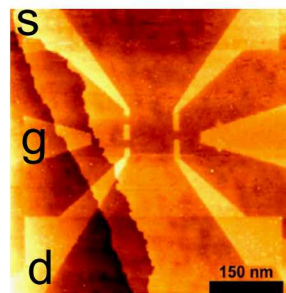
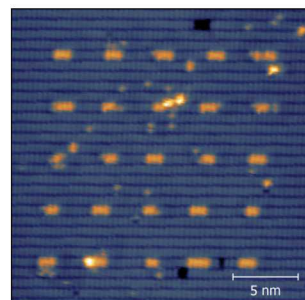


Designing quantum materials in silicon, atom by atom

Shashank Misra

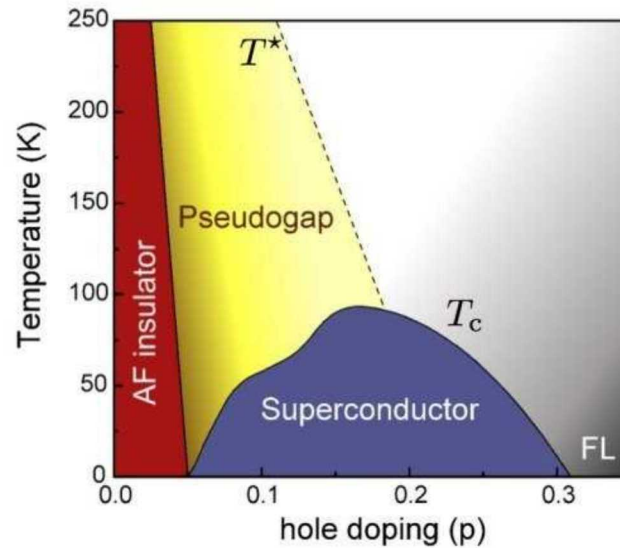
NIST Workshop on Quantum 2D Metamaterials

April 25-26, 2018

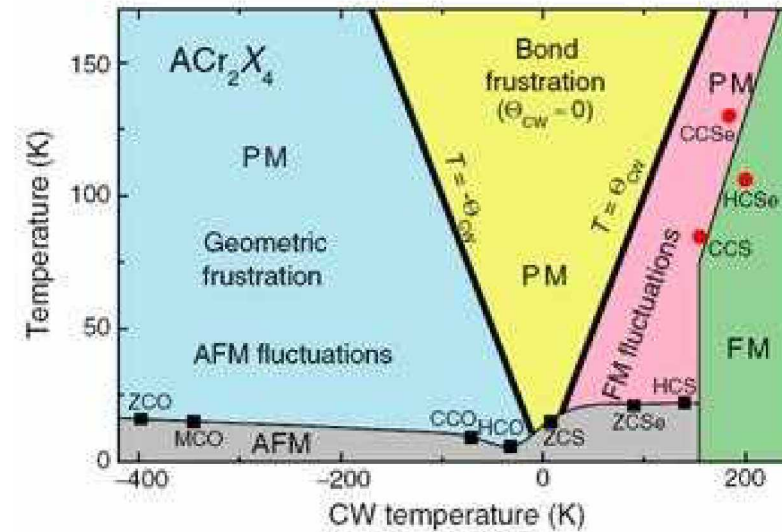


Quantum materials

*Strong correlations-
cuprate superconductors*



*Frustration-
Chromium spinels*



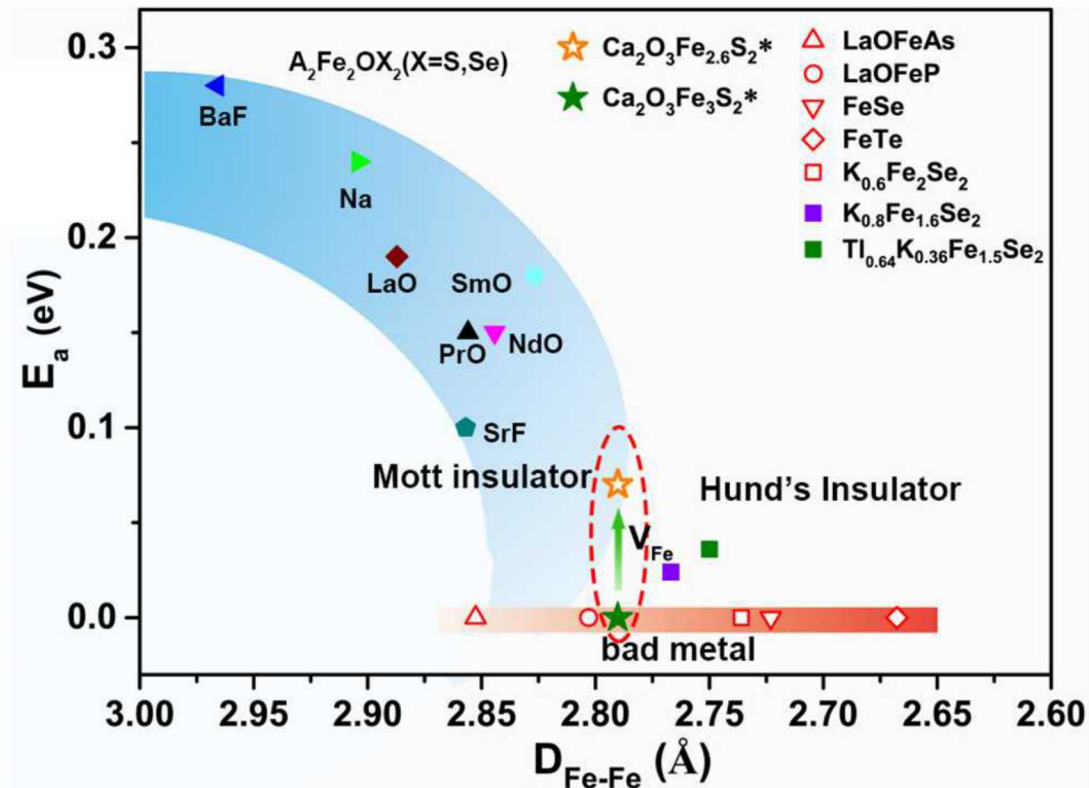
Rudolph, NJP (2007)

Quantum materials have emergent many-body effects that cannot be predicted from single-electron point of view.

What limits experimental progress?

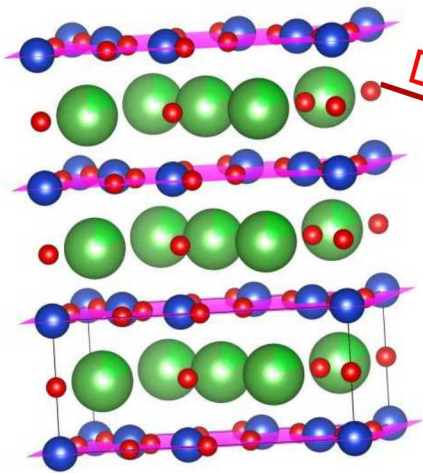


Higgins, Saha (NIST)



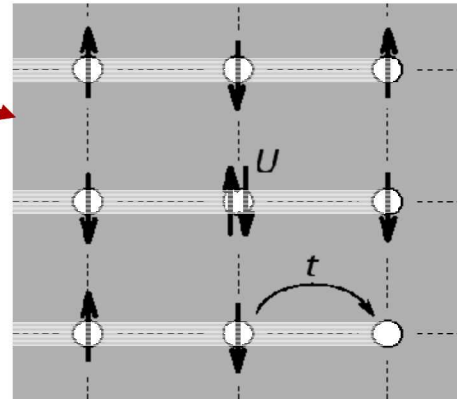
Opportunity limited by what you can synthesize

What limits theory/ modeling progress?



Deep insight

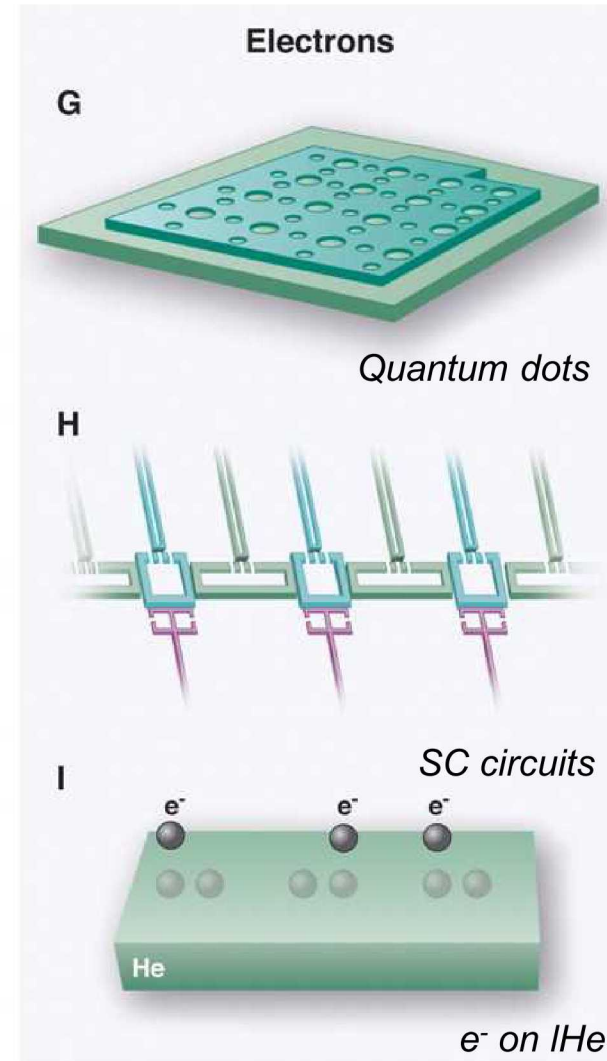
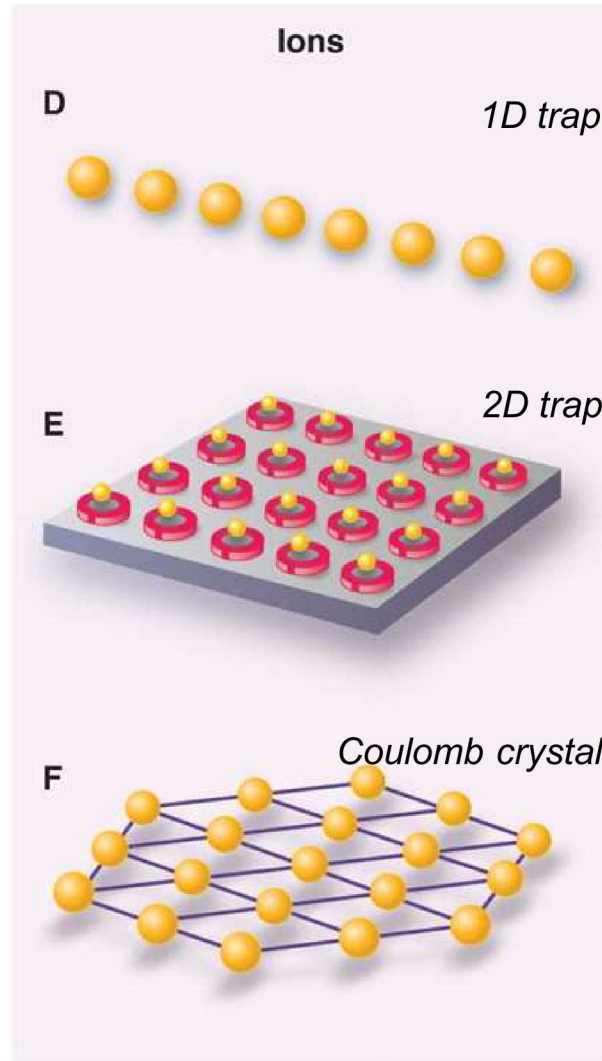
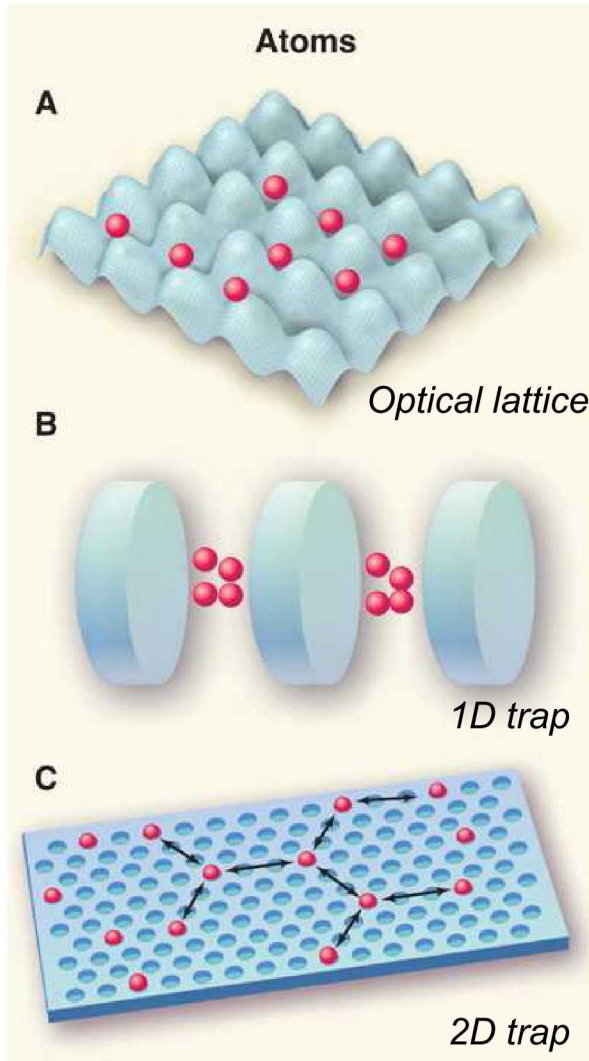
Underlying lattice,
and implied Hamiltonian



- **Classical computation:** Exponential growth (2^N for Ising) of problem with cluster size.
- **Digital quantum computation:** Very few bits available.
- **Quantum simulation:** create a small tunable quantum system to mimic a more complex system.

