# Nonequilibrium Physics of Correlated Electron Materials I:

## Overview and an unconventional view of the physics

**A. J. Millis**

**College de France**
**Sept 21, 2015**

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Columbia University

# Modes of nonequilibrium

- **Transient perturbation**
  - **excite many electrons from ground state**
- **Seady-state drive**
  - **dc (current)**
  - **ac (oscillating field)**



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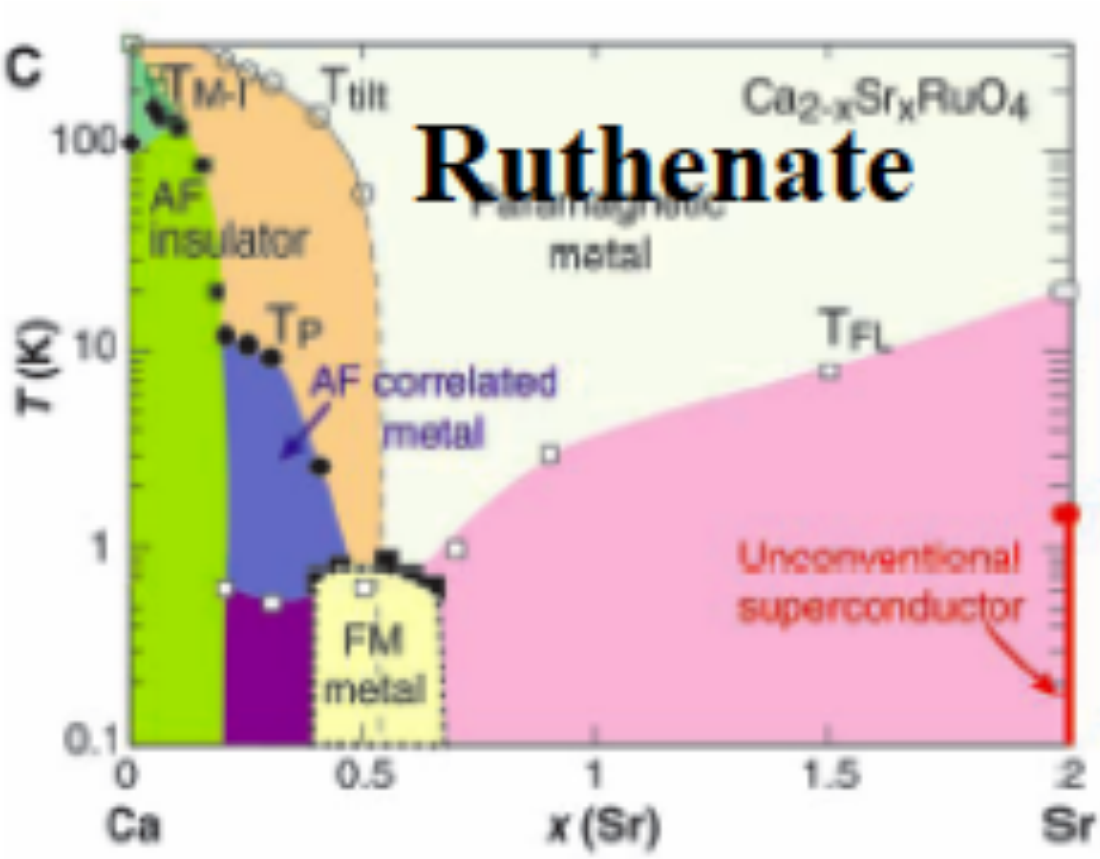
# Current flow

## Example $Ca_2RuO_4$

**Ruthenate**



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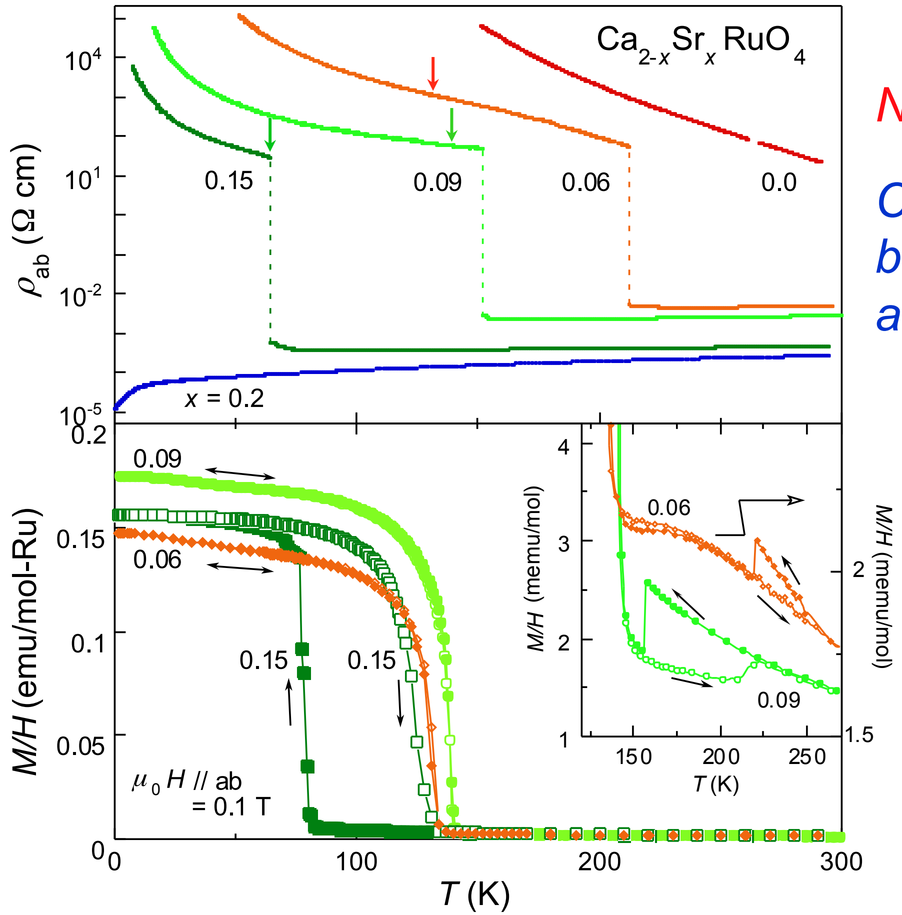
# From Y. Maeno

## Mott transition in $Ca_{2-x}Sr_xRuO_4$




*No anomaly at $T_N$*

*Clear separation between "charge gap" and "spin exitation".*

*Mott insulating ground state*

Nakatsuji *et al.*, PRL.



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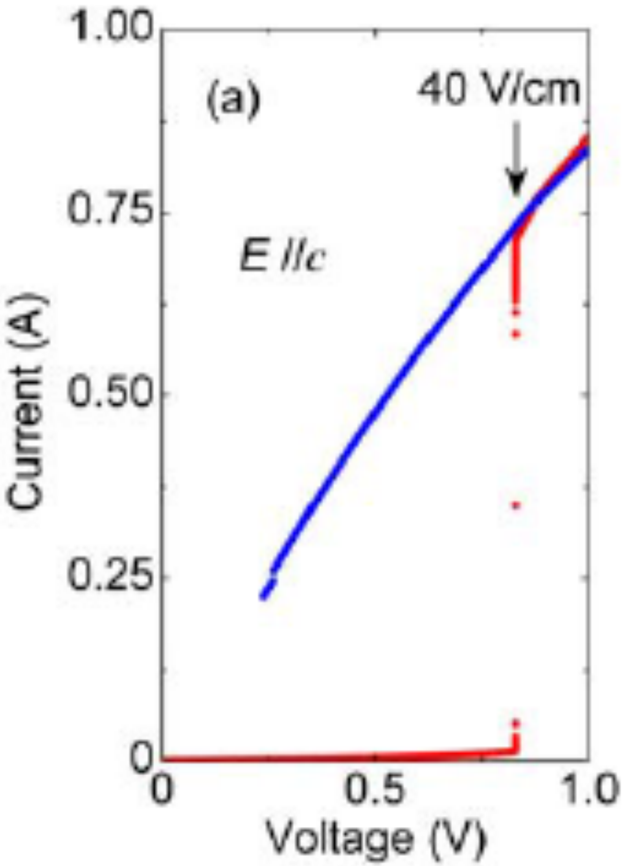
# Metallic state persists to low T under current flow




**Metallic**

$\rho \approx \mathbf{0.4}\ \mathbf{\Omega\text{-cm}}$

**Insulating**

$\rho \approx \mathbf{60}\ \mathbf{\Omega\text{-cm}}$



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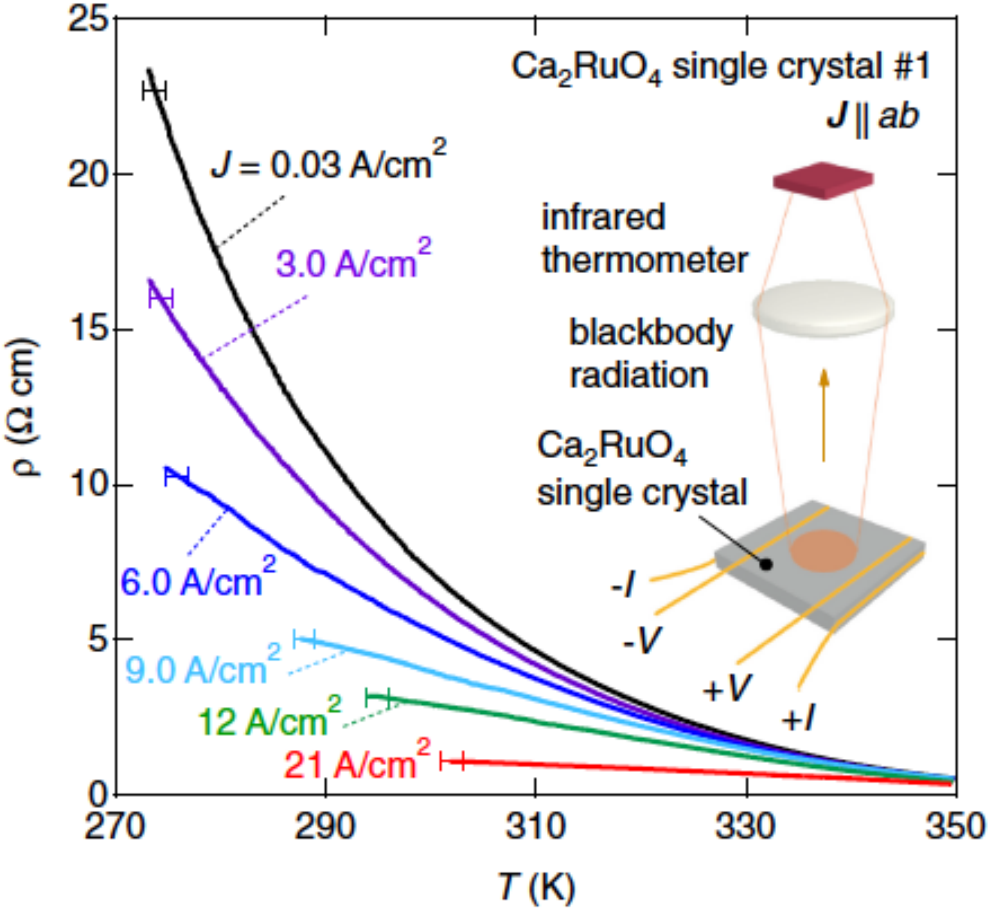
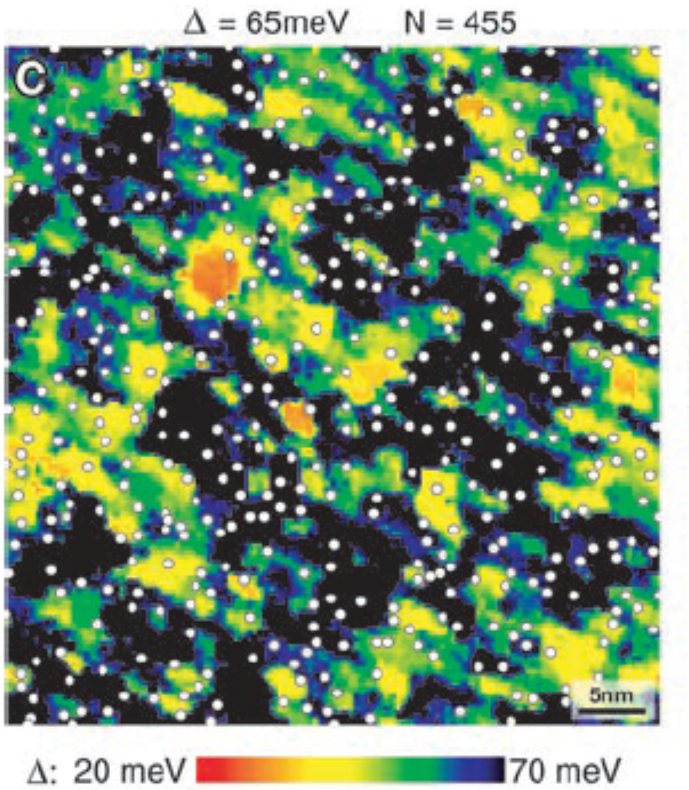
# Heating?




Journal of the Physical Society of Japan **82** (2013) 103702

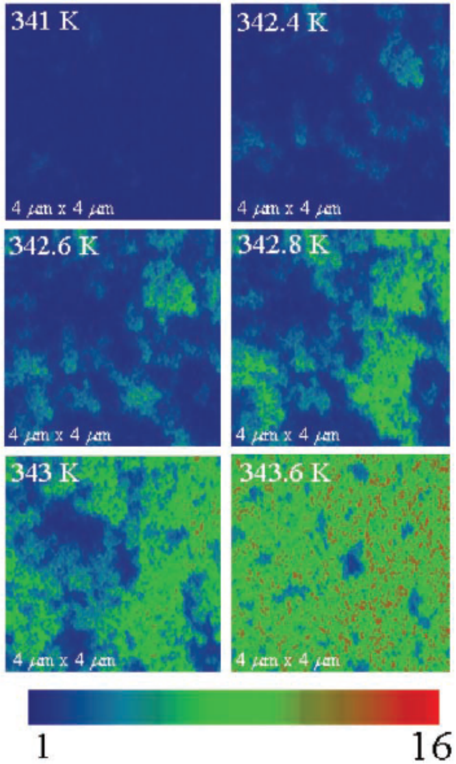
# filamentary conduction

## BSCCO high-$T_c$




K.McElroy et al, Science **309** 1048 (2005)

## $VO_2$




Qazilbash…Basov et al, Science **318** 1750 (2007)

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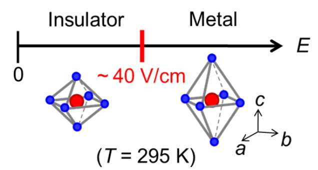
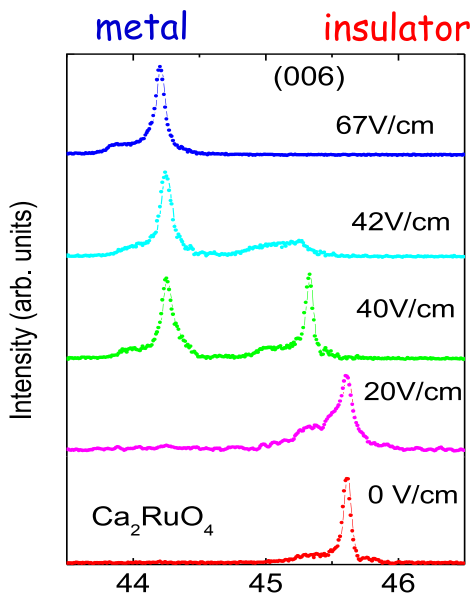
# From Y. Maeno\

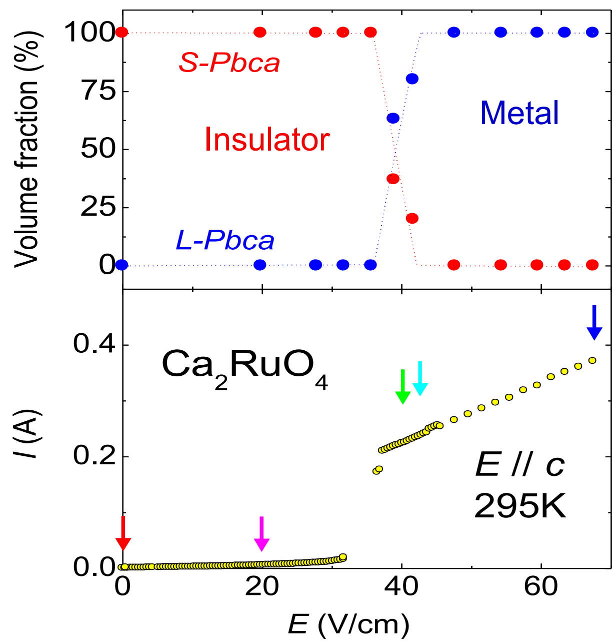
*E*-induced structural transition
from S- to L-Pbca

Dielectric Breakdown in $Ca_2RuO_4$
is the bulk transition.




