A new descriptor for unconventional superconductivity: calibration and testing

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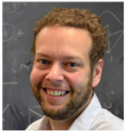
Lucas K. Wagner
UIUC



Awadhesh Narayan
ETH

Brian Busemeyer
UIUC

Supported by the US Department of Energy

1. Unconventional SC may be due to interactions between spin and orbital degrees of freedom
2. Charge-spin susceptibility: a way of estimating spin-orbital coupling from first principles
3. Testing if charge-spin susceptibility can distinguish uSC from non-uSC
    - Test set
    - Charge-spin susceptibility
    - Classifiers for uSC

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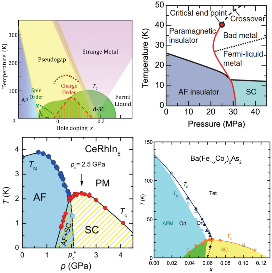
Finding new unconventional (high-T_c) superconductors.

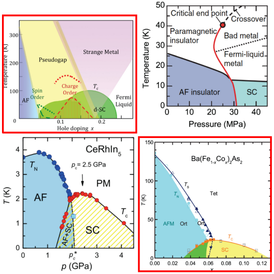
Finding new unconventional superconductors



[Norman, RPP 79, 074502 (2016)]

Heavy fermions, Organic, Cuprates, Iron-based.

Finding new unconventional superconductors



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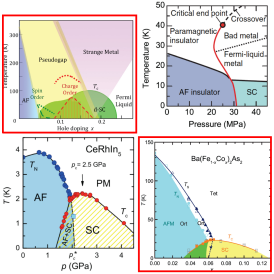

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Data-poor situation!

unlike conventional superconductivity

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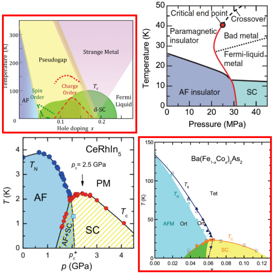

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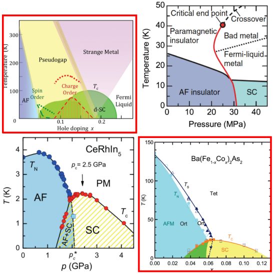

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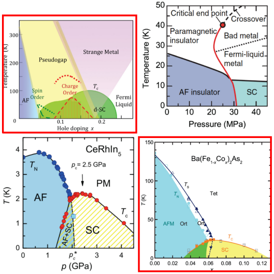
Main issue we will discuss today.

Finding new unconventional superconductors



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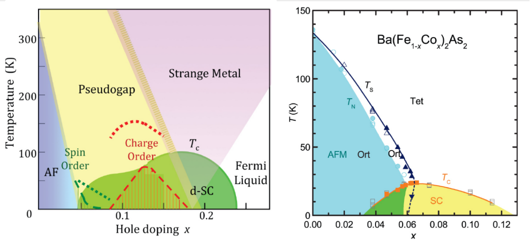
Not clear what to look for!

Up to now, many similarity-based searches.
But no prediction verified so far.

Finding new unconventional superconductors

Commonalities between **cuprates** and **iron-based** superconductors:

- Layered materials with 2D arrays of transition metals.
- Partially filled d-levels $\Rightarrow$ stable local magnetic moments.
- Majority have long-range magnetic order when undoped.
- SC when long-range magn. order $\rightarrow$ paramagnetic.



Adapted from [Norman, RPP 79, 074502 (2016)]

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Crucial role of spin fluctuations in electron pairing.

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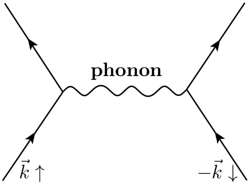
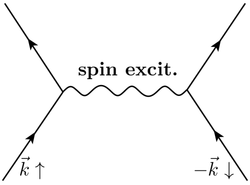
Conventional (BCS) Superconductivity



Unconventional Superconductivity



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Leo Tolstoy in *Anna Karenina*.

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Hypothesis: Some ingredients need to be present for high-T_c uSC.

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Hypothesis: Some ingredients need to be present for high-T_c uSC.

What should those ingredients be?