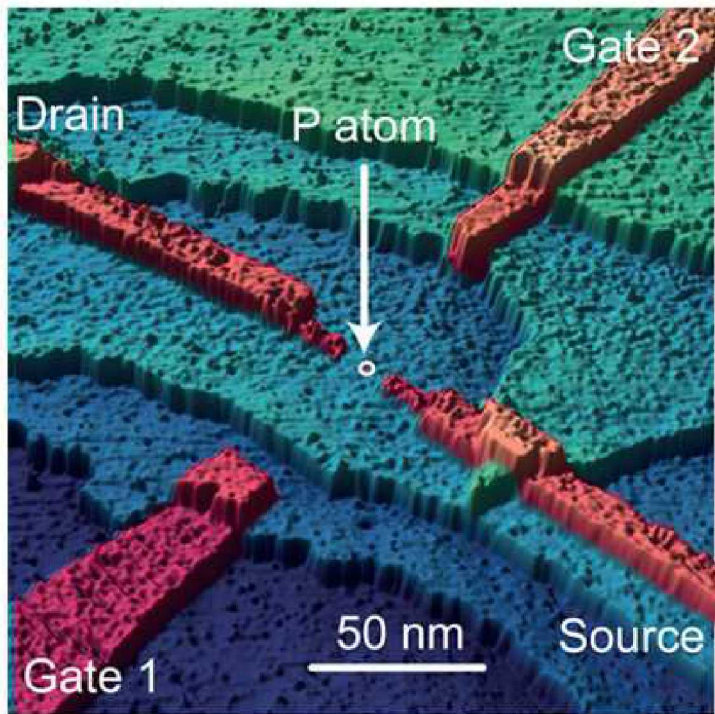
1. **Guide modeling: benchmark modeling approaches**
2. **Guide synthesis: simulate a material before you make it.**

An engineered approach to quantum materials

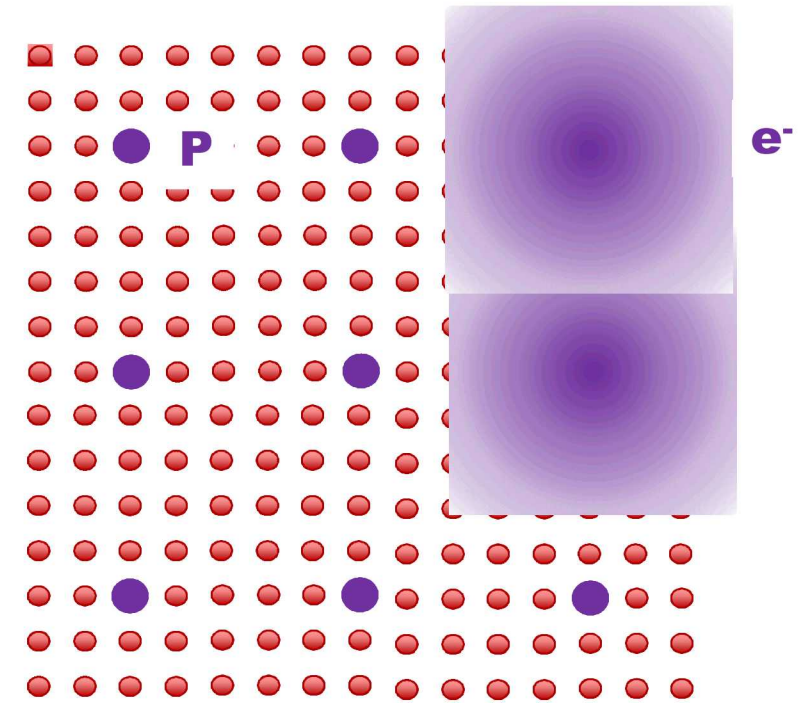
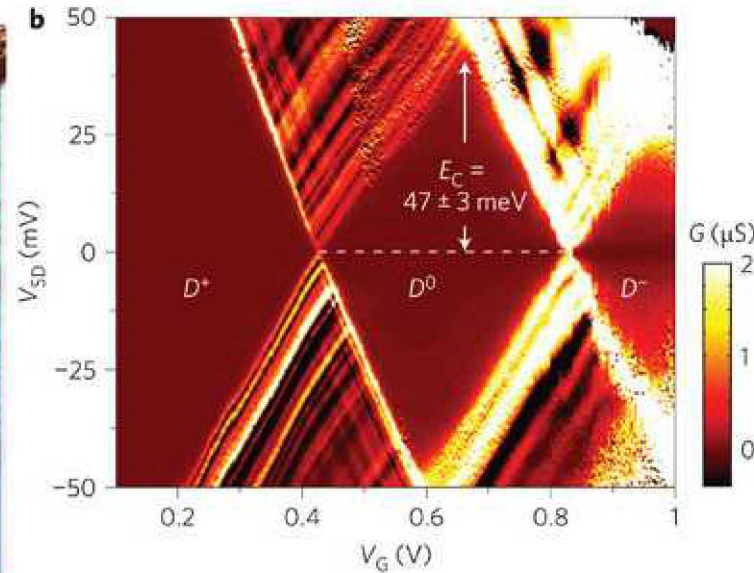


Cheng Chin- *cold atoms*
Alicia Kollar- *SC circuits*
Sjaak van Diepen- *quantum dots*
Ingmar Swart- *assembled structures*
Kaden Hazard- *cold atoms*
Norbert Linke- *ions*
Garnett Bryant- *photons*
Philip Phillips- *twisted bilayer graphene*

Designing quantum materials, using donors in Si



Single P atom in silicon



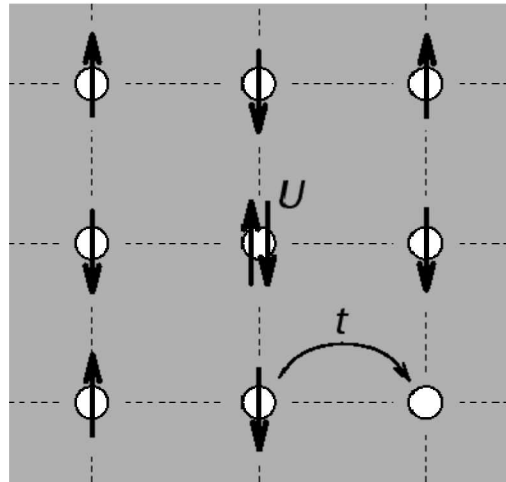
Jon Wyrick & Enrico Prati- donors

Workshop: What role could donor-based arrays play in the study of quantum materials?

This talk: What are opportunities and limitations, as a practitioner.

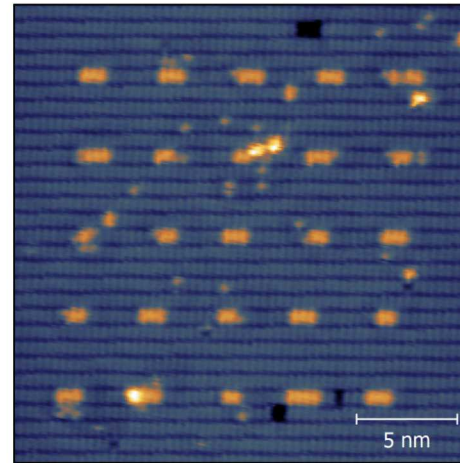
Designing quantum materials, atom by atom

Why emulate quantum materials with donor arrays?



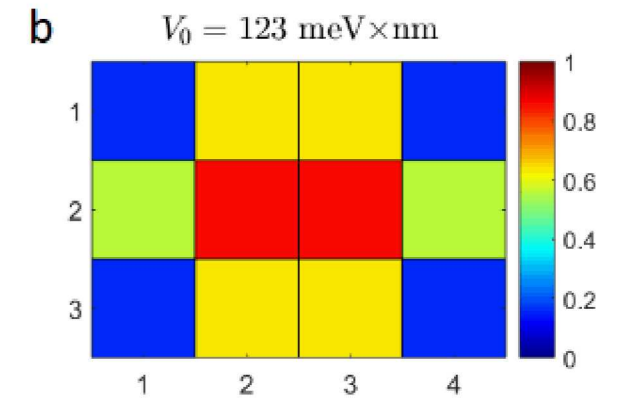
Hubbard model

What donor array can we make?



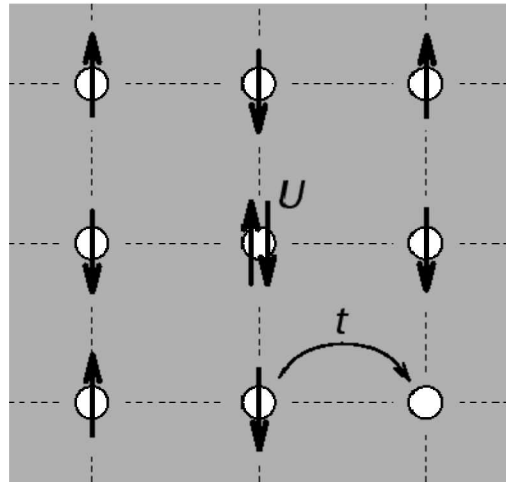
Donor array

What can we measure?



Electrons localized to middle of 3 x 4 array

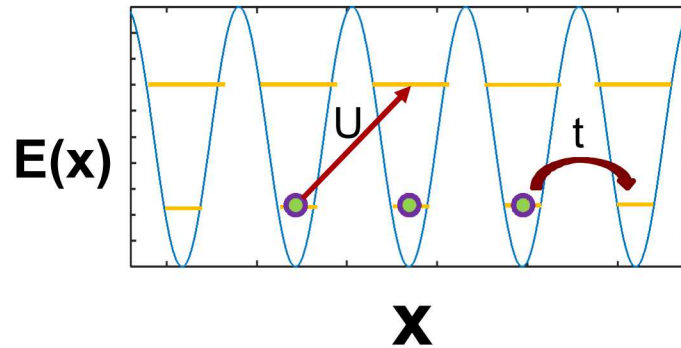
Designing quantum materials, atom by atom



Hubbard model

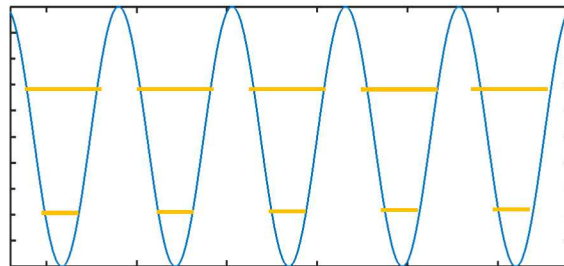
Why emulate quantum materials with donor arrays?