X-ray spectra

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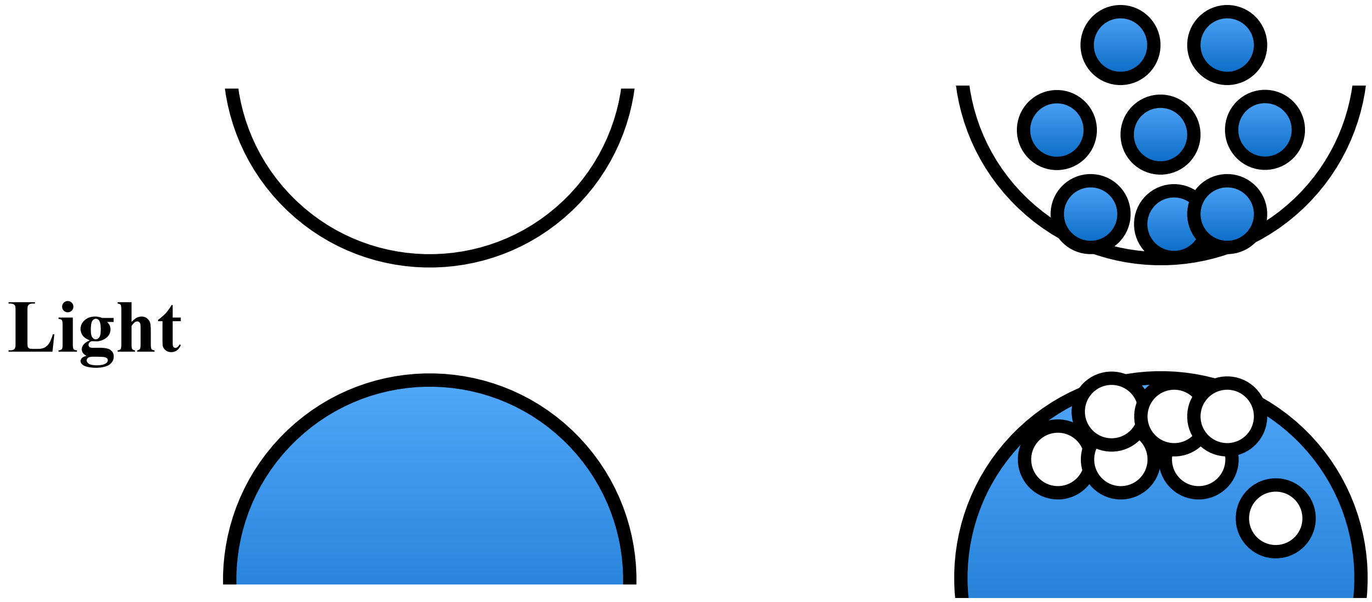
# Current flow

**Genuine nonequilibrium steady state**

**??Properties??**

# Radiation pulse: excite  many carriers




**Issue: if gap is of many-body origin, can exciting the carriers rearrange the electronic structure?**

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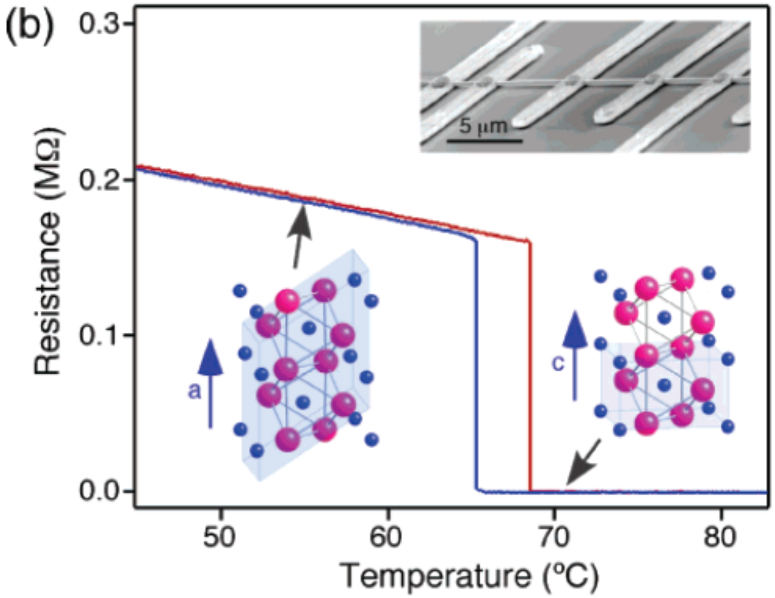
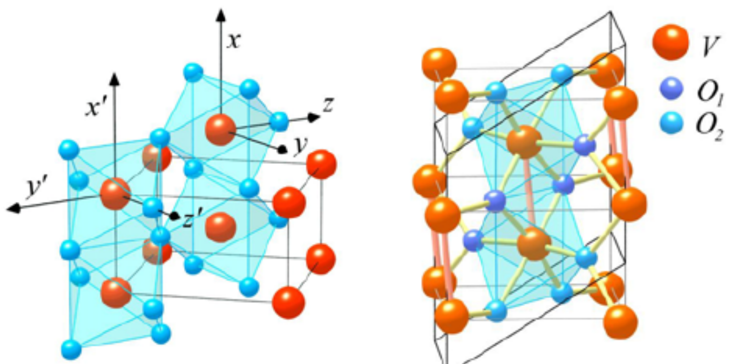
# Long-lived response to pulse

**VO2: high T metal, low T insulator. Low T phase has dimerization and monoclinic distortion, is termed M1**




The metal-insulator transition is accompanied by a structural transition with dimerization of the V atoms and tilting of the pairs out of the z axis.
(From V. Eyert, Ann. Phys. (Leipzig) 11, 650-702 (2002))

Morrison et al Science 346 445

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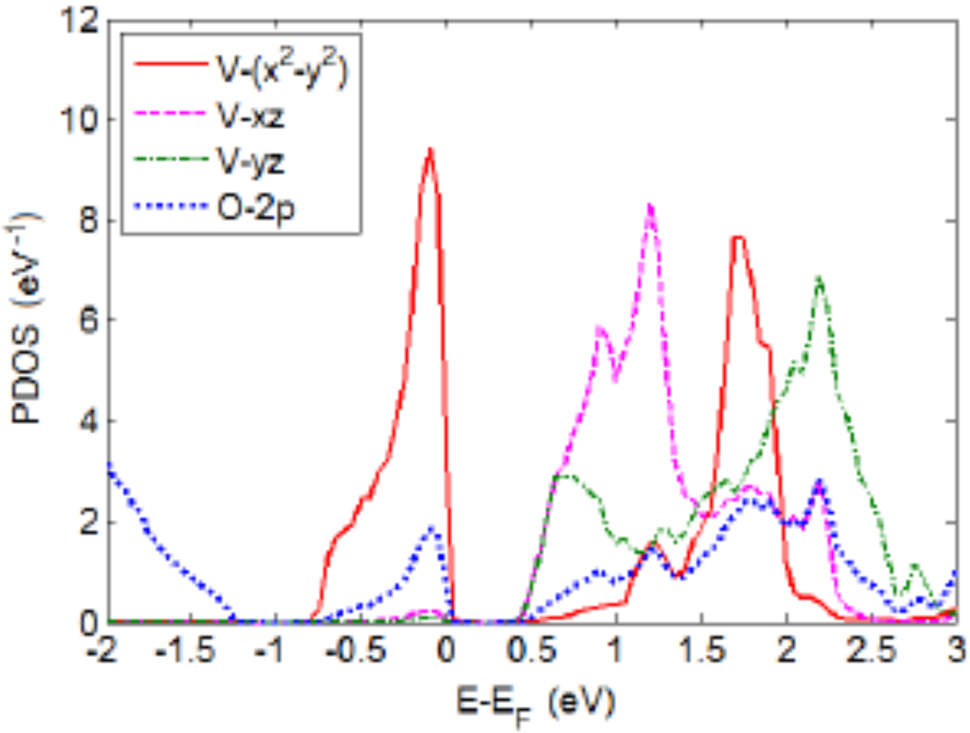
# Hartree-Fock band structure
# Z. He




**Insulating behavior is associated with almost complete 'orbital ordering' ($x^2$-$y^2$ is the orbital that forms the dimer)**



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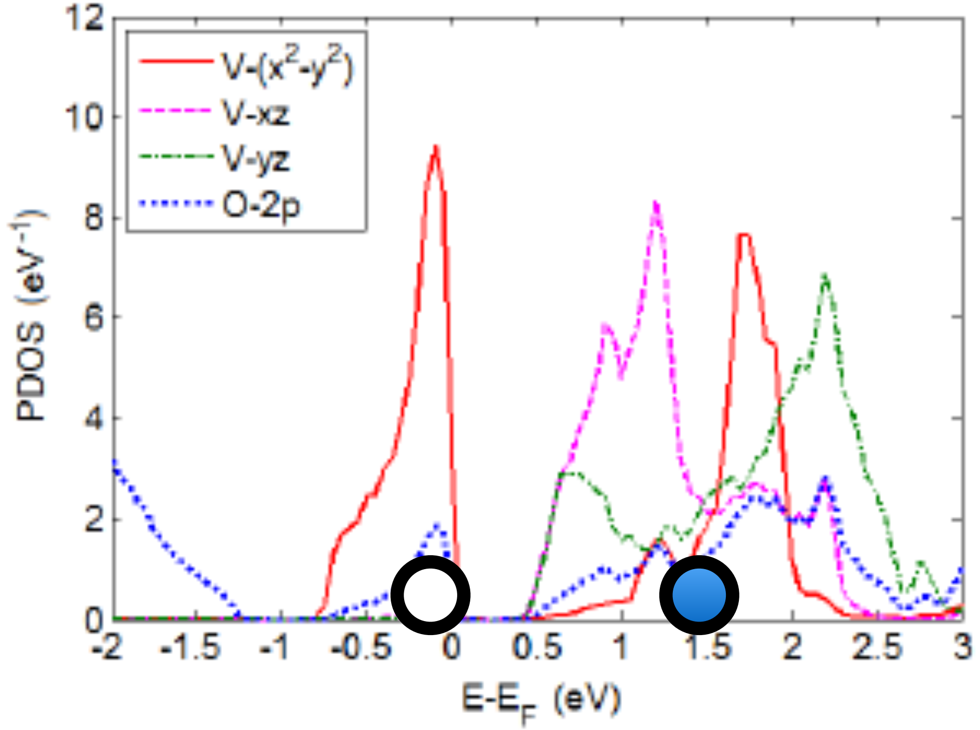
# $VO_2$ experiment

**Apply ~1.5eV pulse.**
**Measure diffraction and reflectivity R**

Morrison et al Science 346 445




**Measure of laser intensity: `fluence’**

**Zhuoran: excitation density ~10%/cell**

Morrison et al Science 346 445

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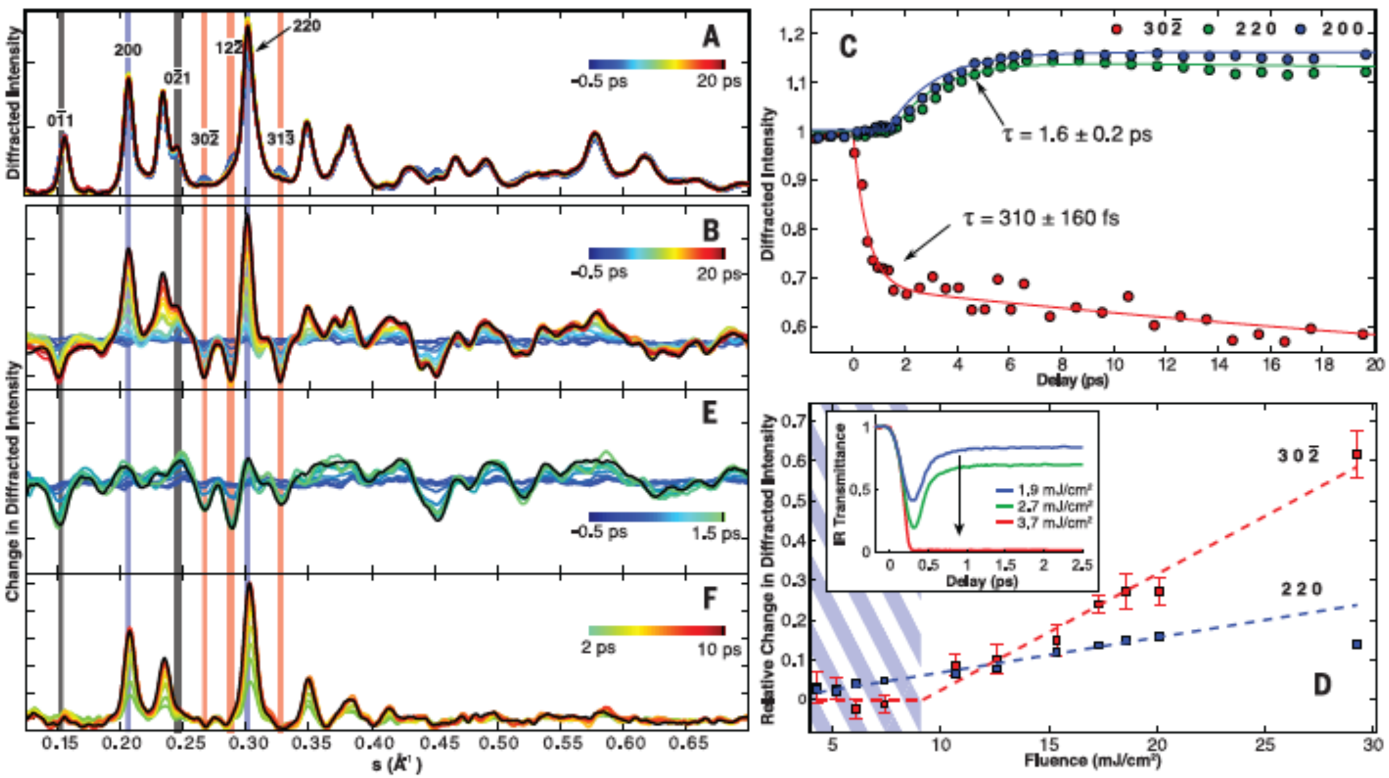
# $VO_2$ experiment

