Charge-spin coupling as a probe of correlated quantum materials

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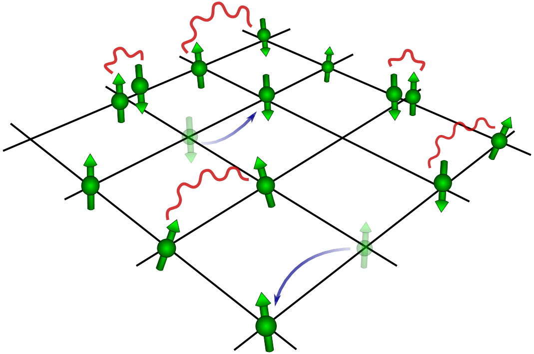
4$^{\text{th}}$ June 2019

Acknowledgments: Center for Emergent Superconductivity, funded by Department of Energy award DEAC0298CH1088. Computational resources of University of Illinois Campus Cluster and Blue Waters funded by awards OCI-0725070 and ACI-1238993.

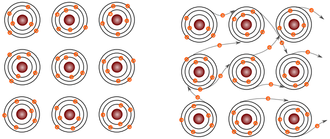
Quantum materials are all about collective behavior



10^{23} interacting particles (electrons and nuclei)
with position and spin

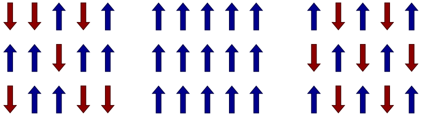
Collective behavior determines materials' properties

Electric conduction



Insulator Metal

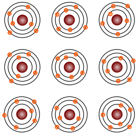
Magnetic ordering



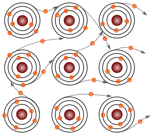
Paramagnet Ferromagnet Antiferromagnet

Collective behavior determines materials' properties

Electric conduction

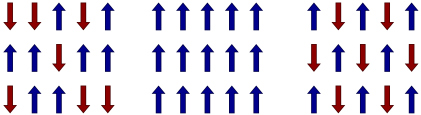


Insulator



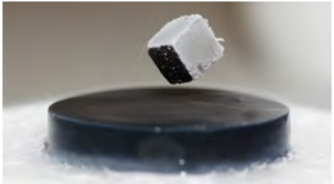
Metal

Magnetic ordering



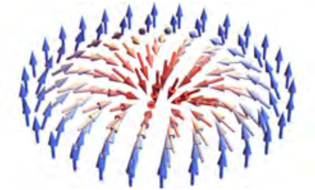
Paramagnet Ferromagnet Antiferromagnet

High temperature superconductivity



Adapted from extremetech.com

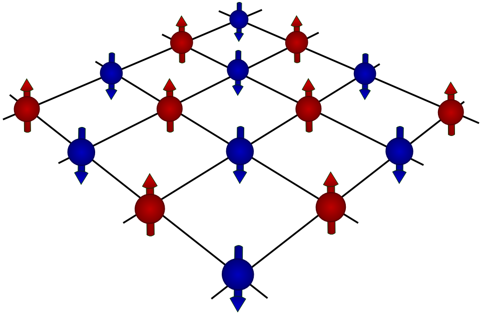
Unconventional magnetic orders



Adapted from asianscientist.com

Some of the materials with such unusual properties seem to have a strong coupling between magnetic and orbital degrees of freedom

Iron-pnictides (FeAs)



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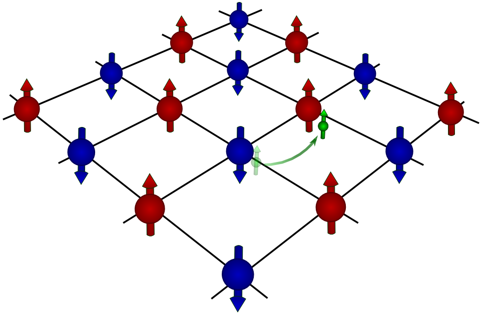