Fermi-Hubbard model

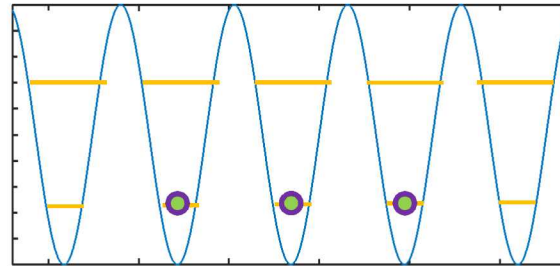


3 parameters: hopping rate t ,
occupancy penalty U , and filling n

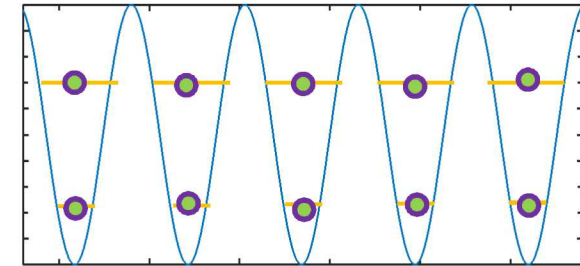
$n=0$ → Band insulator



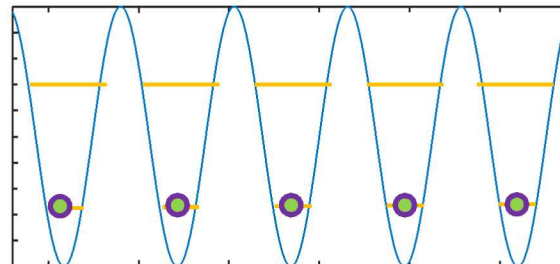
$n \neq 1$ → metal



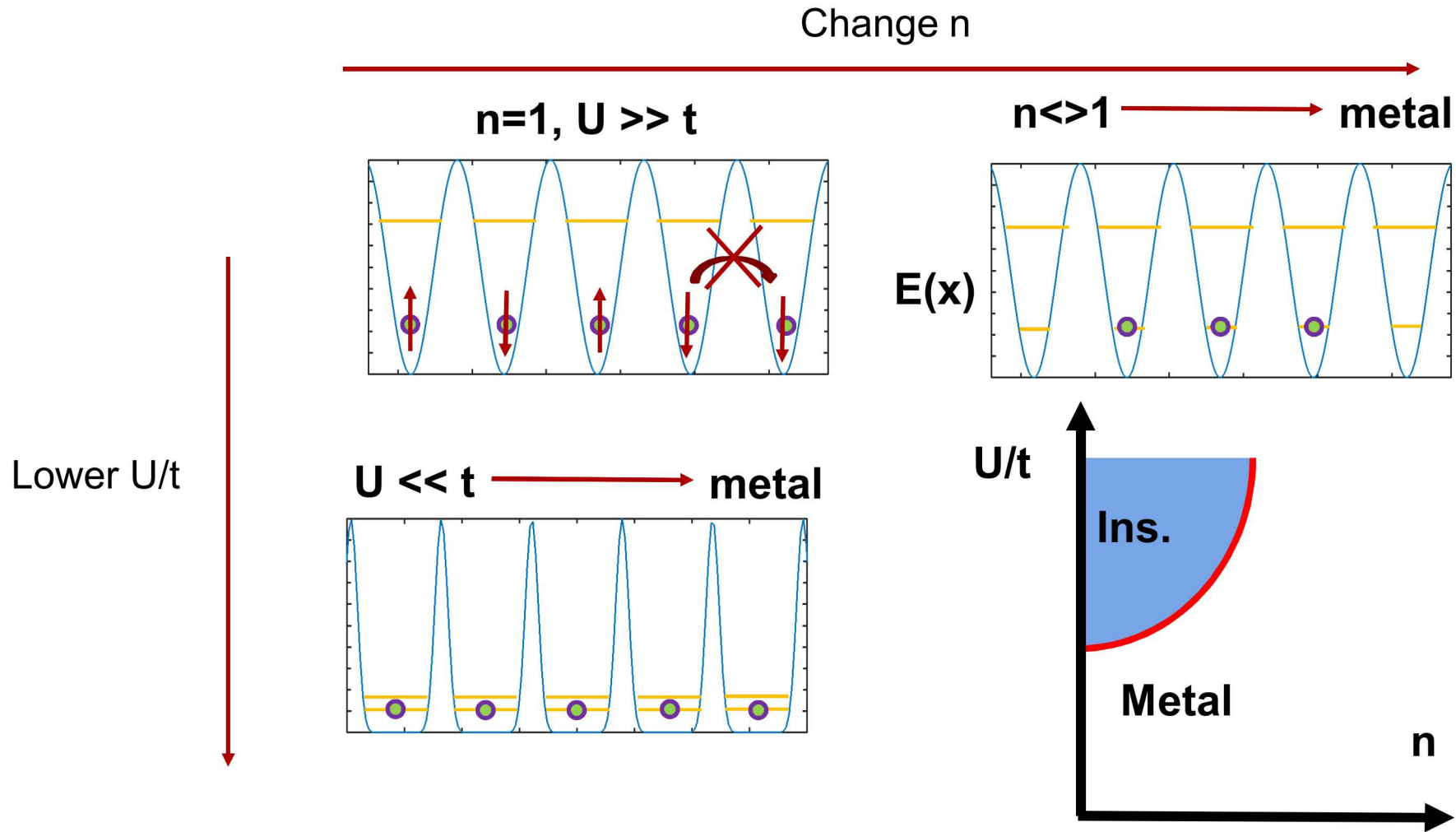
$n=2$ → Band insulator



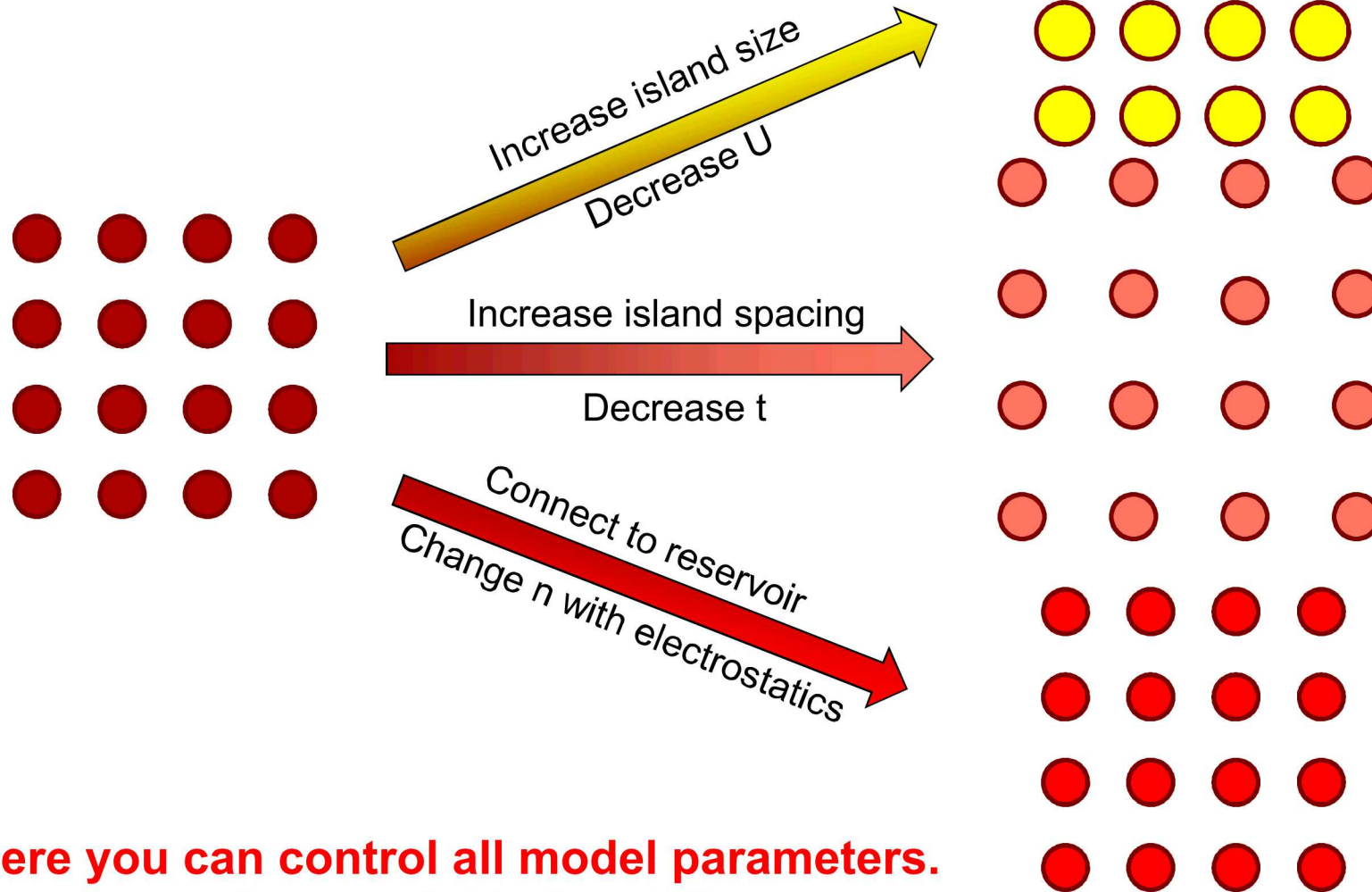
$n = 1$ → insulator



Mott insulator

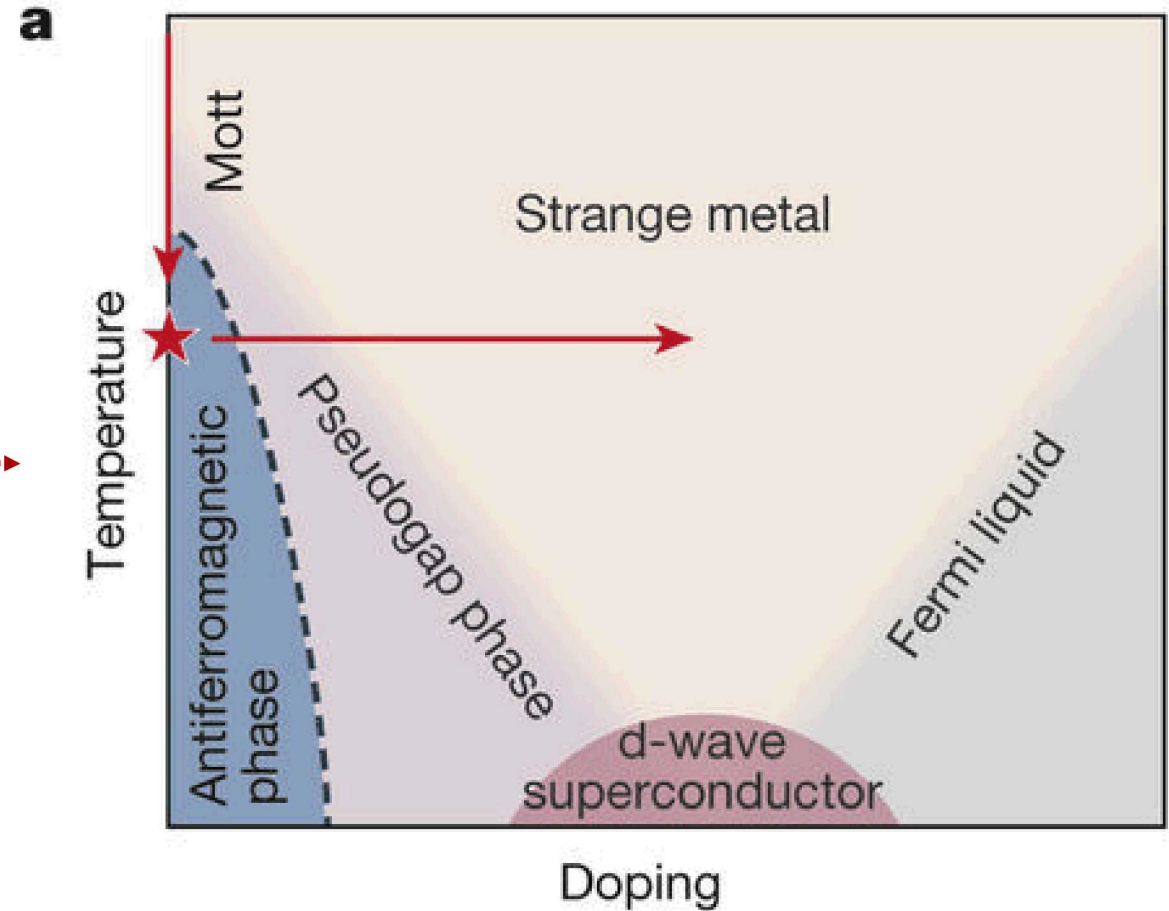
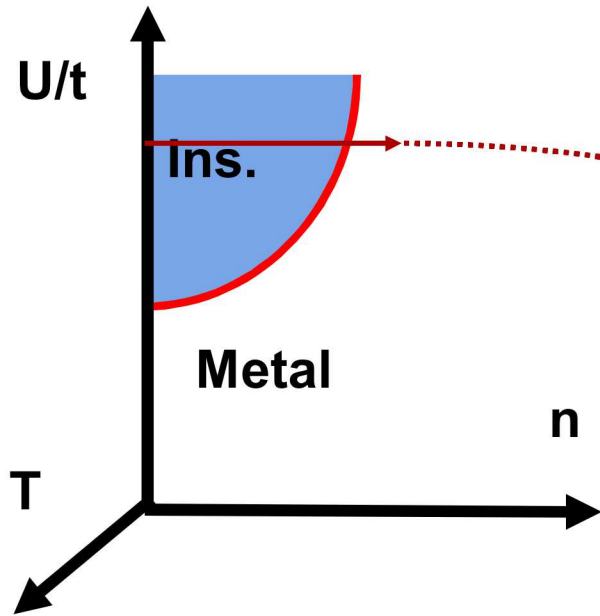


Designer material



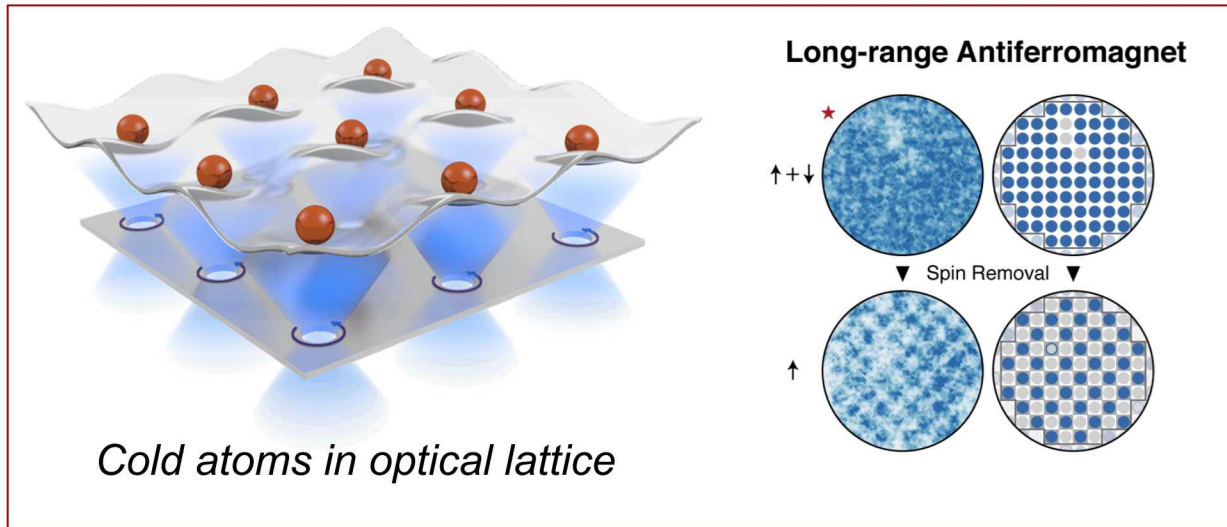
**Array where you can control all model parameters.
Expect crossovers from metal to insulating.**

Strongly correlated system

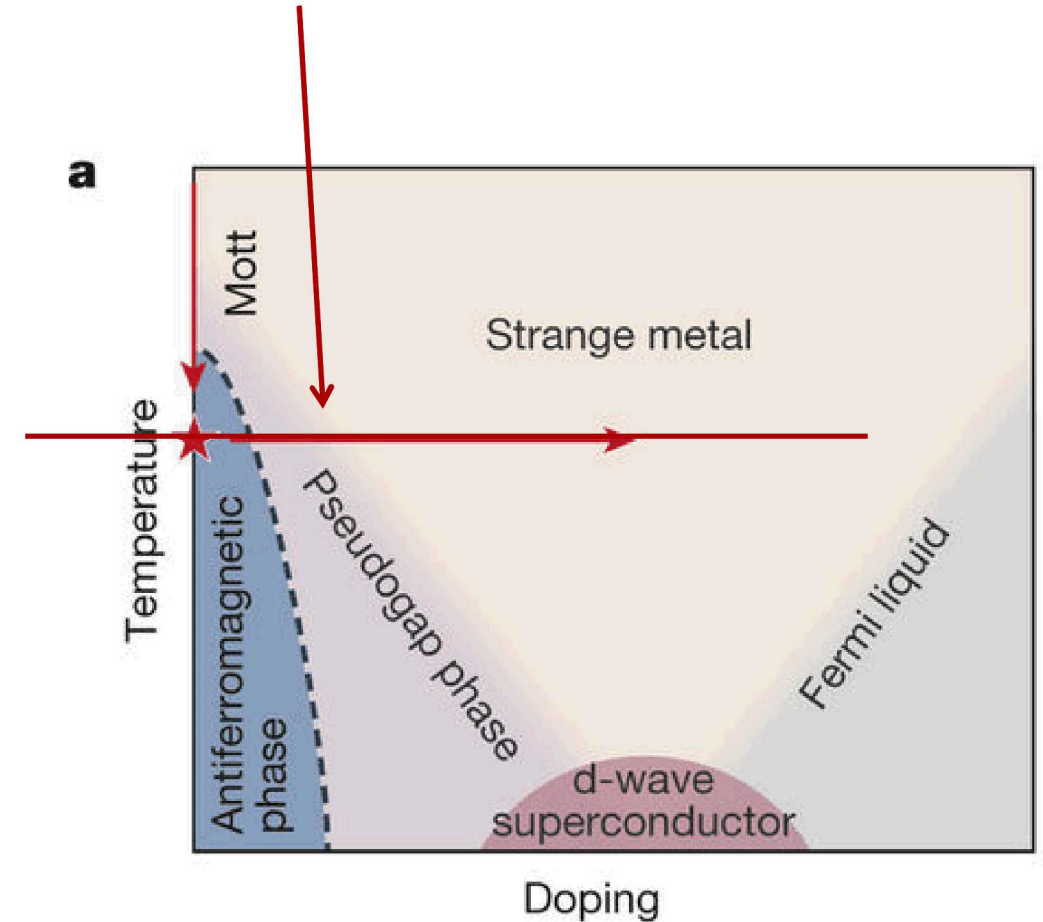


“Interesting” phases at the boundary of the insulator and the metal in quasi-2D.

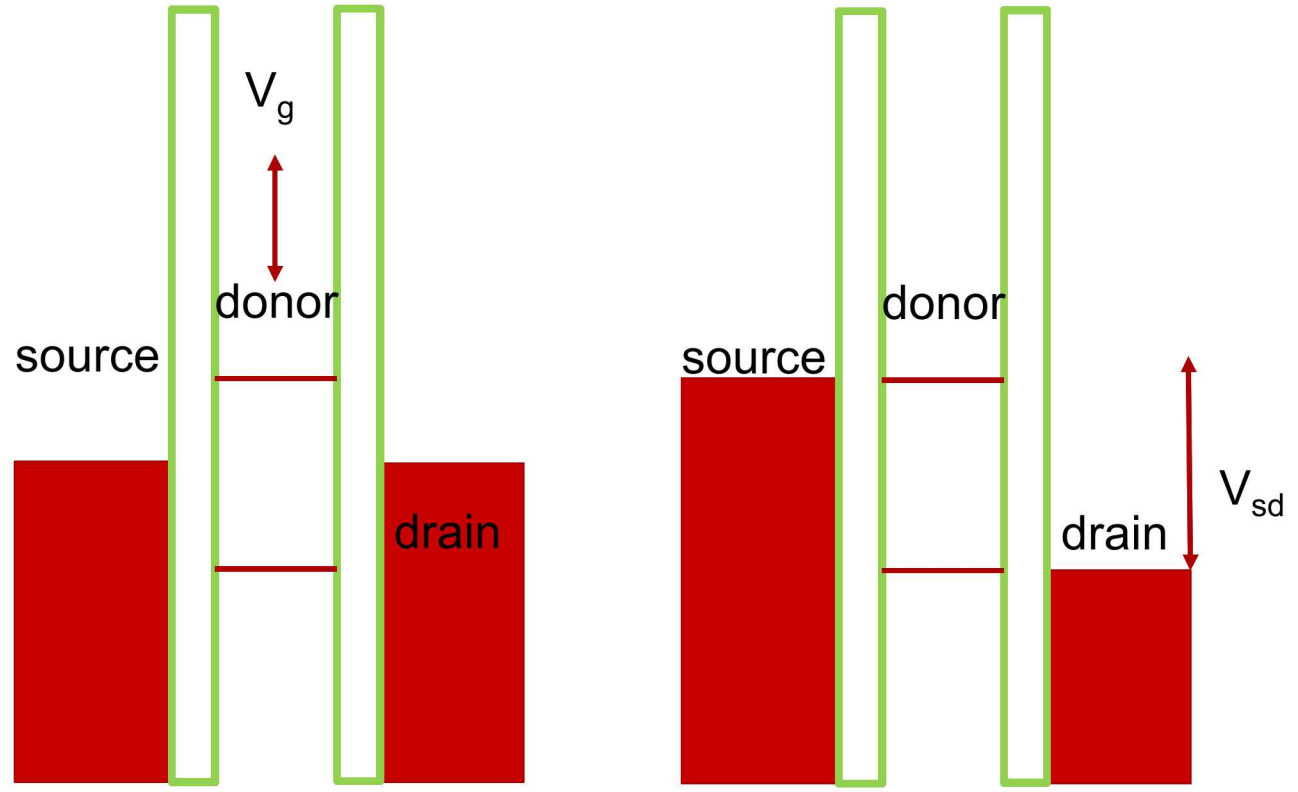
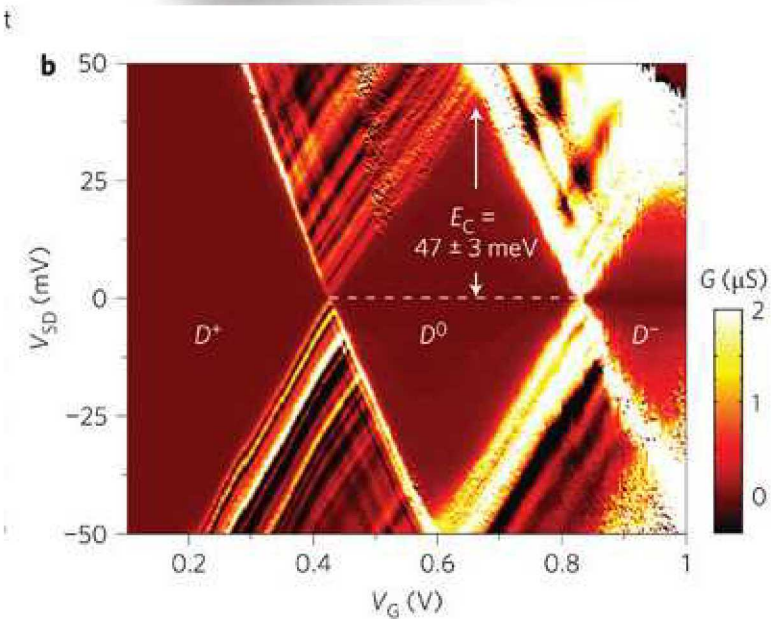
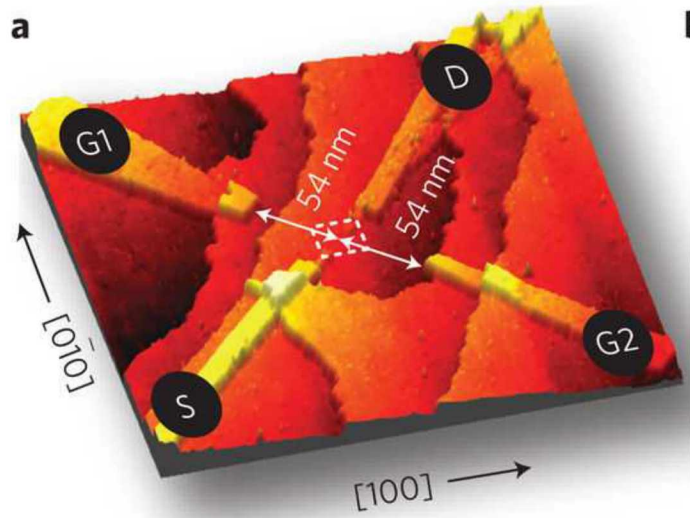
What is new about donors?



