Orbital domain dynamics in magnetite

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NSLS II & CFN Users' Meeting 2018

5/29/2018

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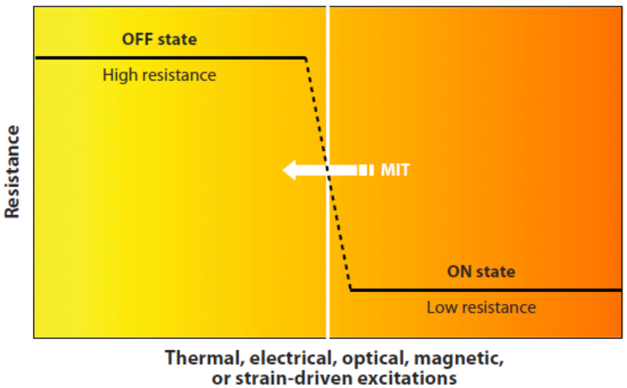
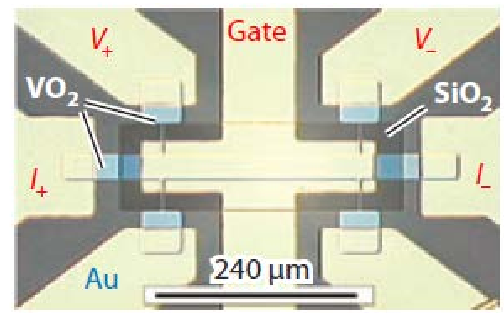
Moving beyond semiconductors

Metal-Insulator transition

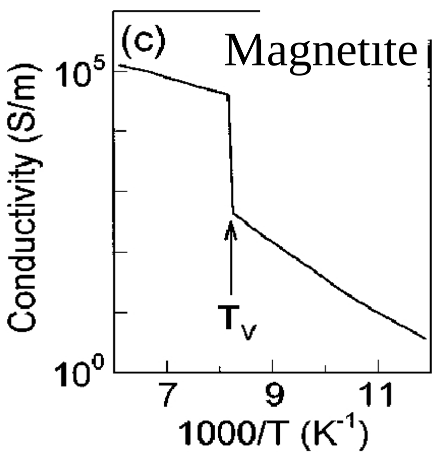
- Thermal
- Electrical
- Magnetic
- Optical
- Strain

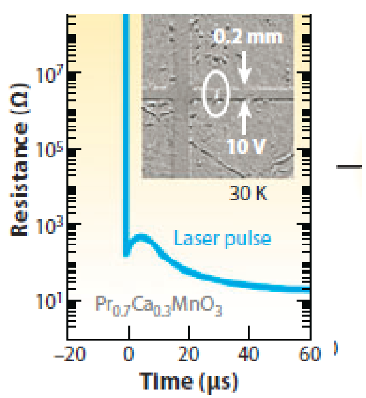


Gated electronic switch (Mott FET)

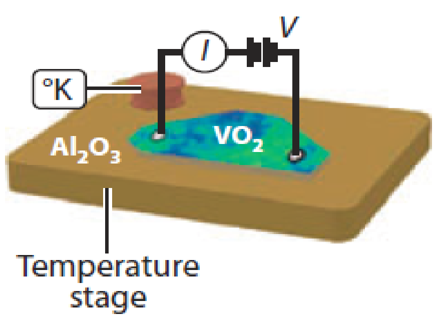


Z. Yang, et al. Annu. Rev. Mater. Res. '11





Memristive device



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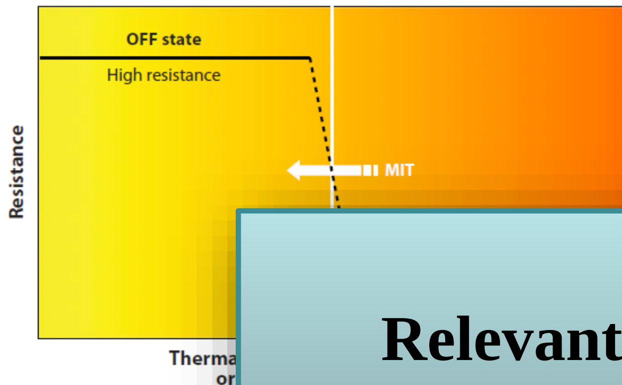
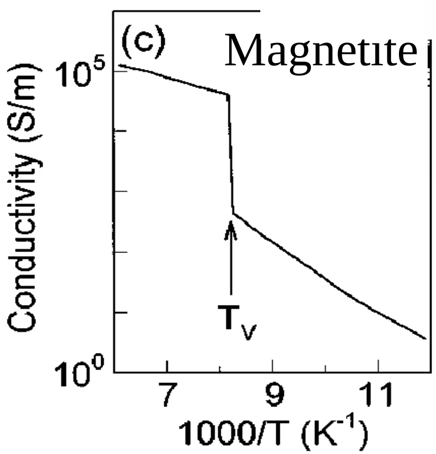


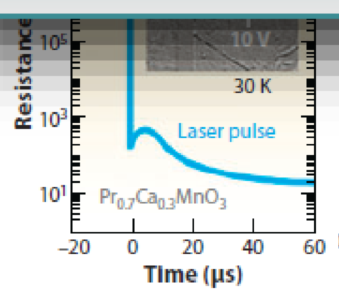
Gated electronic switch (Mott FET)

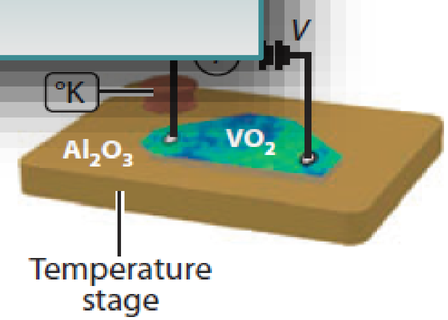
Relevant timescales and lengthscales of switching?

v. Mater. Res. '11

device



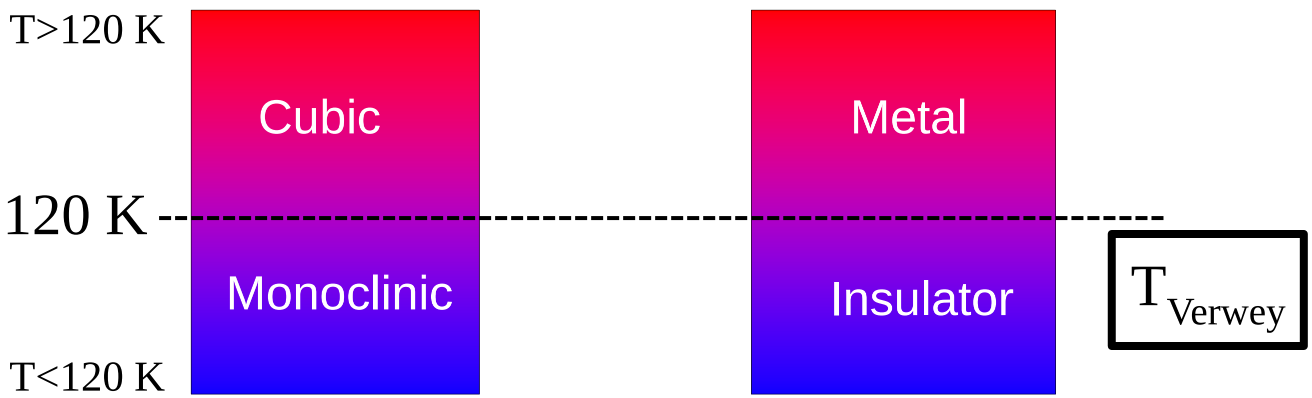




Verwey transition in Magnetite (Fe_3O_4)

