Emergent spin phenomena in the age of artificial intelligence

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Symposium to Honor the Life and Work of Aron Pinczuk
Aron’s Early Influence - Graduate school
Aron’s Early Influence - Graduate school
Opportunities and resources!
Biggest influence – Research career

- Emergent behavior
  - Low dimensional electron systems
  - Spin degrees of freedom
  - Charge degrees of freedom

- Data analytics and artificial intelligence
  - Molecular beam epitaxy (MBE) of graphene story
  - High-dimensional optimization problem?
More Is Different

Broken symmetry and the nature of the hierarchical structure of science.

P. W. Anderson
More Is Different

Broken symmetry and the nature of the hierarchical structure of science.

P. W. Anderson
More Is Different

Broken symmetry and the nature of the hierarchical structure of science.

P. W. Anderson
Spin properties in GaAs quantum wells
GaAs Quantum Well

- 270 Angstroms
- $n = [0.5, 4.2] \times 10^{11} \text{ cm}^{-2}$
- $\text{Mobility}_{\text{max}} = 1.1 \times 10^7 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$
- $V_{\text{gate}}$
2DES in a magnetic field: Classical Hall Effect
2DES in a magnetic field: Quantum Hall Effect

Integer Quantum Hall Effect

2DES in a magnetic field: Fractional Quantum Hall Effect

- Exotic even-denominator state at $\nu = 5/2$
- Topologically protected?
- Implement topological quantum computation?

Milli-Kelvin Spectroscopy

- Dilution fridge: $40\text{mK} < T < 2\text{K}$
- Tilt-angle, $\theta = 20$ degrees
- Titanium:Sapphire laser with fine tunability
Spin properties of 2DES

T.D. Rhone et al, PRL 106, 196805 (2011)
Synthesis of graphene by molecular beam epitaxy

- Grow graphene
- High-dimensional parameter space:
  - Substrate temperature
  - Partial pressure of Argon
  - Target temperature
  - Annealing temp.
  - Annealing time
- Measure quality of grown material
Aron the Influencer – Recently
Two-dimensional Magnetic Materials

Huang et al., Nature 546, 270 (2017)

Gong et al., Nature 546, 265 (2017)
Data-driven study of 2D materials?

CrGeTe₃  
Graphene  
Black phosphorus  
Crl₃  
MoS₂
Data-driven study of 2D magnetic materials

Motivation:
1. Materials discovery
   - 2D magnetic materials
   - Chemical stability
2. Knowledge discovery
   - Magnetic properties of 2D materials

Discover novel magnetic 2D materials using materials informatics
Data-driven study of 2D magnetic materials
Materials Informatics

Materials science + Artificial intelligence

\[ y = f(x_1, x_2, \ldots, x_N) \]

- Magnetic moment
- Number of spin up electrons
- Number of valence electrons
- Electronegativity
Why use artificial intelligence to study materials?

- Materials search space is huge
  - ICSD (inorganic crystal structure database)
    - 200,000 inorganic compounds
  - Chemical Abstracts Service
    - 49,037,297 organic/inorganic entries
  - Virtual Chemistry Space\(^1\)
    - \(\sim 10^{100}\) candidates for materials

- First principles calculations are expensive

- Experiments are slow and expensive

Crystal structure and materials properties

- Structure-property relationships:
The rise of the materials databases
- Data are accessible

Chemical space descriptors exist
- Coulomb Kernel\(^1\), Bag of bonds representation\(^2\)

Data science tools exist
- Scikit learn, Google’s TensorFlow

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Artificial Intelligence for Materials Studies

Magnetic 2D crystals
Artificial Intelligence for Materials Studies

Magnetic 2D crystals

Transition metal trichalcogenides are magnetic 2D atomic crystals

- $\text{CrGeTe}_3$ is a ferromagnet (FM)$^{1,2}$
- Monolayer $\text{CrSiTe}_3$ is a zigzag antiferromagnet (zigzag-AFM)$^1$

$A_2B_2X_6$ crystal structure

1. Sivadas et al., PRB 91 235425 (2015)  
2. C. Gong et al., Nature 546, 265 (2017)
CrGeTe$_3$ is a ferromagnet (FM)$^{1,2}$