Fermi-Hubbard in Li
 $U/t \sim 7$ (strongly interacting)
 $T/t \sim 0.25$ (moderate temperature)



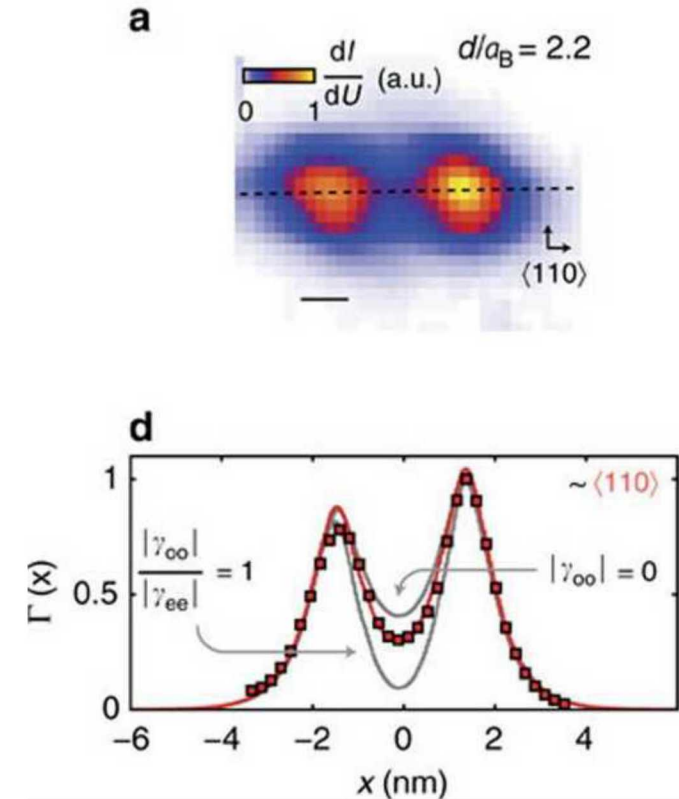
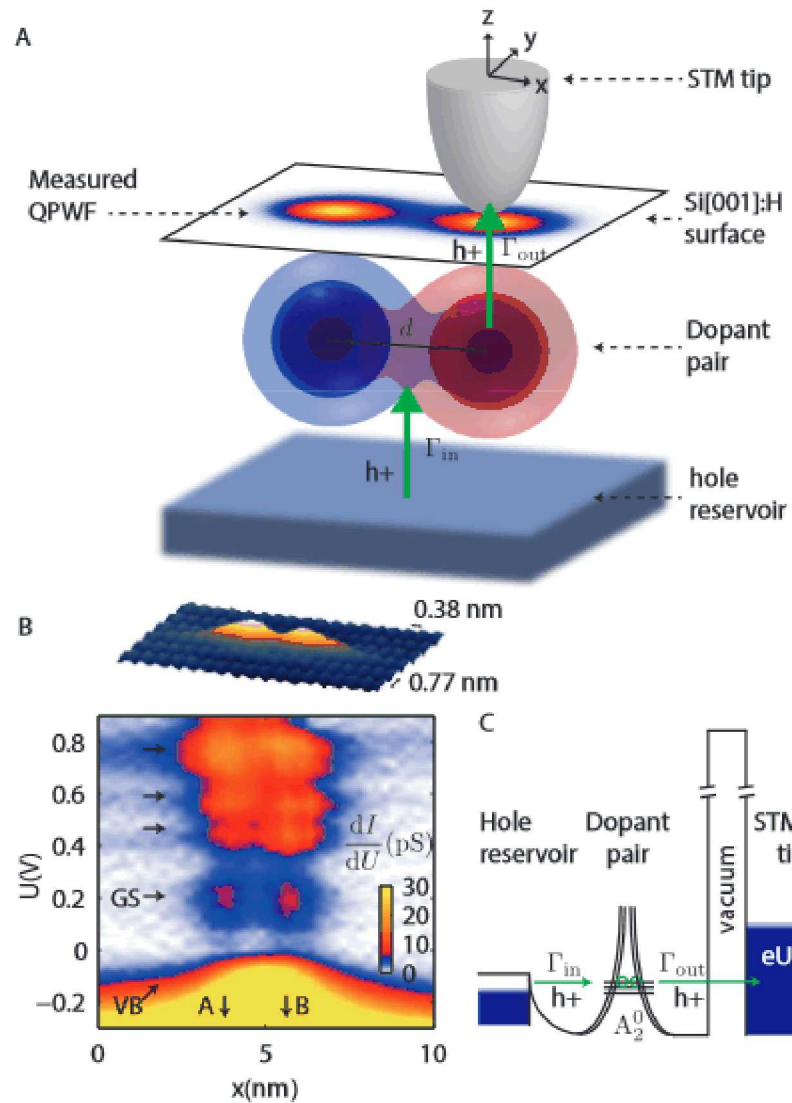
Donors' Hubbard parameters



Single P donor: $U \sim 47 \text{ meV}$

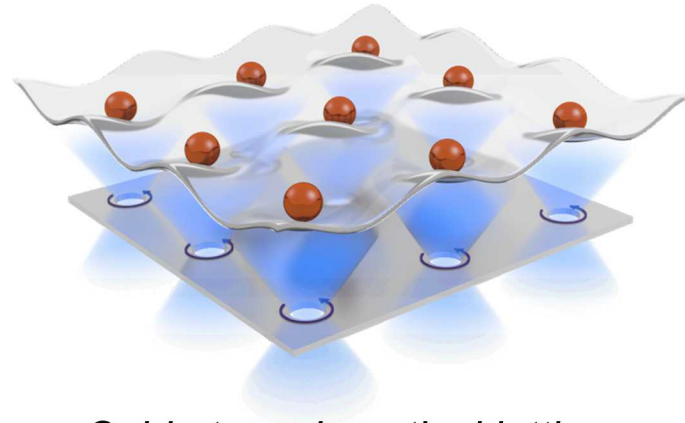
$T_{DR} \sim 0.01 \text{ meV}$

Acceptors' Hubbard parameters



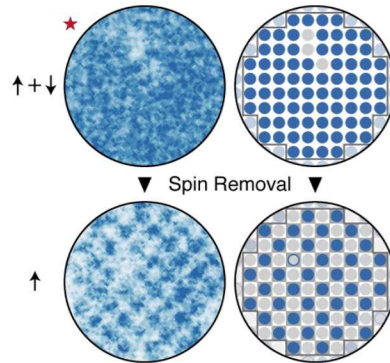
$t \sim 3 - 12$ meV for **Boron** @ separations 2.9 – 4.6 nm
 $T_{DR} \sim 0.01$ meV

What is new about dopants?



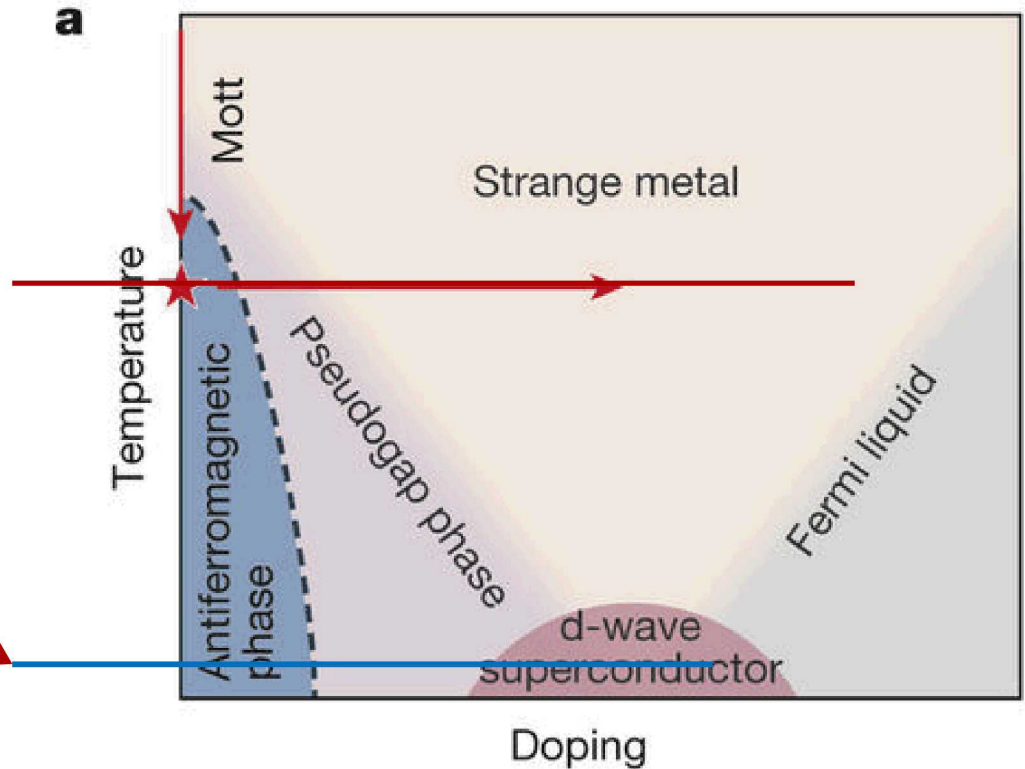
Cold atoms in optical lattice

Long-range Antiferromagnet



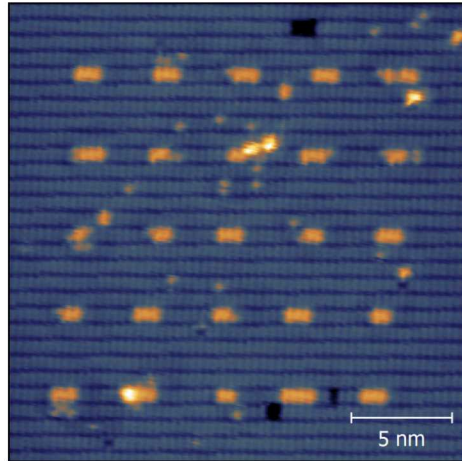
Fermi-Hubbard in Li
 $U/t \sim 7$ (strongly interacting)
 $T/t \sim 0.25$ (moderate temperature)

- Donors**
- Strongly interacting
 $U/t \sim 5 - 500$
 - Low temperature
 $T/t \sim 0.001 - 0.1$



Dopants in Si will give you access to the “interesting” low temperature phases

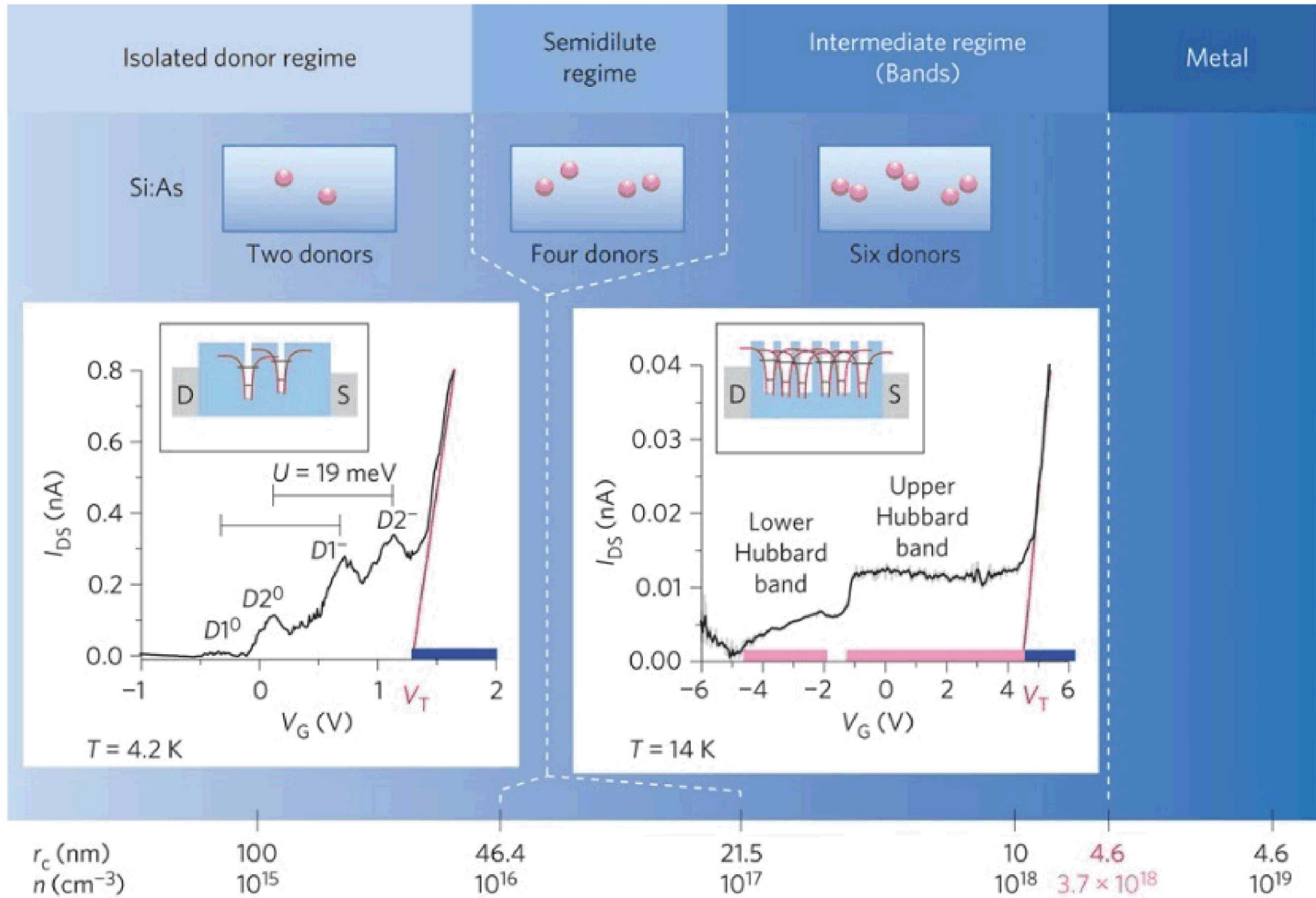
Designing quantum materials, atom by atom



Donor array

What donor array can we make?