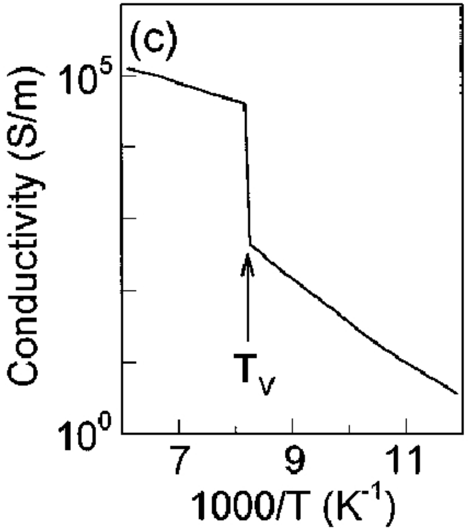
- One of the first known correlated electron systems (1939)
- Shows 100 fold decrease in conductivity below T = 120 K
- Ferrimagnet with full spin polarization – no change in spin alignment during the transition

Lattice **Electronic Order**



Verwey, Nature, '39

Park et. al, PRB 55, 1997



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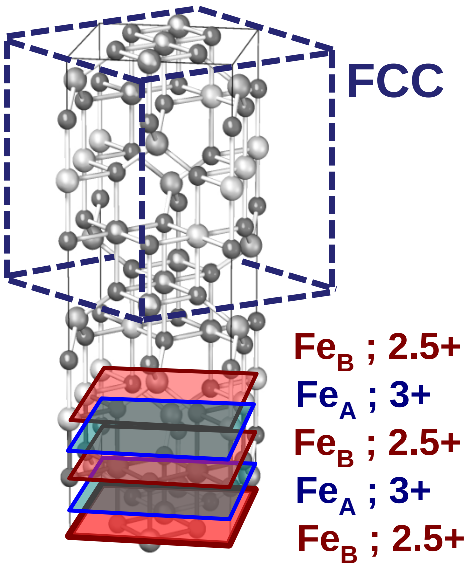
Low vs high temperature structure

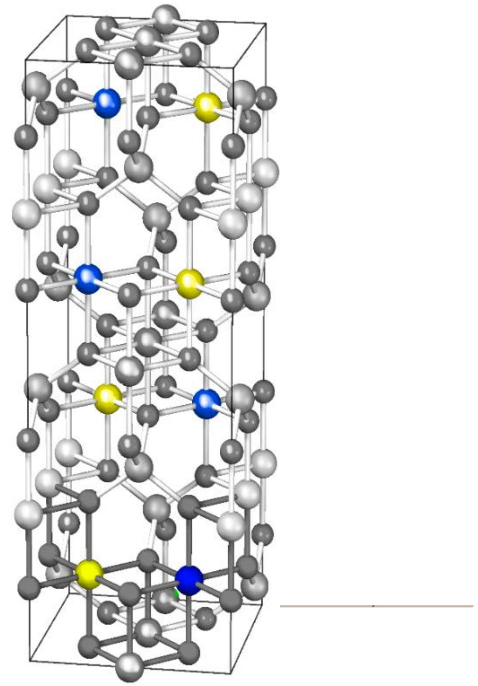
After Wright, Phys. Rev. B '92

High T: FCC cubic

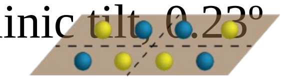


Inverse Spinel
Fe^{+2} and Fe^{+3} in octahedral sites

Low T: Monoclinic



c = 2a, Monoclinic tilt, 0.23°
(001/2), (001)



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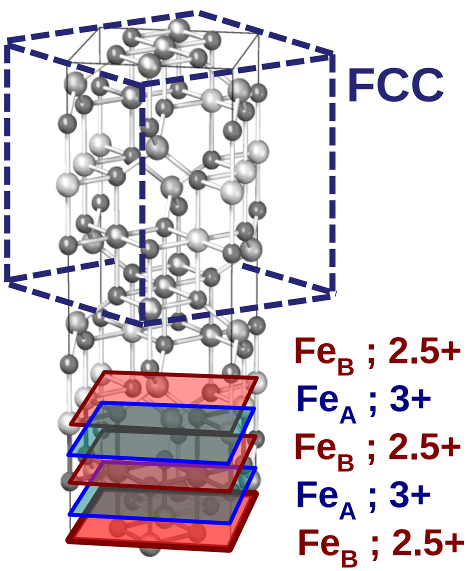

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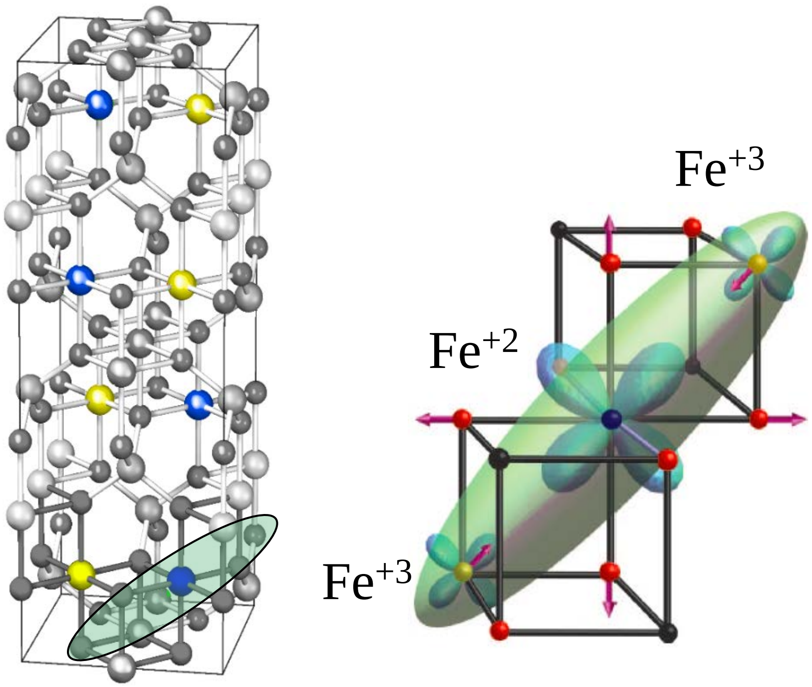

'Trimerons'
Three site Fe^{+3}-Fe^{+2}-Fe^{+3} distortions

After Senn et al. Nature '12

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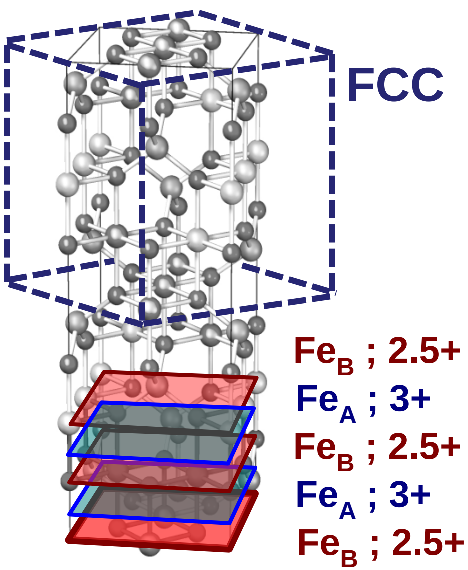
Low vs high temperature structure

After Wright, Phys. Rev. B '92

High T: FCC cubic



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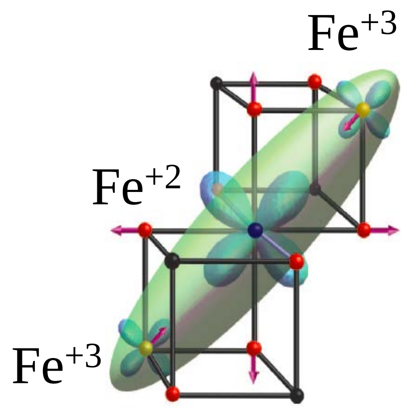