Monolayer CrSiTe$_3$ is a zigzag antiferromagnet (zigzag-AFM)$^1$

Transition metal trichalcogenides are magnetic 2D atomic crystals

Artificial Intelligence for Materials Studies

*Magnetic 2D crystals*

Magnetic structures

1. Sivadas et al., PRB 91 235425 (2015)  
Magnetic van der Waals materials

$A_2B_2X_6$ structures

- Create 198 ABX$_3$ structures using DFT
  - Total # of structures $\sim 10^4$
  - NM, FM and AFM spin configurations
- Extract data:
  - Formation energy
  - Magnetic order
  - Magnetic moment
Magnetic van der Waals materials

$A_2B_2X_6$ structures

Substitutions:

- **A site:**
  - $Cr_{0.5}A_{0.5}$
  - A: Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Y, Nb, Ru

- **B site:**
  - Si, Ge, P combinations
  - B: Si, Ge, P, $Si_{0.5}Ge_{0.5}$, $Si_{0.5}P_{0.5}$, $Ge_{0.5}P_{0.5}$

- **X site:**
  - S, Se, Te
Magnetic van der Waals materials

$A_2B_2X_6$ structures
Magnetic moment of $A_2B_2X_6$

Formation energy of $\text{A}_2\text{B}_2\text{X}_6$
Formation energy of $A_2B_2X_6$

Artificial intelligence in materials science

**Materials descriptors**

- Describe system using easily attainable components
  - Atomic properties, $p$

- Compound Property, $P$
  - $P = \text{mean}(p(A), p(B), p(X))$
  - $P = \text{std. deviation}(p(A), p(B), p(X))$

- Total # of descriptors:
  - 61

- Atomic property, $p$:
  - Number of spin up e’s
  - atomic radius
  - etc.
Machine learning models

- Random forest regression
  - Machine learning model
- Inputs: X
  - Number of spin up e's
- Output: Y
  - Magnetic moment
- Training data
- Test data

- Node m, region $R_m$, $N_m$ observations
- Mean Absolute Error
  - minimize L1 error using median values at terminal nodes

$L'(X_m) = \frac{1}{N_m} \sum_{i \in N_m} |y_i - \bar{y}_m|$

$\bar{y}_m = \frac{1}{N_m} \sum_{i \in N_m} y_i$

Machine learning predictions
Magnetic moment, X=Te

- $N = 262$
- Random forest $R^2 = 0.98$
- Mean absolute error (MAE) = 0.30 $\mu_B$

DFT: first-principles quantum calculations
$\mu$: magnetic moment $\sim$ magnetization

Machine learning predictions of DFT formation energy

Random Forests

$R^2 = 0.913$

DFT Formation Energy

Machine learning predictions of DFT formation energy

**Kernel Ridge**

$R^2 = 0.972$

**Random Forests**

$R^2 = 0.913$

**Neural Network**

$R^2 = 0.978$

Machine learning results

Magnetic moment

Formation Energy
Candidates for chemically stable 2D ferromagnets

- Initial DFT data (198 structures)
- ML predictions (> 4000 structures)

Screening Criteria:
- Magnetic moment > 5 $\mu_B$ & Formation energy < -1.1eV

DFT results:
- $(\text{CrMn})\text{Si}_2\text{Te}_6$
- $(\text{CrMn})\text{Ge}_2\text{Se}_6$
- $(\text{CrFe})(\text{SiP})\text{S}_6$

ML Results:
- $(\text{CrMo})\text{Si}_2\text{Te}_6$
- $(\text{CrW})\text{Si}_2\text{Te}_6$
- $(\text{CrMo})(\text{SiP})\text{Te}_6$

AI-guided materials discovery

Outcomes

- Identified new chemically stable 2D ferromagnets
- AI provides fast estimates of 2D materials’ properties
  - > 4000 $A_2B_2X_6$ composites
  - Magnetic properties
  - Chemical stability
- Non-traditional framework for studying emergent spin phenomena in 2D materials

Summary

- Aron’s invaluable guidance shaped the lives and careers of many
- A collection of people, under the right conditions, can give rise to ‘emergent phenomena’
- AI-guided identification of emergent spin phenomena in materials
Acknowledgements

- This work used the Extreme Science and Engineering Discovery Environment (XSEDE), which is supported by National Science Foundation grant number ACI-1548562.