**Apply ~1.5eV pulse. Measure diffraction and reflectivity R**

Morrison et al Science 346 445

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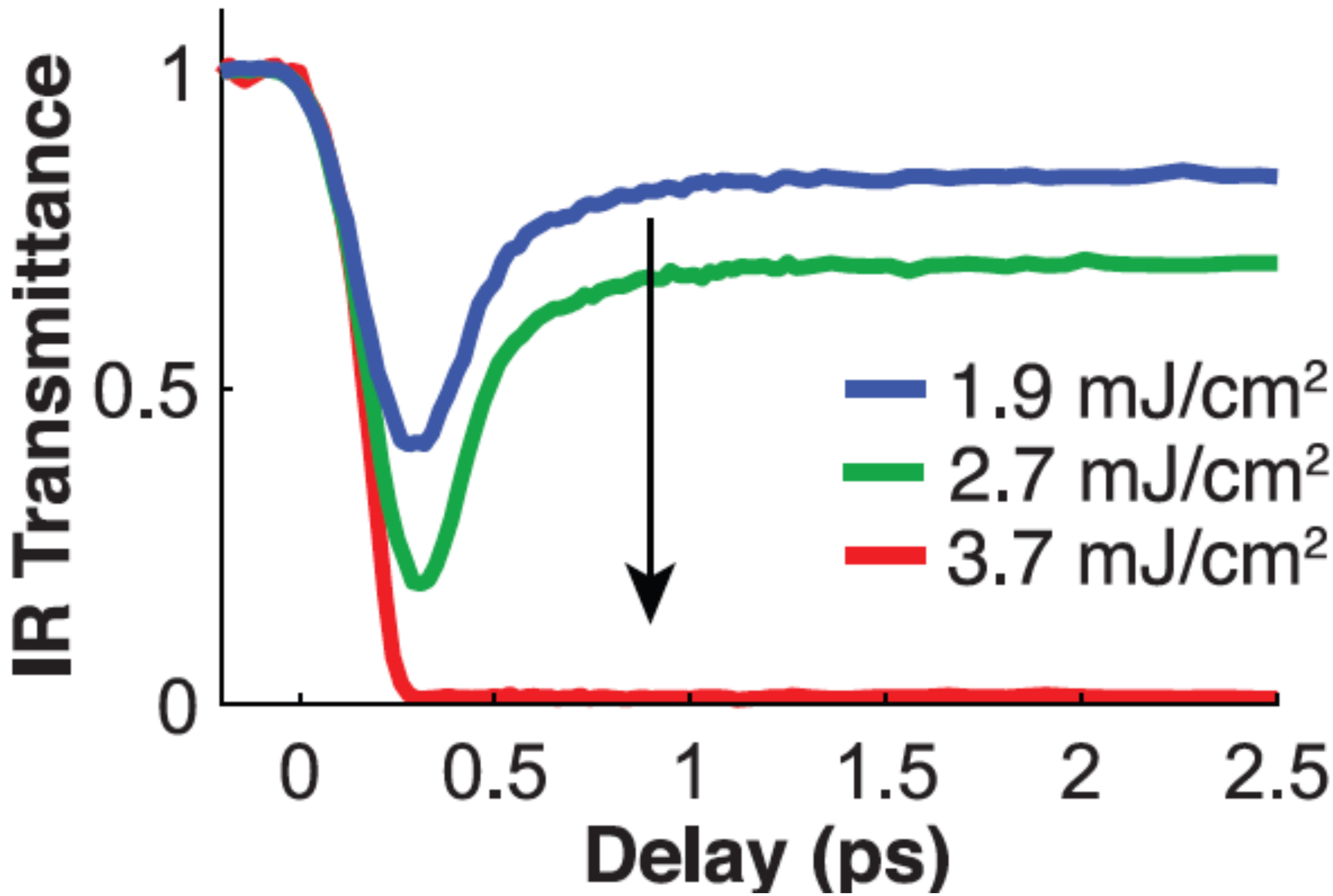
# Modest fluence: long-lived change of state

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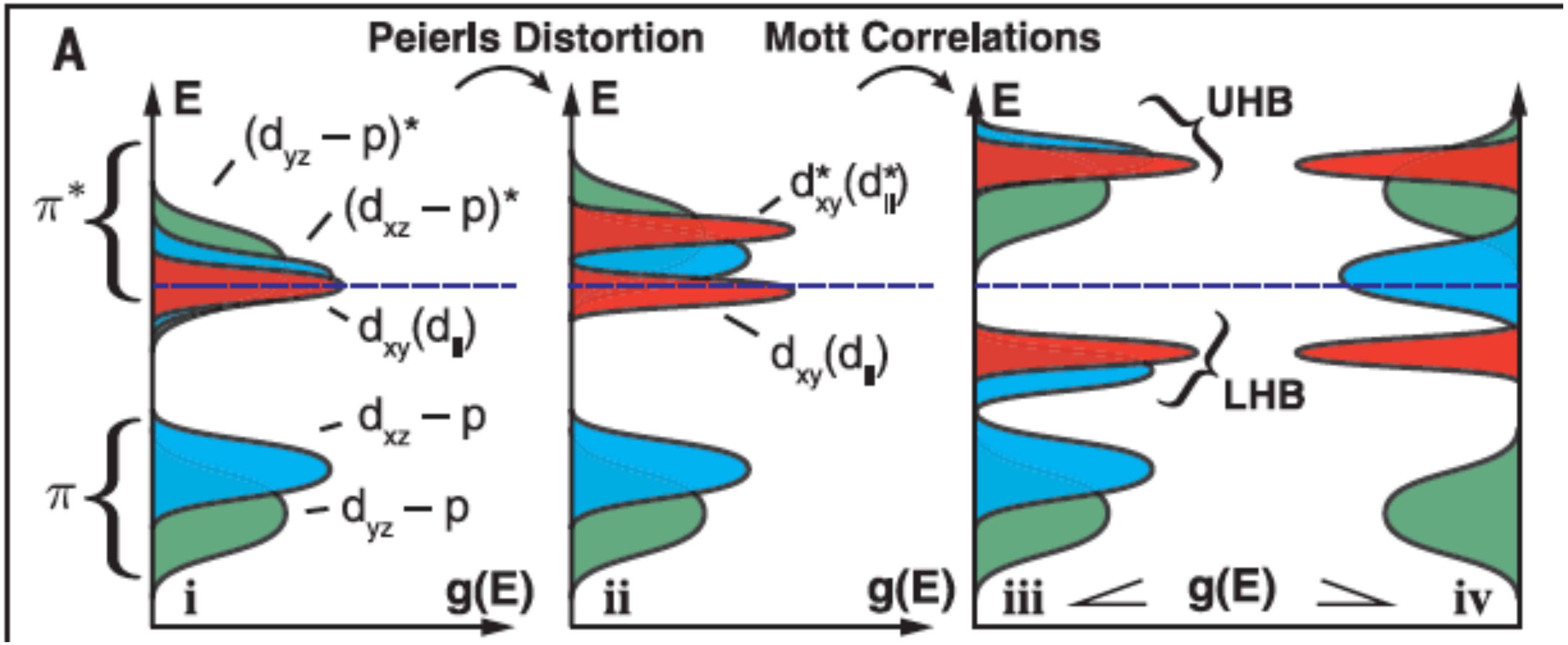
# $VO_2$ experiment

**Interpretation**

Morrison et al Science 346 445




**Story: IR pulse=>drives long-lived orbital rearrangement without obvious change in lattice**

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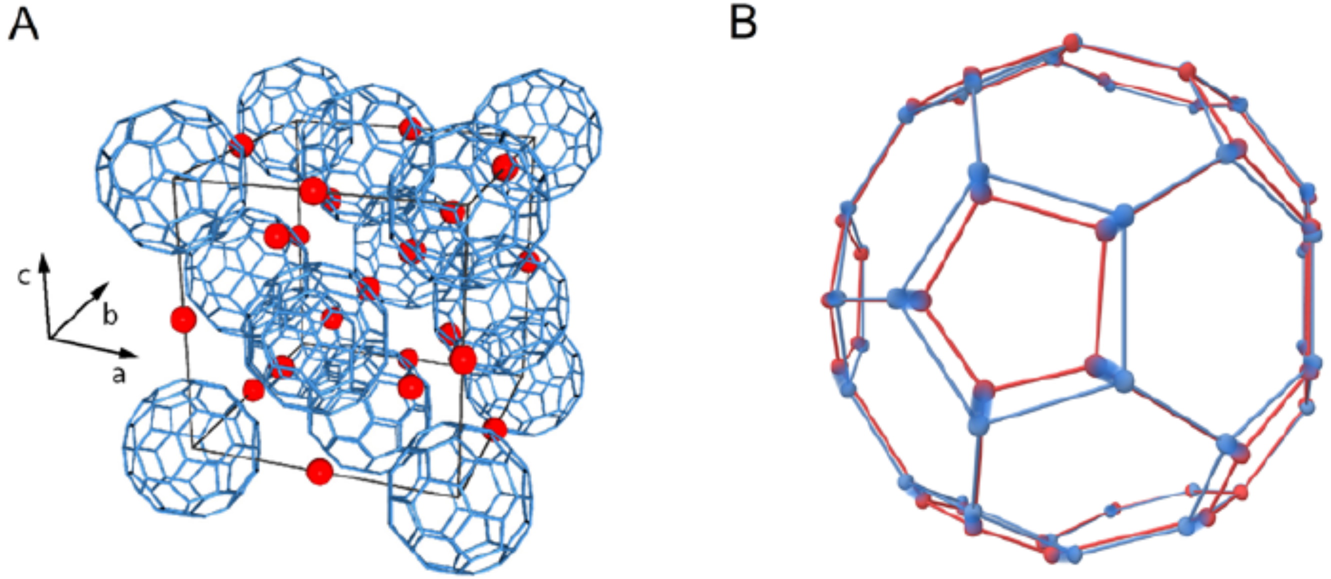
# AC Drive

## A. Cavalleri et al: excite phonon in $K_3C_{60}$




arXiv:1505.04529

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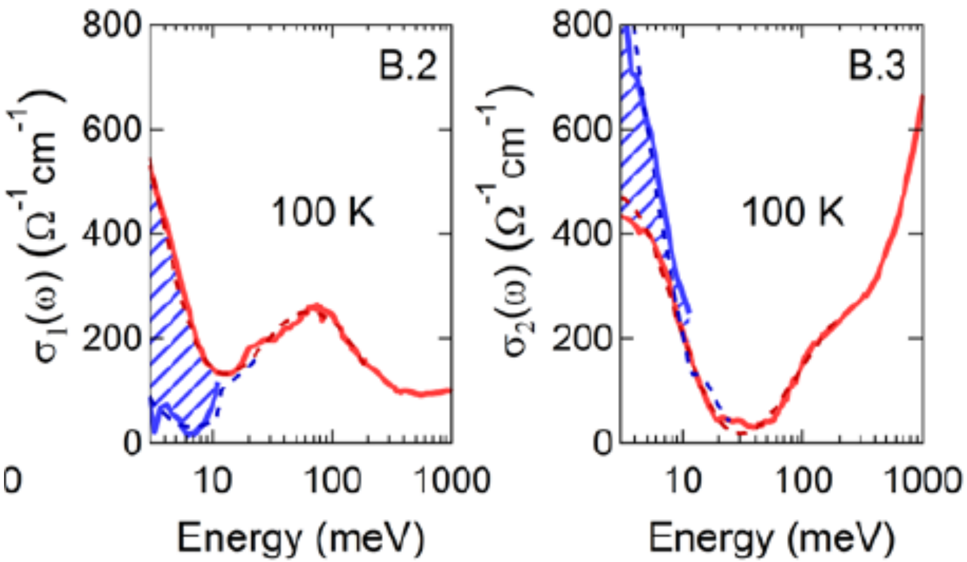
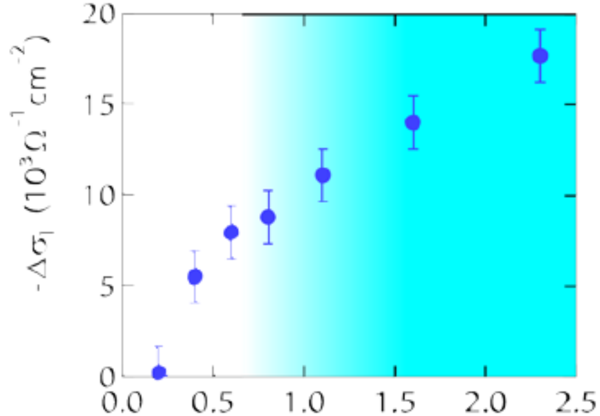
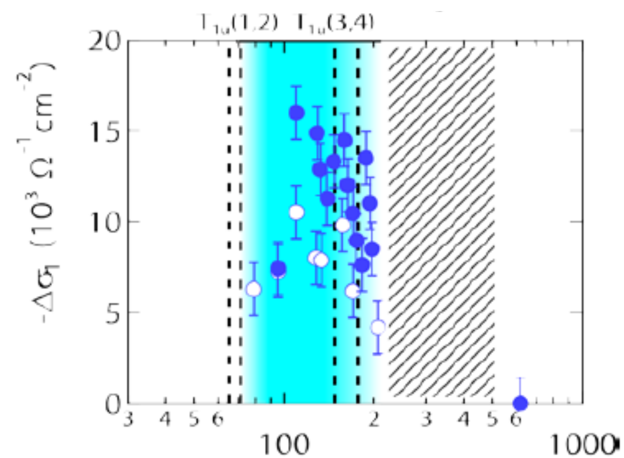
# Substantial change in conductivity





**decrease in real part, increase in im part strongly suggestive of superconducivity**

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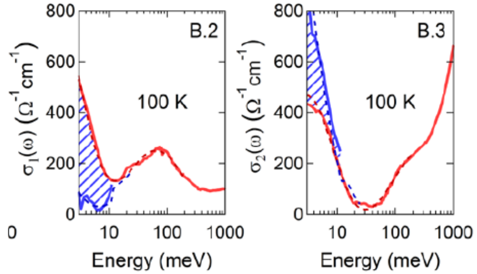
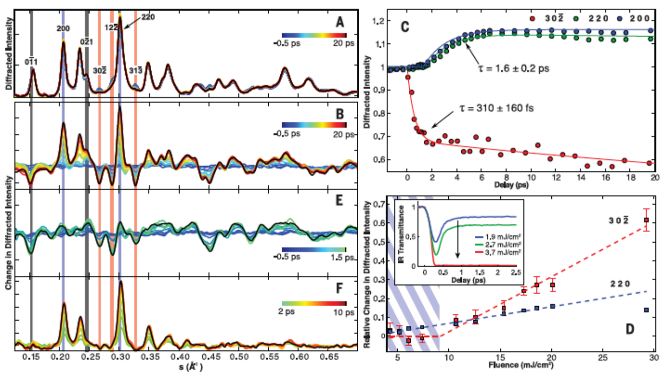
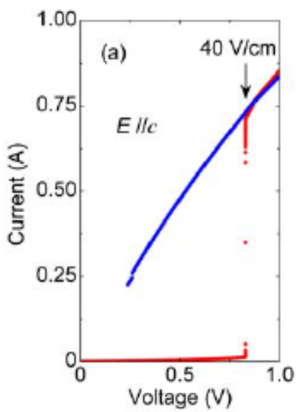
# Summary




- **Modest-amplitude nonequilibribum drives can change the state of a correlated electron material**

**Challenge: physical understanding**

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# Key point

**Each class of experiment leads to interesting long-lived regimes, different from known equilibrium phases**

## Questions

- **What new regimes can we discover by applying strong dynamical perturbations?**
- **Are these new regimes actual phases**
- **What are the generic properties?**



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## the subject presents two difficulties:

- **The correlated electron part**
- **The nonequilibrium part**



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# Some very simple remarks

**Hamiltonian**

$$\mathbf{H} = \sum_i \frac{-\nabla_i^2}{2m_e} + \sum_i V_{ext}(r_i) + \frac{1}{2}\sum_{i \neq j} \frac{e^2}{|r_i - r_j|}$$

**Single particle part**

**Interaction part**



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# Some very simple remarks

**Modification: incorporate average effect of interactions into single particle potential**

$$H = \sum_i -\frac{\nabla^2}{2m_e} + \sum_i V_{eff}(r_i) + \frac{1}{2}\sum_{i\neq j} : \frac{e^2}{|r_i - r_j|} :$$

**Mean field part**

**Residual interactions**



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# Correlations

**``Weakly correlated": renormalized single particle picture applies, with residual interactions treated perturbatively:**

**``Strongly correlated": crucial aspects of physics are outside of renormalized single particle description**

# Equilibrium

**Key quantity: partition function, expressed as imaginary time integral**

$$\mathbf{Z} = \sum_{\mathbf{n}} \mathbf{e}^{-\frac{\mathbf{E_n}}{\mathbf{T}}} \equiv \mathbf{Tr}\left[\mathbf{e}^{-\int_0^{\frac{1}{\mathbf{T}}} \mathbf{d}\tau \mathbf{H}(\tau)}\right]$$

- **Powerful physically-motivated understanding of general structure**
- **Numerics: estimation of combinations of decaying exponentials**



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# Non-equilibrum

**Key quantity: density matrix, expressed as two-contour real-time integral**

$$\hat{\rho}(\mathbf{t}) = \mathbf{e}^{-\mathbf{i}\int_0^t \mathbf{dt'H(t')}}\hat{\rho}(\mathbf{t}=\mathbf{0})\mathbf{e}^{\mathbf{i}\int_0^t \mathbf{dt''H(t'')}}$$

- **Very little understanding of general structure: what you do is integrate forward from initial condition**
- **Numerics: estimation of combinations of oscillating exponentials**