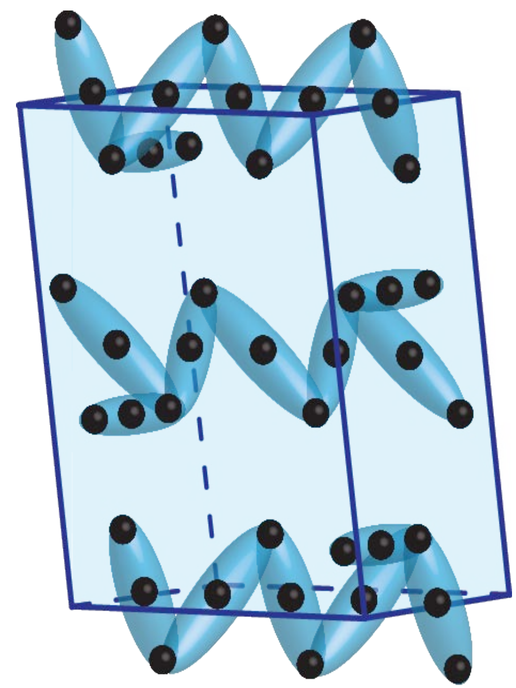

'Trimerons'
Three site Fe^{+3}-Fe^{+2}-Fe^{+3} distortions

After Senn et al. Nature '12

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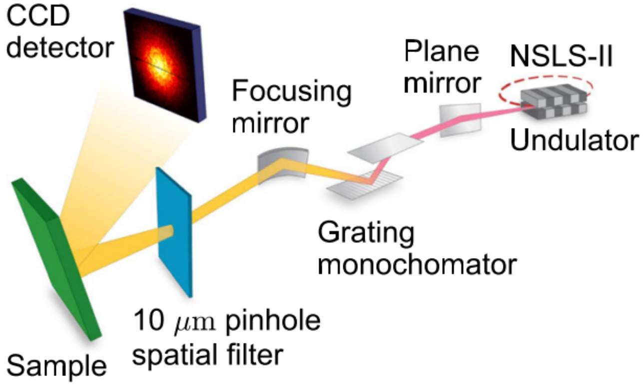
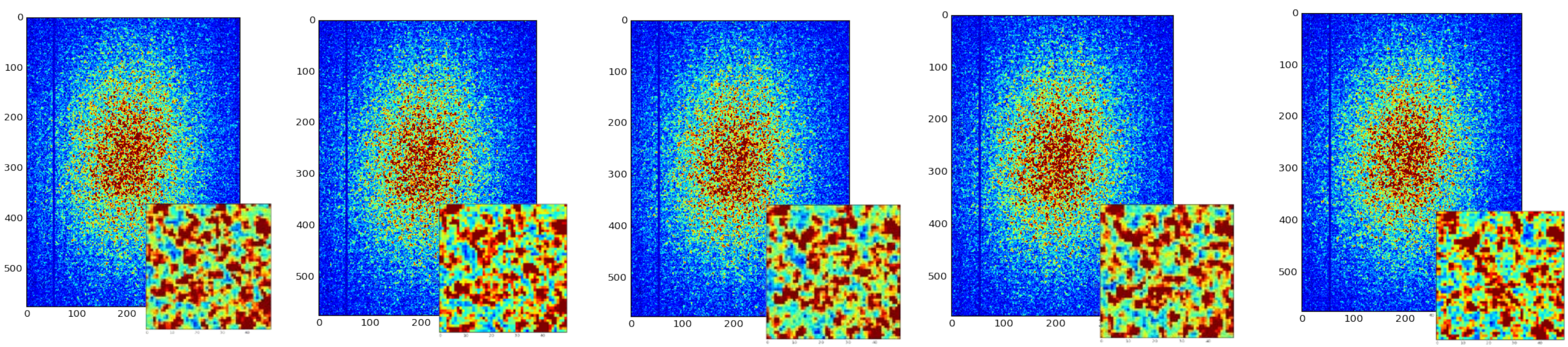
Orbital dynamics using coherent x-rays



Chen *et al*. PRL **117**, 167001 (2016)

- X-ray Photon Correlation Spectroscopy measurements at the CSX-1 beamline (NSLS-II)
- *Speckle pattern* at Fe L-edge to access fluctuations in orbital ordering

T = 90 K



t = 0 t = 10 s t = 50 s t = 100 s t = 300 s

(001/2) at Fe L-edge

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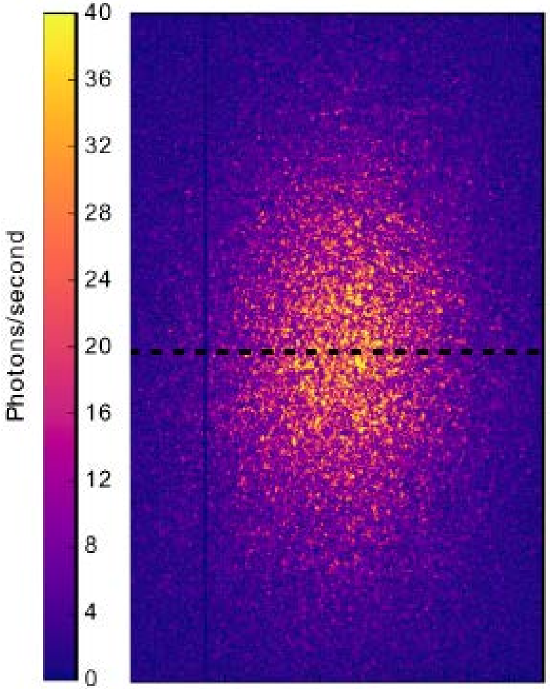
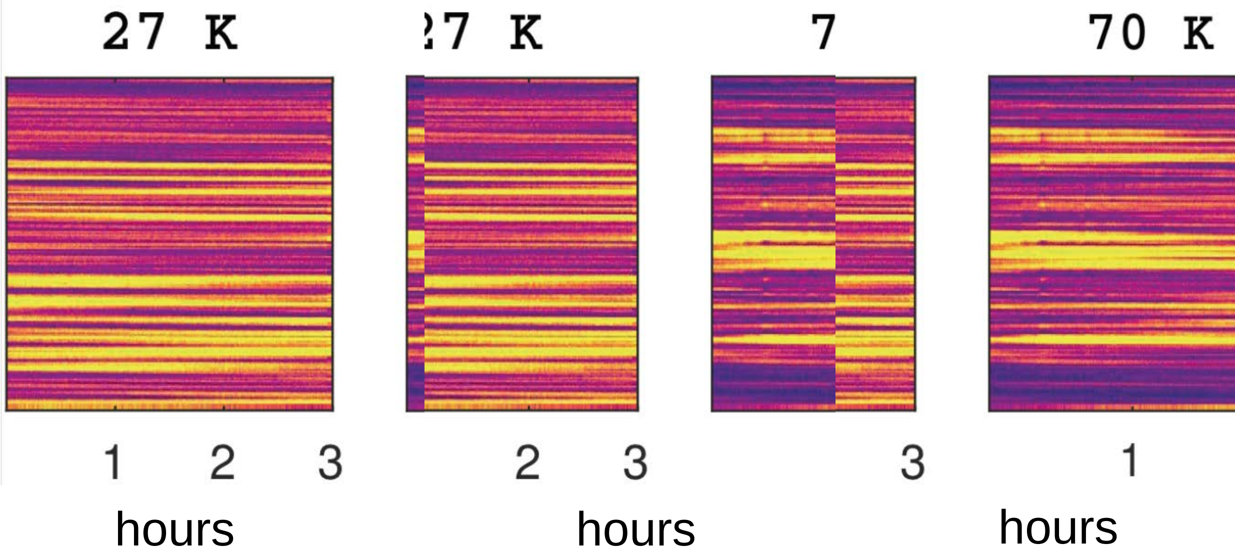
Domain dynamics near transition