- Some of the computations in this paper were run on the Odyssey cluster supported by the FAS Division of Science, Research Computing Group at Harvard University.

- This research used resources of the Argonne Leadership Computing Facility, which is a DOE Office of Science User Facility supported under Contract DE-AC02-06CH11357.

- This material is based upon work supported by the National Science Foundation CAREER award under Grant No 2044842.
Aron’s Early Influence – Postdoctoral studies
Postdoc at NTT, Japan
Charge properties of electrons in GaAs quantum wells
NMR probes Wigner Solids: *In-plane local density variations at* $\nu \sim 2$

NMR profiling of quantum electron solids in high magnetic fields

1) L. Tiemann(†), T.D. Rhone(†), N. Shibata, K. Muraki, Nature Physics, doi:10.1038/nphys3031

(†: equal contribution)
Nuclear Magnetic Resonance (NMR) Knight Shift

\[ \text{Knight Shift} \]

\[ K_S \propto P \]

Nuclear spin \( I = 3/2 \)

Electron spin \( P = 0 \)

\( P > 0 \)

\( P = 0 \)

AlGaAs

GaAs

AlGaAs

GaAs (100)
Simulating RD-NMR Spectra

\[ I_v (f - f_0) = \int_{-w/2}^{w/2} g(f - f_0 + \alpha_v |\psi_v(z)|^2) |\psi_{\text{read}}(z)|^2 \, dz \]

(spins polarization \( \alpha_v \)) \( \times \) (electron density)

NMR signal

Frequency (MHz)

\( v = 1/3 \)
Materials descriptors: data visualization

- Compound Property, $P$
  - $P = f(p(A), p(B), p(X))$
- $p$:
  - Enthalpy
  - Polarizability ($dipole_{std}$)
  - Ionization energy ($std_{ion}$)
  - # valence electrons ($nvalence_{avg}$)
Quantum Well

\[ \omega_c = \frac{eB}{m^*c} \]

Spin gap is the Zeeman energy

\[ \nu = \frac{n}{(B/\phi_0)} \]
What is Materials Informatics?

Teaching computers materials science using machine learning!
2DES in varying LL regimes

N = 0 LL, \( \nu = \frac{1}{3} \)

N = 1 LL, \( \nu = 3 \)
NMR probes Wigner Solids: *In-plane local density variations at* $\nu \sim 2$

1) L. Tiemann*, T.D. Rhone*, N. Shibata, K. Muraki, Nature Physics, doi:10.1038/nphys3031

(*: equal contribution)
Dataset: Transition Metal Dichalcogenides (TMDs)

MX$_2$

Dataset: Transition Metal Dichalcogenides (TMDs)

MX$_2$

Heat of Formation of TMDs

Data visualization: TMDs

MX$_2$ materials ‘features’
- Atomic properties:
  - $\Delta x = x(M) - x(X)$
- $x$:
  - Polarizability
  - Ionization energy
  - Covalent radius
  - Electronegativity
Machine Learning models

- Linear regression is a type of machine learning model
- Inputs: \( X \)
  - Dipole polarizability of A, B
- Output: \( Y \)
  - Heat of formation
- Training data
- Test data

- Algorithm estimates \( m \) & \( c \)
  - Reduce test error
  - \( \min \{ \sum_i (y_i - \hat{y}_i)^2 \} \)

\[
f(x) \approx Y = mX + c + \varepsilon \\
Y = \{y_1 + y_2 + \ldots\} \\
X = \{x_1 + x_2 + \ldots\}
\]
Machine learning TMD data to predict stability

$$y = f(x_1, x_2, \ldots, x_N)$$

- Heat of formation
- Dipole polarizability difference
- # valence electrons difference
- Electronegativity difference
Machine learning TMD data to predict stability

Heat of formation of Boron Nitride

- Predicted value: \(-2eV\)
- Expected value: \(-1.56eV\)
Machine Learning models: Kernel ridge regression

- # data points = 89
- # features = 30

Test score, $R^2 = 0.94$
Train score, $R^2 = 0.99$

\[
C = \frac{1}{2} \sum_{i} (y_i - w^T x_i)^2 + \frac{1}{2} \lambda ||w||^2
\]
Machine learning predictions
Magnetic moment, X=Te