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# Weakly correlated materials out of equilibrium

$$\hat{\rho}(\mathbf{t}) = \mathbf{e}^{-\mathbf{i}\int_0^t \mathbf{dt}'\mathbf{H}(\mathbf{t}')}\hat{\rho}(\mathbf{t}=\mathbf{0})\mathbf{e}^{\mathbf{i}\int_0^t \mathbf{dt}''\mathbf{H}(\mathbf{t}'')}$$

**Physics is (renormalized) independent particles plust perturbative interactions: integration forward in time is manageable**

**??Strongly correlated nonequilibrium??**



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# Equilbrium physics: conceptual framework

- **Partition function <=> path integral**

$$\mathbf{Z} \text{``} = \text{''} \int \mathcal{D}\{\phi\} \; \mathbf{e}^{-\mathbf{S}[\{\phi(\tau)\}]}$$

- **In many cases: path integral dominated by saddle point +gaussian fluctations**

$$\mathbf{Z} \rightarrow \mathbf{e}^{-\mathbf{S}^*} \int \mathcal{D}\psi_{\mathbf{a}} \mathcal{D}\psi_{\mathbf{b}} \ldots \; \mathbf{e}^{-\frac{1}{2} \int \mathbf{d}\tau_{\mathbf{1}} \mathbf{d}\tau_{\mathbf{2}} \sum_{\mathbf{ab}} \psi_{\mathbf{a}}(\tau_{\mathbf{1}}) \chi^{-\mathbf{1}}(\tau_{\mathbf{1}} - \tau_{\mathbf{2}}) \psi_{\mathbf{b}}(\tau_{\mathbf{2}})}$$



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# thus

$$S_{\mathbf{gaussian}} = -\frac{1}{2}\int \mathbf{d}\tau_{\mathbf{1}}\mathbf{d}\tau_{\mathbf{2}} \sum_{\mathbf{ab}} \psi_{\mathbf{a}}(\tau_{\mathbf{1}})\chi^{-1}(\tau_{\mathbf{1}}-\tau_{\mathbf{2}})\,\psi_{\mathbf{b}}(\tau_{\mathbf{2}})$$

- **Identify fixed point (`phase')**
- **Identify important fluctuations  (quasiparticles)**
- **quasiparticle propagators <=> linear response susceptibilies**

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# Criticality

- **Nonlinearities important near second order phase transition**
- **Consequence: change functional form of susceptibilibity**


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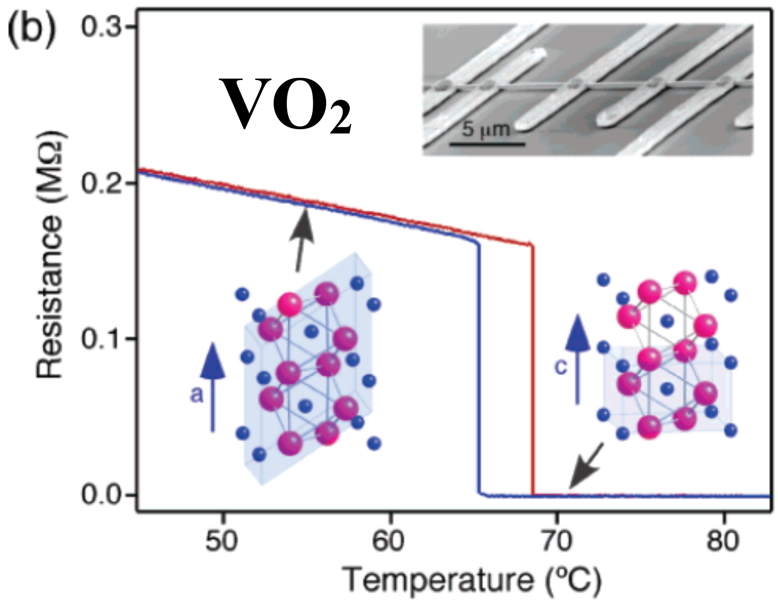
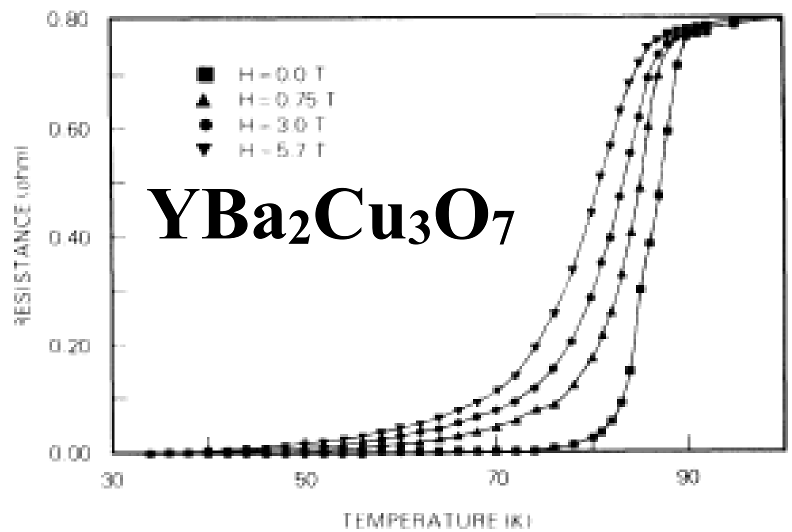
# Correlated electron materials: striking electronic behaviors

## metal-insulator transition




Wu et al, Nanoletters 6 2313 (2006)

## High transition temperature superconductivity




Wu et al, PRL 58 (1987)

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# In these and many other cases

**It seems that the gaussian fluctuation/ quasiparticle picture does not tell us what we need to know**



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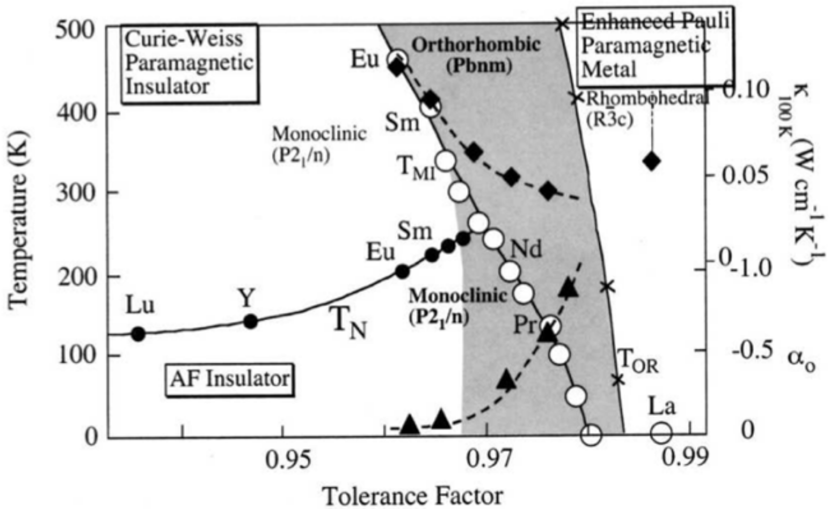
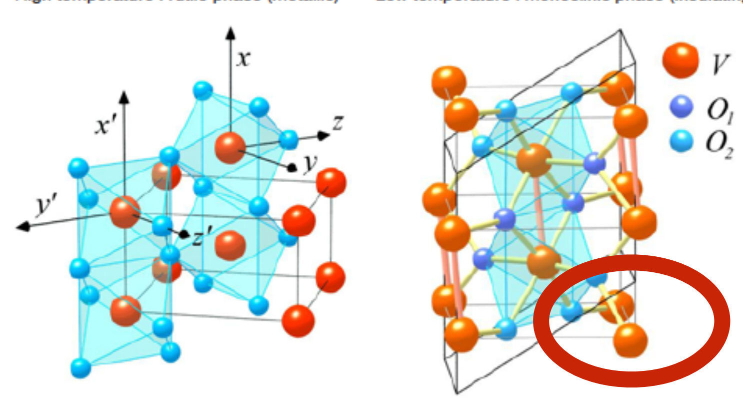
# **Nontrivial behavior often associated with lattice distortion**

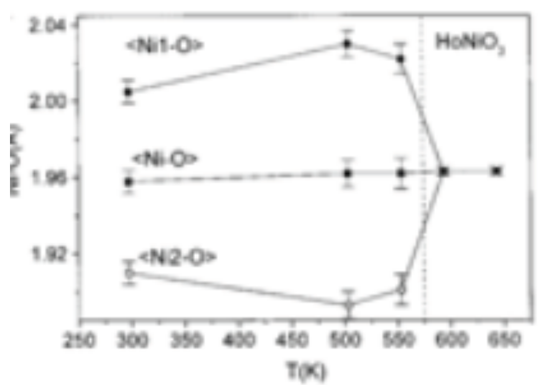



# $VO_2$




The metal-insulator transition is accompanied by a structural transition with dimerization of the V atoms and tilting of the pairs out of the z axis.
(From V. Eyert, Ann. Phys. (Leipzig) 11, 650-702 (2002))

# $RNiO_3$




J A Alonso et al PRL 82 3871 (1999)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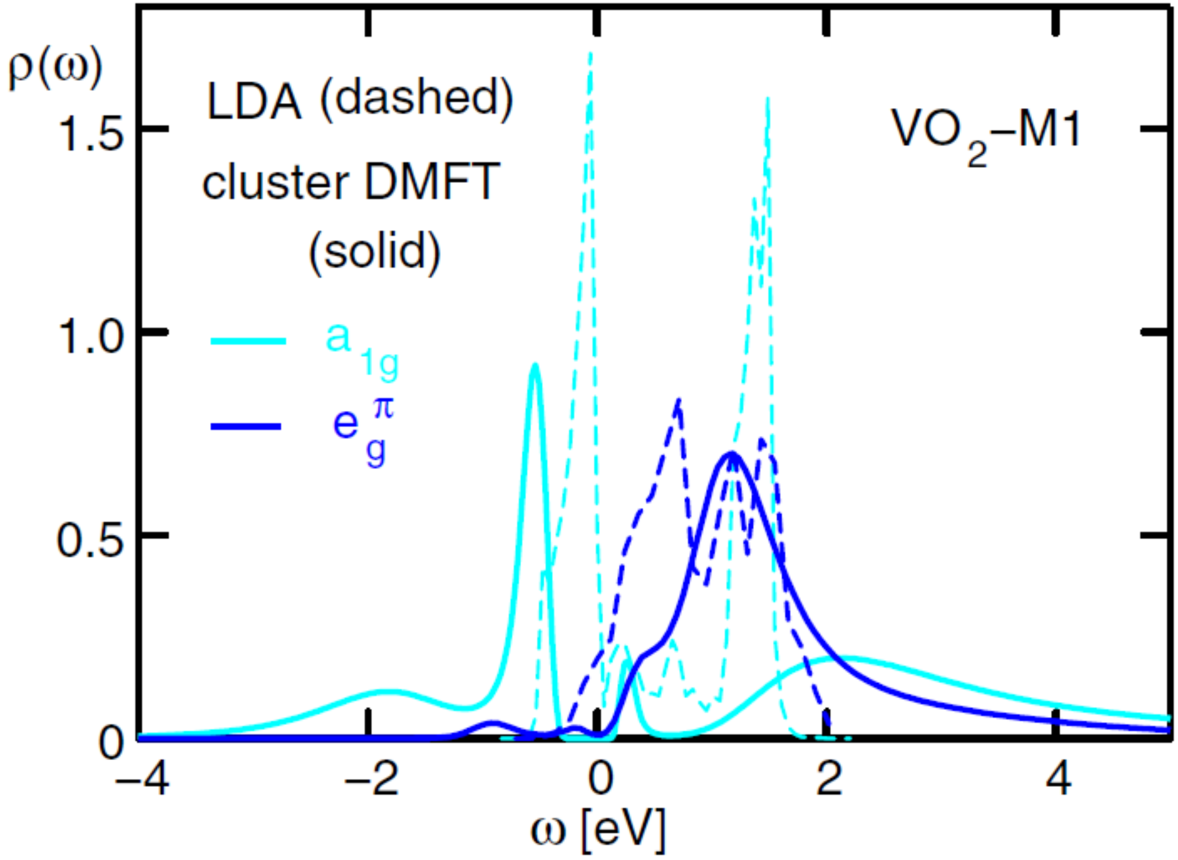
# Lattice distortion by itself not enough to produce interesting behavior




Biermann_PRL 94, 026404 (2005)

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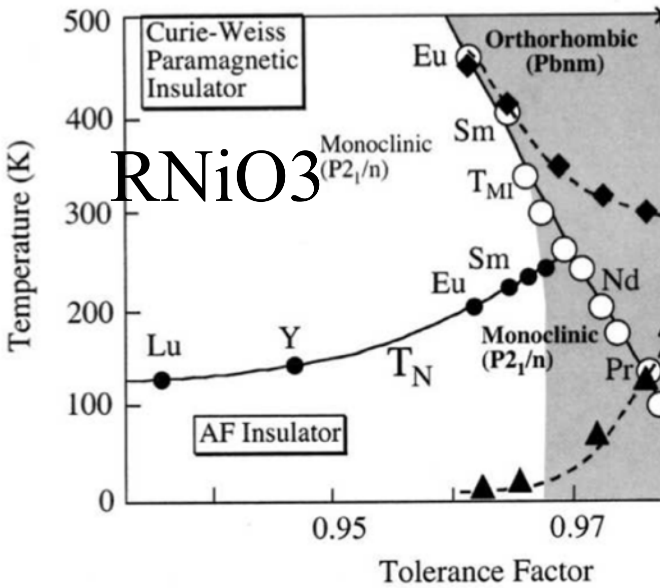
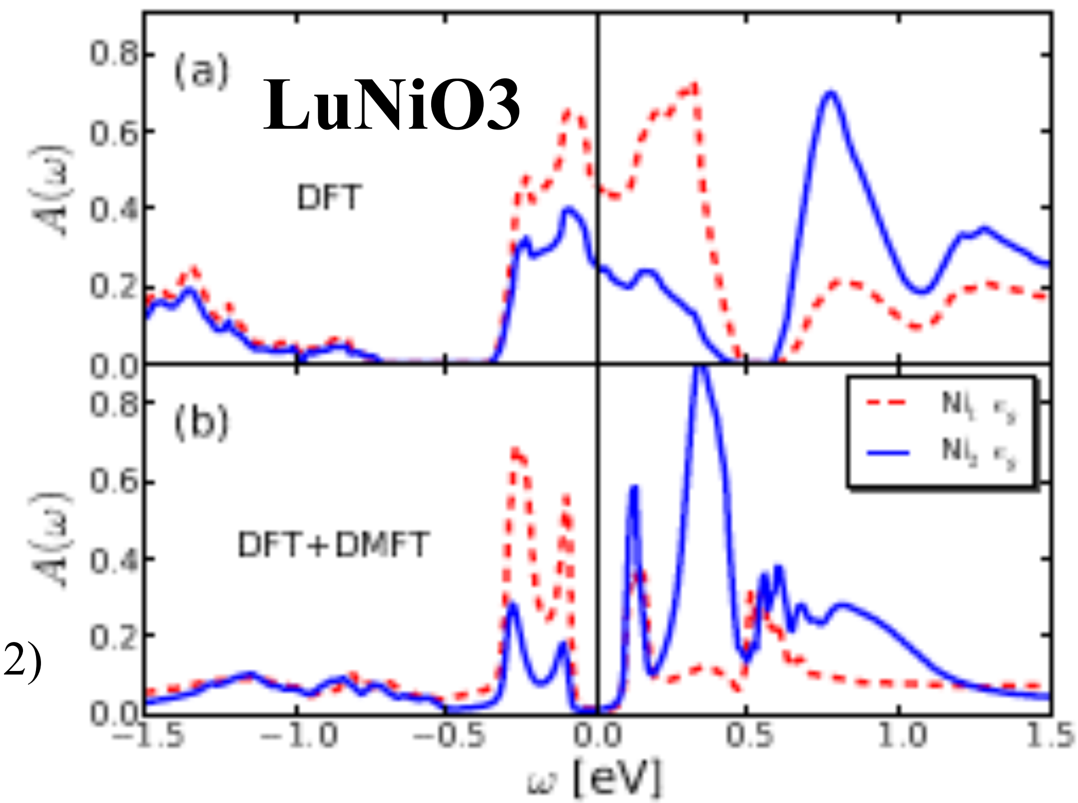
# Lattice distortion not enough




