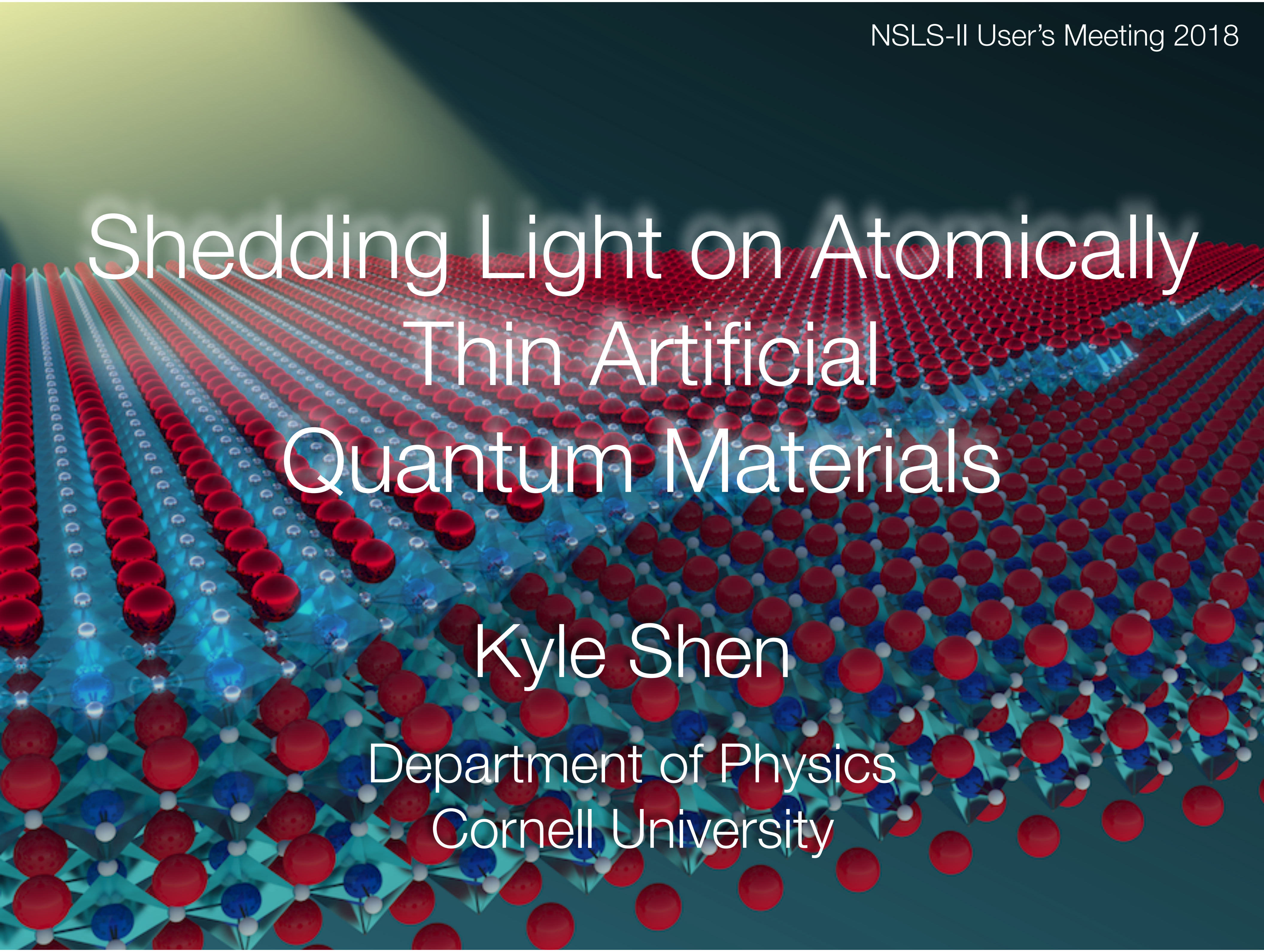


NSLS-II User's Meeting 2018

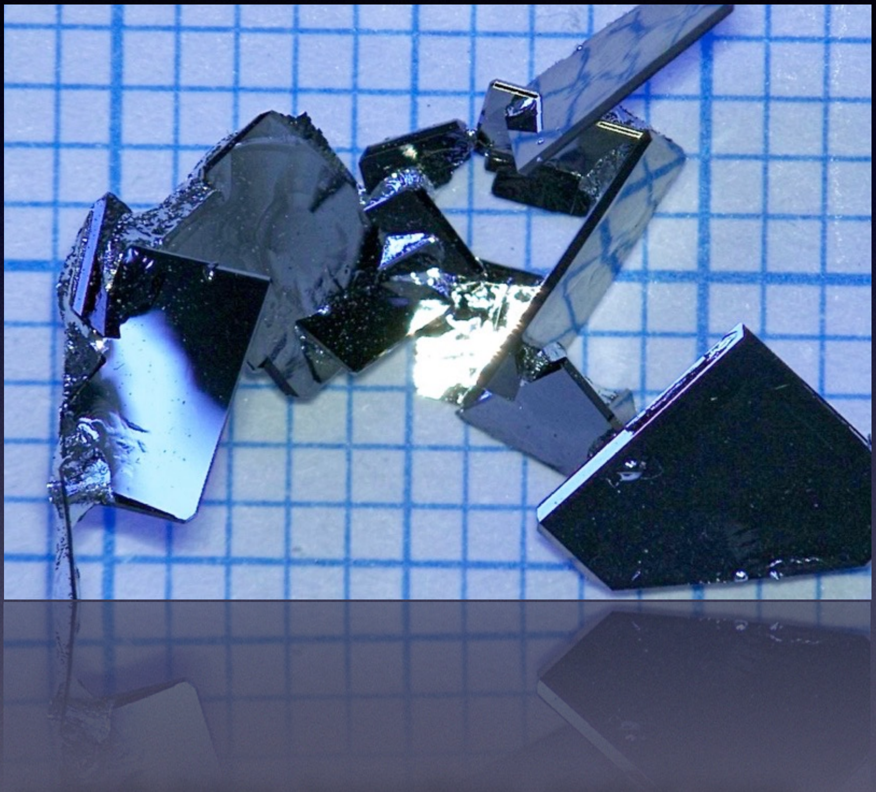
Shedding Light on Atomically Thin Artificial Quantum Materials

Kyle Shen

Department of Physics
Cornell University

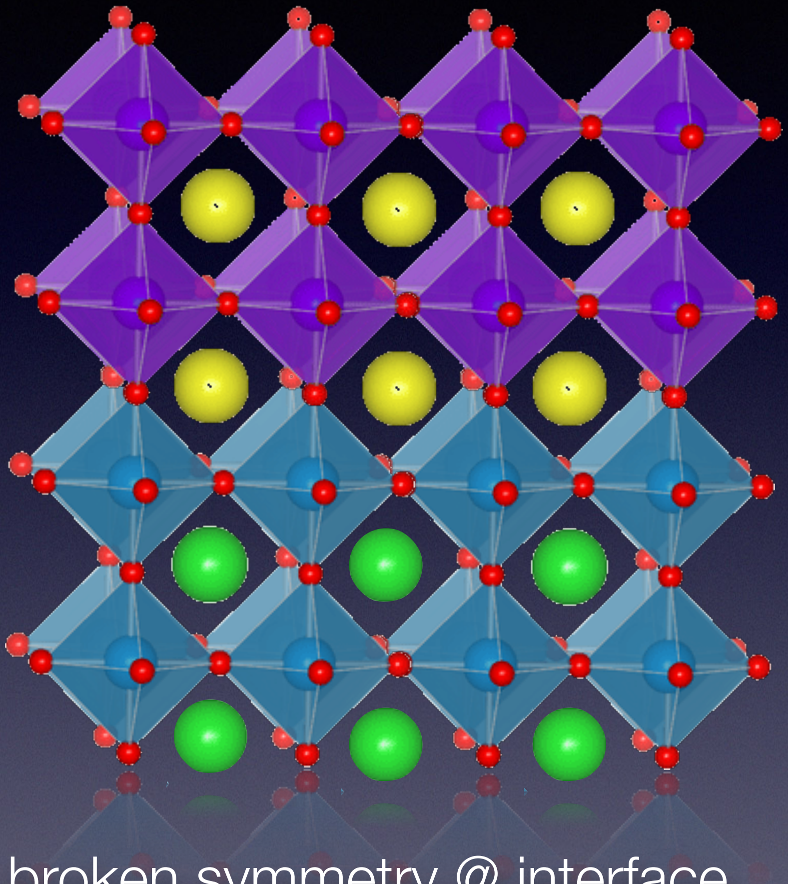
transition metal oxide interfaces : a new frontier

bulk materials



- switching transition metal ions
- chemical doping (cation substitution)
- lattice parameters (cation radius)

interfaces



- broken symmetry @ interface
- lowered dimensionality
- chemical potential offsets
- epitaxial strain (lattice constants)

interfaces made from complex quantum materials

- control of electron density
- dimensional confinement

} conventional materials (Si, GaAs, Ge...)

- strength of interactions
- mass of the carriers
- magnetic interactions
- relativistic or classical

} more exotic building blocks with tunable properties and interactions!