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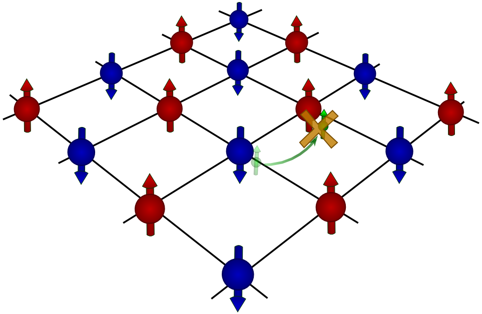

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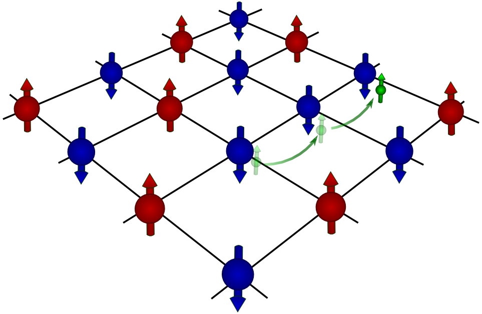

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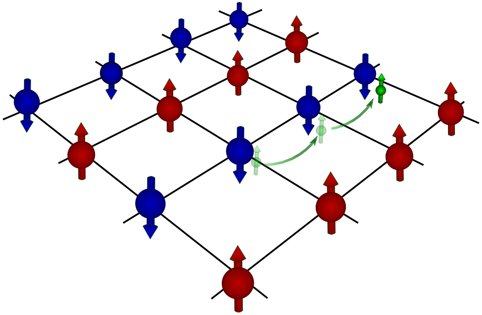

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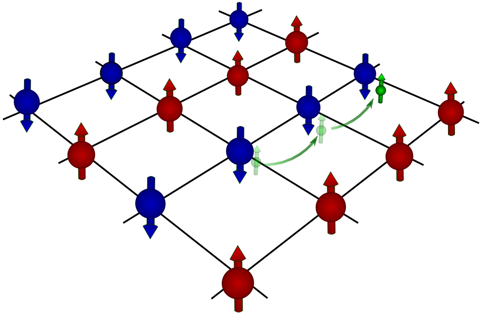
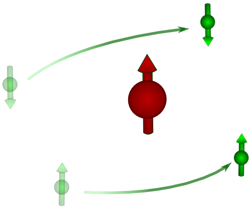


Superconducting pairing mediated by charge-spin interactions?



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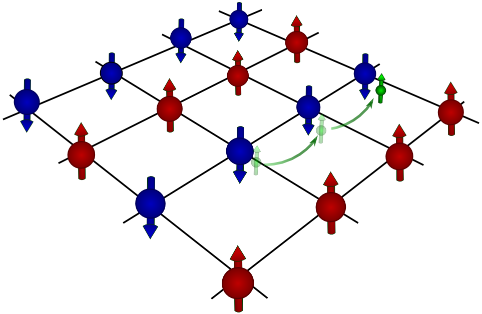

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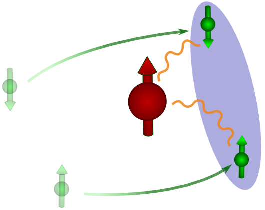
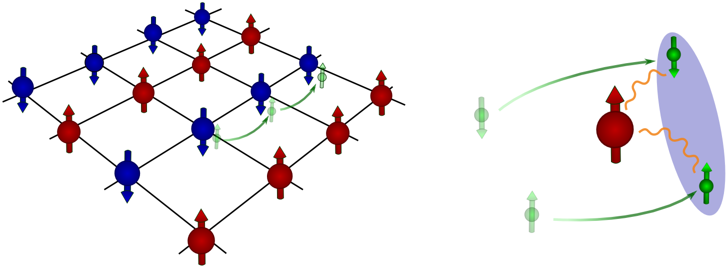