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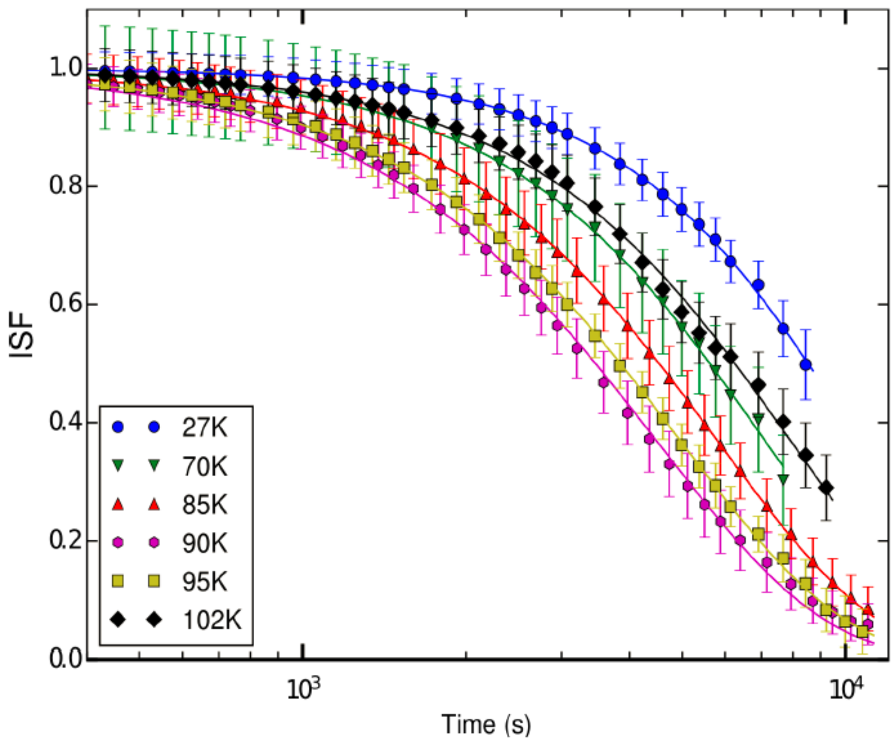
Orbital dynamics near Verwey transition



Autocorrelation function

$$g_2(t) = \frac{\langle I(\tau) I(\tau + t) \rangle_\tau}{\langle I(\tau) \rangle_\tau^2}$$

Intermediate Scattering Function

ISF = g_2 - 1

$g_2(t) = 1 + A \exp[(-t/\tau)^\beta]$

β* - *stretching exponent, compressed shape

τ* - *relaxation time scales vs temperature

A* - *speckle contrast

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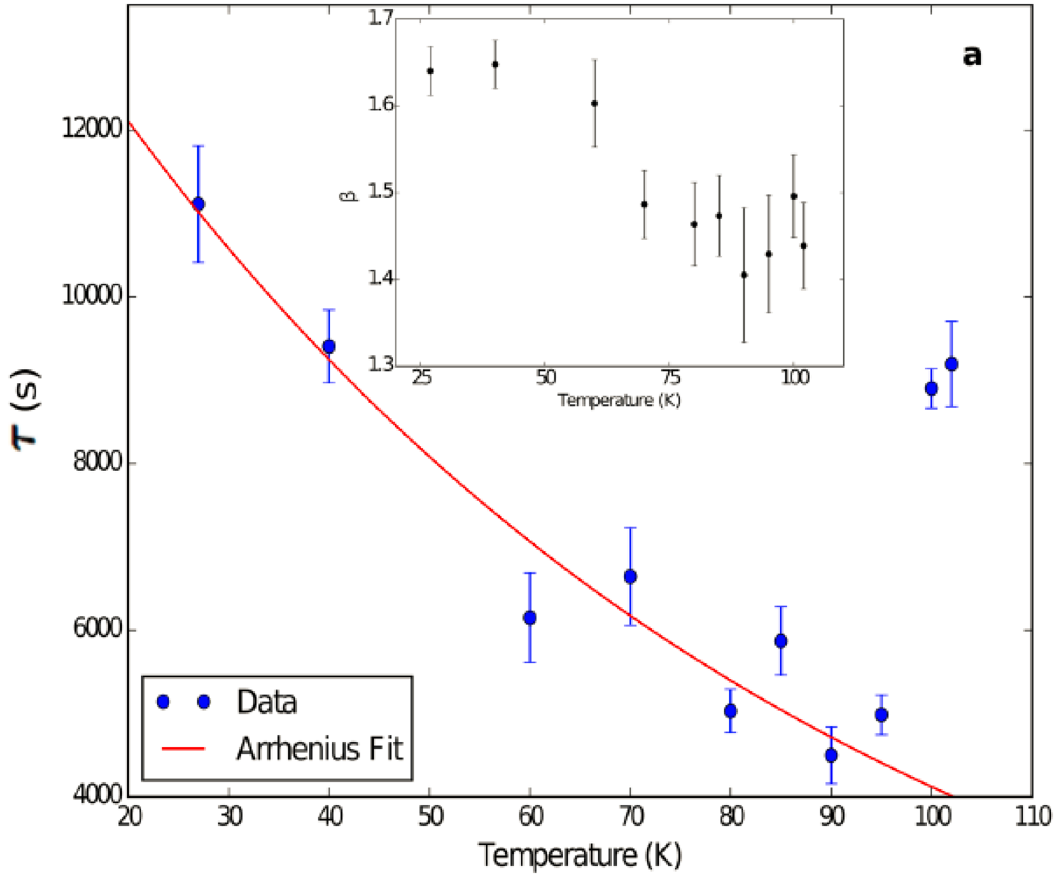
Orbital dynamics near Verwey transition



$g_2(t) = 1 + A \exp[(-t/\tau)^{\beta}]$

- β ~1.5, *stretching exponent, compressed shape,*
- $\boldsymbol{\tau}$ *- relaxation time scales vs temperature* $1/\tau = f \exp(-\Delta E/k_B T)$

First regime shows thermally activated Arrhenius behavior with an activation energy of be **$\Delta E/k_B$ = 32 ± 5 K**

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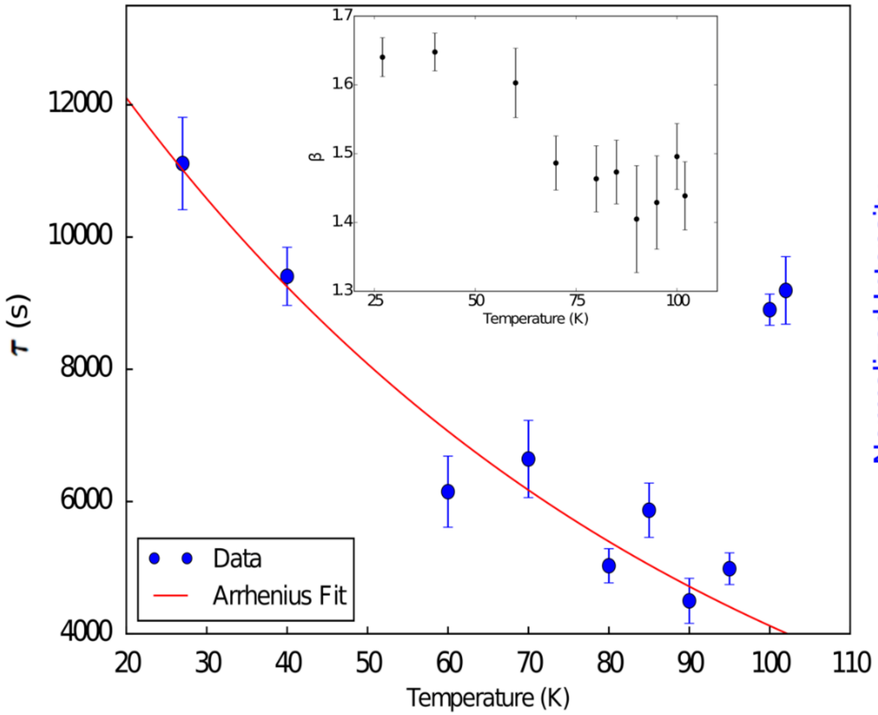
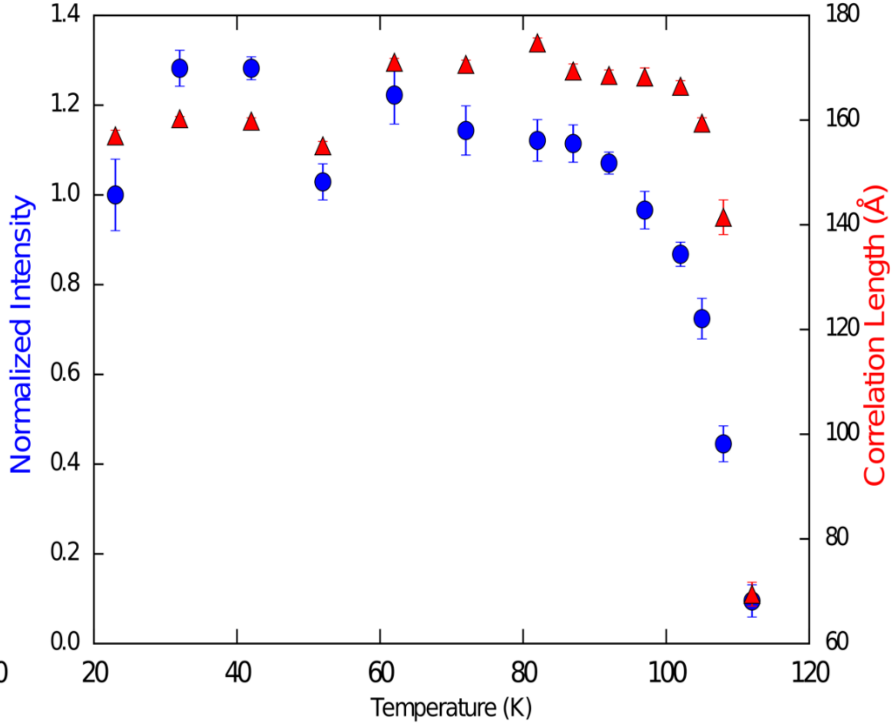
Orbital dynamics near Verwey transition