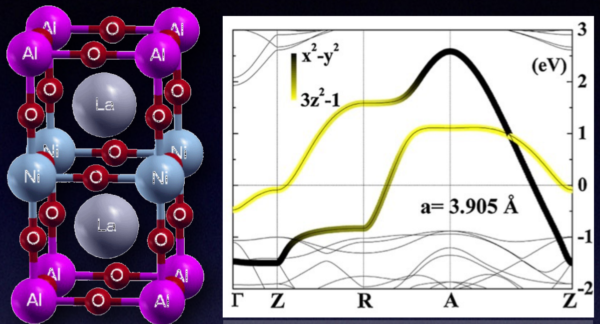
conventional semiconductors

“correlated” quantum materials

the grand challenge : controlling & optimizing quantum materials

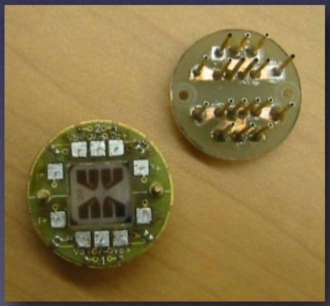
prediction

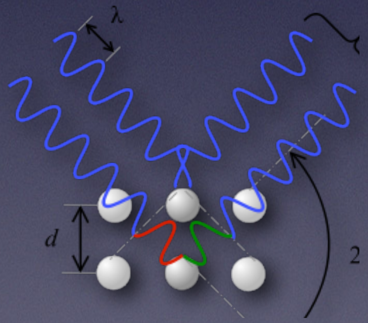
Superconductivity?