Traditional fabrication route- ion implantation

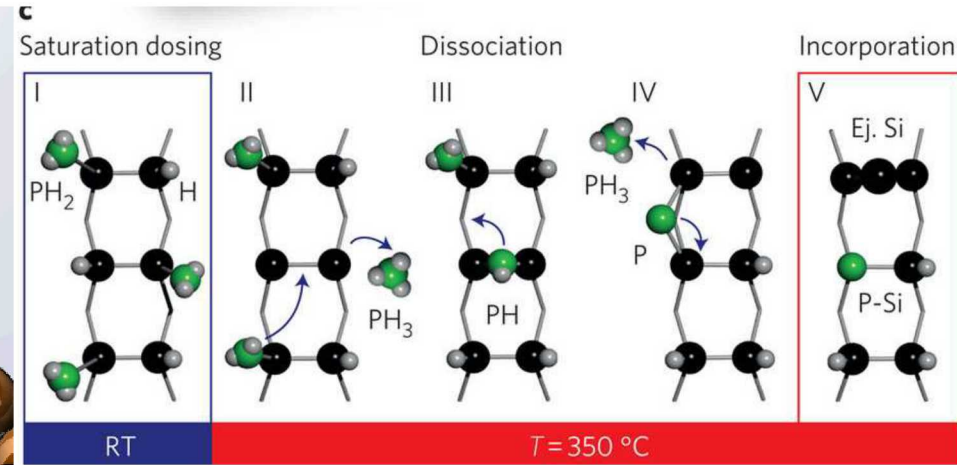
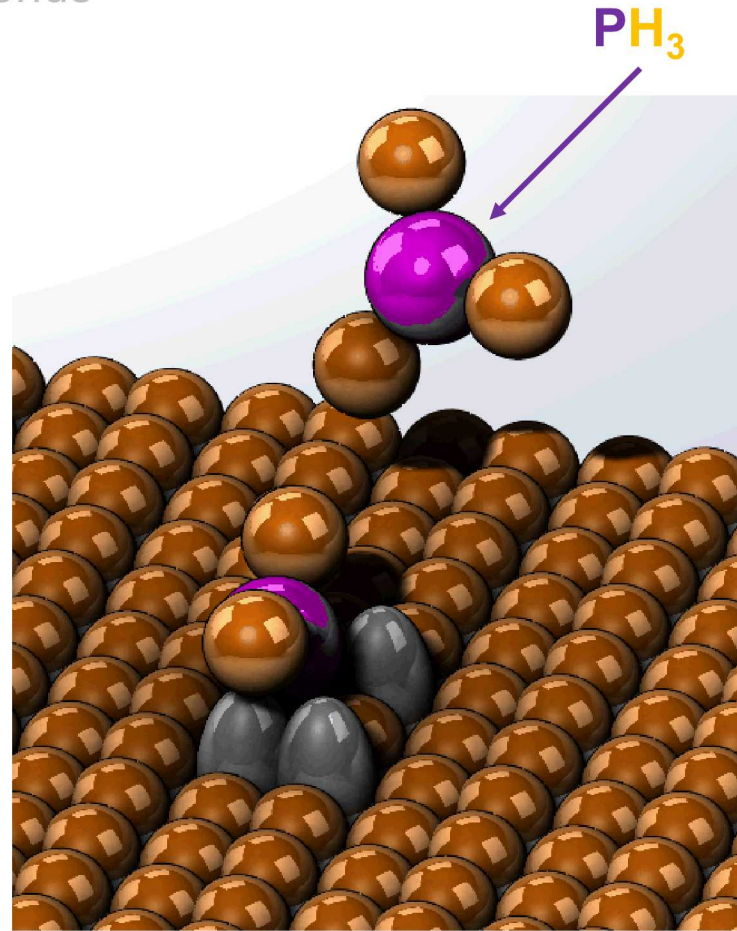
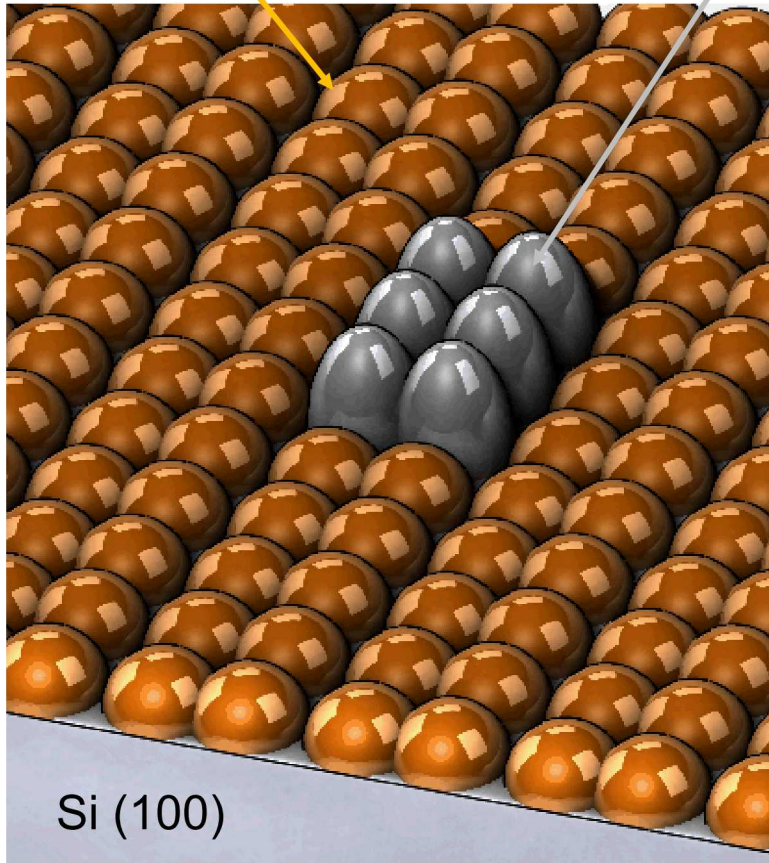


Ion implantation has straggle ~ 10 nm

Enrico Prati

A different route: hydrogen lithography

H-terminated Si Si dangling bonds

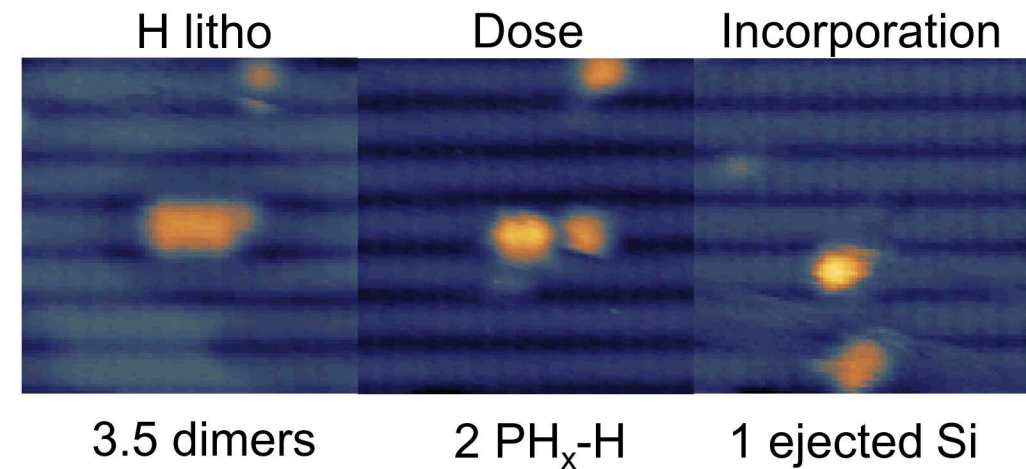
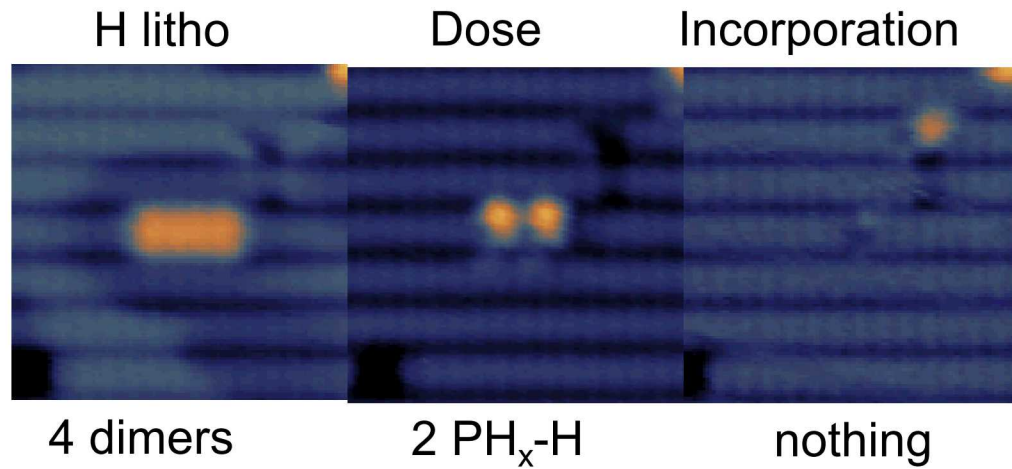


3 by 1 dimer window yields 1 P donor.

Make an array of P donors with STM & hydrogen lithography.

Jon Wyrick

Surface chemistry of single donor incorporation



$$\Gamma_{A \rightarrow B} = A_{A \rightarrow B} \exp(\Delta_{A \rightarrow B}/k_B T)$$

Start with kinetics in which PH₃ *never* desorbs

Chemistry is most sensitive to PH₃ desorption rate

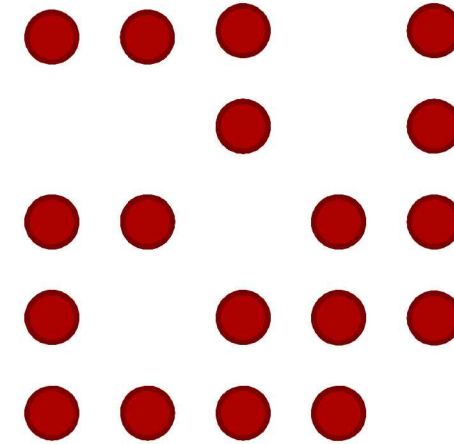
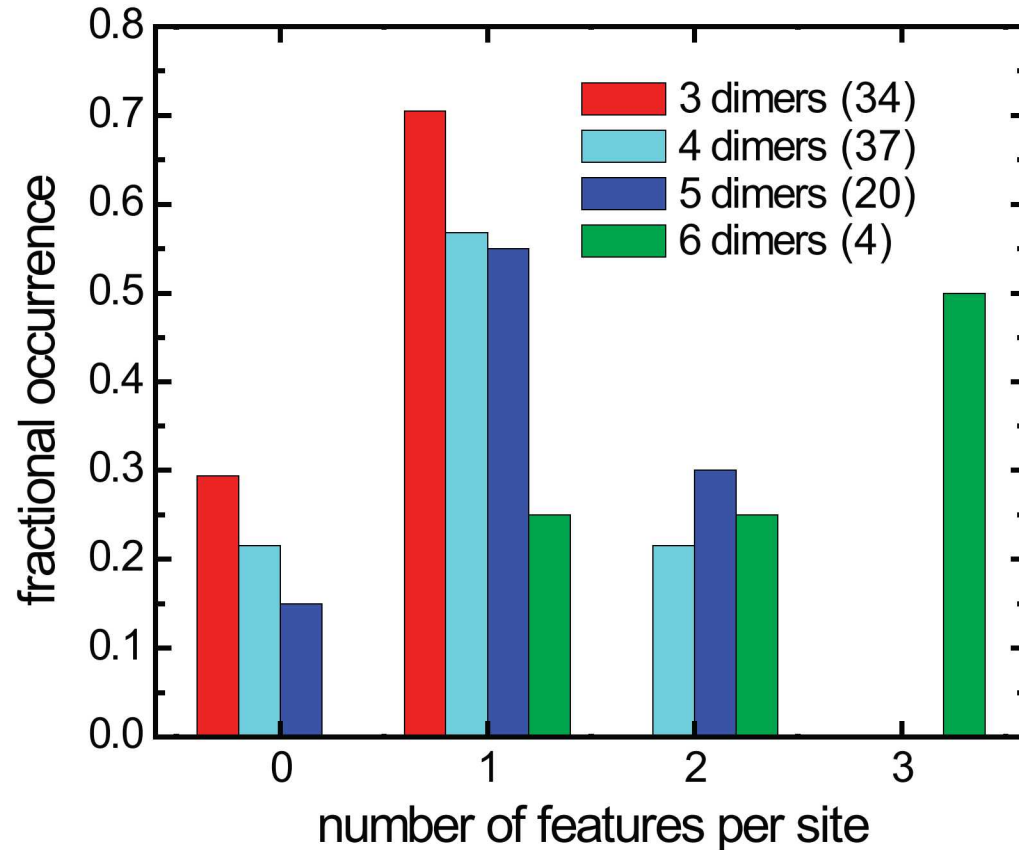
Almost back to reality,
fails at predicting intermediates

Peter Schultz (SNL) has a beautiful heuristic
Forget desorption! Incorporation
happens *quickly* through a concerted
reaction

	0	1	2
Expt.	0.7	0.3	0.0
KMC 1	1.0	0.0	0.0
KMC 2	0.7	0.3	0.0
Schultz	0.67	0.33	0.0

New understanding of surface decomposition of PH₃

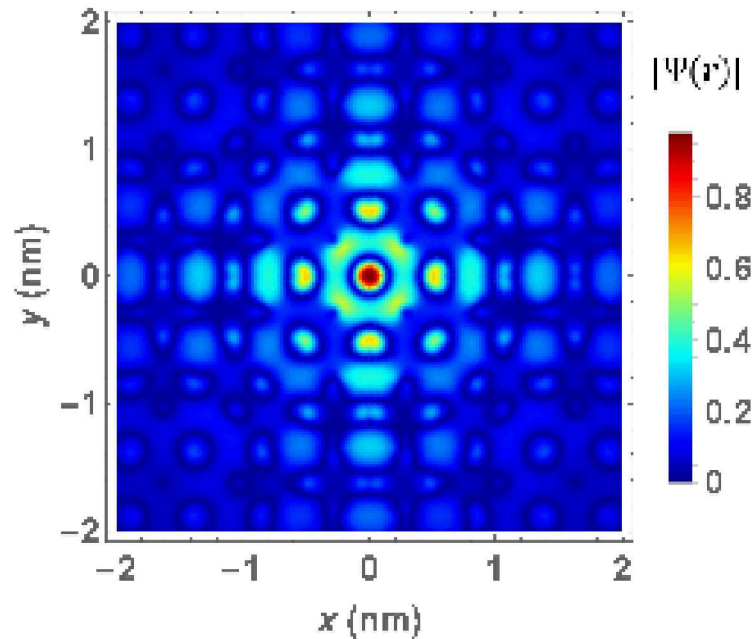
Chemistry is stochastic.



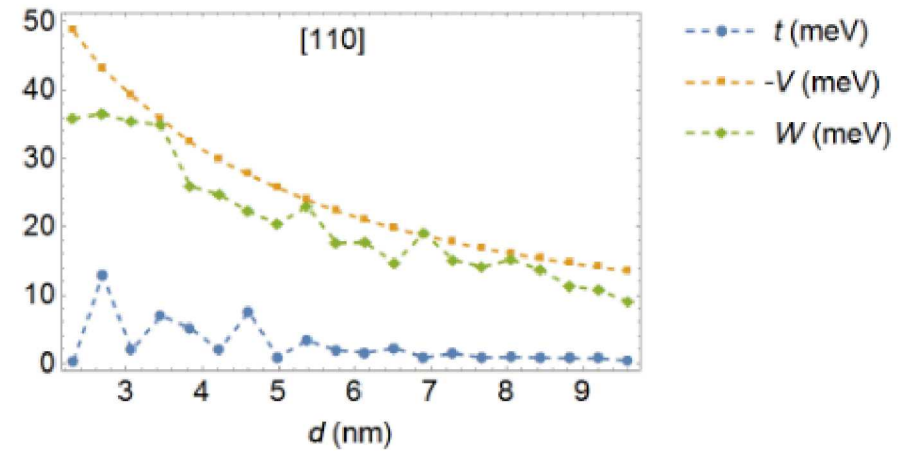
Odds that all 25 elements have one donor, assuming perfect 1 x 3 dimer lithography, is $(0.7)^{25} = 0.01\%$.