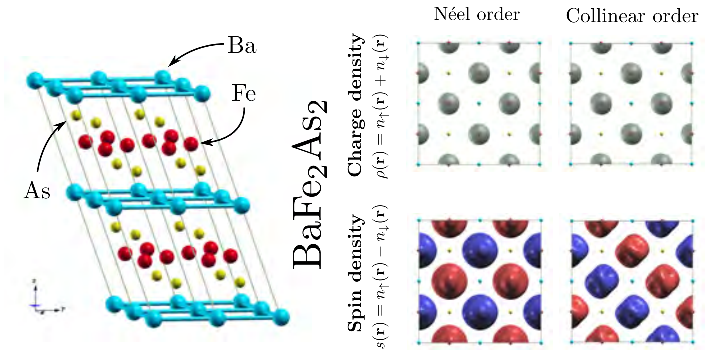
$\Longrightarrow$ High-temperature Superconductivity

We are looking for more materials that have this kind of coupling between charge and spin degrees of freedom



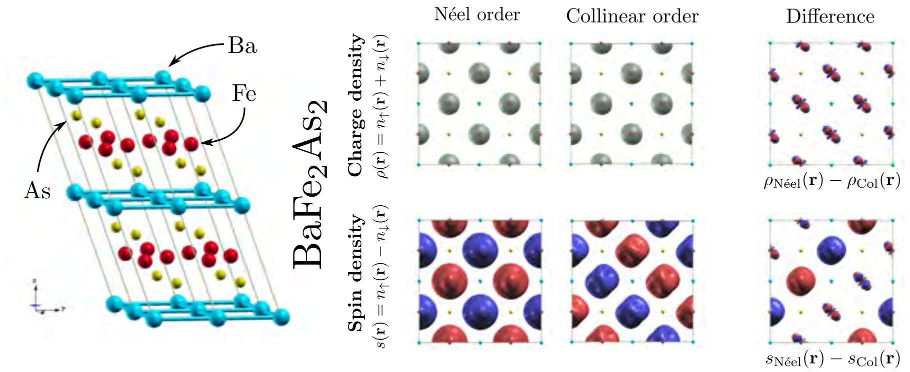
Is there a cheap way of estimating this coupling in a material?

A cheap way of estimating charge-spin coupling



Density Functional Theory (DFT) to calculate charge and spin densities.

A cheap way of estimating charge-spin coupling



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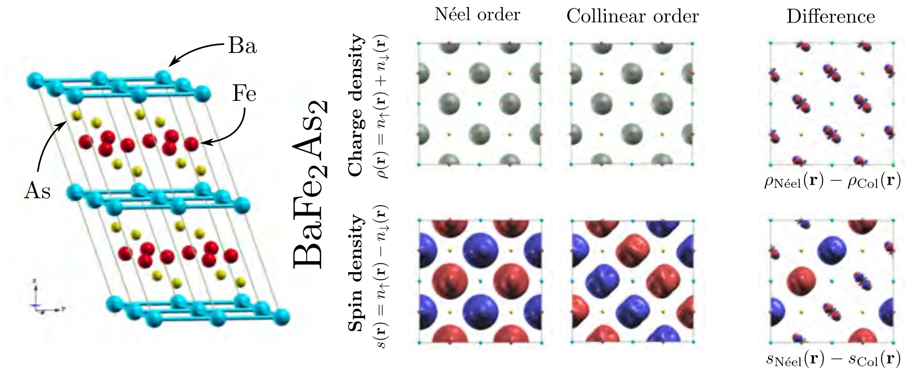
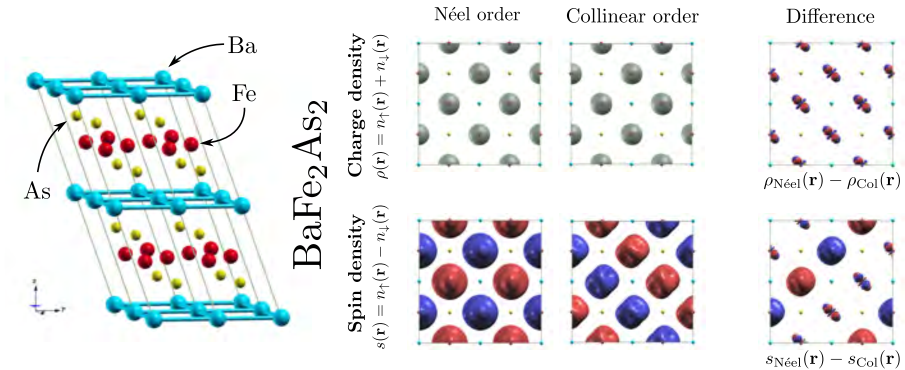