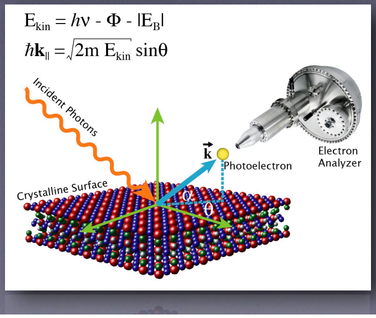


P. Hansmann *et al.*, *Phys. Rev. Lett* **103**, 016401 (2009)

characterization



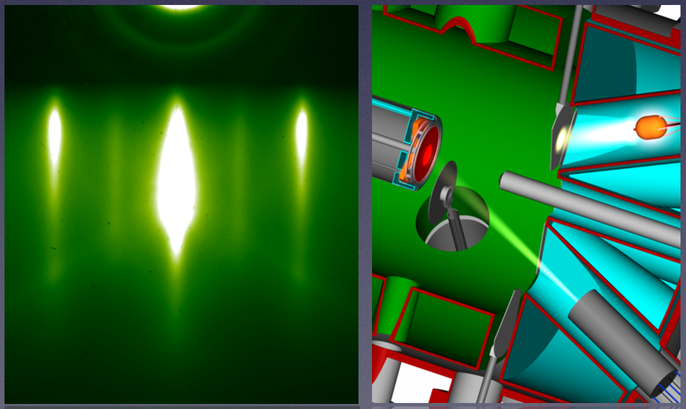




$E_{kin} = h\nu - \Phi - |E_B|$

$\hbar \mathbf{k}_{\|} = \sqrt{2m\, E_{kin}}\, \sin\theta$

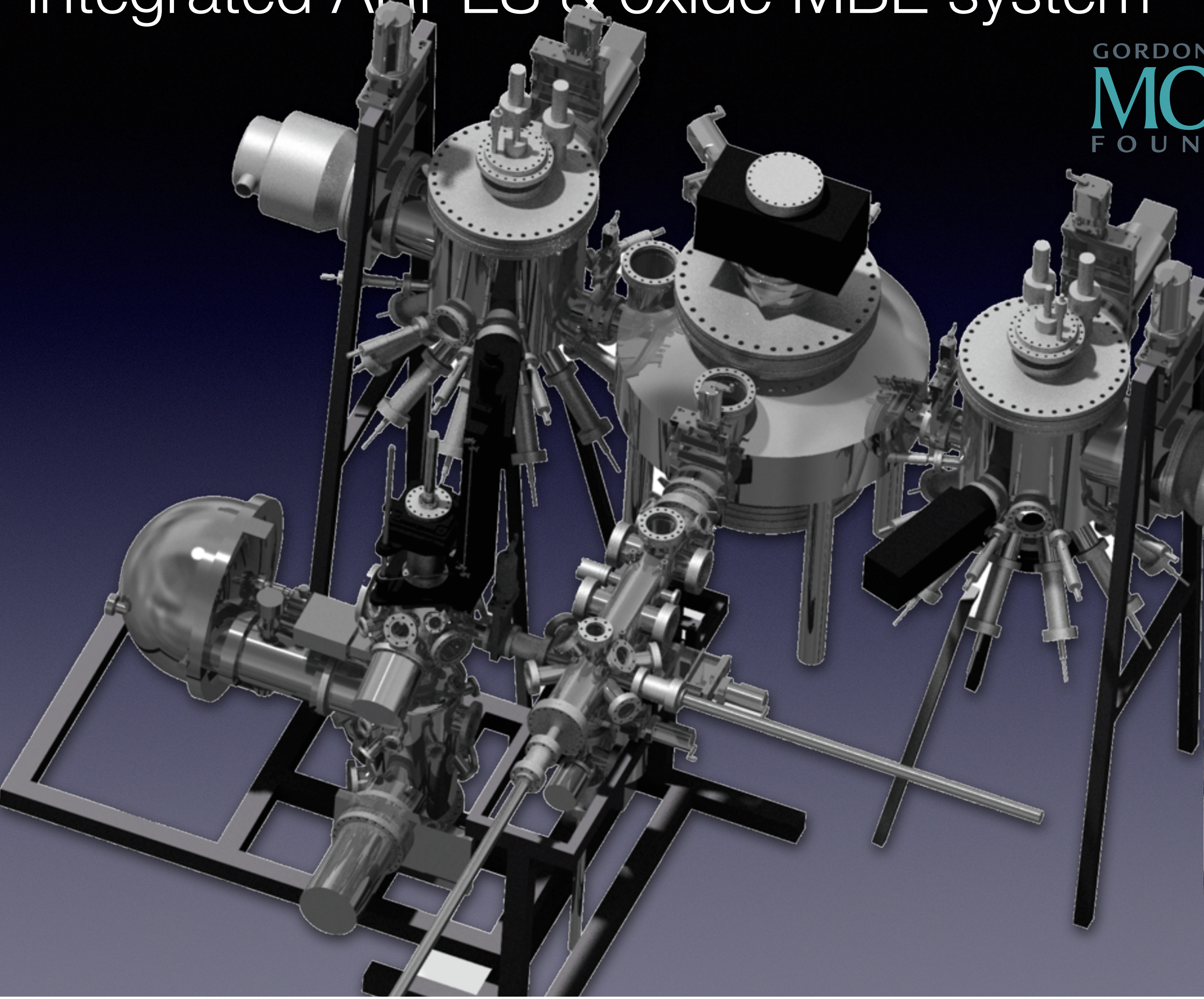
synthesis

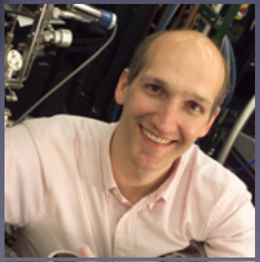


Courtesy of Schlom Group

integrated ARPES & oxide MBE system

GORDON AND BETTY
MOORE
FOUNDATION

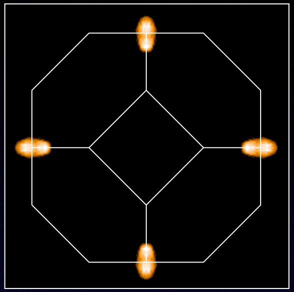




Darrell Schlom

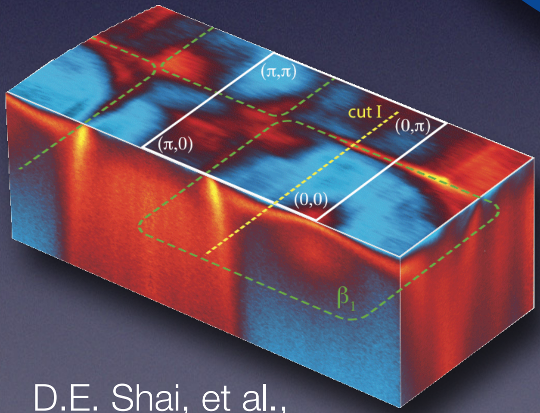
expanding the palette of available materials

EuO



D.E. Shai, et al.,
PRL 108, 267003
(2012)

SrRuO$_3$



D.E. Shai, et al.,
PRL 110, 087004
(2013)

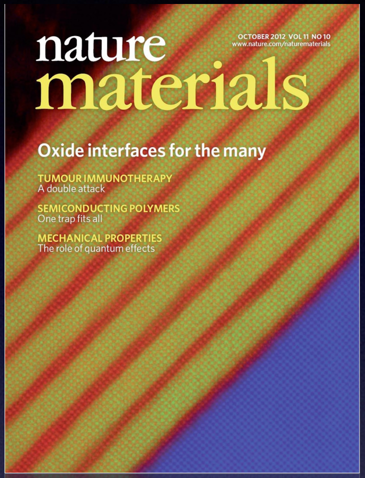
Bulk materials

Cleavable
single
crystals

Artificial
Heterostructures
&
Thin Films

Metastable
Phases not
available
in bulk

oxide
superlattices



nature materials

OCTOBER 2012 VOL 11 NO 10
www.nature.com/naturematerials

Oxide interfaces for the many

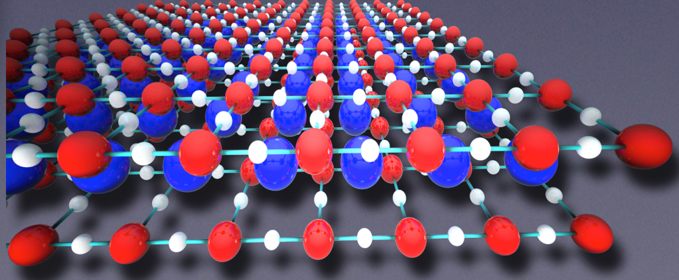
TUMOUR IMMUNOTHERAPY
A double attack

SEMICONDUCTING POLYMERS
One trap fits all

MECHANICAL PROPERTIES
The role of quantum effects

E.J. Monkman et al.,
Nature Materials 11, 855
(2012)