Yield of single donor array is zero.

How necessary is it to have true atomic precision?



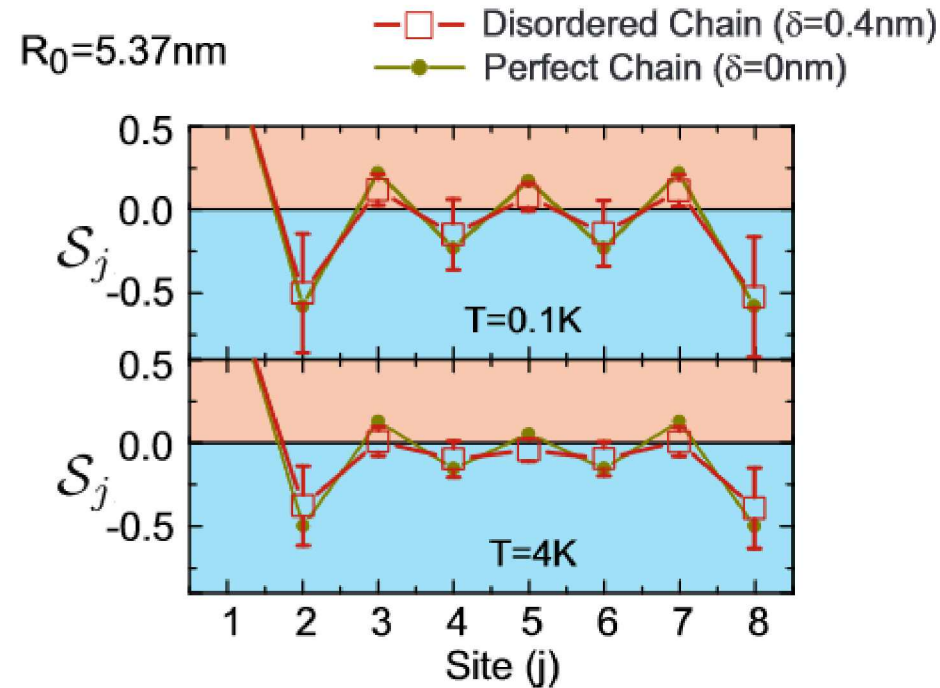
Donor electron density



Tunnel coupling oscillations

Significant variation in tunnel coupling from +/- 1 lattice site uncertainty.

Do oscillations matter?



Yes!

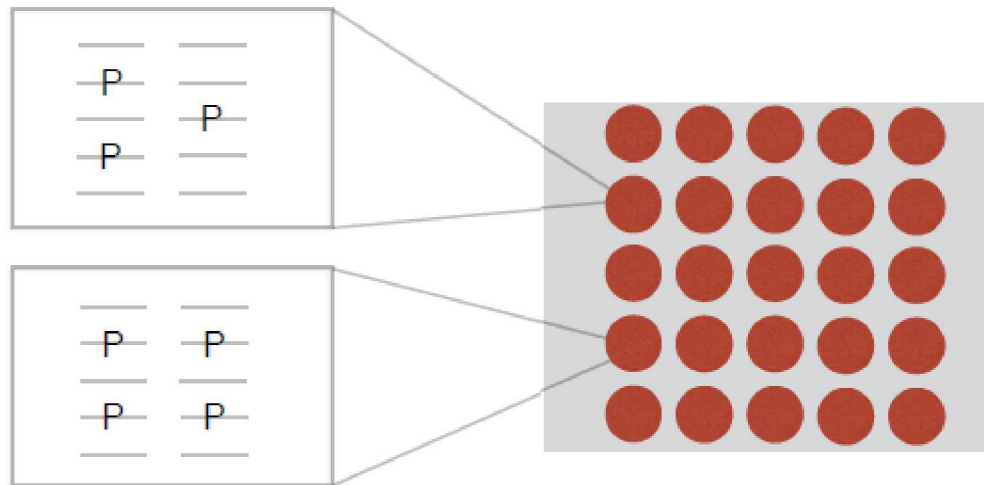
See-saw = antiferromagnetic chain

With 1 lattice site disorder = marginal

You want ~~single donors~~ clusters of donors

*Per node: pair of 4 or 5 by 1 windows.
P(0 donors) on 1 node: 2-4%*

Yield of 5 x 5 array: 35-55%

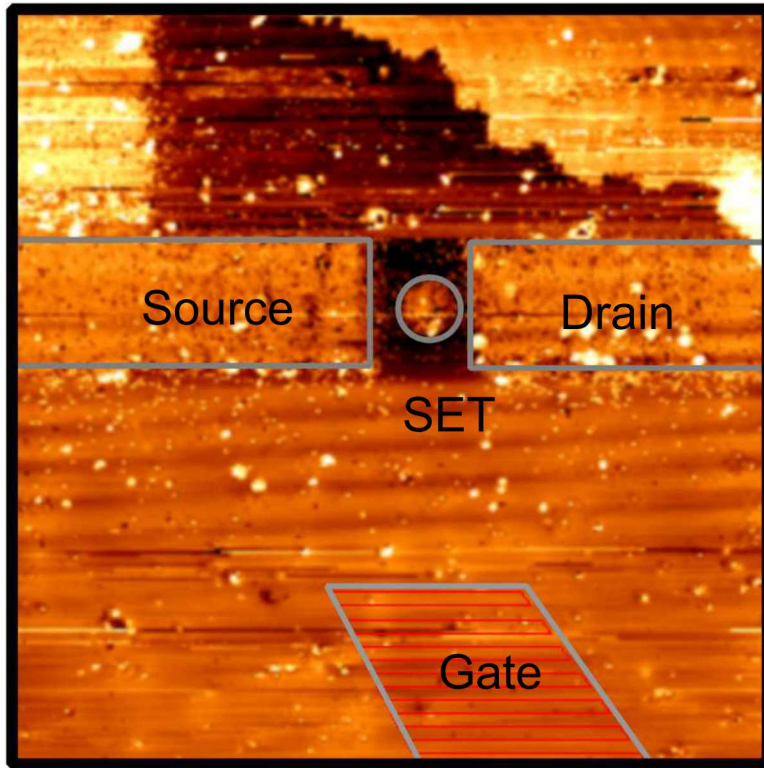


Accept that each island is going to have Poisson distribution of donors.

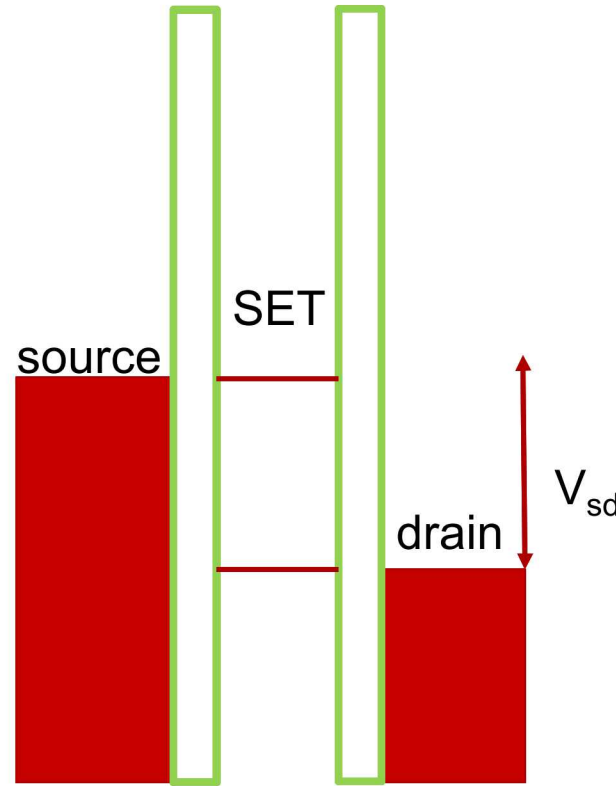
Individual donor wave-functions will smear out- t oscillations suppressed

What's a good cluster size?

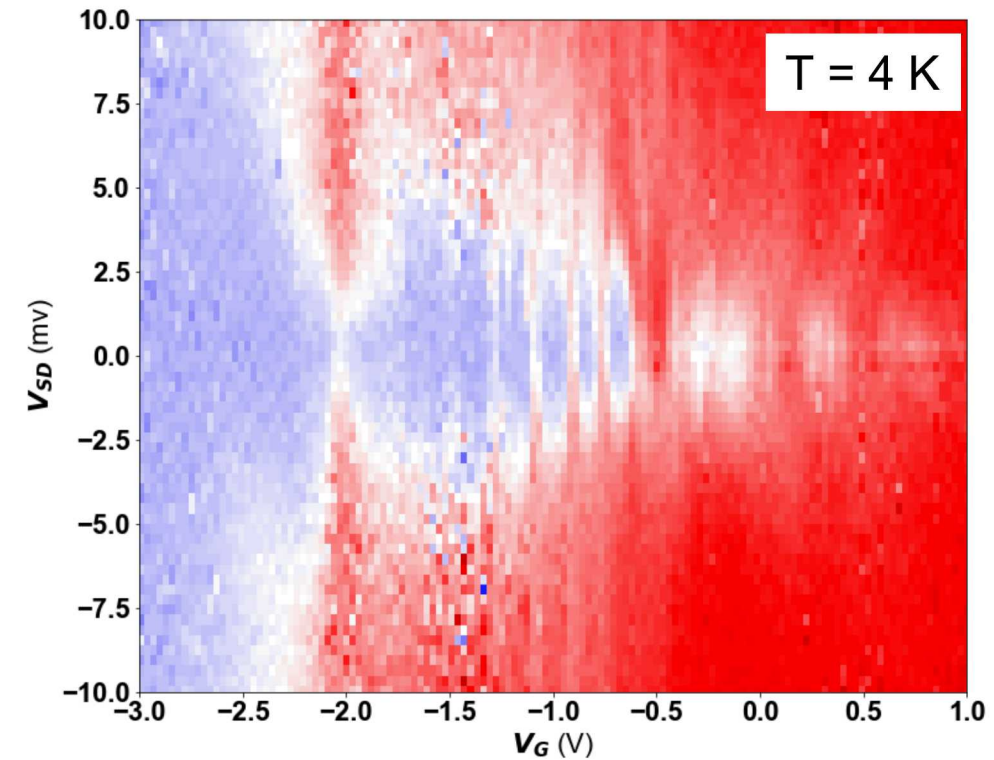
How big should cluster be?



8 nm diameter island
25-40 donors

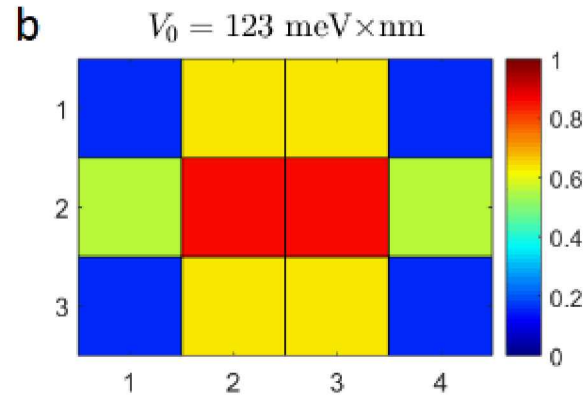


~ 5 nm islands.
Working on spacing now (~ 5 nm spacing)



$U \sim 5 \text{ meV}$
 $T_{DR} \sim 0.01 \text{ meV}$

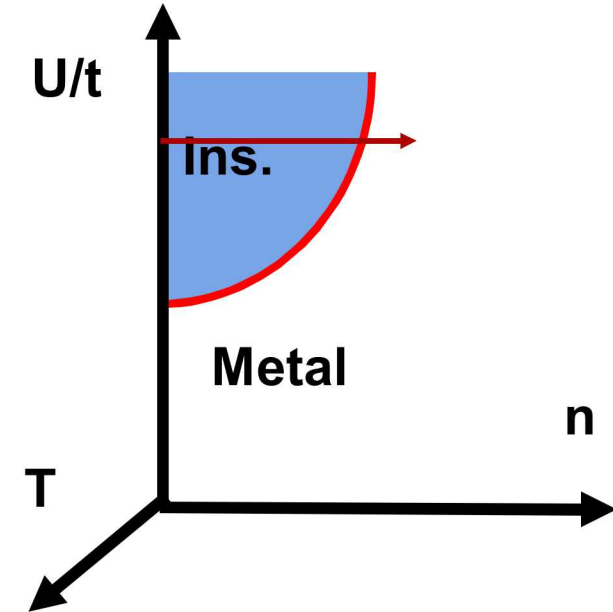
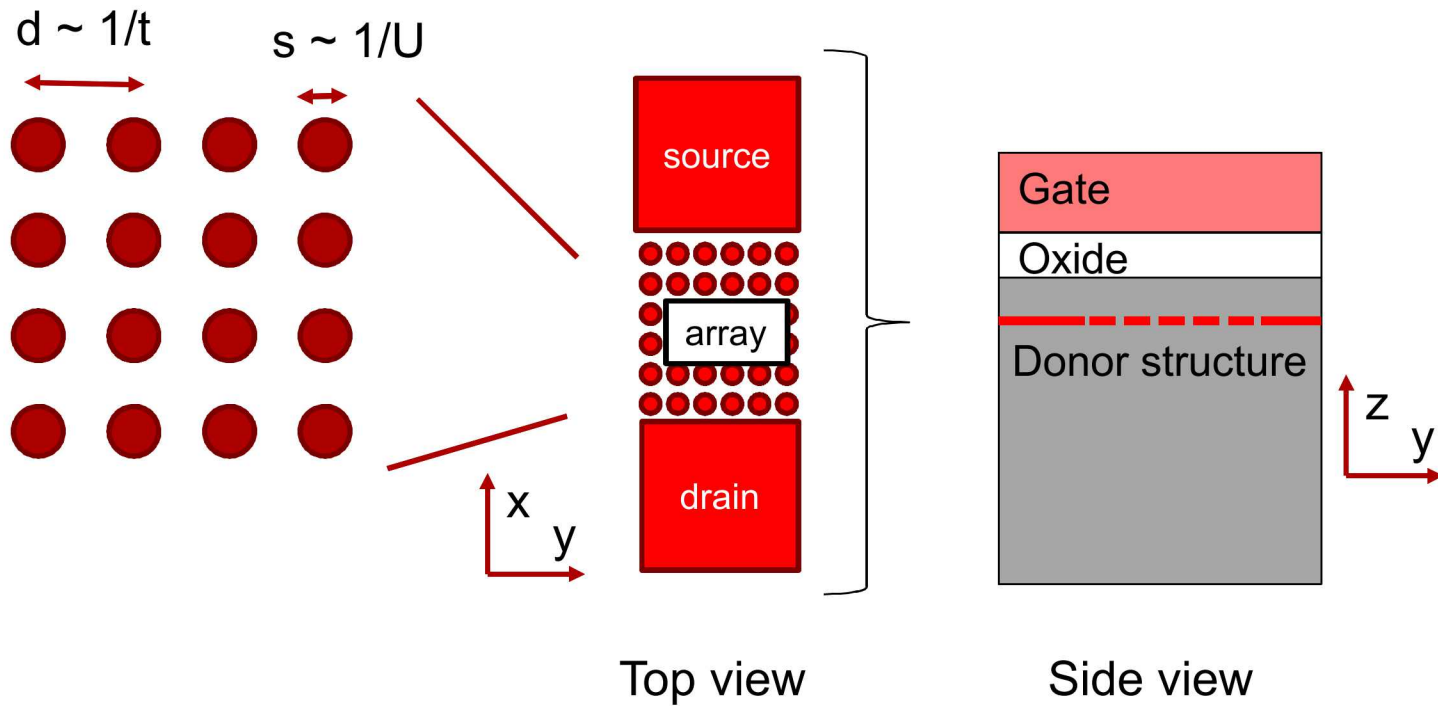
Designing quantum materials, atom by atom



*Electrons localized to
middle of 3 x 4 array*

What can we measure?