Charge-spin susceptibility, χ, as an estimator of charge-spin coupling:

$$\chi = \frac{\Delta\rho}{\Delta s} = \frac{\int \mathrm{d}\mathbf{r}\, |\rho(\mathbf{r}) - \rho_0(\mathbf{r})|}{\int \mathrm{d}\mathbf{r}\, |s(\mathbf{r}) - s_0(\mathbf{r})|}$$

A cheap way of estimating charge-spin coupling



Charge-spin susceptibility, χ, as an estimator of charge-spin coupling:

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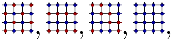
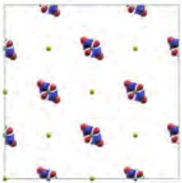
Average over magnetic textures: $\boxed{\chi_{cs} \equiv \frac{1}{N} \sum_i \frac{\Delta \rho_i}{\Delta s_i}}$ with $i =$ , ...

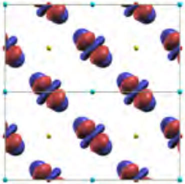
Charge-spin response **qualitatively different** across materials.

$BaCr_2As_2$



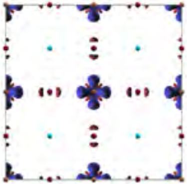
$\rho_{\mathrm{Col}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

$BaFe_2As_2$



$\rho_{\mathrm{Néel}}(\mathbf{r}) - \rho_{\mathrm{Col}}(\mathbf{r})$

$SrCuO_2$



$\rho_{\mathrm{FM}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

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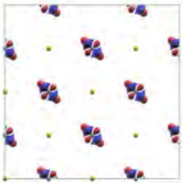
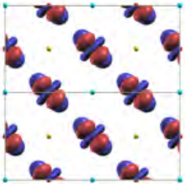


$\rho_{\mathrm{Col}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

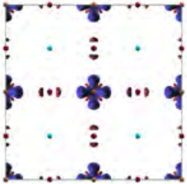
Weaker charge-spin response

$BaFe_2As_2$



$\rho_{\mathrm{Néel}}(\mathbf{r}) - \rho_{\mathrm{Col}}(\mathbf{r})$

$SrCuO_2$



$\rho_{\mathrm{FM}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

Stronger charge-spin response

Charge-spin response **qualitatively different** across materials.

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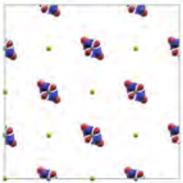

$\rho_{\mathrm{Col}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

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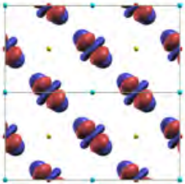

$\rho_{\mathrm{Néel}}(\mathbf{r}) - \rho_{\mathrm{Col}}(\mathbf{r})$

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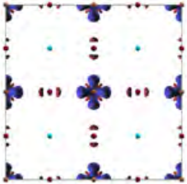

$\rho_{\mathrm{FM}}(\mathbf{r}) - \rho_{\mathrm{Néel}}(\mathbf{r})$

Stronger charge-spin response

The charge-spin susceptibility χ_{cs} gives a sense of how strong is the charge-spin response.

Can this coupling differentiate materials?

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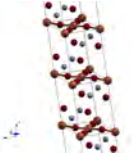
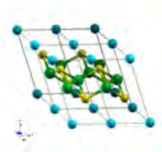
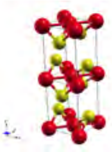
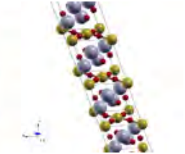
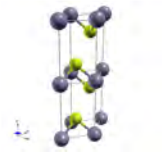
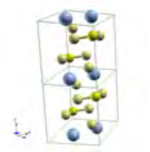
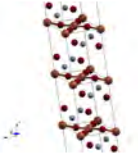
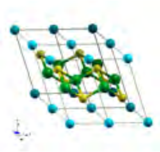
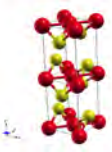
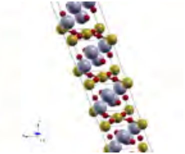
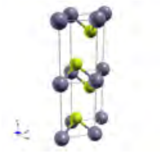
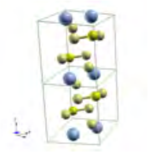
Our test set