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- are **layered** with arrays of transition metals.
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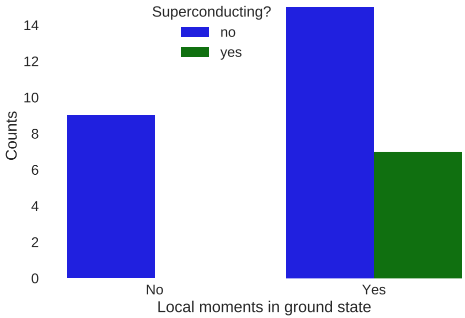

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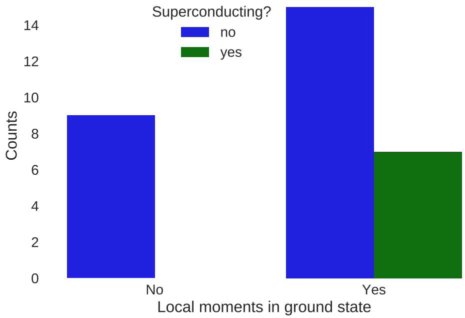

Bad at distinguishing uSC from other materials.

Detecting spin-orbital coupling

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For concreteness consider the model Hamiltonian

$$H = H_o + H_S + \lambda H_{oS}$$

with λ coupling orbital and deep spin levels.

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Hole-doped cuprates:

- conduction holes mostly oxygen-p;
- half-filled Cu-d as local magn. moments;

Iron-pnictides:

- minority spin levels as conduction electrons;
- majority spin levels as local magn. moments;

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Can spin-orbital coupling separate cuprates
and iron-based SCs from other materials?

Detecting spin-orbital coupling

Approximate spin-orbital coupling $\frac{\lambda}{w}$ by charge-spin susceptibility χ_{cs}

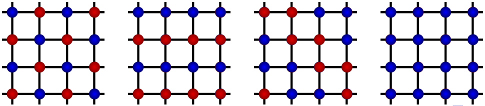
$$\chi_{cs} \equiv \frac{1}{N}\sum_{i=1}^{N}\chi_i = \frac{1}{N}\sum_{i=1}^{N}\frac{\Delta\rho_i}{\Delta s_i}$$

N is the number of different magnetic textures considered, $\Delta\rho_i$ (Δs_i) is stands for the spatial fluctuations in charge (spin) density relative to that of lowest-energy magnetic order.The former are given by

$$\Delta\rho_i = \int \mathrm{d}\mathbf{r}\,\left|\rho_i(\mathbf{r}) - \rho_0(\mathbf{r})\right|$$

$$\Delta s_i = \int \mathrm{d}\mathbf{r}\,\left|s_i(\mathbf{r}) - s_0(\mathbf{r})\right|$$

where $\rho_0(\mathbf{r})$ and $s_0(\mathbf{r})$ are the charge and spin distributions of the lowest-energy state.



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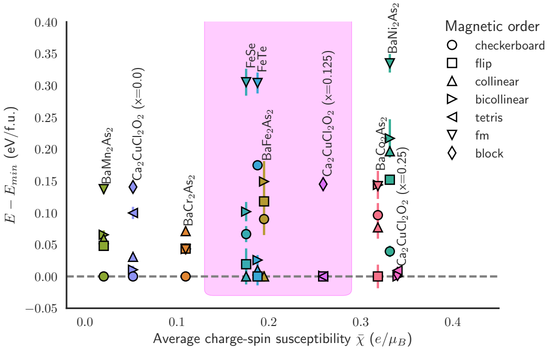

[Narayan *et al.*, arXiv:1705.01008]

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How are we going to calculate these properties?

(Local magnetic moments and charge-spin susceptibility)

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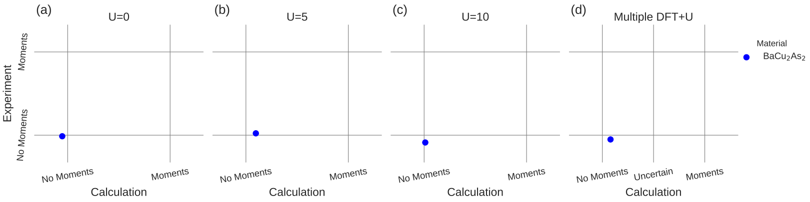
Multiple-DFT calculations to control errors.

Example: Find if there are stable local magnetic moments with multiple DFT+U. [Cococcioni and Gironcoli PRB 71, 035105 (2005)]

Lowest-energy state:

- is non-magnetic ⇒ stable magnetic moments absent.
- is magnetic ⇒ stable magnetic moments exist.