H. Park, AJM, and C. Marianetti, Phys. Rev. Lett. 109, 156402 (2012)




**DFT (LDA or GGA)=>metal, even in expt structure**

**Minimizing DFT energy=>no distortion**

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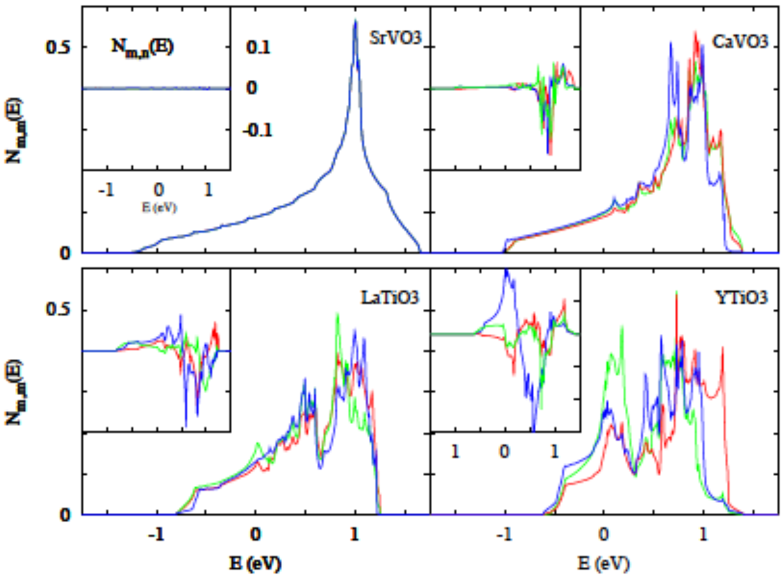
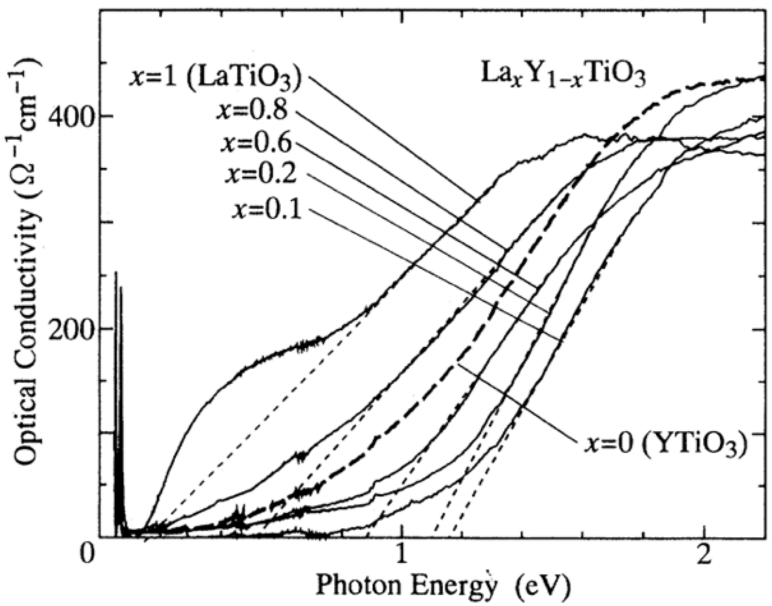
# (LaY)TiO3:
# Canonical `Mott' Insulator




**Pavarini et al 2004**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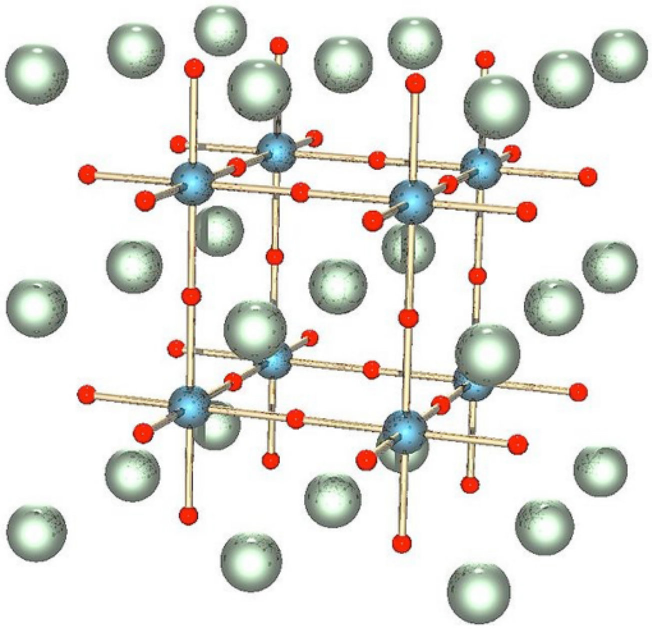
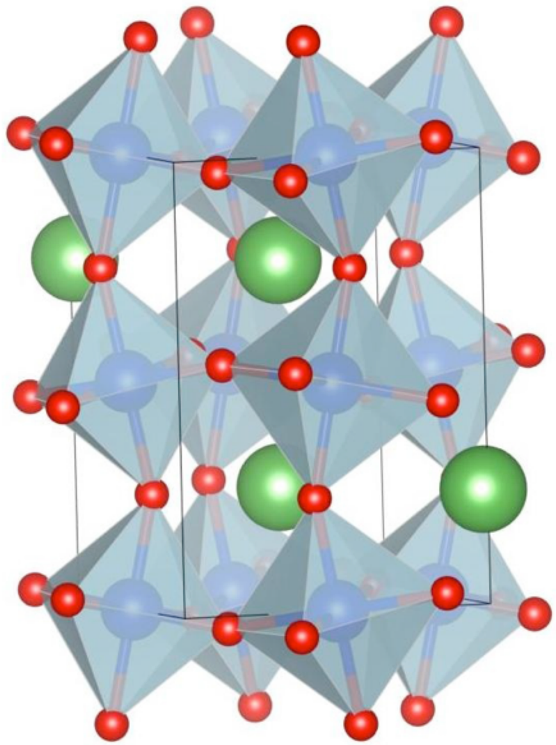
# $GdFeO_3$-rotation

**Cubic perovskite**



**`tilted' structure**



**Typical rotations: 10-20 degrees=>modest change in electronic structure**

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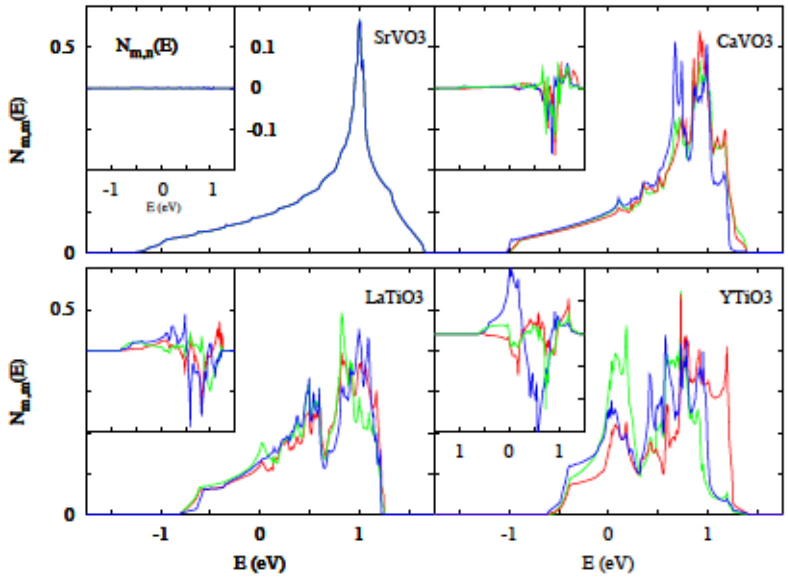
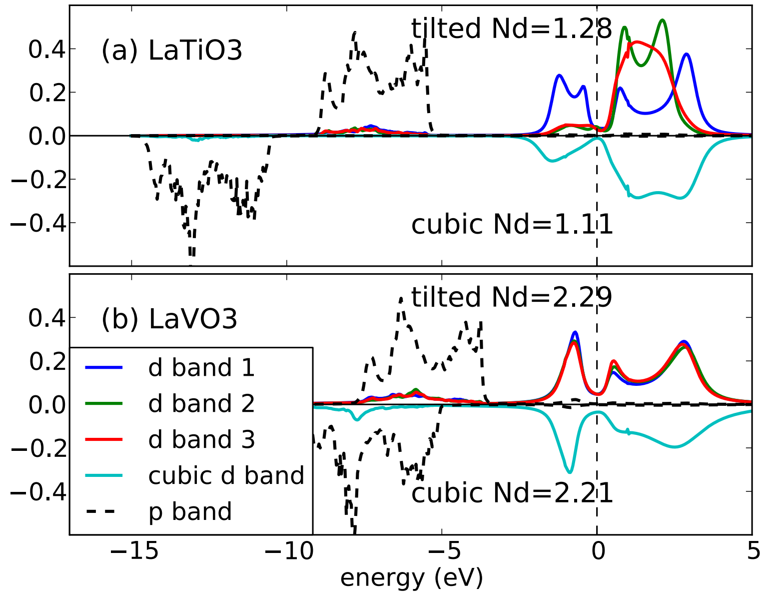
# GdFeO3-rotation necessary for insulating behavior

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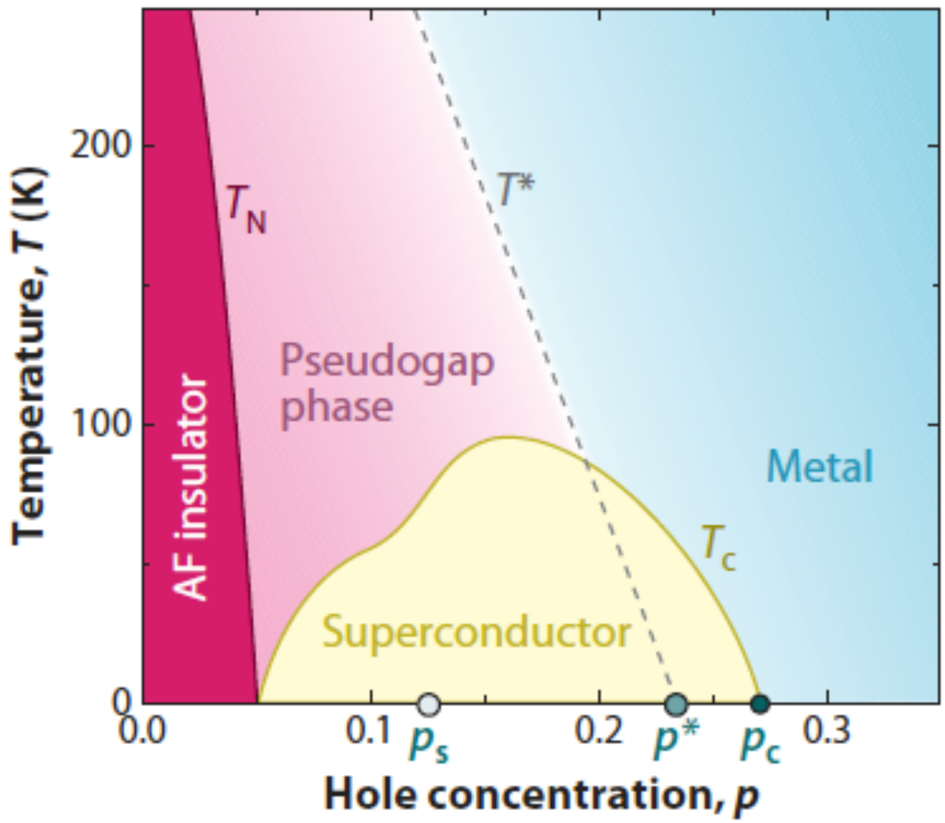
# High-Tc (copper oxide) superconductivity

## Qualitative phase diagram

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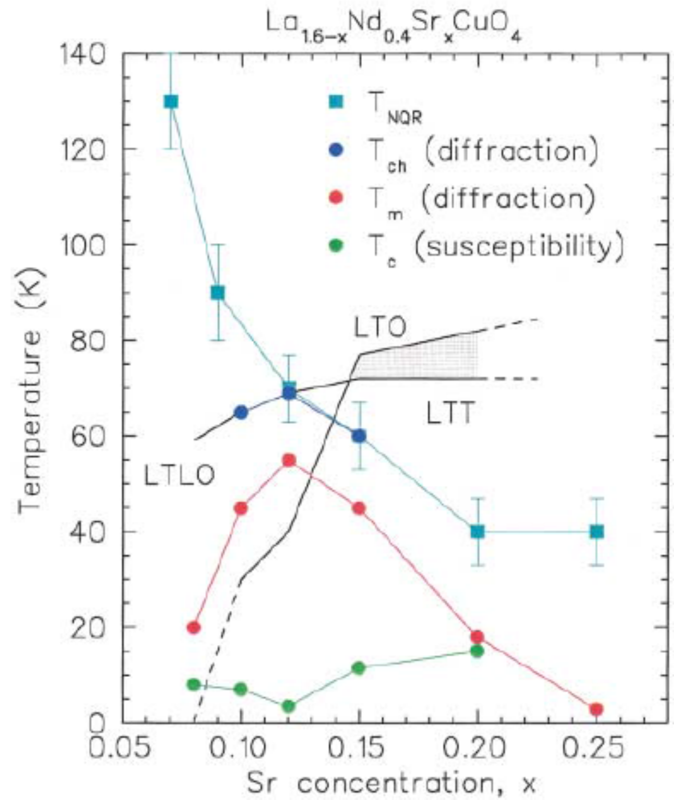
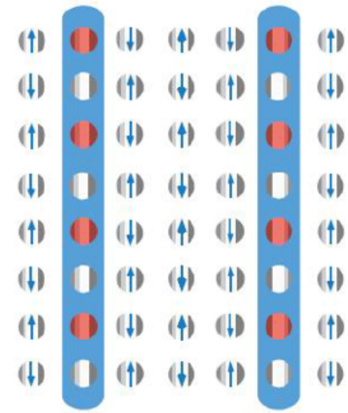
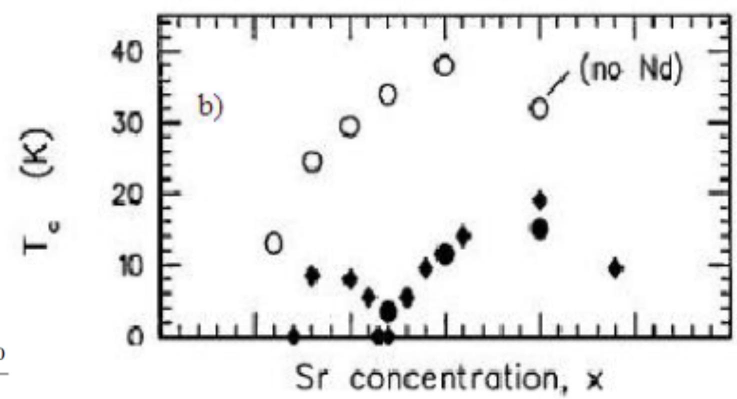
# Stripes vs superconductivity in cuprates






VOLUME 85, NUMBER 8 PHYSICAL REVIEW LETTERS 21 AUGUST 2000

**Local Magnetic Order vs Superconductivity in a Layered Cuprate**

N. Ichikawa,[1,*] S. Uchida,[1] J. M. Tranquada,[2] T. Niemöller,[3] P. M. Gehring,[4] S.-H. Lee,[4,5] and J. R. Schneider[3]

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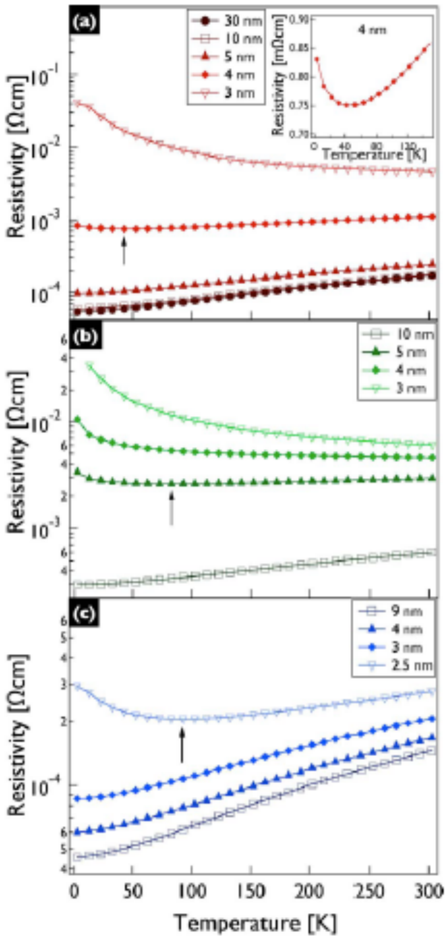
# Strain control of metal-insulator transition in rare earth nickelates