- $N = 262$
- Random forest $R^2 = 0.98$
- Mean absolute error (MAE) = 0.30 $\mu_B$

DFT: first-principles quantum calculations
$\mu$: magnetic moment $\sim$ magnetization
Machine learning predictions
Magnetic moment, X=Te

- Predicted \( \mu \)
- Training data
- Test data

Top 6 descriptors

Machine Learning models: Kernel ridge regression

Overfitting
Machine Learning models: Random forest regression

Random forest regression is based on decision trees

- # data points = 89
- # features = 39

Test score, MSE = 0.095
Random Forest model and Decision Trees

Is a customer at a company likely to buy a computer or not?
Machine Learning models: Random forest regression

**Feature Importances**

Top ten descriptors:

1. Dipole dif
2. Dif in ionization E
3. atomic radius avg
   - num valence dif
   - atomic vol dif
   - e_negativity avg
covalent rad avg
vdw radius dif
dipole avg
10 covalent rad dif
Challenges

1. Can our ML models generalize to other 2D materials?
2. Are our descriptors sufficient?
3. Develop descriptors to describe crystal structure?

Top ten descriptors

1. Dipole dif
2. Dif in ionization E
3. atomic radius avg
   . num valence dif
   . atomic vol dif
   . e_negativity avg
   covalent rad avg
   vdw radius dif
   dipole avg
10. covalent rad dif
Summary

- Materials informatics
  - Identified and exploited trends in materials’ properties data for vdW materials
  - Predicted heat of formation of TMDs

- Materials databases + access to machine learning tools makes materials informatics possible
Outlook

- Discovery of new 2D materials?
- Tailor 2D materials to have desired properties?
- Use statistical inference to guide the discovery of new physics?
Outlook

- Academic research
  - Universities
  - Materials Project (Materials Genome Initiative)
  - NIMS

- Industry
  - Citrine
  - Google
  - Applications:
    - Photovoltaics
    - Thermoelectrics
    - Organic LEDs
    - Magnetocalorics
2DES in a magnetic field: Integer Quantum Hall Effect

Integer Quantum Hall Effect

Fractional Quantum Hall Effect


Emergent Quasiparticles: Composite Fermions

IQHE
\[ \Psi_{\nu=1} = \prod_{j<k} (z_j - z_k) \exp \left[ -\frac{1}{4\gamma^2} \sum_l |z_l|^2 \right] \]

FQHE
\[ \Psi_{\nu/(\nu+1)} = \hat{P}_{LLL} \prod_{j<k} (z_j - z_k) \Psi_{\nu=p} \]

\[ \nu^* = n \phi_o / |B^*| \quad \nu = \frac{\nu^*}{\phi \nu^* + 1} \]

Spin properties of 2DES

T.D. Rhone et al, PRL 106, 196805 (2011)
Resistively detected NMR (RDNMR) as a Local Density Probe

\[
\text{(effective field exerted on nuclear spins)} \propto \text{(electron spin polarization)} \times \text{(local electron density)}
\]
Resistively Detected NMR (RD-NMR)

Electrically Controlled Nuclear Spin Polarization and Relaxation by Quantum-Hall States

K. Hashimoto, K. Muraki, T. Saku, and Y. Hirayama

\[ E_z = g \mu_B (B_0 + b_n \langle I_z \rangle) \]

\[ \Delta E_z \propto \Delta \langle I_z \rangle \]

if \( \Delta R_{xx} \propto \Delta E_z \), then \( \Delta R_{xx} \propto \Delta \langle I_z \rangle \)

only applicable to those states with:

- sensitivity to \( \Delta E_z \)
- finite \( R_{xx} \)
Resistively Detected NMR (RD-NMR)

\[ E_z = g \mu_B (B_0 + b_n \langle l_z \rangle) \]

\[ \Delta E_z \propto \Delta \langle l_z \rangle \]

if \( \Delta R_{xx} \propto \Delta E_z \), then \( \Delta R_{xx} \propto \Delta \langle l_z \rangle \)

only applicable to those states with:
- sensitivity to \( \Delta E_z \)
- finite \( R_{xx} \)

Our Method: Modified RD-NMR

Key idea:
Use gate voltage to go to a different filling factor $v_{\text{read}}$ for the readout