Cuprates	Ba-122	FeX	214s	TMDCs	MPX_3
					
$SrCuO_2$, $CaCuO_2$, T-La_2CuO_4, T'-La_2CuO_4	BaM_2As_2 with M=Ni,Mn, Fe,Cr,Co,Cu.	FeX with X=Se,S,Te	La_2MO_4 (M=Co,Ni), Sr_2MO_4 (M=V,Cr, Mn,Fe,Co) and K_2MF_4 (M=Co,Ni,Cu)	MSe_2 (M=Ti, Nb,Ta,W) and MS_2 (M=Mo,Ta)	VPS_3, $NiPSe_3$, $CdPSe_3$, $CrGeTe_3$

Can this coupling differentiate materials?

Our test set

Cuprates	Ba-122	FeX	214s	TMDCs	MPX_3
					
$SrCuO_2$, $CaCuO_2$, T-La_2CuO_4, T'-La_2CuO_4	BaM_2As_2 with M=Ni,Mn, Fe,Cr,Co,Cu.	FeX with X=Se,S,Te	La_2MO_4 (M=Co,Ni), Sr_2MO_4 (M=V,Cr, Mn,Fe,Co) and K_2MF_4 (M=Co,Ni,Cu)	MSe_2 (M=Ti, Nb,Ta,W) and MS_2 (M=Mo,Ta)	VPS_3, $NiPSe_3$, $CdPSe_3$, $CrGeTe_3$

Unconventional high-temperature superconductors, strange metals, non-trivial magnetic ground states.

Can we compute the charge-spin response accurately yet cheaply?

Can we compute the charge-spin response accurately yet cheaply?

Use density functional theory (DFT)
(some quantum Monte Carlo for benchamark)

DFT+U functional (which simulates strong electron-electron interactions in transition metal atoms d-orbitals)

Multiple DFT+U calculations to control errors.

($U = 0, 5$ and 10 eV [details in arXiv:1810.03014])
[Cococcioni and Gironcoli PRB 71, 035105 (2005)]

High-throughput calculations with **Blue Waters**

Multiple DFT+U

$+$

Several magnetic orders

$\simeq$

1000 node-hours per material.

High-throughput calculations with **Blue Waters**

Multiple DFT+U

$+$

Several magnetic orders

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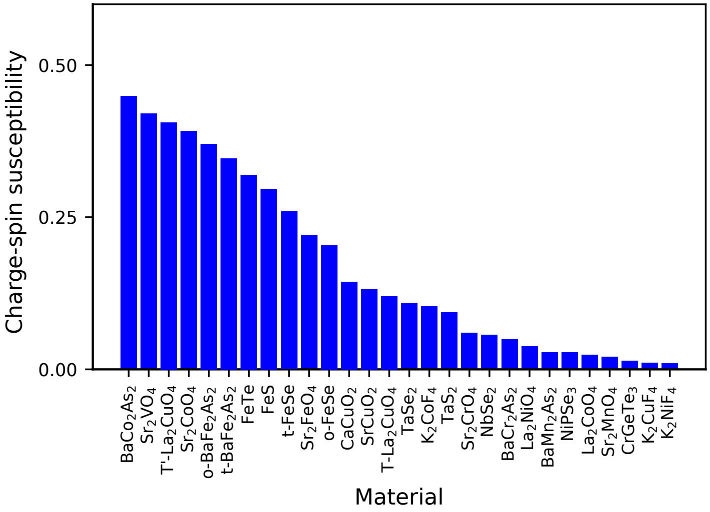
1000 node-hours per material.

Checking accuracy of charge-spin susceptibility (from DFT+U) with diffusion Monte Carlo on small set of materials ($\simeq$ 40000 node-hours per material).