STM-based approach

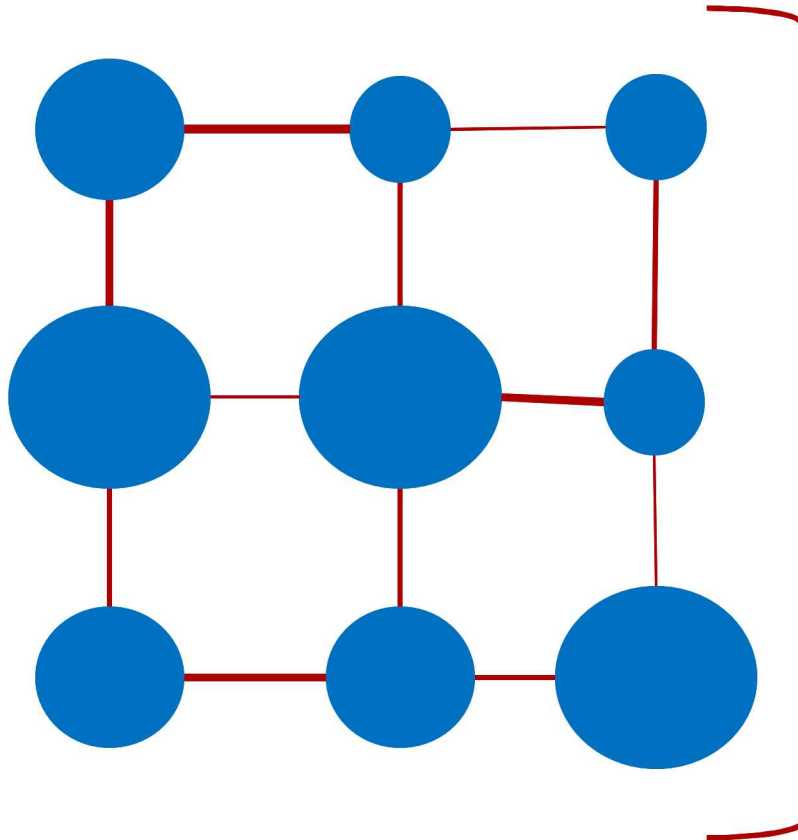


Is the array metallic or insulating?

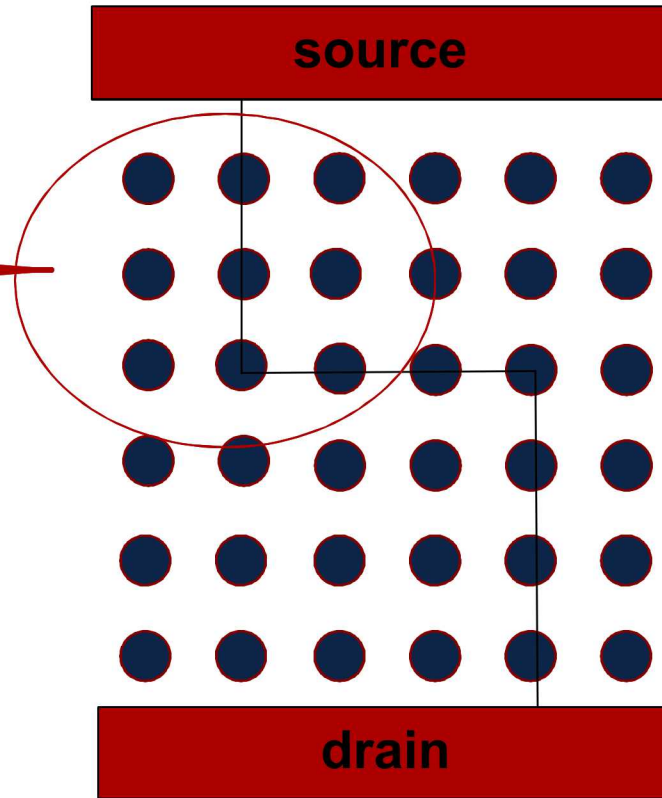
- n (increase V_g)
- T (shrunk array spacing)
- U (shrink island size)

How much does disorder and finite size matter?

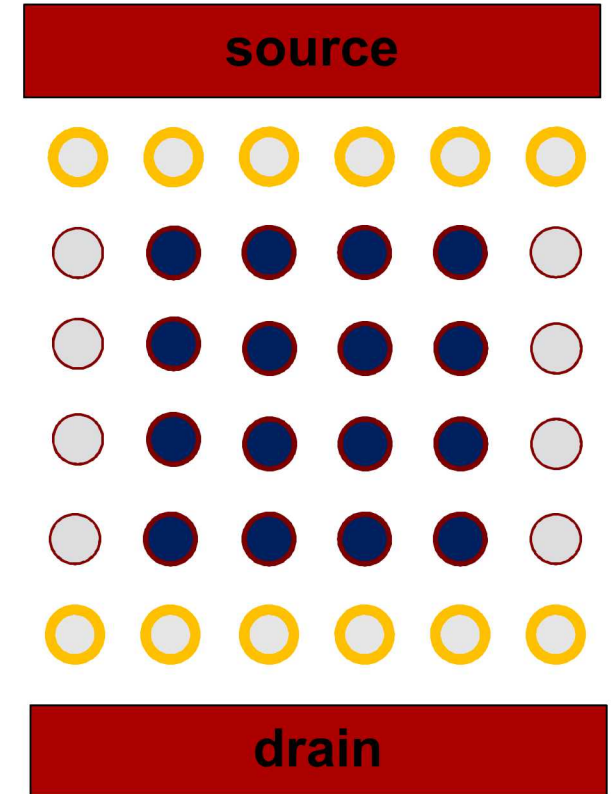
$N \pm \sqrt{N}$ donors per site.
Distribution of U's, t's.



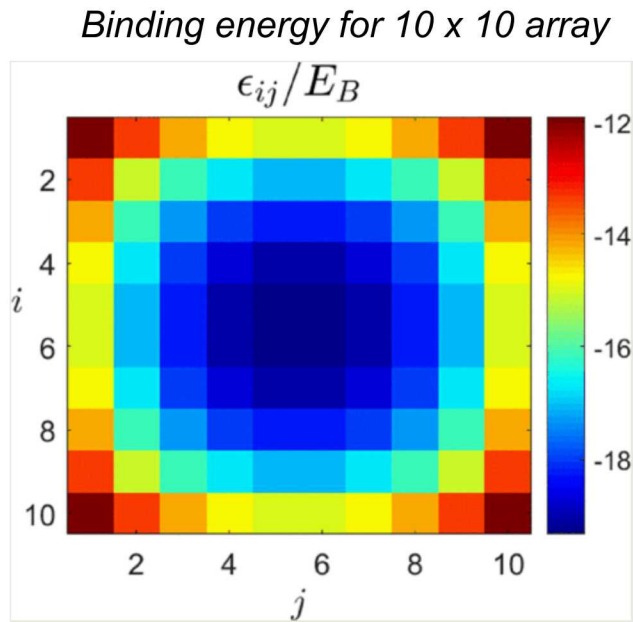
Disorder



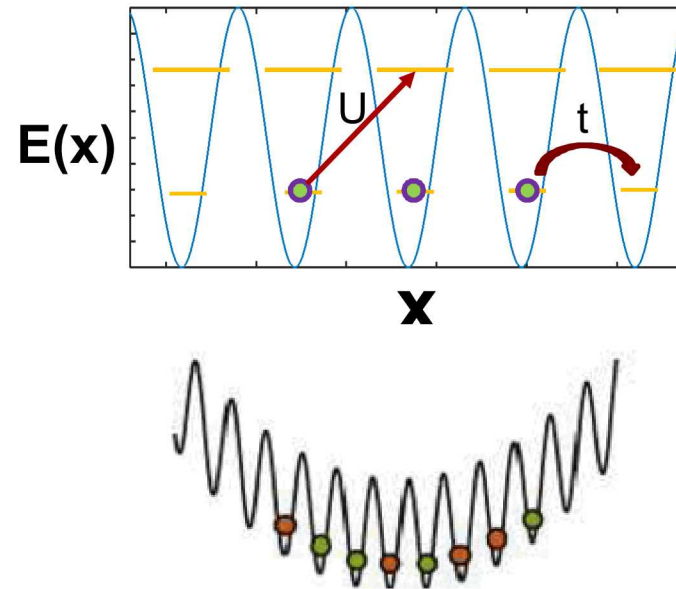
Finite size



Finite size effects.



Net attraction from overlap of nuclear potentials to middle of array

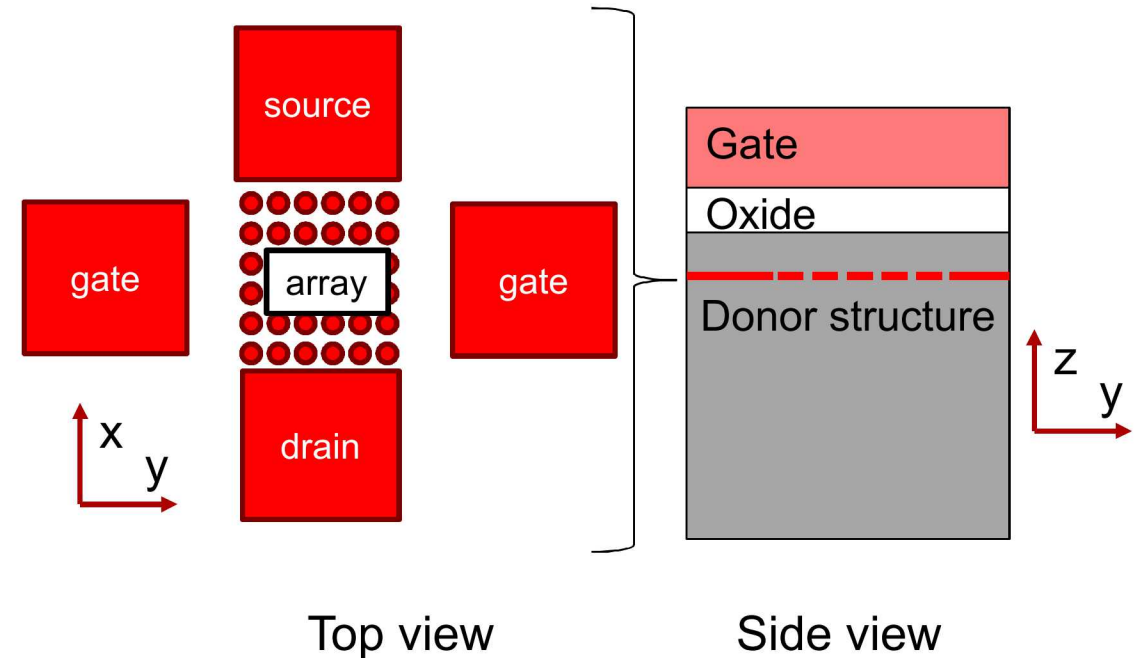
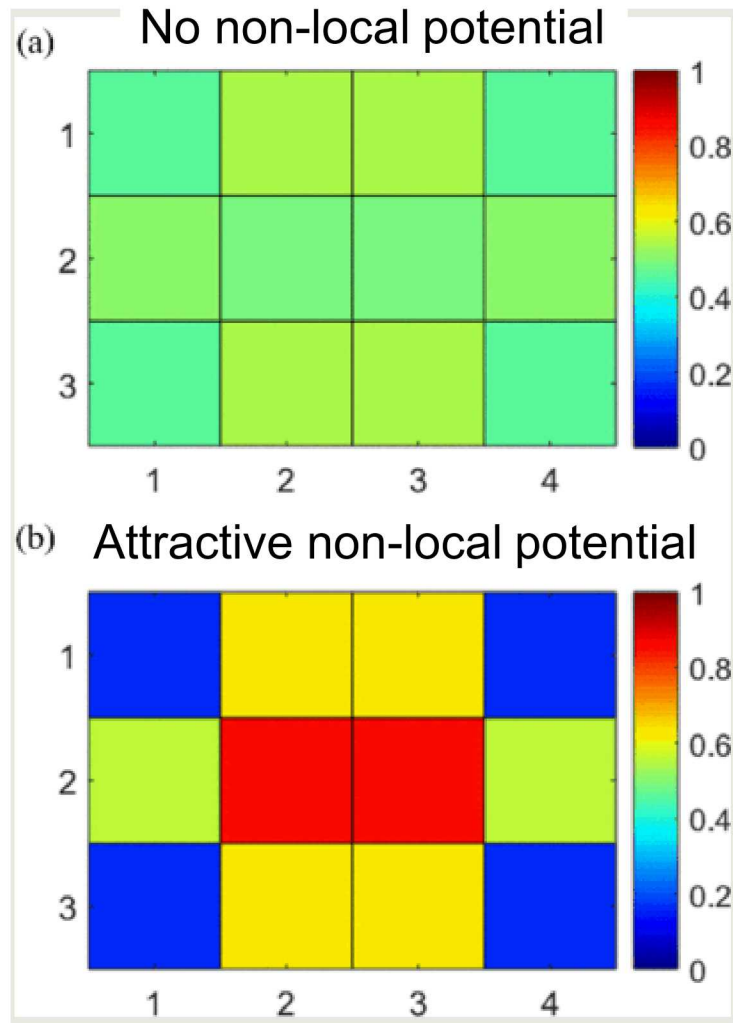


Nuclei not far enough apart for the potential to recover.

→ “Saggy couch”

Extended Hubbard Model.

3 x 4 donor array

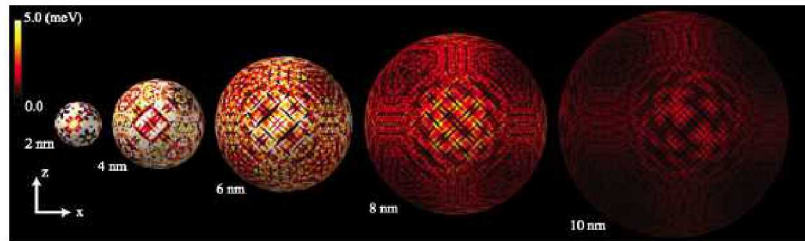


Calculation does not account for source or drain, or potential shaping using in-plane gates.

Remains a significant open question!