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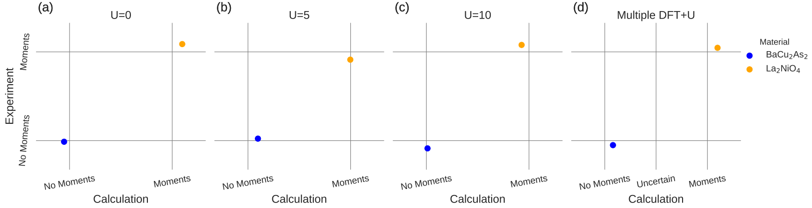

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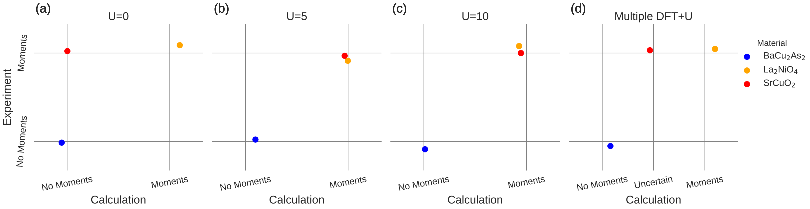

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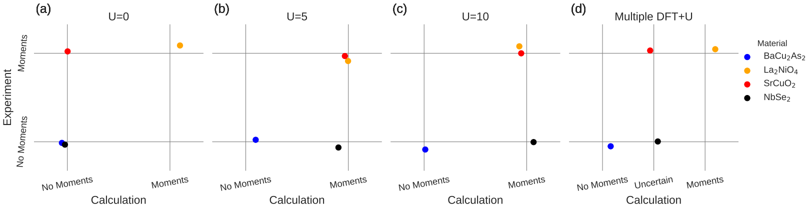

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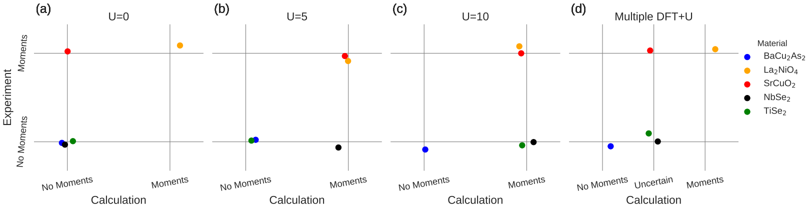

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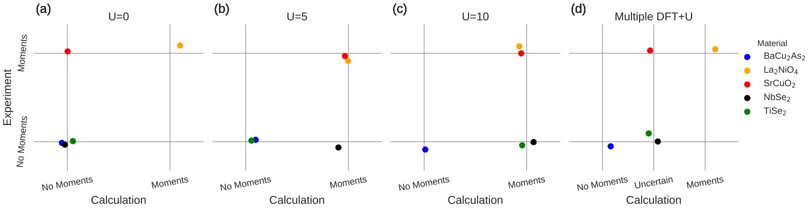

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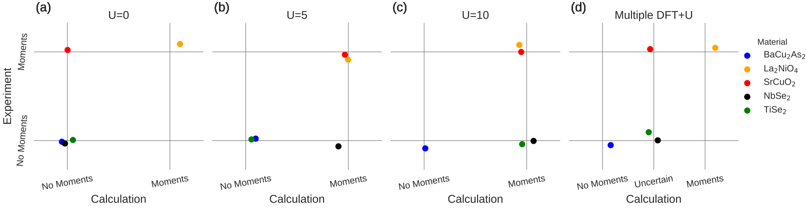
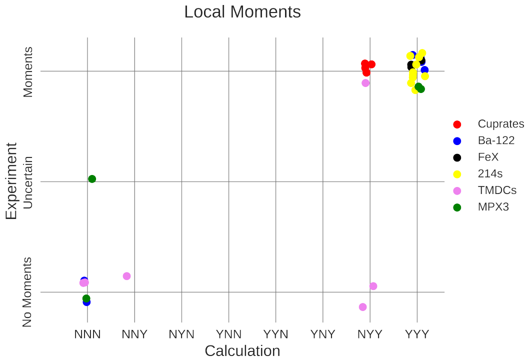


Lower rate of false positives and false negatives.

But large rate of uncertain.