LAO: 1.3% compressive strain
LSAT: 0.8% tensile strain
DSO: 2.5% tensile strain

**Critical thickness for metal-insulator transition depends on magnitude and sign of strain**

Son et al Appl. Phys. Lett. 96 062114 (2010)



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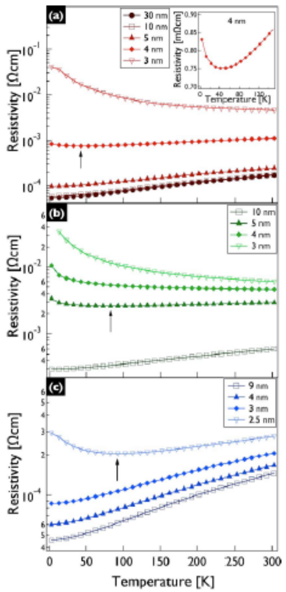
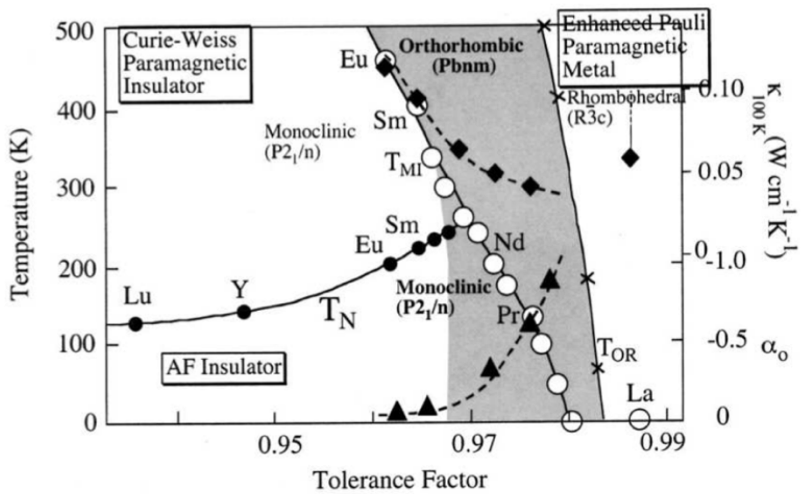
# modest changes drive phase transitions

## =>very large NONLINEAR response



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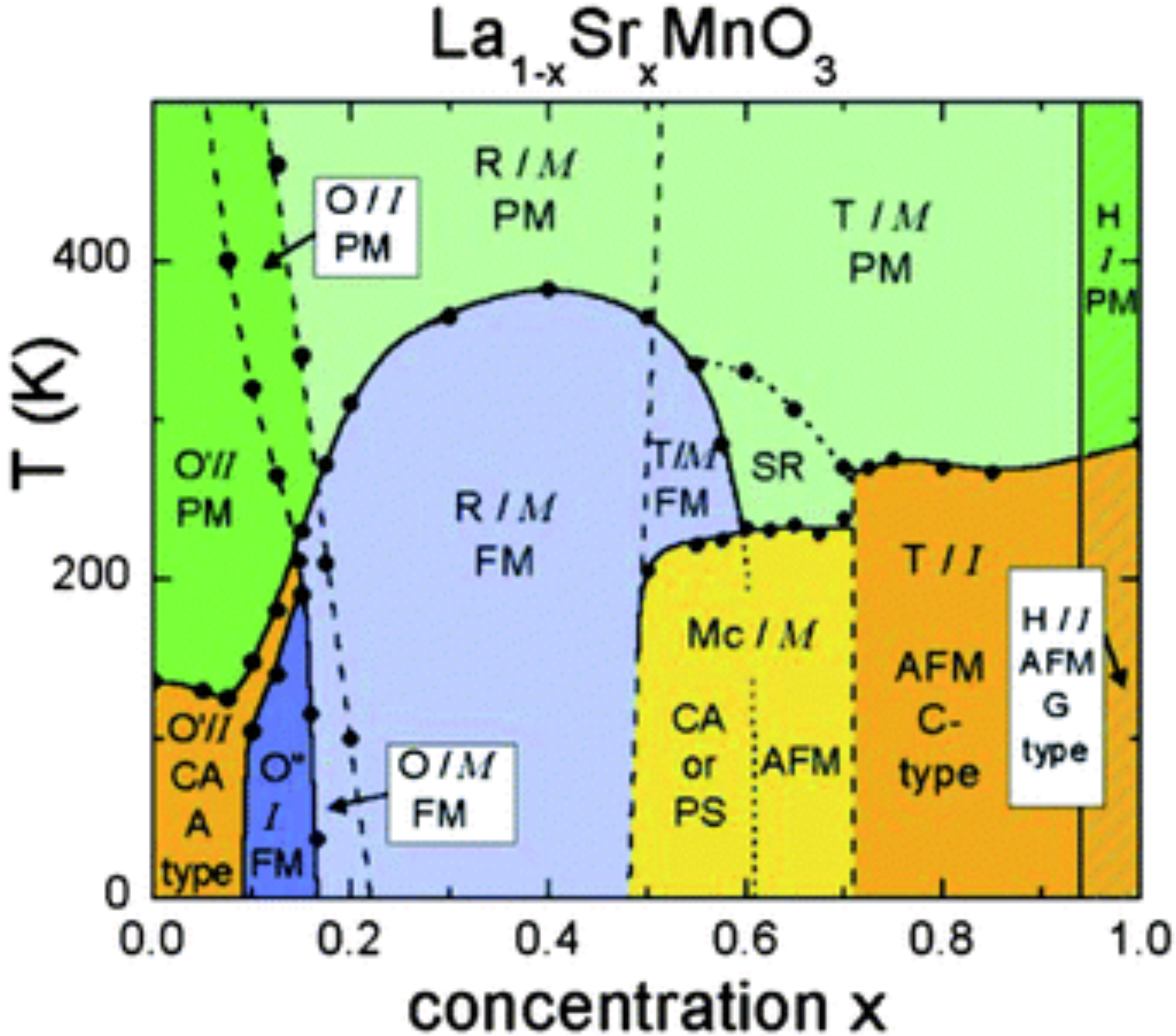


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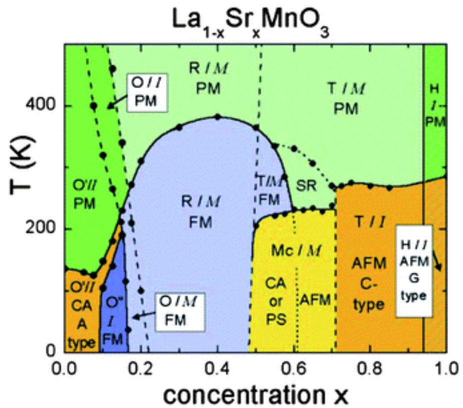
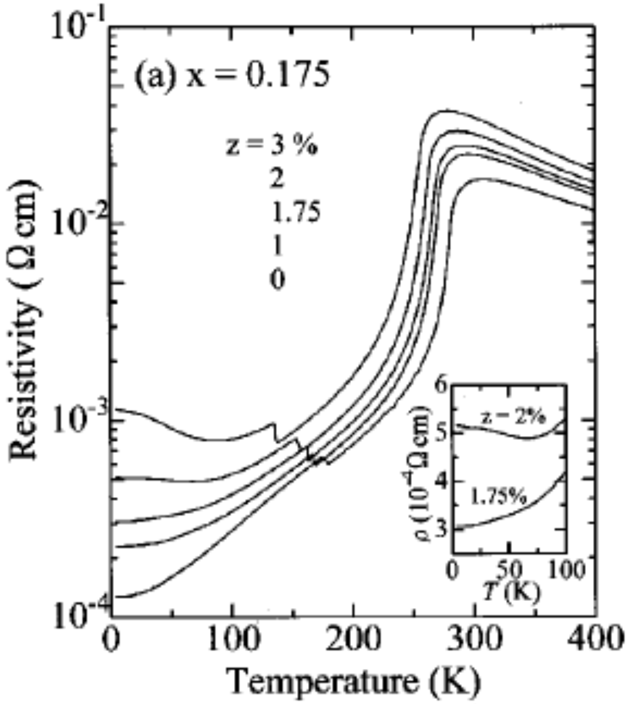
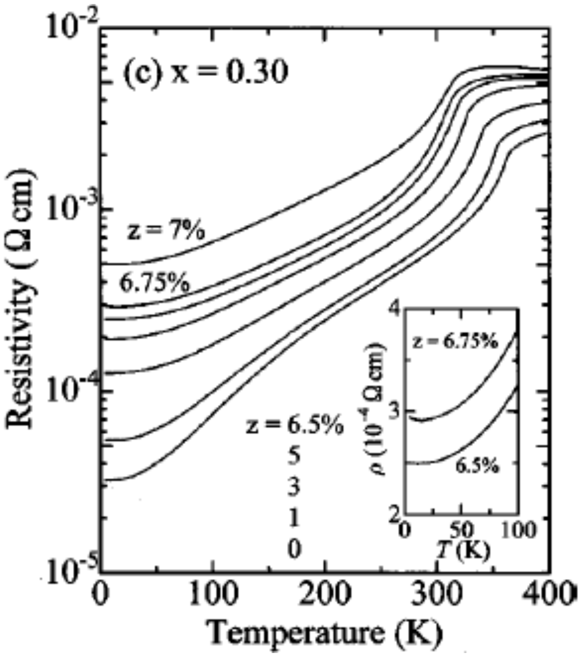
# $La_{1-x}Sr_xMnO_3$: Reponse to impurities




PRB 61 11 588

# $La_{1-x}Sr_x(Mn_{1-z}Al_z)O_3$

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# Residual resistivity vs Al

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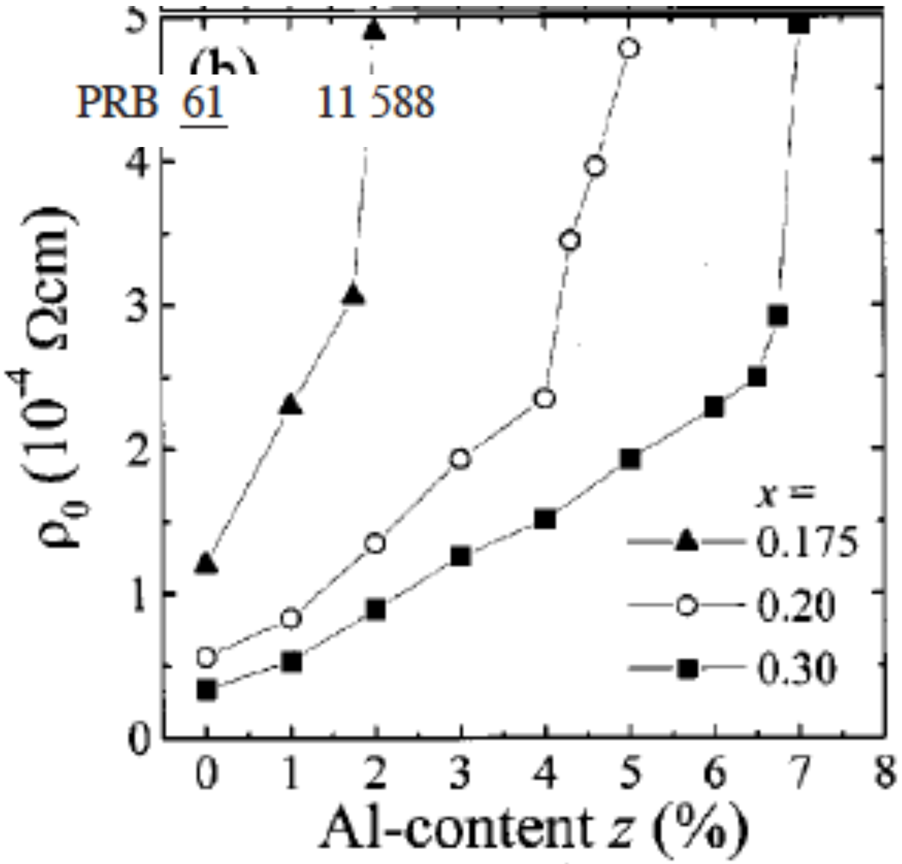


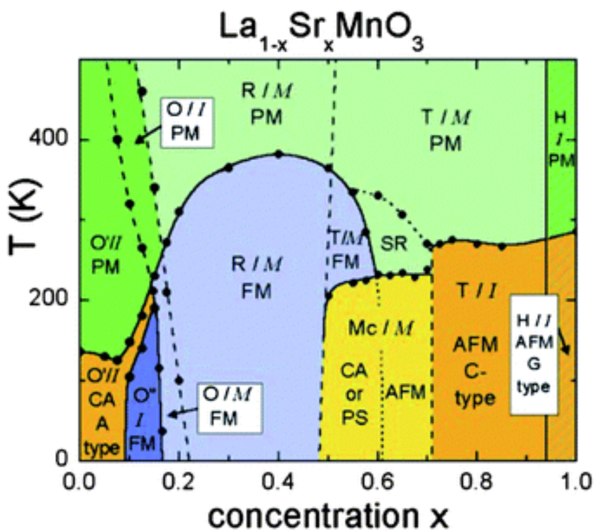


# Small amount of Al: new phase

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# Transition Metal Oxides: Summary

1. **Interesting electronic behavior**

2. **Tightly coupled to lattice**

3. **Many `control knobs'**

4. **Modest perturbations lead to change of electronic phase**

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# Presumably related issue: large nonlinear fluctuations

## SDW order and fluctuations in Iron Arsenide Superconductors.

**With Abhay Pasupathy & Rafael Fernandes**



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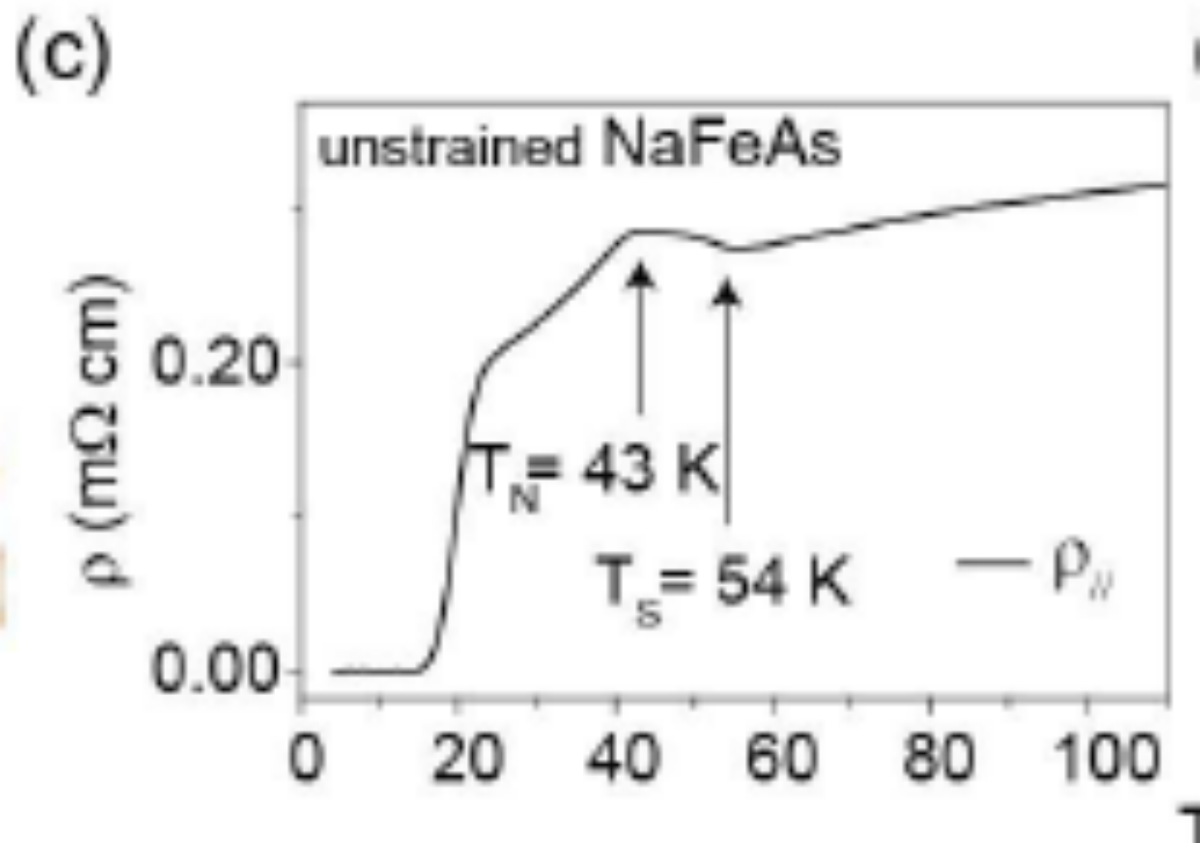
# NaFeAs: `stripe' (0,Pi) order below 43K `nematic' order below 54K