Detect resonant rf absorption at $v$ as a change in $R_{xx}$ at $v_{\text{read}}$

- Nuclear spins interact with electron spins at $v_{\text{read}}=2/3$
- $R_{xx}$ at $v_{\text{read}}$ can be used to measure spin degrees of freedom
**NMR Line Shape**

\[ K_s(r) \text{ (Knight shift of nucleus at position } r) = \alpha_v \left( \text{spin polarization} \times n \right) \times \rho(r) \text{ (local probability density)} \]

\[ P = \frac{\alpha_v}{\alpha_{\text{max},v}} \]

- For \( P > 0 \), NMR line shape \( \cong \) mirror image of local density histogram
- spectral mapping of subband wavefunction
**NMR probes Wigner Solids: *In-plane local density variations at ν ~ 2***

1) L. Tiemann(*), T.D. Rhone(*), N. Shibata, K. Muraki, Nature Physics, doi:10.1038/nphys3031

(*: equal contribution)
"I Keep Six Honest Serving Men ..."

I keep six honest serving-men
(They taught me all I knew);
Their names are What and Why and When
And How and Where and Who...

by Rudyard Kipling
Crystal structure and materials properties

- Structure-property relationships:

Graphene

Boron Nitride
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Reduced set of descriptors

1 covalentrad_max_dif  11 nvalence_std_dif
2 hardness_var       12 dipole_std
3 hardness_mean      13 covalentrad_std_dif
4 covalentrad_avg    14 dipole_max_dif
5 sum_ion            15 nvalence_sum_dif
6 Nup_mean           16 atomic_vol_std_dif
7 Nup_var            17 nvalence_max_dif
8 atomic_rad_std_dif 18 atomic_vol_sum_dif
9 atomic_vol_avg     19 nvalence_std
10 atomic_vol_max_dif 20 nvalence_avg
Random Forests Regression

Decision Tree Regression

```
data
max_depth=1
```

```
target
0.0
0.5
1.0
1.5
```

```
data
0
1
2
3
4
5
```

```
Random Forests Regression

Decision Tree Regression

- Blue line: max_depth=1
- Green line: max_depth=2

Data points are shown in orange circles.
Random Forests Regression

Decision Tree Regression

- max_depth=1
- max_depth=2
- max_depth=3

Data points are plotted against the target variable, with different colors indicating the maximum depth of the decision trees.
Random Forests Regression

Decision Tree Regression

- max_depth=1
- max_depth=2
- max_depth=3
- max_depth=5
- data
Extra forest regression: Feature extraction

Feature Importance Ranking:

1. Ionization energy dif.
2. # valence electrons avg.
3. Atomic radius dif.
4. Dipole polarizability dif.
5. Num of electrons avg.
7. Covalent radius dif.
8. Atom E_AB
9. Dipole polarizability avg
10. Ionization energy sum
11. # of valence electrons dif
Test score vs training set size
Where and who of Materials Informatics

- You are an alchemist living in ancient Greece
- You have a small random set of elements
Where and who of Materials Informatics

- What is a good descriptor?
- What are trends in the data?
Where and who of Materials Informatics

- Descriptor:
  - Atomic number
- Apply a model:
  - Sort atomic numbers
Where and who of Materials Informatics

- Descriptor:
  - Atomic number
- Apply a model:
  - Sort atomic numbers
Where and who of Materials Informatics

- Descriptor:
  - Atomic number

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Machine Learning for Materials studies

Data

Materials Project

AFLOW

Automatic - FLOW for Materials Discovery

MatNavi

NIMS Materials Database

MATDAT.COM

Material Properties Database and Estimation Tools

Inorganic Crystal Structure Database
Kitchen sink method:
  • Use statistical analysis to learn relevant descriptors

Domain knowledge:
  • Construct features that are important for describing a system

Example:
  • Ionic compounds have atoms in different columns of periodic table
  • Descriptor: column # of the periodic table
Machine Learning for Materials studies

Data science tools

Statistical models

- MongoDB
- Spark
- Python
- pandas
- MySQL
- SQL

Joins are what RDBMS’s do for a living
If you change the way you look at things, the things you look at change.
AI usually beats natural stupidity
Don’t trust everything you see, even salt looks like sugar