Materials in our test set according to their charge-spin susc (for $U = 5$ eV)



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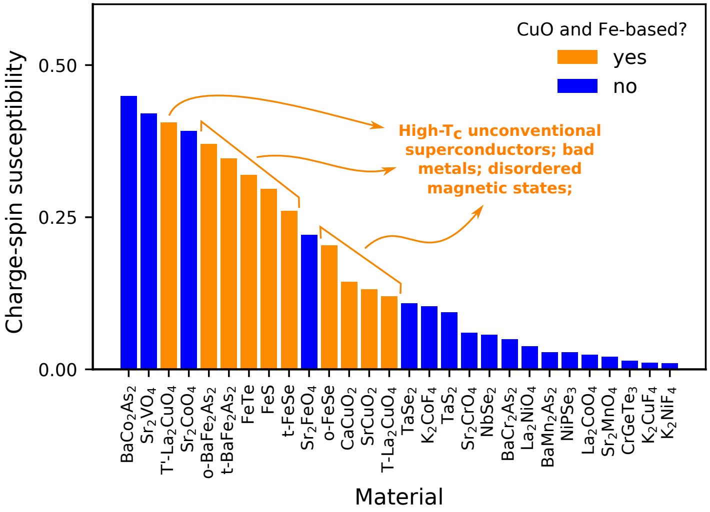

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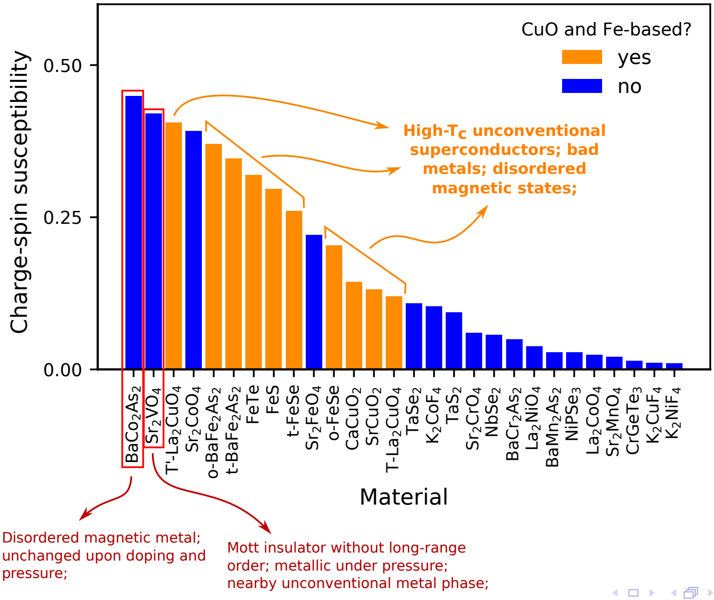

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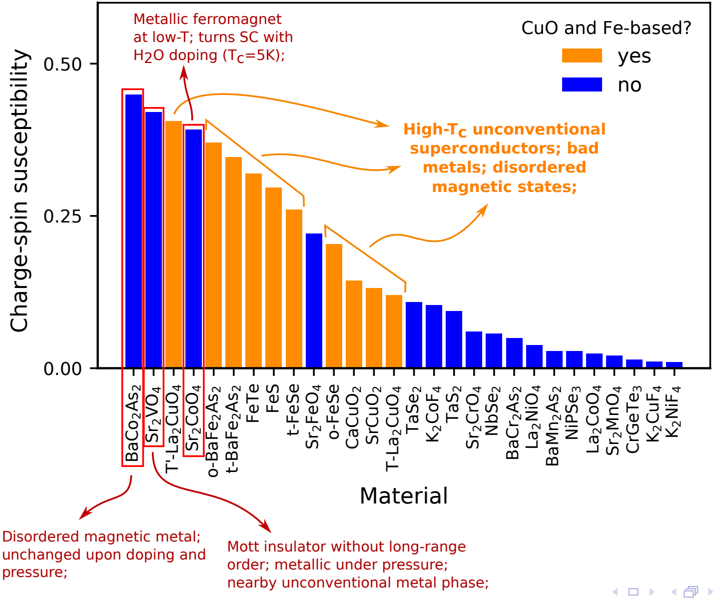