R. Kukreja, N. Hua et al., in review PRL (2018)

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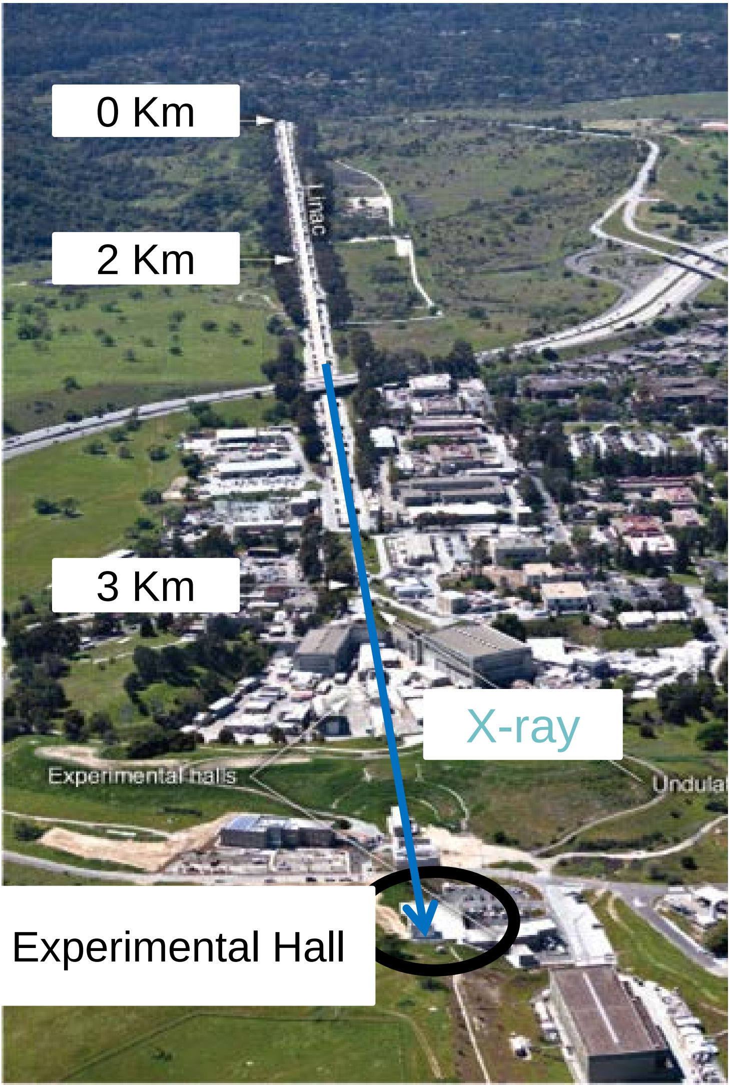
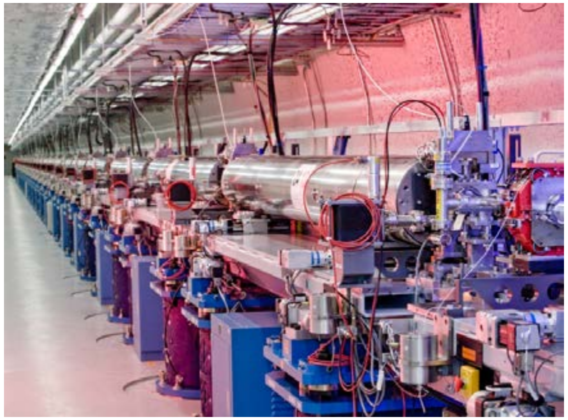
Time Resolved Experiment at LCLS