Y. Zhang,[1] C. He,[1] Z. R. Ye,[1] J. Jiang,[1] F. Chen,[1] M. Xu,[1] Q. Q. Ge,[1] B. P. Xie,[1] J. Wei,[2] M. Aeschlimann,[2] X. Y. Cui,[3] M. Shi,[3] J. P. Hu,[4] and D. L. Feng[1,*]



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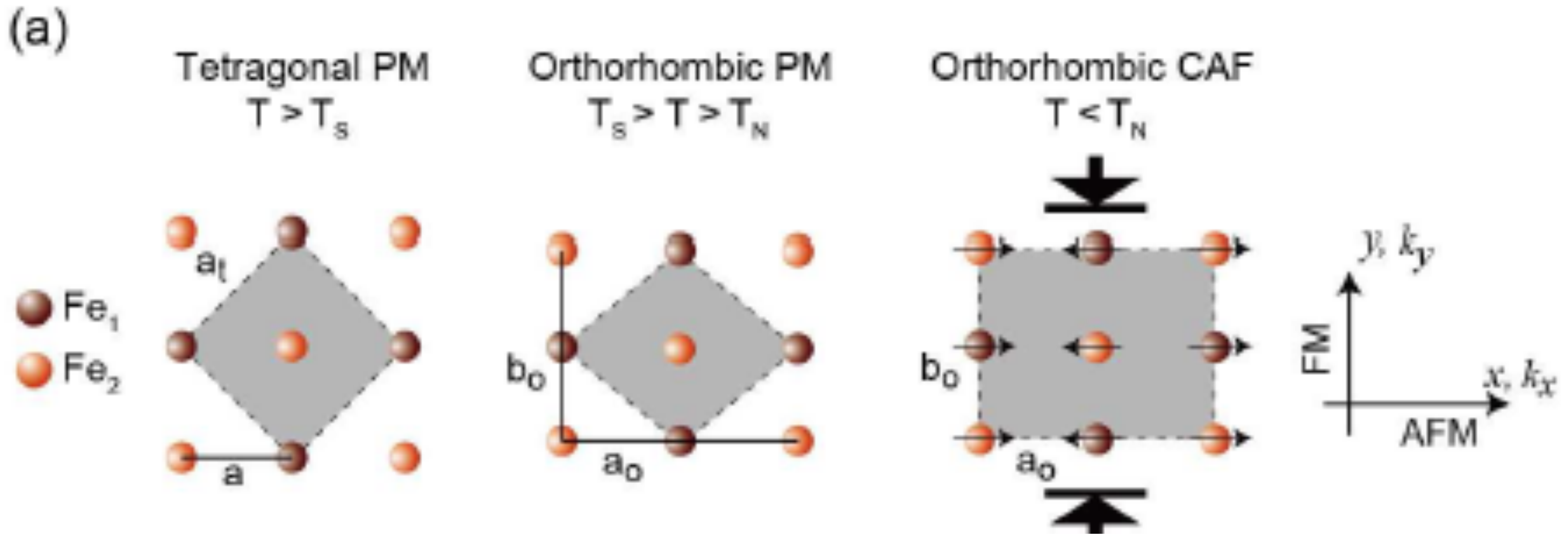
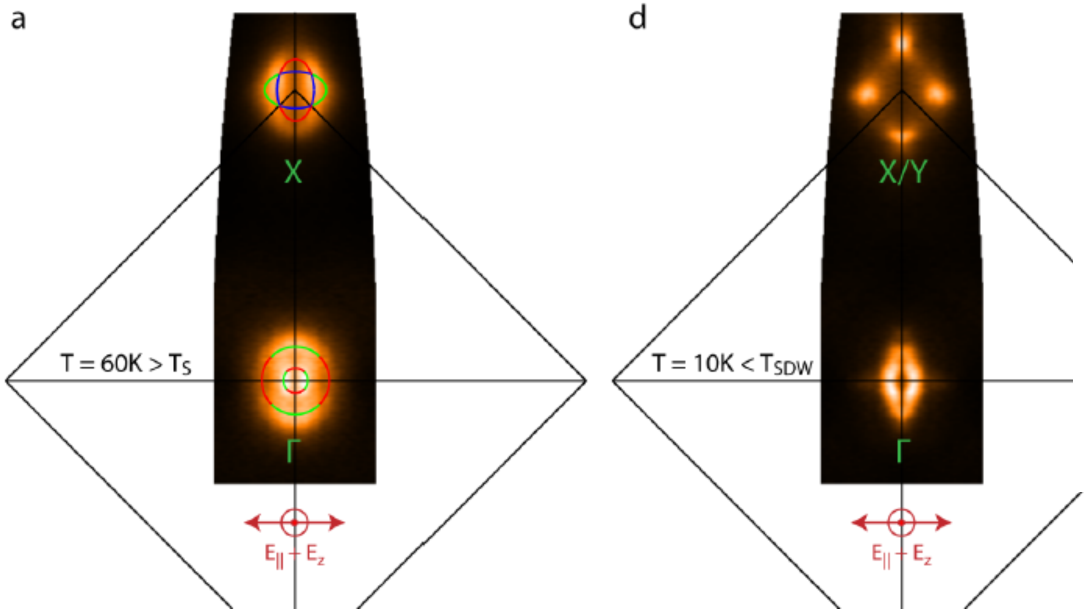
# NaFeAs: `stripe' (0,Pi) order below 43K `nematic' order below 54K



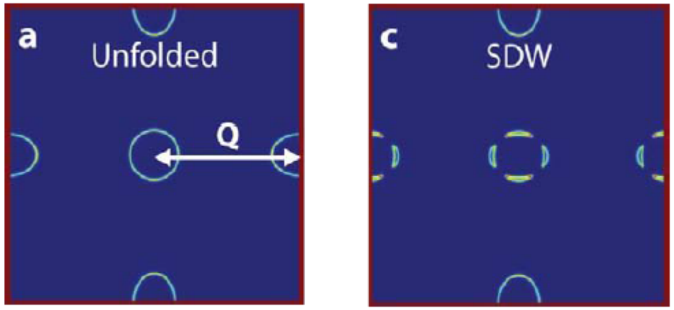
# SDW rearranges the Fermi surface

## Data:

M Yi[1,2], D H Lu[3], R G Moore[1], K Kihou[4,5], C-H Lee[4,5], A Iyo[4,5], H Eisaki[4,5], T Yoshida[5,6], A Fujimori[5,6], Z-X Shen[1,2]*

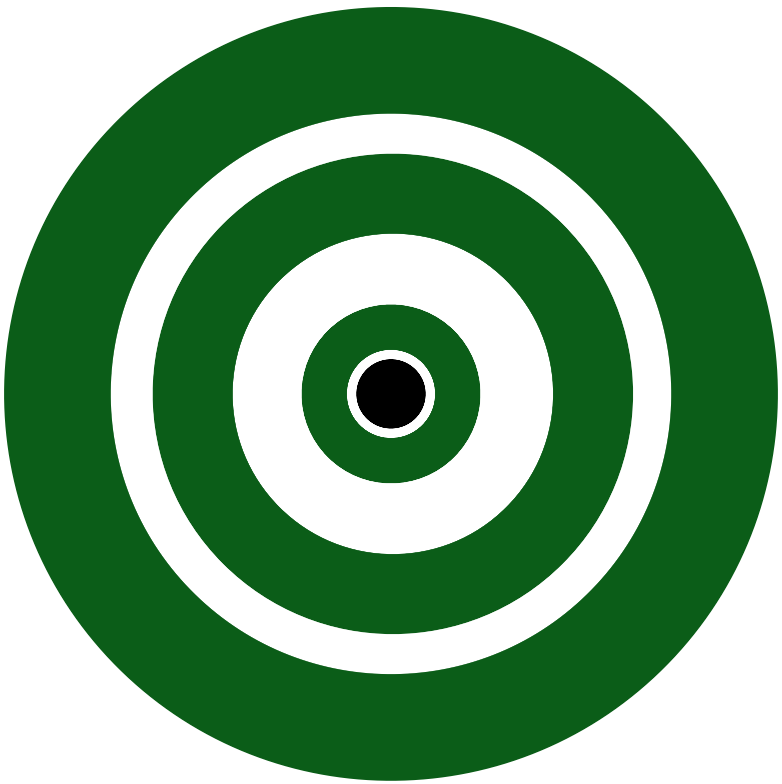



## Calculations



# Quasiparticle Interference

**Impurity=>standing wave of electron density**



**Standing wave period and spatial structure related to fermi surface**

**Many impurities: more complicated analysis but same conclusion**



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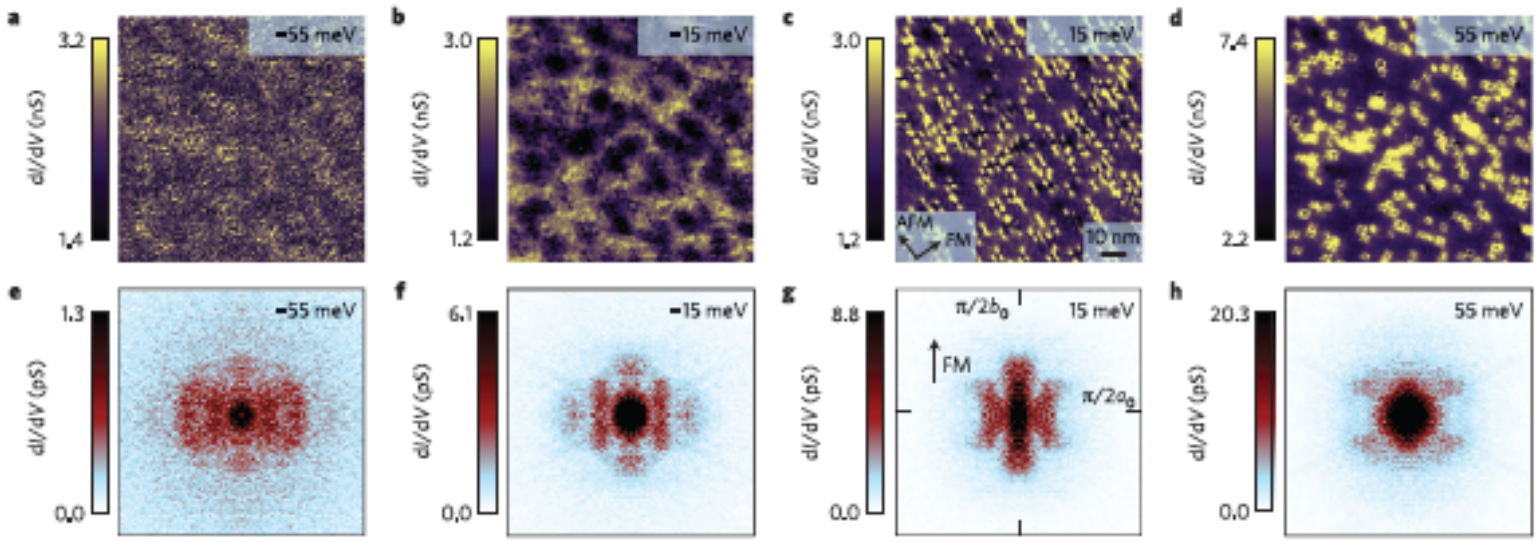
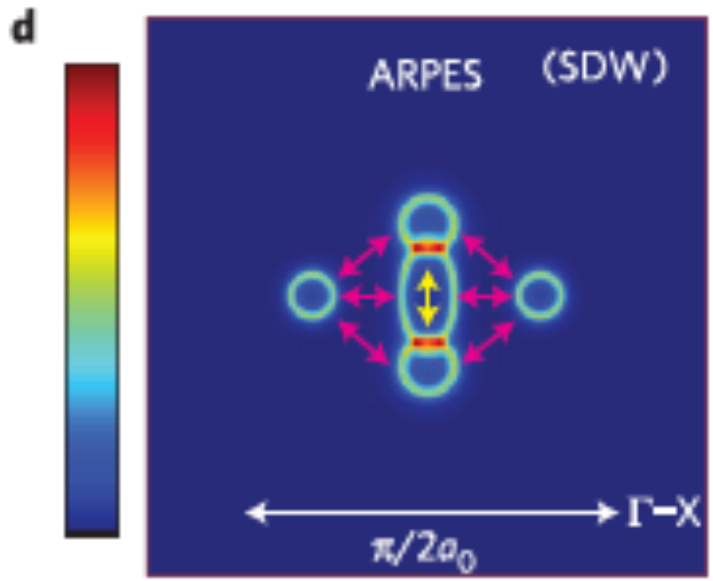
# Quasiparticle Interference Reveals the Reconstruction

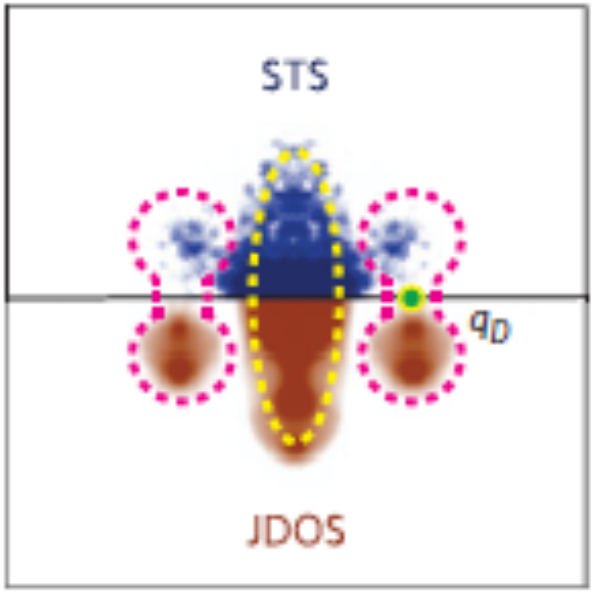
E. P. Rosenthal[1], E. F. Andrade[1], C. J. Arguello[1], R. M. Fernandes[2], L. Y. Xing[3], X. C. Wang[3], C. Q. Jin[3], A. J. Millis[1] and A. N. Pasupathy[1*]

N. Phys. 10 225 (2014)



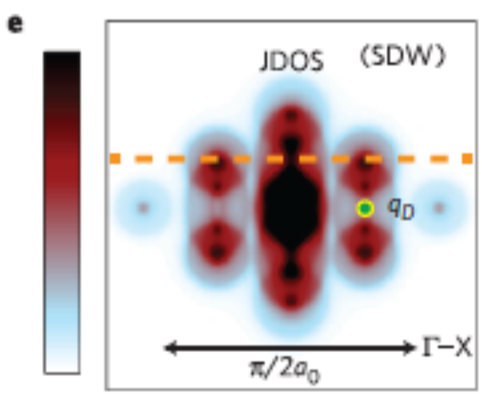
# Structure in QPI reveals fermi surface