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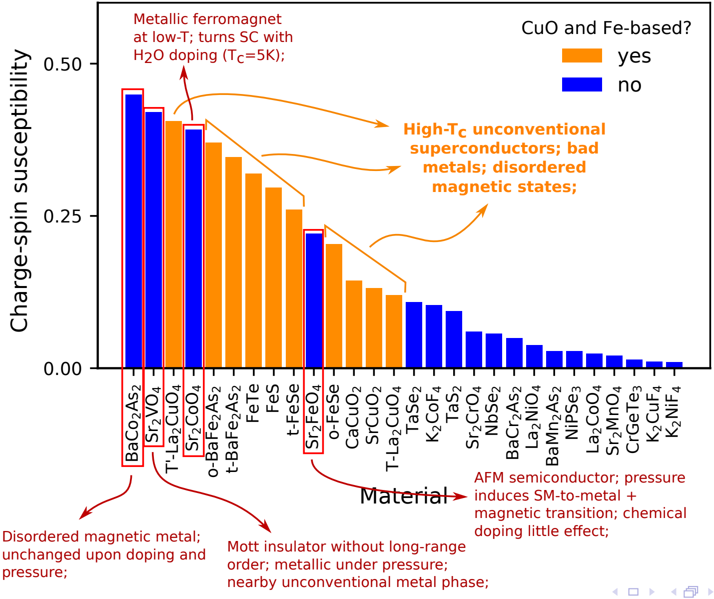

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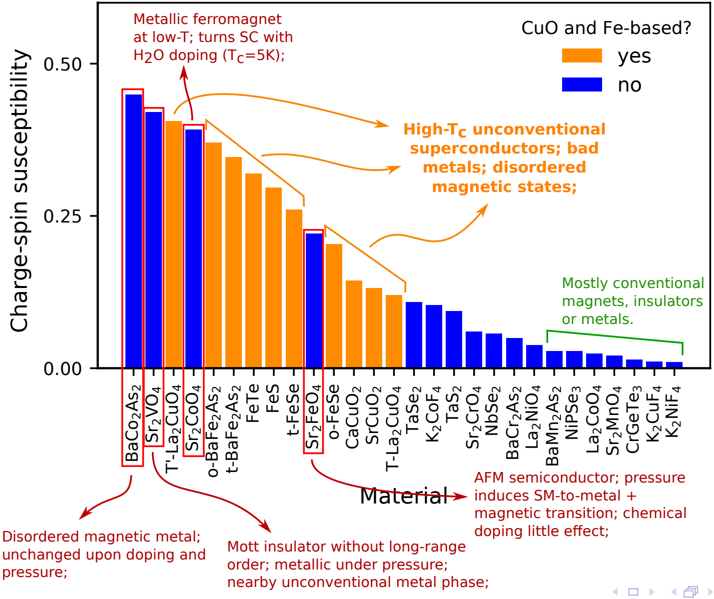


Accomplishments: Solid evidence that materials with **medium-to-large charge-spin coupling** generally exhibit uncommon correlated phases of matter.

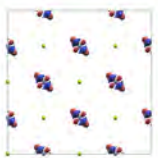
Broader impacts: New computational probe of electronic correlations in quantum materials.
Accelerate material discovery $\Rightarrow$ computational-guided searches.

Currently looking into a set of scarcely studied materials

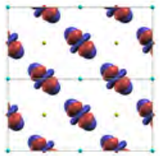
Charge-spin susceptibility identified some interesting ones

Working with experimental group at UIUC which is interested in exploring these materials

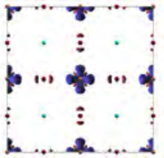
Summary



$BaCr_2As_2$



$BaFe_2As_2$



$SrCuO_2$

Charge-spin susceptibility is easy to compute. Probes charge-spin coupling.

Materials with large charge-spin coupling generally present uncommon correlated pases.

Working with experimental group to explore new materials

arXiv:1810.03014