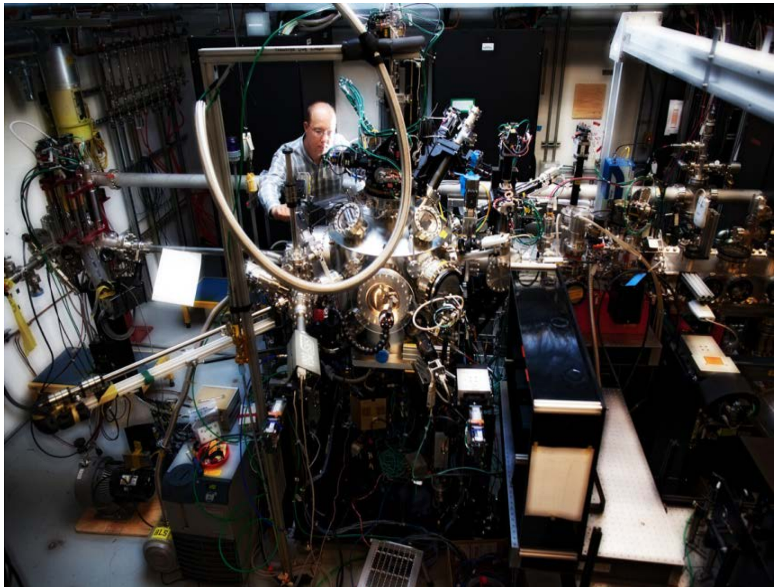




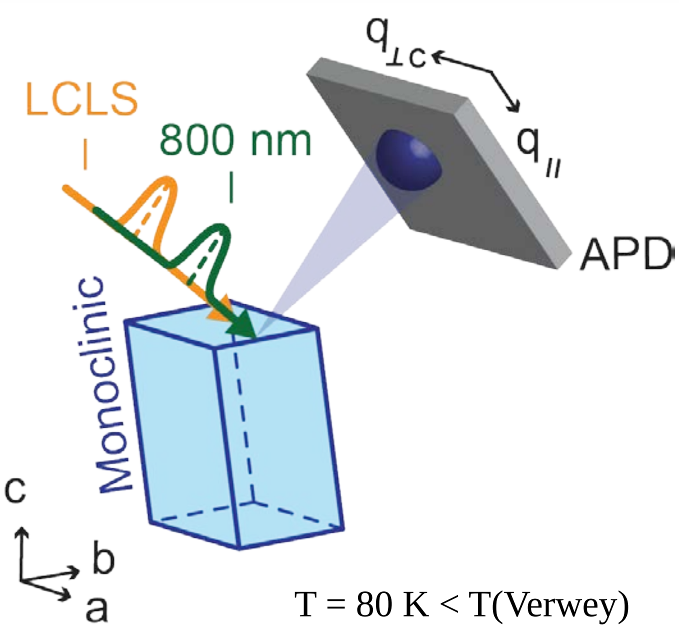
Undulator Hall

Peak brightness increase ~10^{12}
fs pulse length ~10'fs

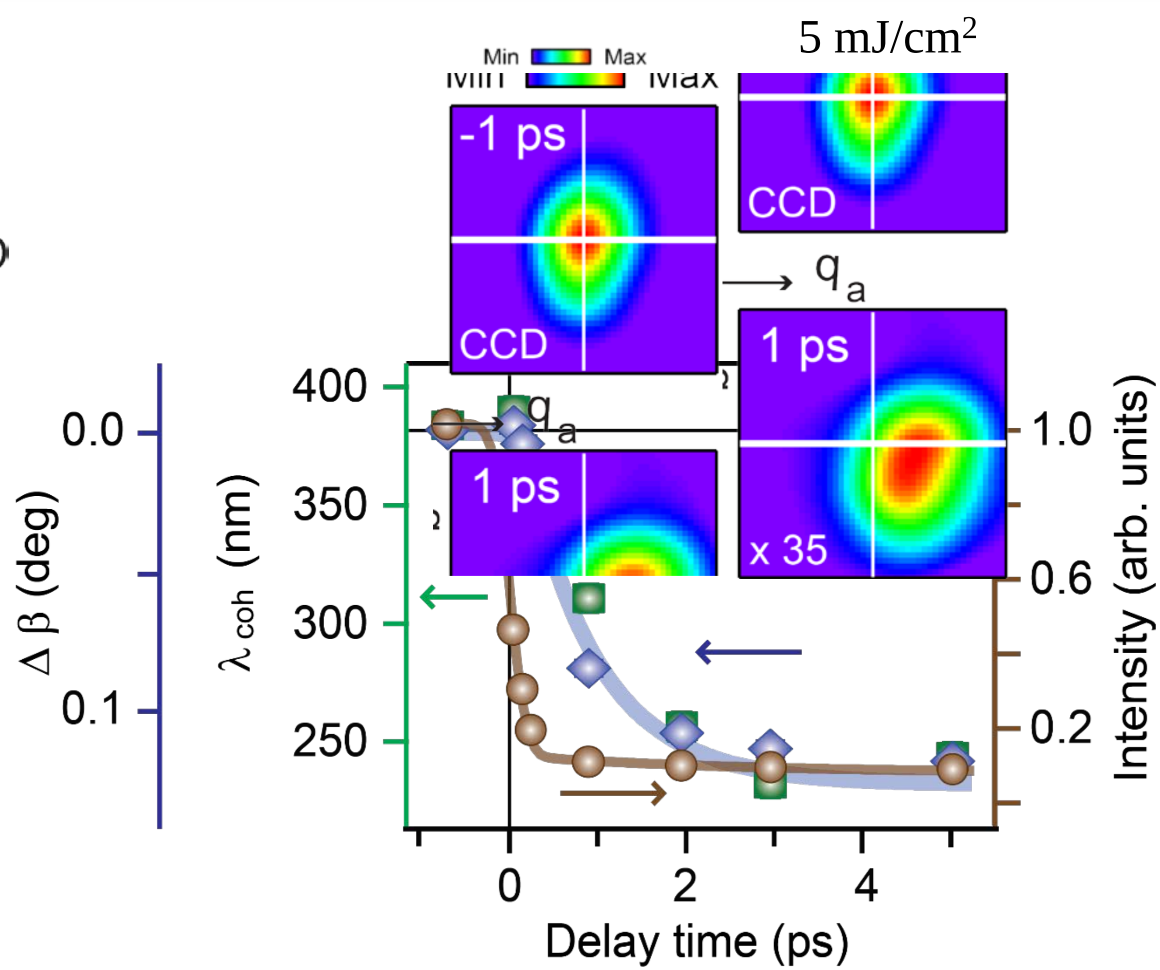
Resonant soft x-ray diffraction (RSXD) endstation



Coherence Length and Monoclinic Tilt



T = 80 K < T(Verwey)



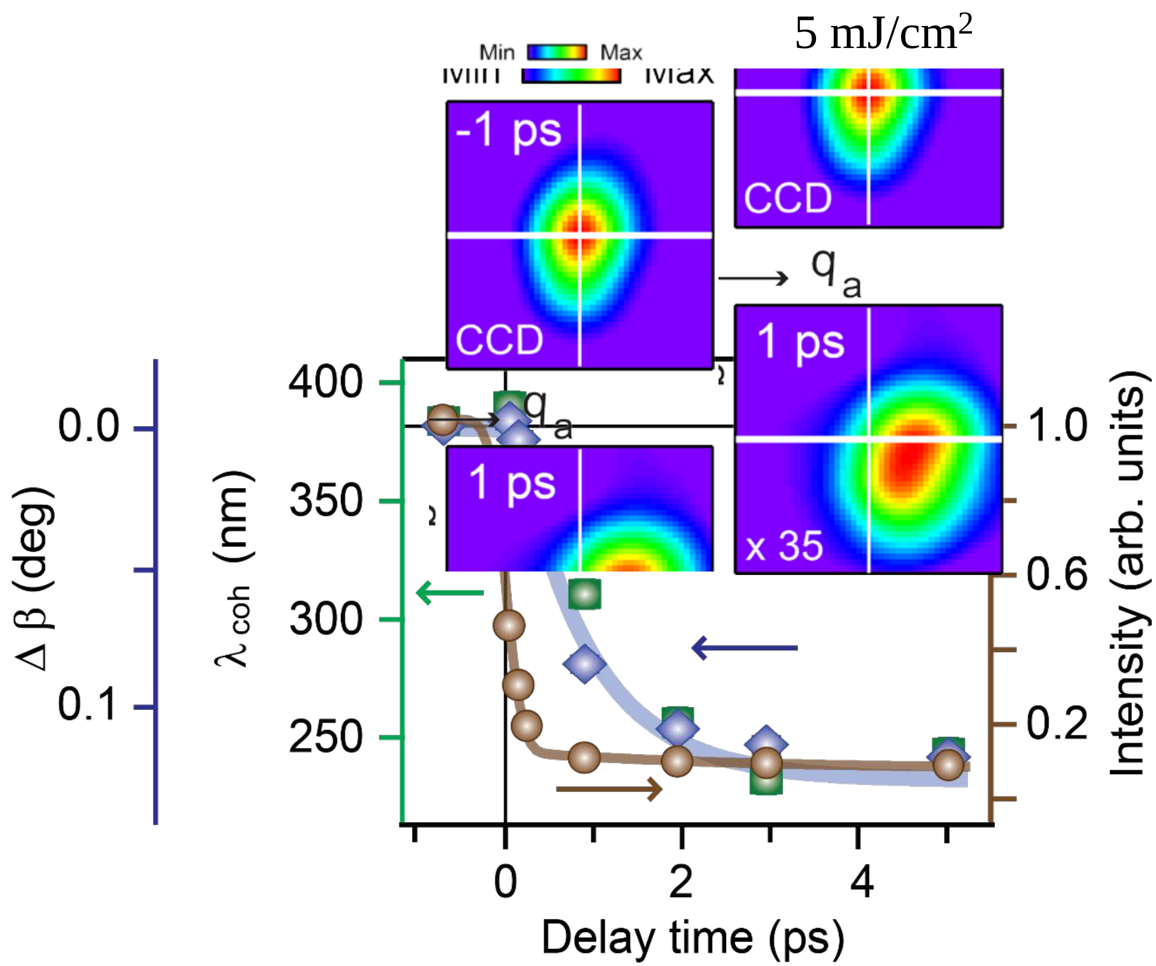
Coherence Length and Monoclinic Tilt

Peak Intensity

- drops to less than 10% within first 300 fs

λ_{coh} and $\Delta\beta$

- Slower ps timescale (1.5 ps)
- λ_{coh} decreases – correlation length scales for low temperature ordering
- $\Delta\beta$ relaxes towards high temperature cubic values