SrCuO$_2$

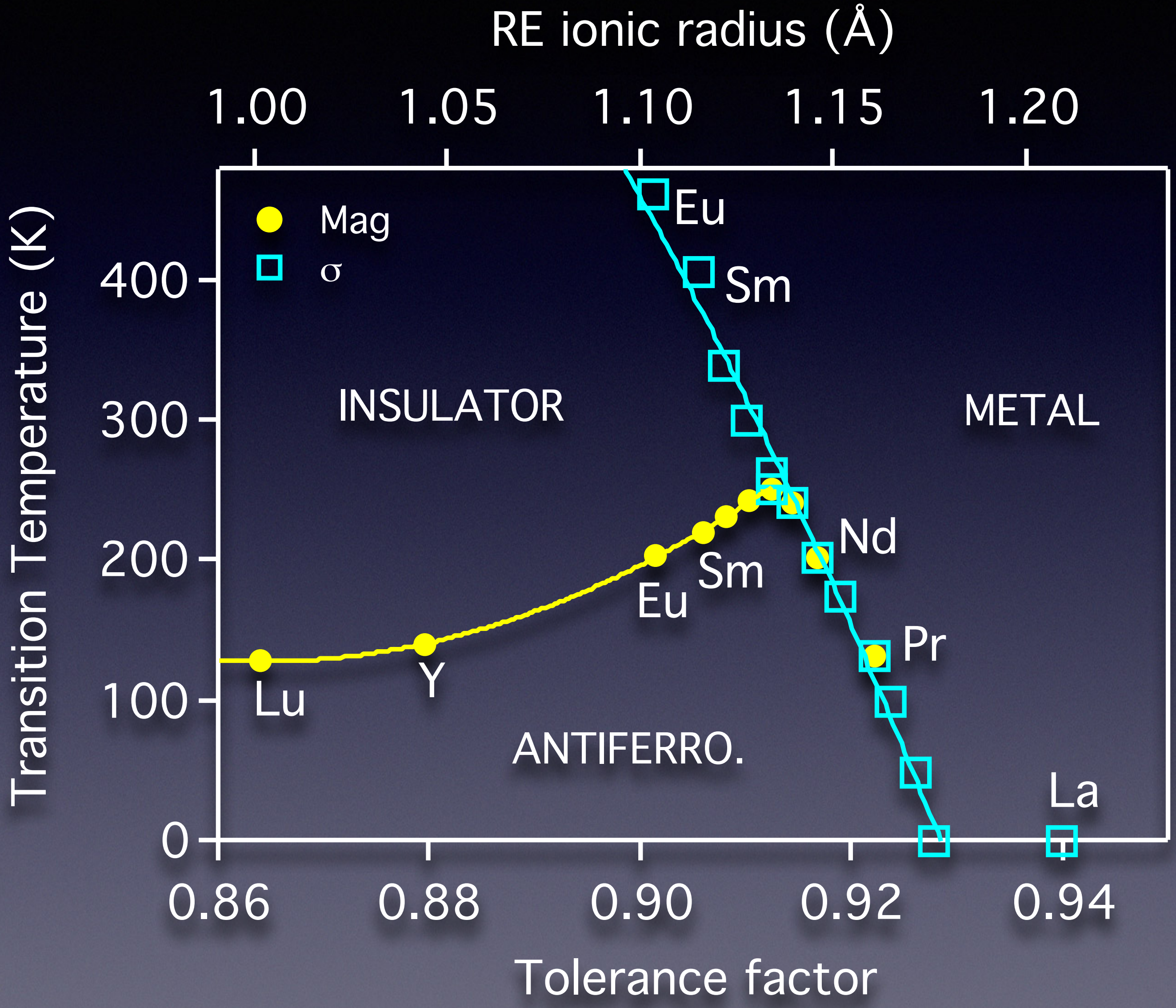


J.W. Harter et al., *PRL* 109, 267001 (2012)

recent examples in atomically thin interface materials

- quantum materials in the ultrathin limit : atomically thin $LaNiO_3$
- enhanced high-T_c superconductivity in monolayer FeSe grown on $SrTiO_3$