Improved estimator for existence of magnetic moments (multi DFT+U):



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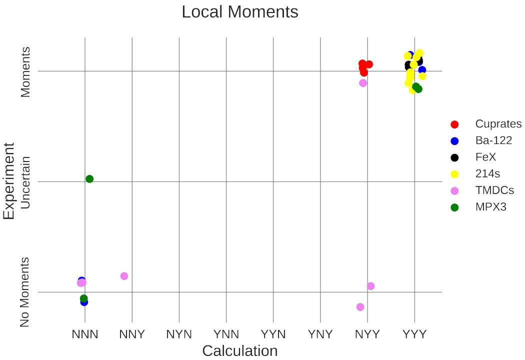

$$P(M|NNN \vee NNY) \approx 0.0\,,$$
$$P(M|NYY) \approx 0.6\,,$$
$$P(M|YYY) \approx 1.0\,.$$

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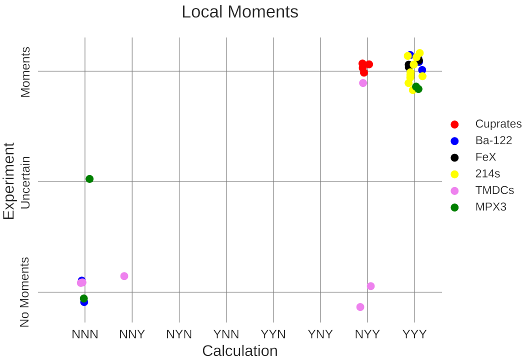

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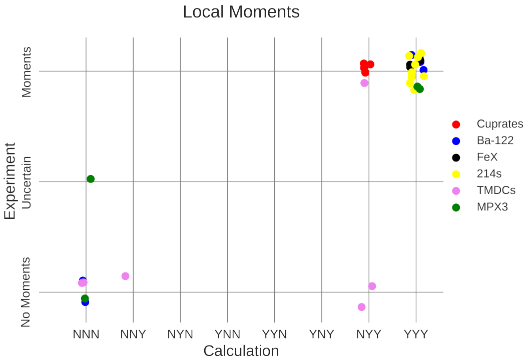

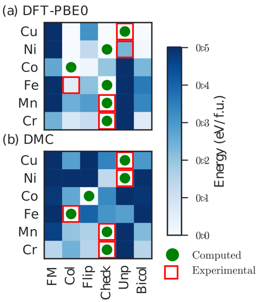

This is our biggest source of error.

Can use QMC in those with most uncertainty.

[Narayan *et al.*, arXiv:1705.01008]

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Test set

35 pure materials (all layered, some magnetic, some uSC):

Cuprates: $CaCuO_2$, $SrCuO_2$, T-La_2CuO_4, T'-La_2CuO_4.

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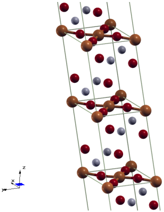

All are antiferromagnetic (AFM) insulators.
All turn superconductor upon doping.

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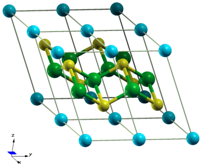
35 pure materials (all layered, some magnetic, some uSC):

Cuprates: $CaCuO_2$, $SrCuO_2$, T-La_2CuO_4, T'-La_2CuO_4.
Ba-122s: BaM_2As_2 (M=Cr,Mn,Fe,Co,Ni,Cu).



Typically AFM or paramagnetic metals.
$BaFe_2As_2$ turns superconductor under pressure or doping.

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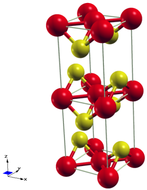
FeX: FeSe, FeTe, FeS.



All magnetic metals.
Turn superconductor upon charge doping. FeSe turns SC in pure form.

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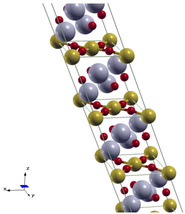
214s: La_2MO_4 (M=Co,Ni), Sr_2MO_4 (M=V,Cr,Mn,Fe,Co) and K_2MF_4 (M=Co,Ni,Cu).



All are AFM insulators.
Were never made superconducting.

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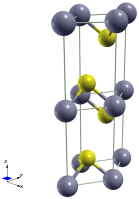
TMDCs: MSe_2 (M=Ti,Nb,Ta,W) and MS_2 (M=Mo,Ta).



Nonmagnetic metals and insulators.
Conventional superconductor under charge doping or pressure.