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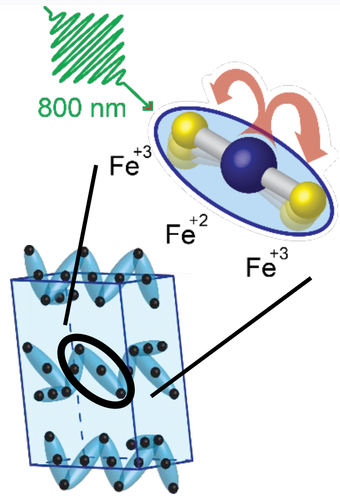
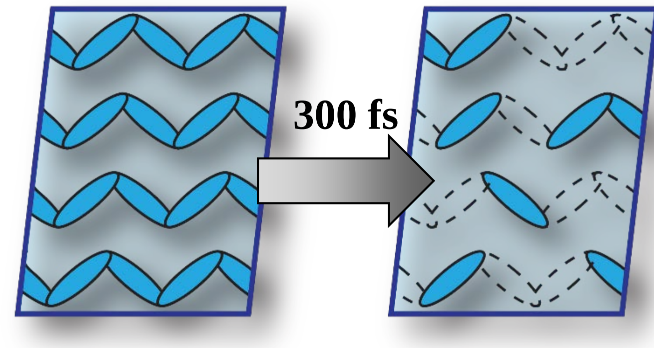
Coexisting insulating and metallic phase

Peak Intensity

- drops to less than 10% within first 300 fs
- *shooting holes in 'trimeron' lattice*





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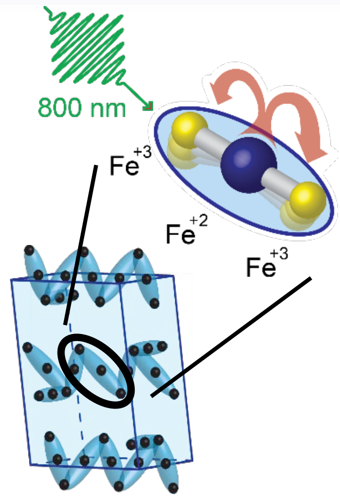
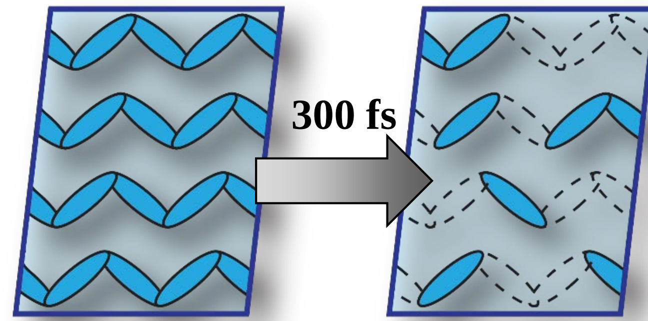


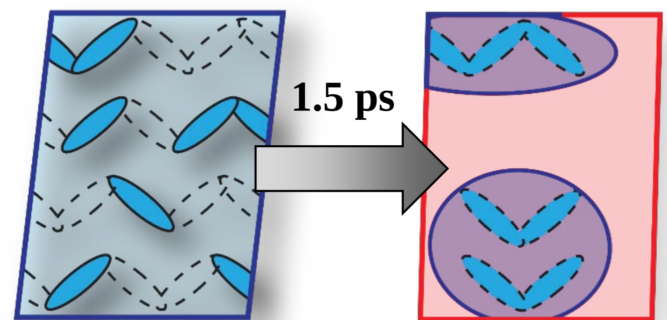


Peak Intensity

- drops to less than 10% within first 300 fs
- *shooting holes in 'trimeron' lattice*

λ_{coh} and $\Delta\beta$

- Slower ps timescale (1.5 ps)
- λ_{coh} decreases and relaxation of $\Delta\beta$ towards high temperature cubic values
- *Phase separation into insulating and metallic regions*



Blue – low temperature monoclinic phase
Red – emerging metallic phase

S de Jong, R. Kukreja et al. Nat. Mater. '13

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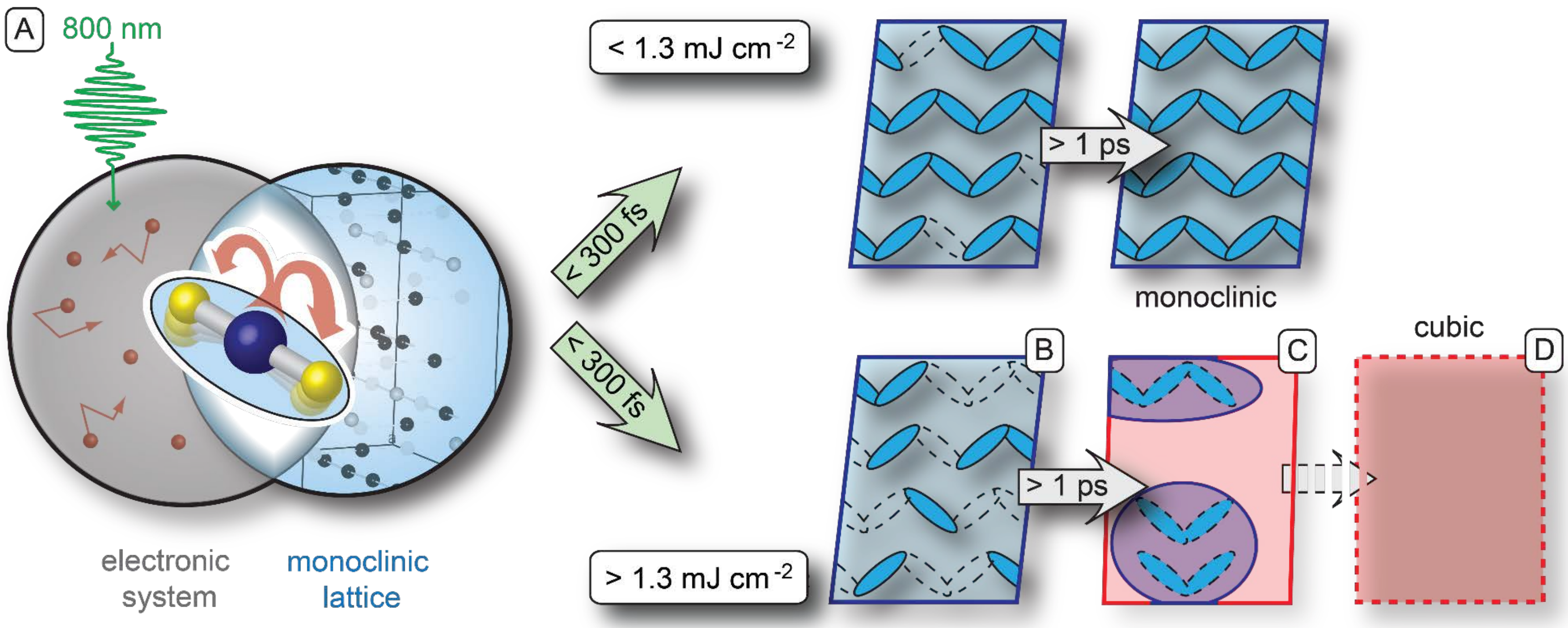
Presence of a Threshold

Below Threshold



Above Threshold

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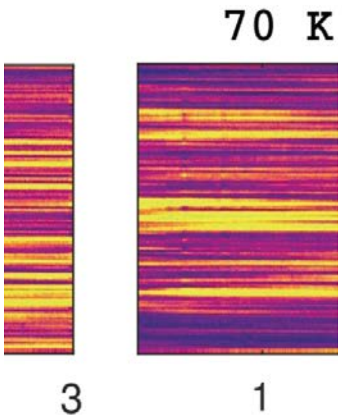
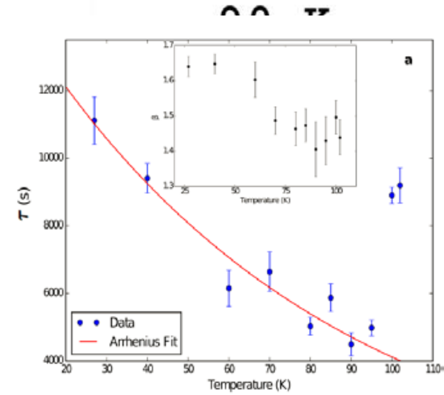
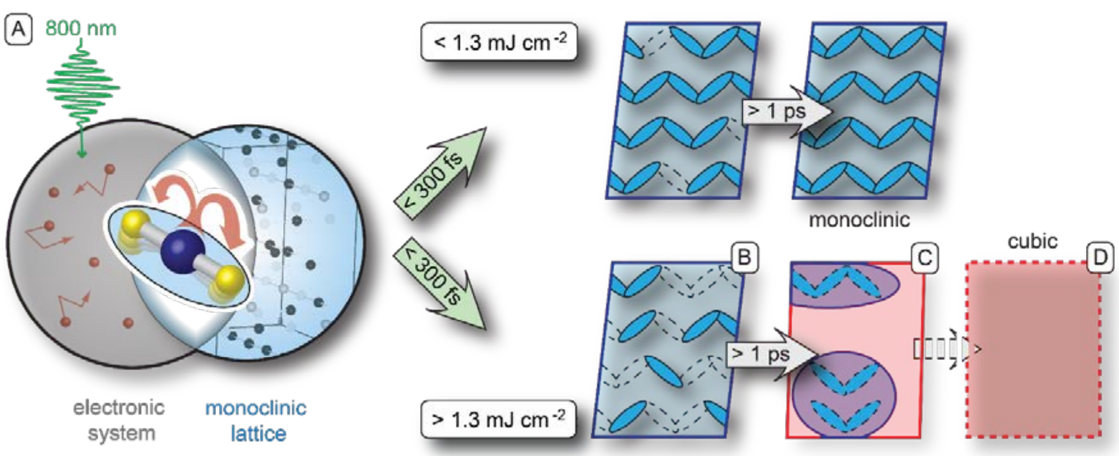
Summary