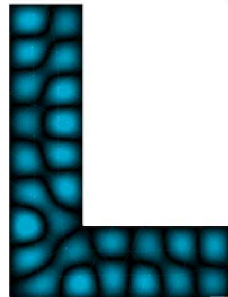
Extended Hubbard/Transport

- Multi-valley effective mass theory is:
1. Quantitatively accurate relative to atomistic tight-binding
 2. Fast enough to do statistics

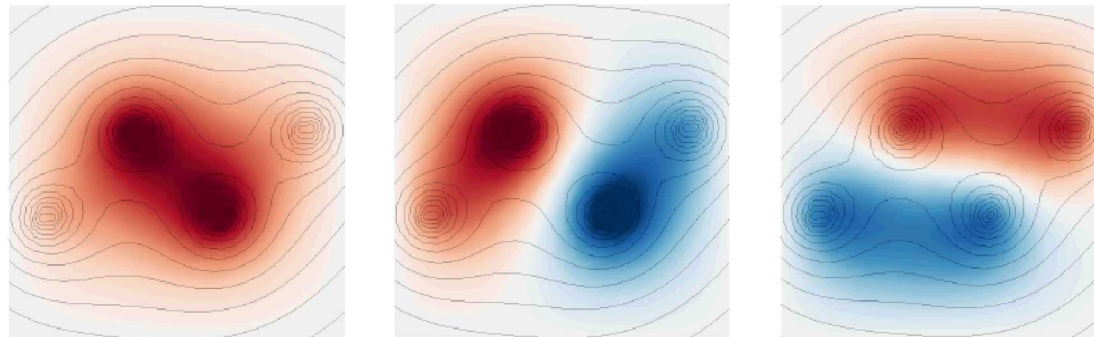


Gamble and Jacobson, et al., *Phys. Rev. B* (2015)

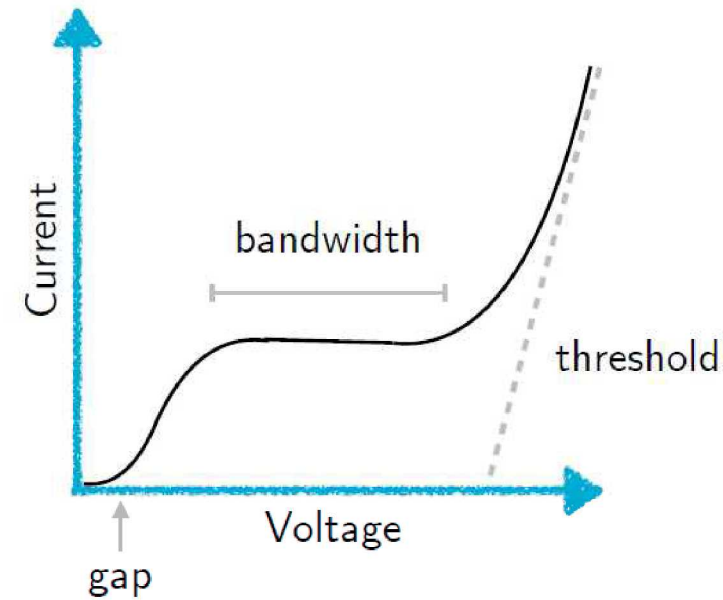
$$EF_l(\mathbf{r}) = [\hat{T}_l + U(\mathbf{r})]F_l(\mathbf{r}) + \sum_{j \in \pm\{x,y,z\}} V_{lj}^{VO}(\mathbf{r})F_j(\mathbf{r})$$



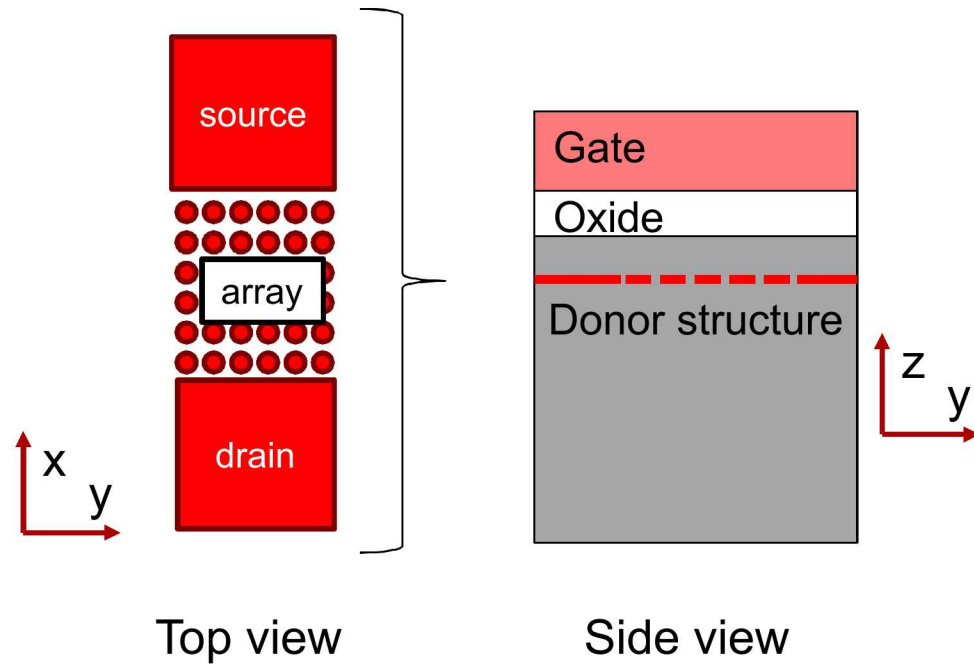
Laconic
(spectral element code)



Envelopes of the first 3 single electron orbitals on a 4P STM cluster



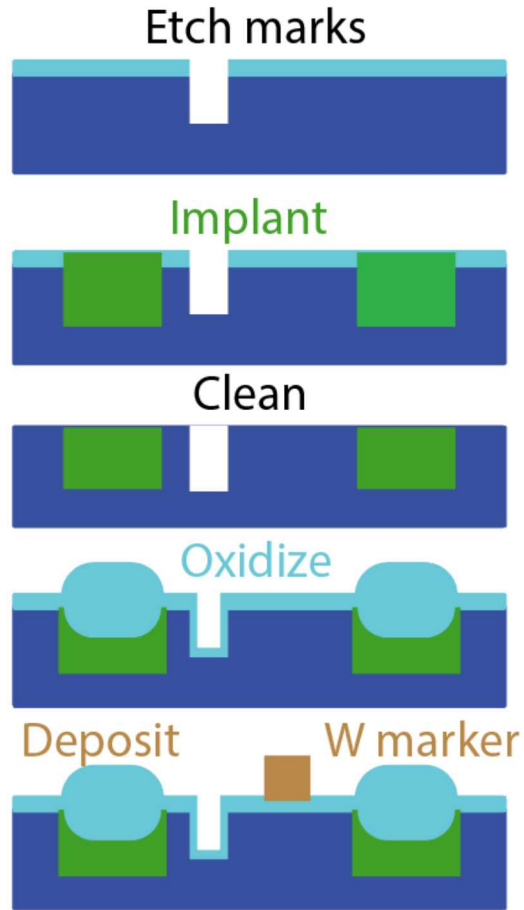
Electrical transport is limiting



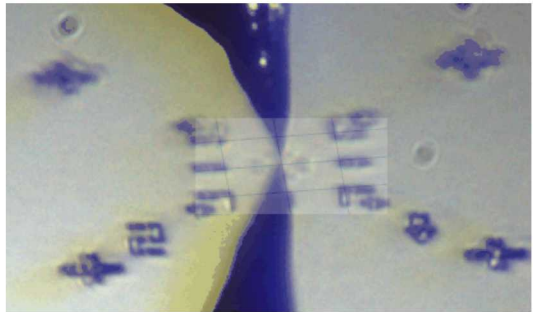
Can only ask questions about whether transport is ohmic or not as a function of state variables.

How to measure other quantities?

CMOS compatible STM "module"

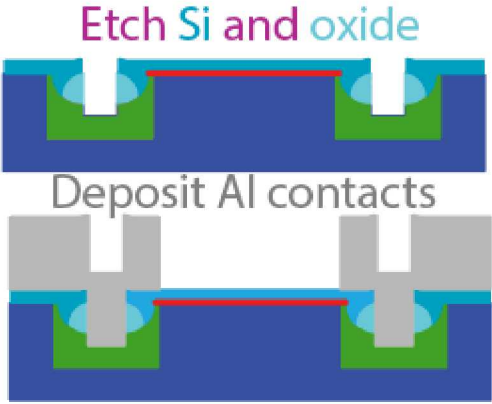


Open local window
Heat sample to 1200°C
800°C



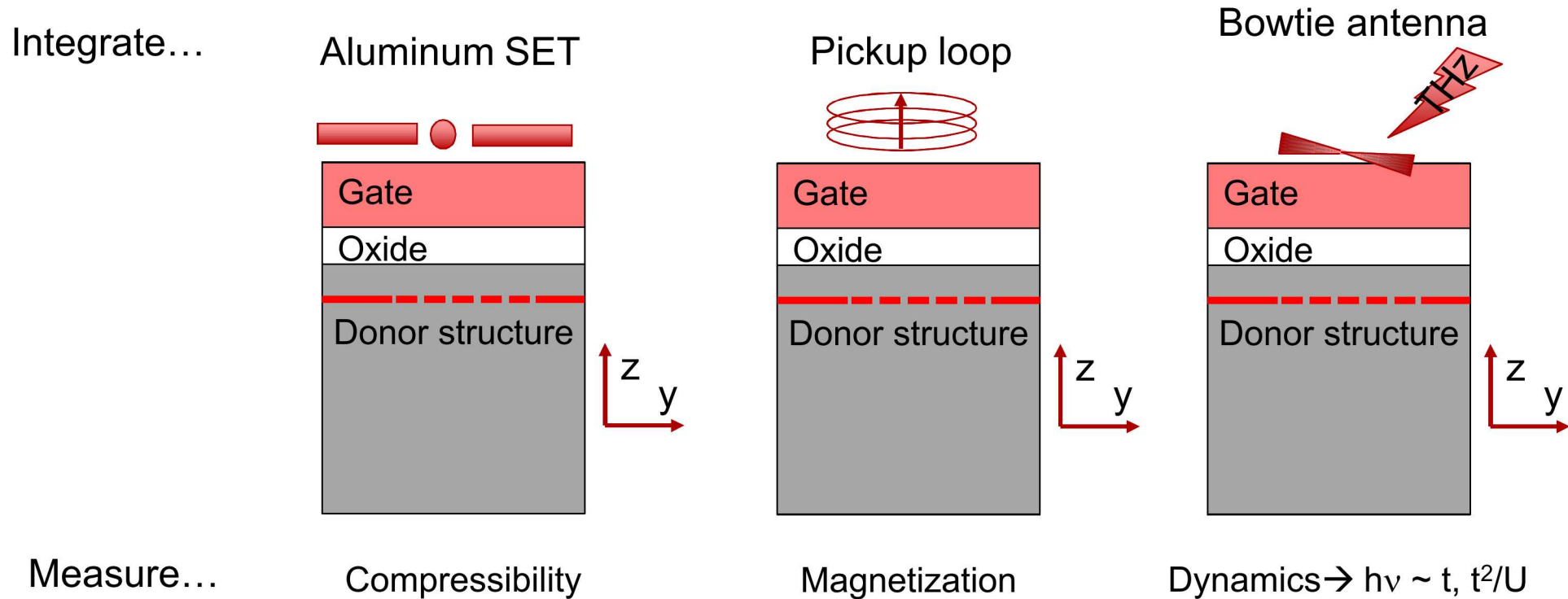
Donor pattern can survive heating to 500°C

Deposited oxides



CMOS integration & more complicated structures now possible

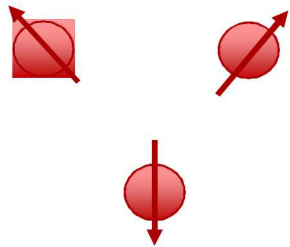
CMOS enables different measurement schemes



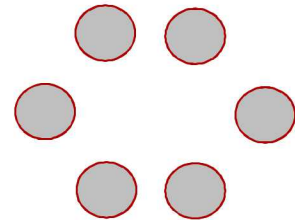
More speculative opportunities...

Change lattice symmetry

Frustration



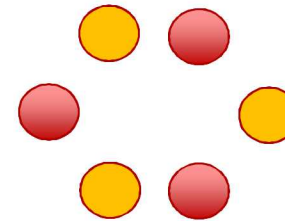
Dirac electrons?



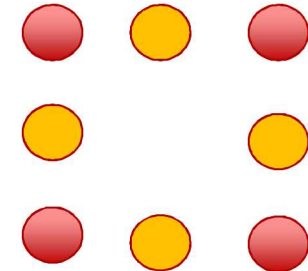
Simple, based on existing lithography capabilities

Add acceptor dopant species

Dichalcogenide-like



Lieb-like



Requires some Hydrogen lithography- compatible Boron or Aluminum precursor to be discovered.

Thanks to the folks who do all the real work...

We've run 600 samples in 3.5 years to get this far.

I'm not a chemist, or an electrical engineer, or a materials scientist, or...

STM	Fabrication	Measurement	Modeling
Mike Marshall	DeAnna Campbell	Tzu-Ming Lu (DEAL)	Leon Maurer (DEAL)
Justin Koepke	Dan Ward	Lisa Tracy (DQM)	Andrew Baczewski (DQM)
Shashank Misra		David Scrymgeour	
		Willie Luk	

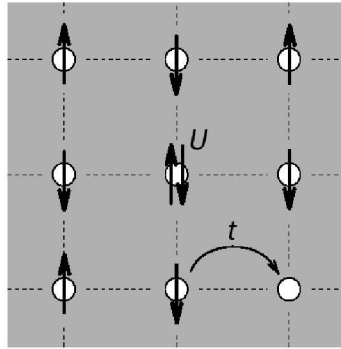
DEAL = transistor work
DQM = Hubbard lattice work

A million thanks to: Malcolm Carroll, Ezra Bussmann, Joe Simonson, Ed Bielejec, Rick Muller, Peter Schultz, Bob Butera, Esmeralda Yitamben, Martin Rudolph, Steve Carr, Zyvex Labs

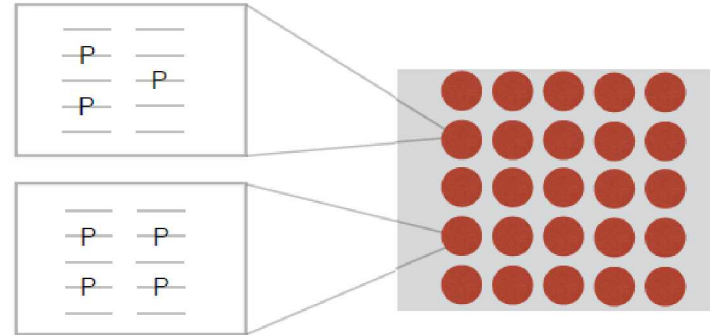
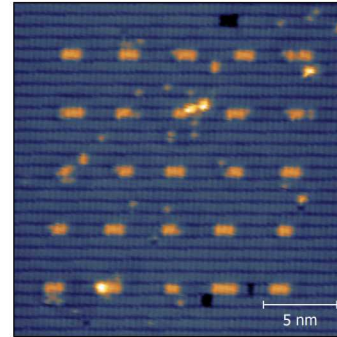
Some of this work is supported by the Laboratory Directed Research and Development Program at Sandia National Laboratories, and was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525

Engineering quantum materials, atom by atom

Why emulate quantum materials with donor arrays?

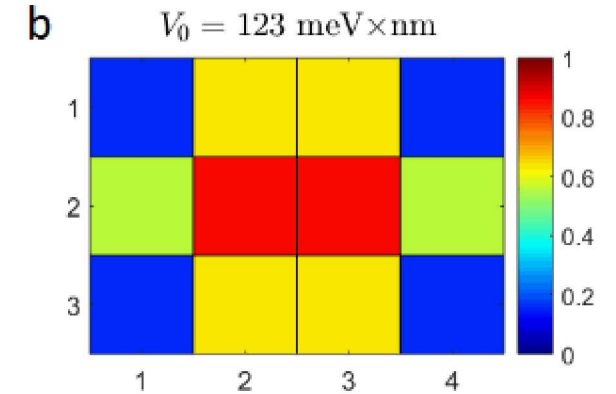


What donor array can we make?



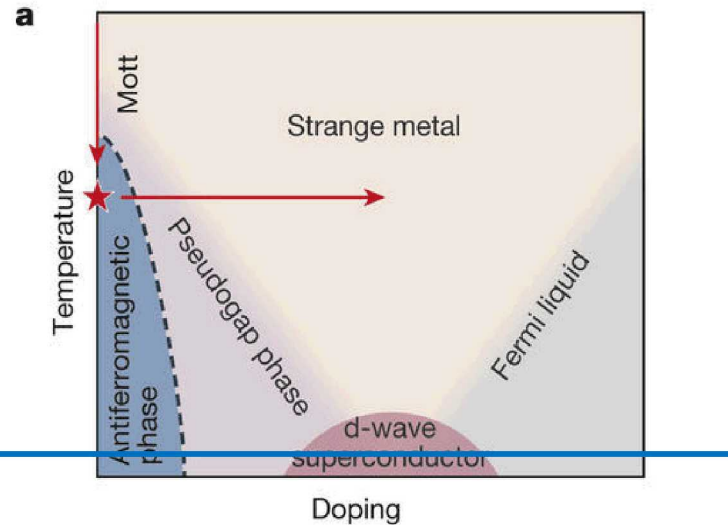
Lithographically easy:
5 nm diameter islands,
5 nm spacing

What can we measure?



Electrical transport.
Effects of disorder & finite
size need to be sorted out

CMOS integration gives you:
Magnetization?
Compressibility?
Dynamics?



Donors have big energies relative to T