Rare-earth nickelates *RE*NiO$_3$

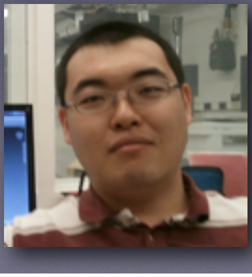


Torrance *et al.*, PRB 45 (1992) 8209

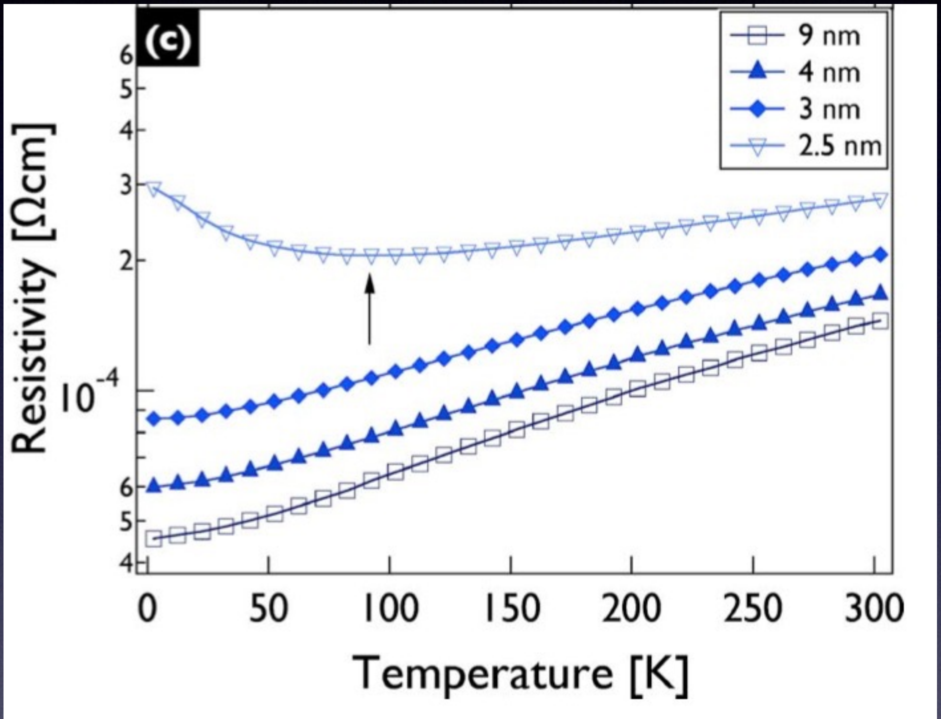
Phil King
(Kavli Fellow;
now faculty @ St.
Andrews Univ)

Masaki Uchida
(JSPS Fellow;
now Asst. Prof.
U. Tokyo)