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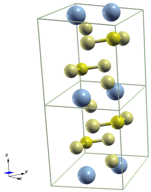
MPX_3: VPS_3, $NiPSe_3$, $CdPSe_3$, $CrGeTe_3$.



Magnetic and non-magnetic insulators. **Superconductivity unknown.** We suspect they **cannot** be made uSC.

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Charge-spin susceptibility

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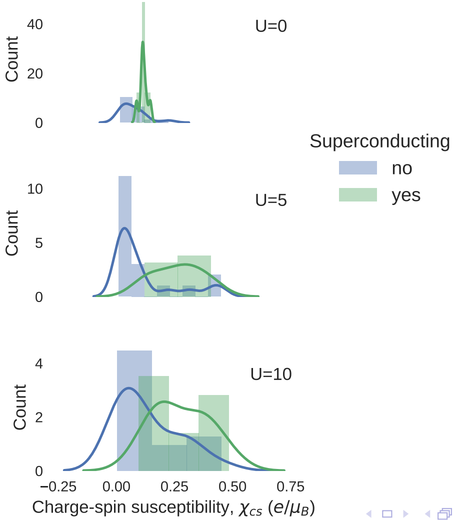

Charge-spin susceptibility

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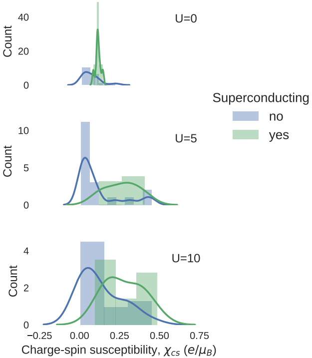

χ_{cs} distinguishes cuprates and iron-based SCs from other materials!

uSC seems to require **intermediate** spin-orbital coupling.
(similar to e-ph coupling conventional SCs. [Esterlis *et al.*, arXiv:1806.00488])

1. Unconventional SC may be due to interactions between spin and orbital degrees of freedom
2. Charge-spin susceptibility: a way of estimating spin-orbital coupling from first principles
3. Testing if charge-spin susceptibility can distinguish uSC from non-uSC
   - Test set
   - Charge-spin susceptibility
   - Classifiers for uSC

Classifiers for unconventional superconductivity

Classifiers for uSC based on three ingredients:

- layered structure (with TMs);
- stable local magnetic moments;
- charge-spin susceptibility;

Quantified as: $P(SC|\chi_{cs}^{U}, M, 2D) \approx P(SC|\chi_{cs}^{U})\, P(M)$.

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$$P(SC|\chi_{cs}^U) = \frac{\rho_{sc}^U}{\rho_{sc}^U + \rho_{\neg sc}^U}$$

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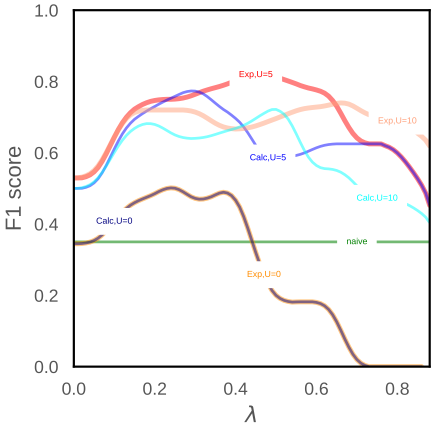
Probability local moments exist $P(M)$:

From experiment	From DFT+U
$P(M_{\mathrm{exp}}) = 1 \,\mathrm{or}\, 0$	$P(M_{\mathrm{calc}}) \equiv \begin{cases} P(M\|NNN \vee NNY) \approx 0.0 \\ P(M\|NYY) \approx 0.6 \\ P(M\|YYY) \approx 1.0 \end{cases}$

Classifiers for unconventional superconductivity

Assessing quality of different classifiers: $\mathrm{F1} = \frac{2\mathrm{TP}}{2\mathrm{TP}+\mathrm{FP}+\mathrm{FN}}$.



Best classifier is $P(SC|\chi_{cs}^{U=5})\,P(M_{\mathrm{exp}})$.