Domain dynamics near thermally induced Verwey transition. First regime shows thermally activated Arrhenius behavior with an activation energy of be $\Delta E/k_B = 32 \pm 5$ K. Second regime indicates phase separation into metallic and insulating domains.







Imaging optically induced phase separation of magnetite into metallic and insulating regions with timescale of 1.5 picoseconds.

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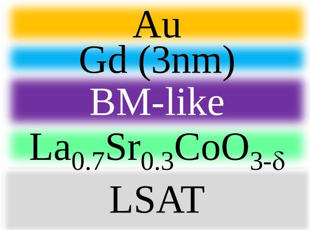
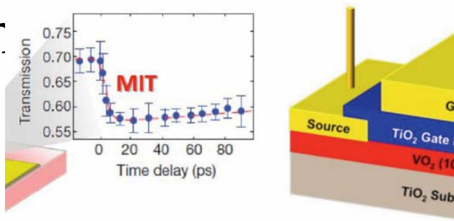
Future Prospects

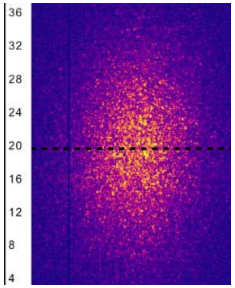
- Controlling nanoscale morphology – heterostructures, epitaxial strain, doping etc.



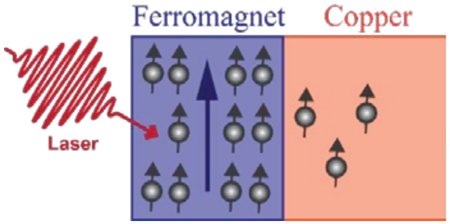
- Role of nanoscale heterogeneities in phase transition



- Imaging spin fluctuations – novel magnetic ordering, cluster phases



- Transport dynamics across heterostructures



Acknowledgements

Domain dynamics in magnetite

UC Davis – Jianheng Li

UC San Diego - Nelson Hua, Oleg Shpyrko, Eric Fullerton

NSLS II – Andi Barbour, Wen Hu, Claudio Mazzoli, Stuart Wilkins

Ultrafast phase separation in magnetite

SLAC – S. de Jong, W.S. Lee, D. H. Lu, M. Yi, R. Moore, M. Trigo, Hermann Dürr

Cologne - C. Trabant , C. F. Chang, M. Döhler, M. Buchholz and C. Schüßler-Langeheine

HZB Berlin - N. Pontius, T. Kachel, M. Beye and A. Föhlisch

LCLS - W. Schlotter, J. J. Turner and O. Krupin

Nanodiffraction studies of phase separation in Gd/LSCO

UC Davis – Ian Rippy, Jianheng Li, Yayoi Takamura

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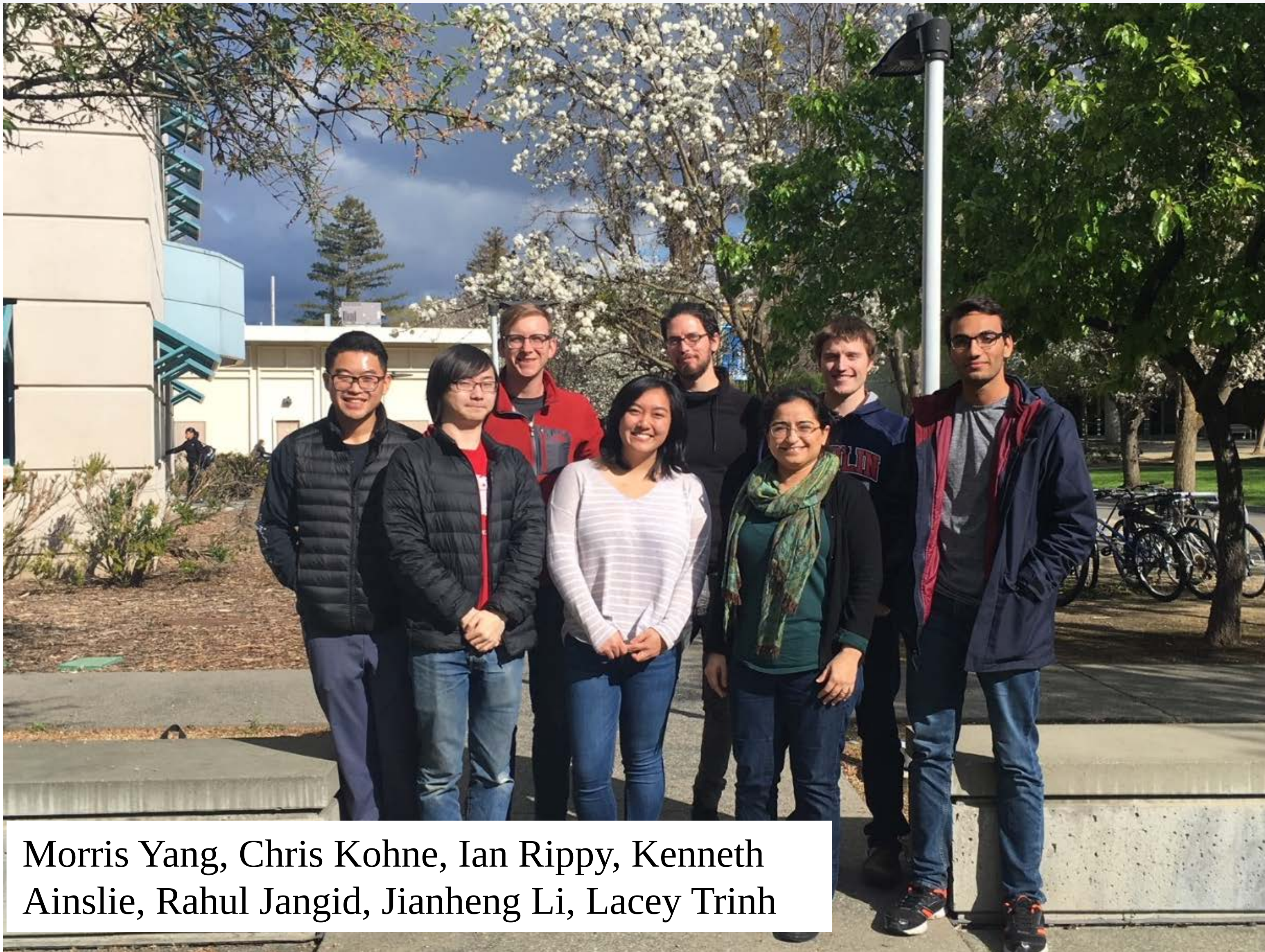
Group at UC Davis



Morris Yang, Chris Kohne, Ian Rippy, Kenneth Ainslie, Rahul Jangid, Jianheng Li, Lacey Trinh

Thank You