Maksov et al., npj Computational Materials 5, 12 (2019)
Beam induced reactions of Si atoms on the edge of graphene

Reconstructing Si impurity configurations at graphene edge

Experimental data

Network’s output

Chemical transformations on the edge

Derived classes of Si-C edge configurations

- Gaussian mixture model
- Discrete rotation symmetry + structural similarity algorithm

Transition probabilities matrix

- Markov state analysis

“What I cannot create, I do not understand.”
— Richard Feynman

Is There a Third Way?
We induce local amorphous/crystalline transition in a defined area

- No crystallization observed away from interface
- Electron-beam induced solid phase epitaxy
- Control of beam position and speed
- We chose to explore, exploit, and understand this behavior

- Formation of 3D structures inside solid
- Epitaxial registration
- MD simulations have confirmed experimental observations
- Foundation for making atomically precise, multi-component, multi-layer systems
Beam Induced Transformations in Solids: Feedback

Start inside crystal, Fast advance

Amorphous/x-line interface

Growth of new crystalline atomic layer

Beam advances to next atomic layer
Manufacturing at the Atomic Scale with Beams

Directed Crystal Growth in STO

~16 u.c. wide
Atomic manipulations

Moving Si dopants

Assembling primitive structures

Custom platform for dopant atom movement with real-time image updates during manipulation

What I cannot create, I do not understand
R. Feynman

**Need to find out:**
- What are local atomic functionalities
- Why do atoms do it?
- How we direct them to do what we want?

**X-ray and neutron scattering:** where the atoms are on average

**Electron and probe microscopy:** where exactly are the atoms

**Dynamic microscopies:** what the atoms do

**Present**

- Nanotechnology
- Beyond Moore
- Molecular machines
- Materials design

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*Original Text*

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**STEM: The Lab on the Beam**
Concluding:

• Data matters (but only if physics and chemistry is analyzed!)
• Building physics and chemistry from atomic level up
• E-beam atomic fabrication

See cnms.ornl.gov for more information.