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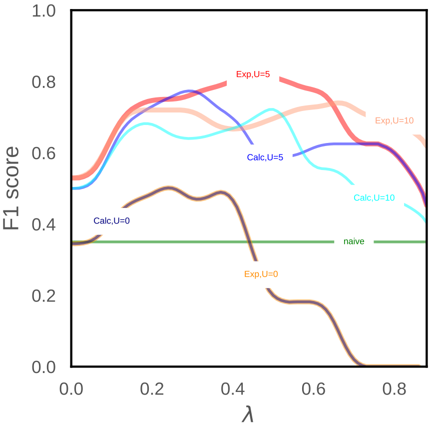
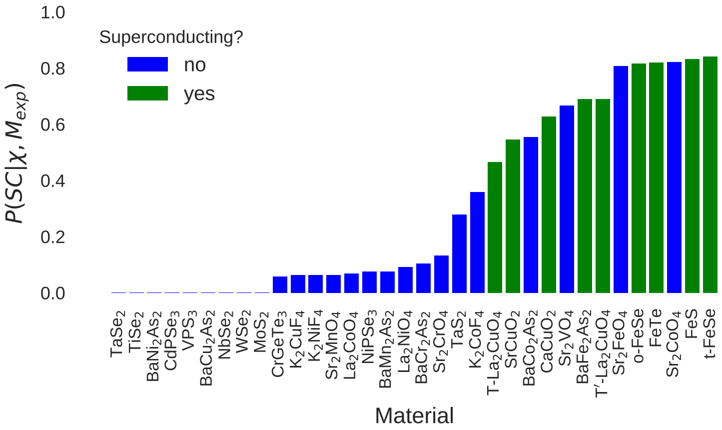


Best classifier is $P(SC|\chi_{cs}^{U=5})\,P(M_{\mathrm{exp}})$.

$P(M_{\mathrm{calc}})$ with DFT+U is inaccurate. **QMC can help.**

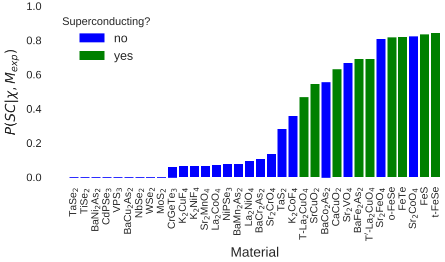
Ranking test set materials

Ranking test set according to **best classifier**: $P(SC|\chi_{cs}^{U=5})\,P(M_{\mathrm{exp}})$.



Clearly distinguishes known uSCs from other materials!

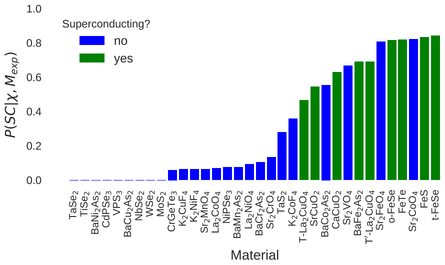
Ranking test set materials



A few comments on highly ranked non-uSCs:

- DFT+U too inaccurate $\Rightarrow$ false prediction. **Clarifiable with QMC**.

Ranking test set materials



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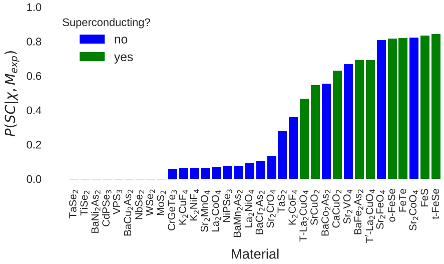
Ranking test set materials



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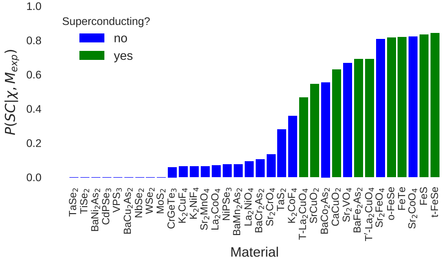

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- ∃ necessary ingredient (that we didn't consider) and material lacks.
- Good spin-orbital coupling but other instability dominates over SC.
- Charge doping issue:
  - Charge doping degrades spin-orbital coupling.
  - Material not amenable to doping for chemical reasons.
  - **Can be made uSC** (with correct doping) but never attempted.

Summary

- Classifier for uSC with 3 ingredients: layered, LMs and spin-orbital.
- LMs prediction introduces most inaccuracies. **QMC-improvable?**
- Sufficiently specific to distinguish cuprates and iron-based SCs.
- Singles a few non-uSC. **Experimentalists should look at them**.