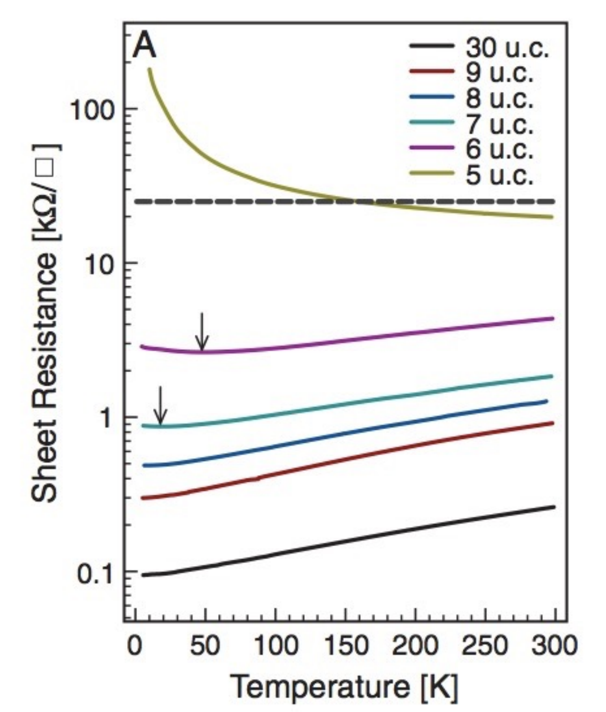
Haofei Wei



metal-insulator transitions in ultrathin $LaNiO_3$ films



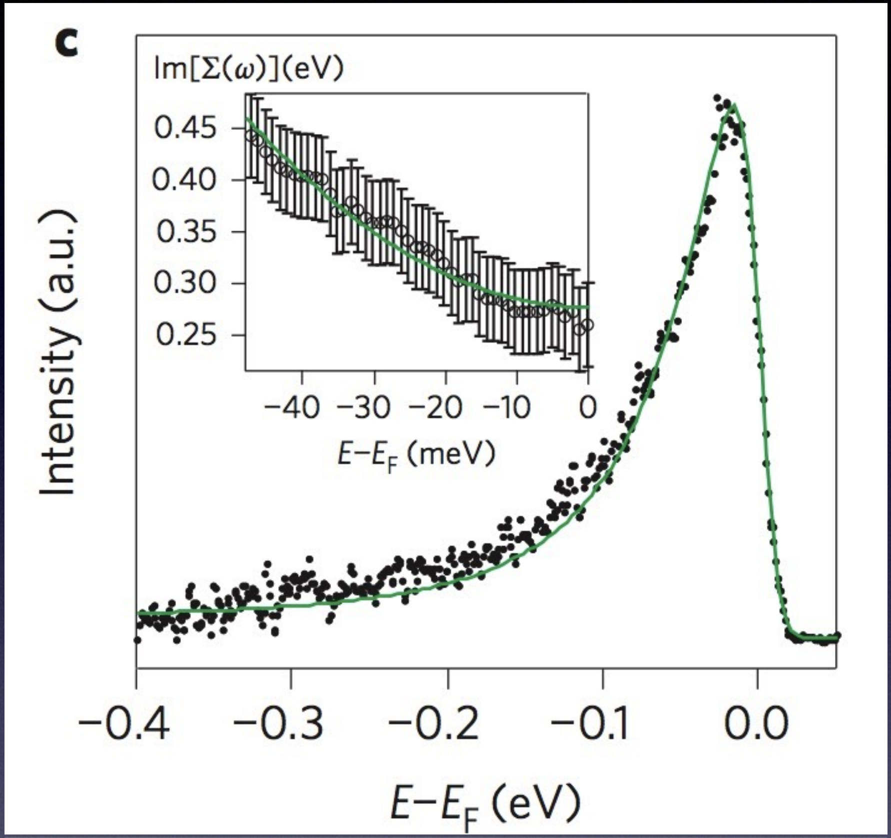
J. Son *et al*, Appl. Phys. Lett. 96, 062114 (2010)



J. Scherwitzl *et al*, Phys. Rev. Lett. 106, 246403 (2011)

$LaNiO_3$ is a correlated metal in bulk, but reducing its thickness below a few unit cells turns the system progressively more insulating

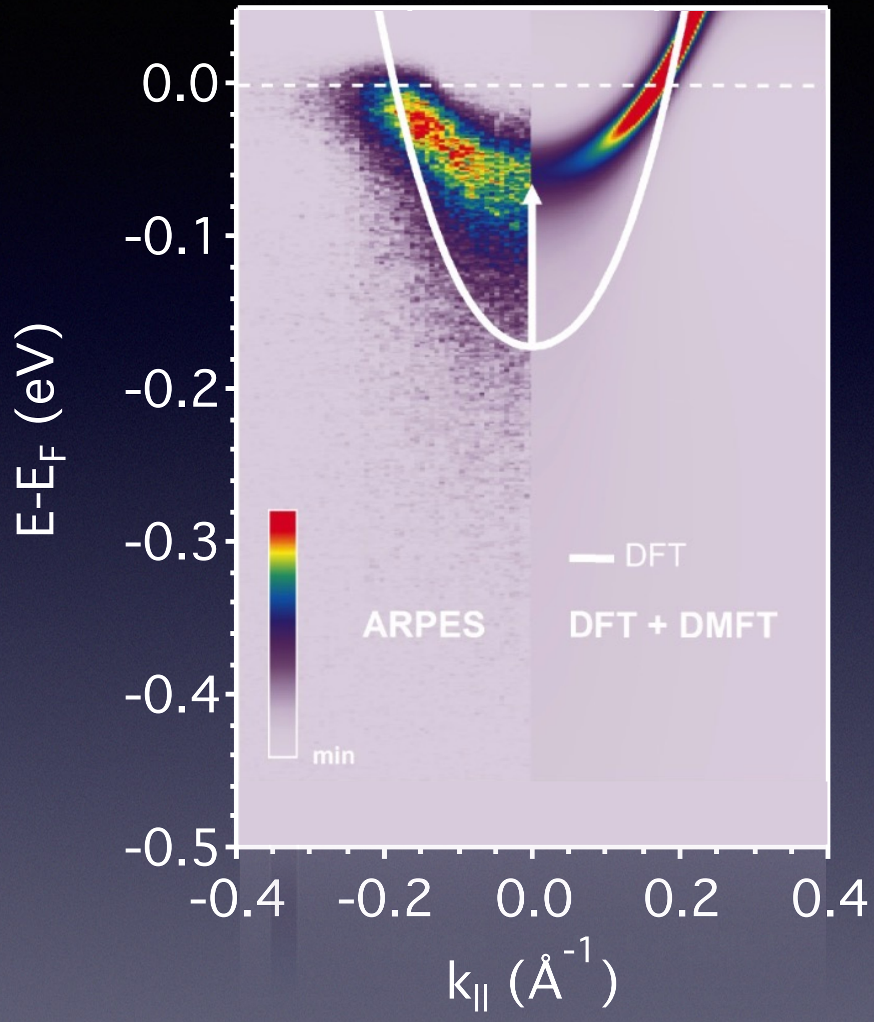
bulk $LaNiO_3$ exhibits a large mass renormalization