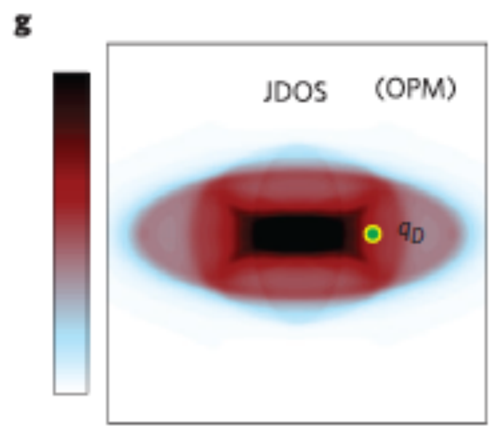



# As you raise the temperature, expect the SDW-derived features to go away

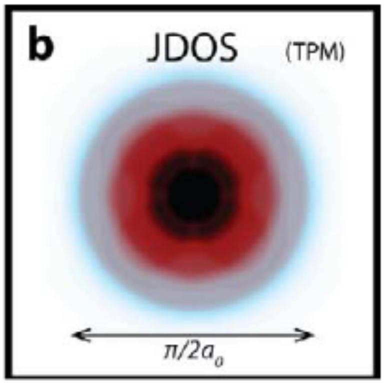
$T<T_N\sim 43K$




$T_N<T<T_{nematic}$




$T_{nematic}<T$




**Here I show you joint DOS for simplicity.**
**Full QPI calculations give the same physics**



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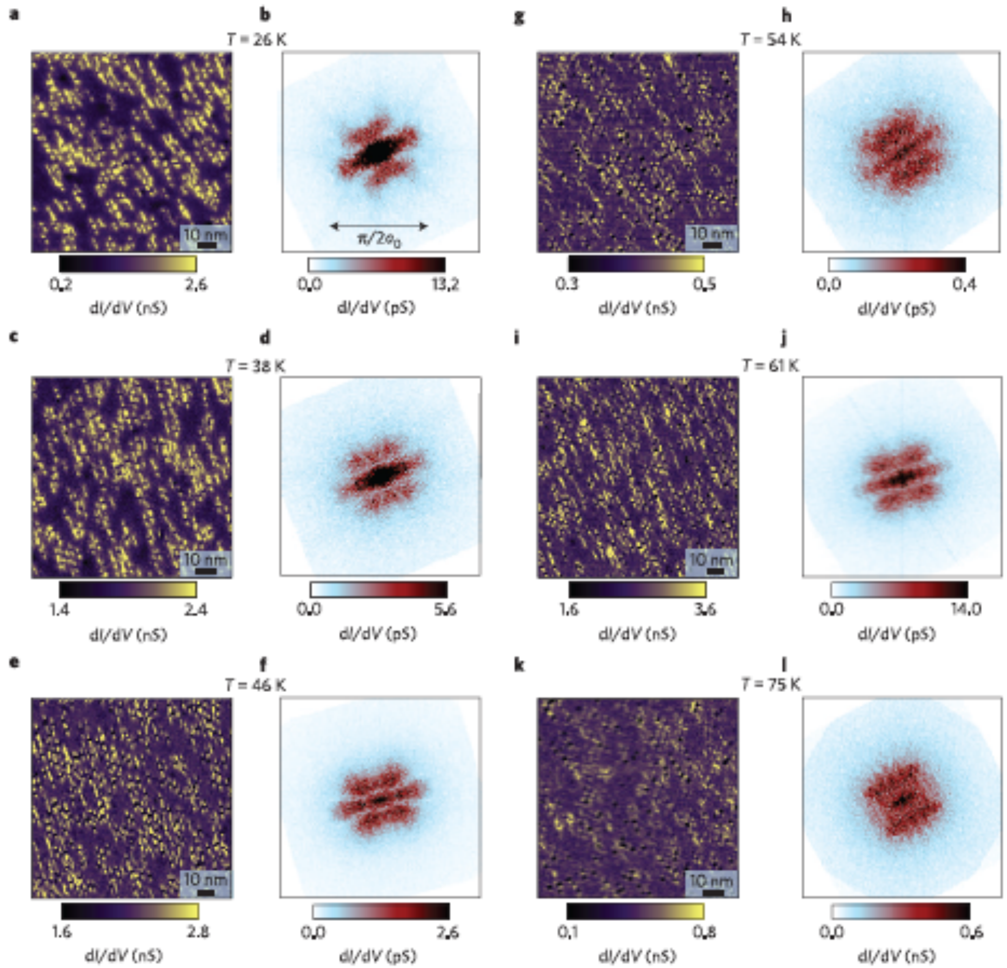
# This is not what happens



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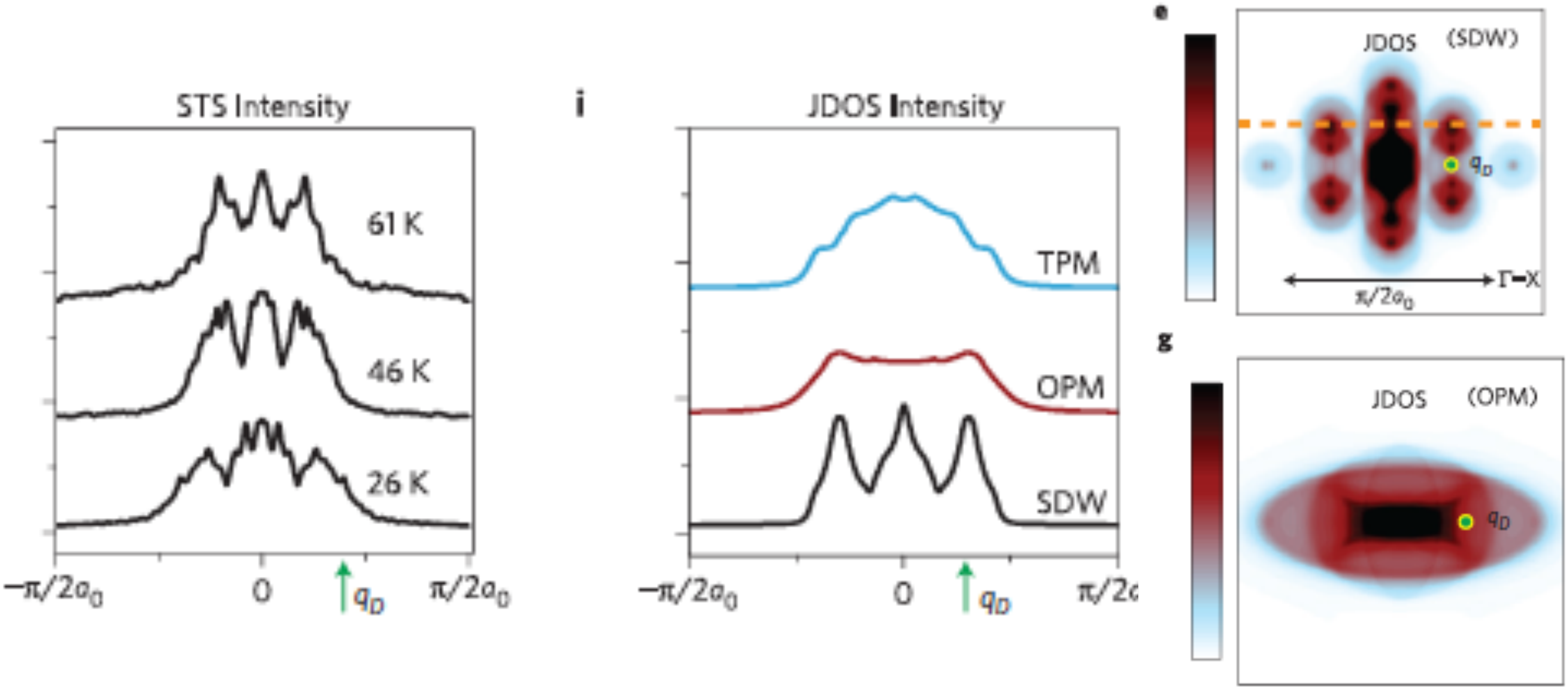
# More easily visualized as a line cut




# Key Result: SDW-like features persist to high T, in fact up to $T=2T_N$



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# Physics idea: finite size regions of SDW order, which last for finite time

Region of size $\xi$

Local SDW amplitude => gap $\Delta$

Requirement: $\frac{v_F}{\xi} > \Delta$

Then electrons 'feel' gap before exiting region.



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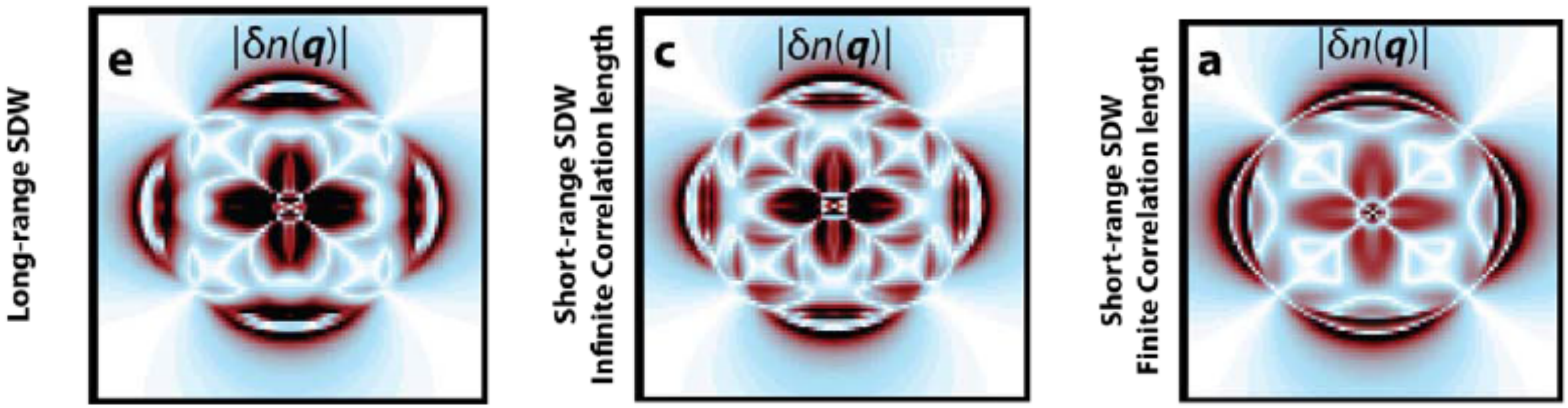
# Model
# ‘Lee-Rice-Anderson’ ansatz

$$\mathbf{\Sigma}(\omega, \mathbf{k}) = \frac{\mathbf{\Delta^2}}{\omega - \varepsilon_{\mathbf{k+Q}} - \frac{\mathbf{i}}{\xi}}$$

**This is broadened backscattering (no `coherence factors in normal state)**

# Model QPI Calculations




**The short ranged SDW calculations use the standard QPI formula but with the Lee-Rice-Anderson G in a simplified 3-band approximation to the pnictide bands**



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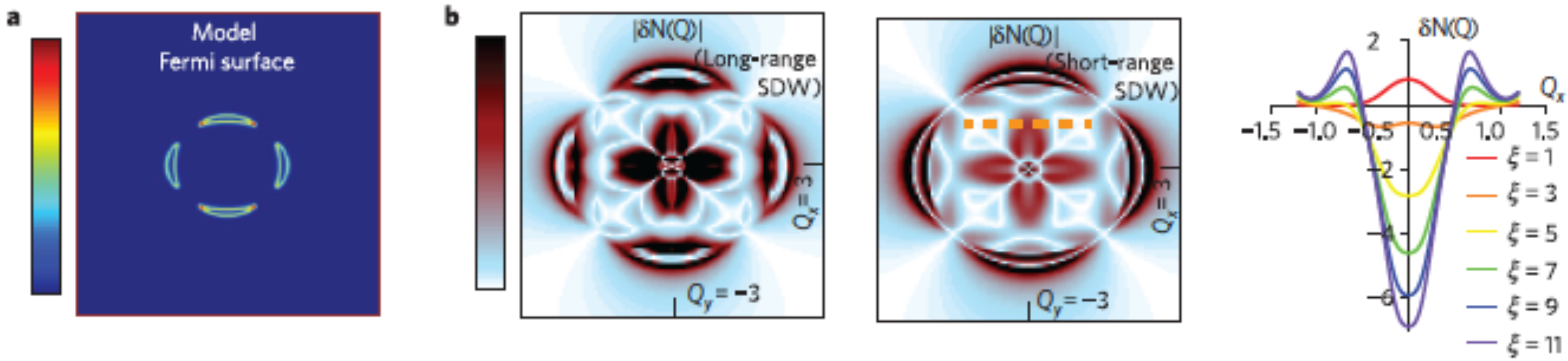
# Vary correlation length

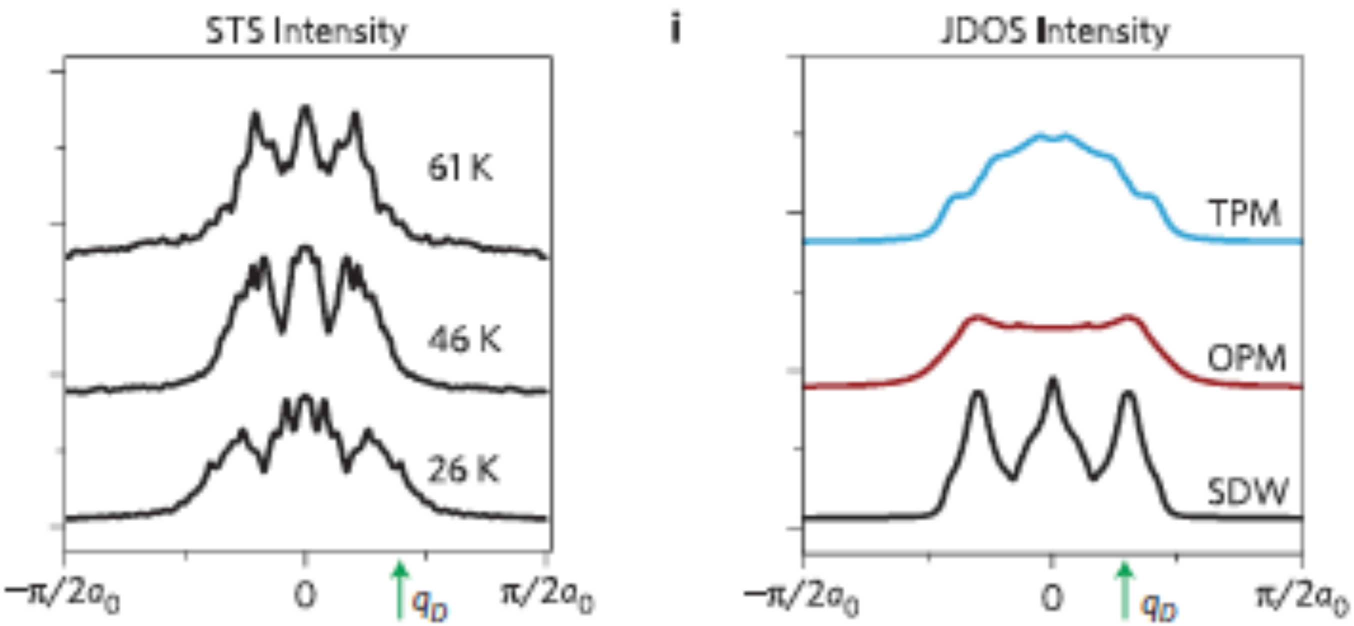



$$\mathbf{\Sigma}(\omega, \mathbf{k}) = \frac{\mathbf{\Delta}^2}{\omega - \varepsilon_{\mathbf{k}+\mathbf{Q}} - \frac{\mathbf{i}}{\xi}}$$

**To get peaks in line cuts need to keep Delta at approximately the T=0 value., have correlation length not too short.**

# In other words

**Data imply large amplitude, slow fluctuations of density wave order, persisting up to ~2x observed transition temperature**




**Paramagnetic phase has hidden structure**



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# Poetically

# Poetically

**We used to think of the fermi sea as a (relatively) placid lake with modest ripples (RPA fluctuations).**

# Poeticallly

**We used to think of the fermi sea as a (relatively) placid lake with modest ripples (RPA fluctuations).**

# Poetically

**Pasupathy's results suggest an alternative picture**

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**Pasupathy's results suggest an alternative picture**

# Poetically

**Pasupathy's results suggest an alternative picture: A stormy sea with giant amplitude, slowly moving waves.**

# Poetically

**Pasupathy's results suggest an alternative picture: A stormy sea with giant amplitude, slowly moving waves.**

**(a) Pasupathy experiments indicate large amplitude response to impurities (local reconstruction of FS)**

**(b) This situation implies large nonlinear response**

# Context for nonequilibrium correlated electron physics

- **Multiplicity of electronic phases**
- **Important coupling to lattice**
- **Large nonlinear response**
- **associated with change of electronic phase**



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