$$\frac{m^*}{m} = 3.3 \pm 0.5$$

in agreement with optical & thermodynamic measurements

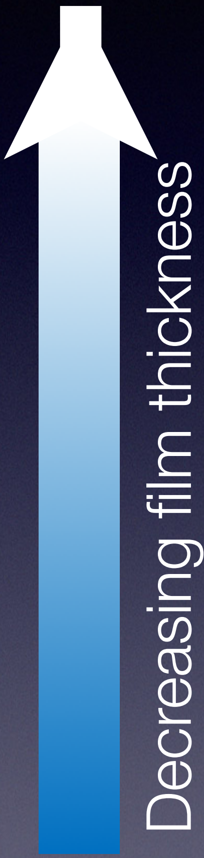
$LaNiO_3$

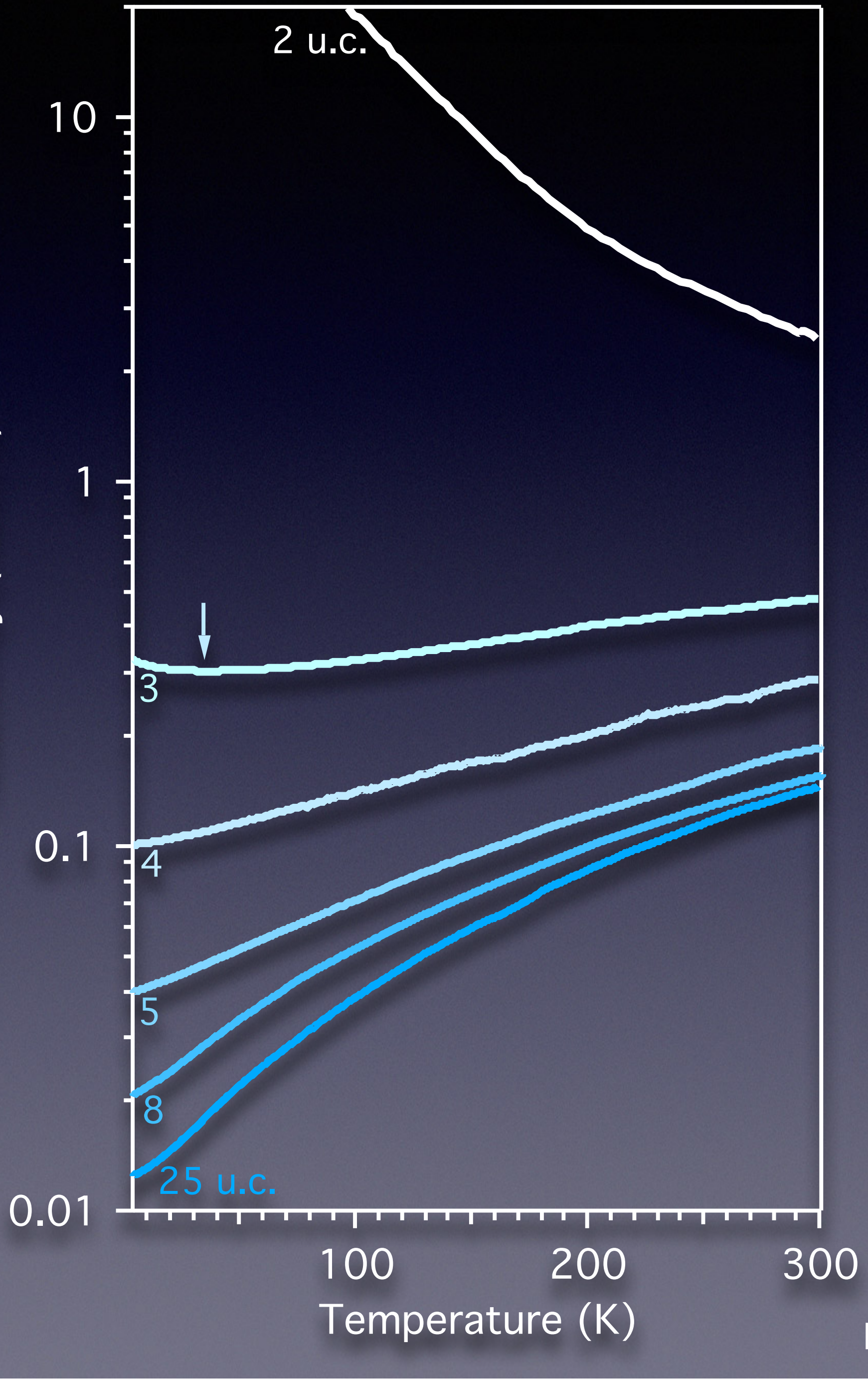


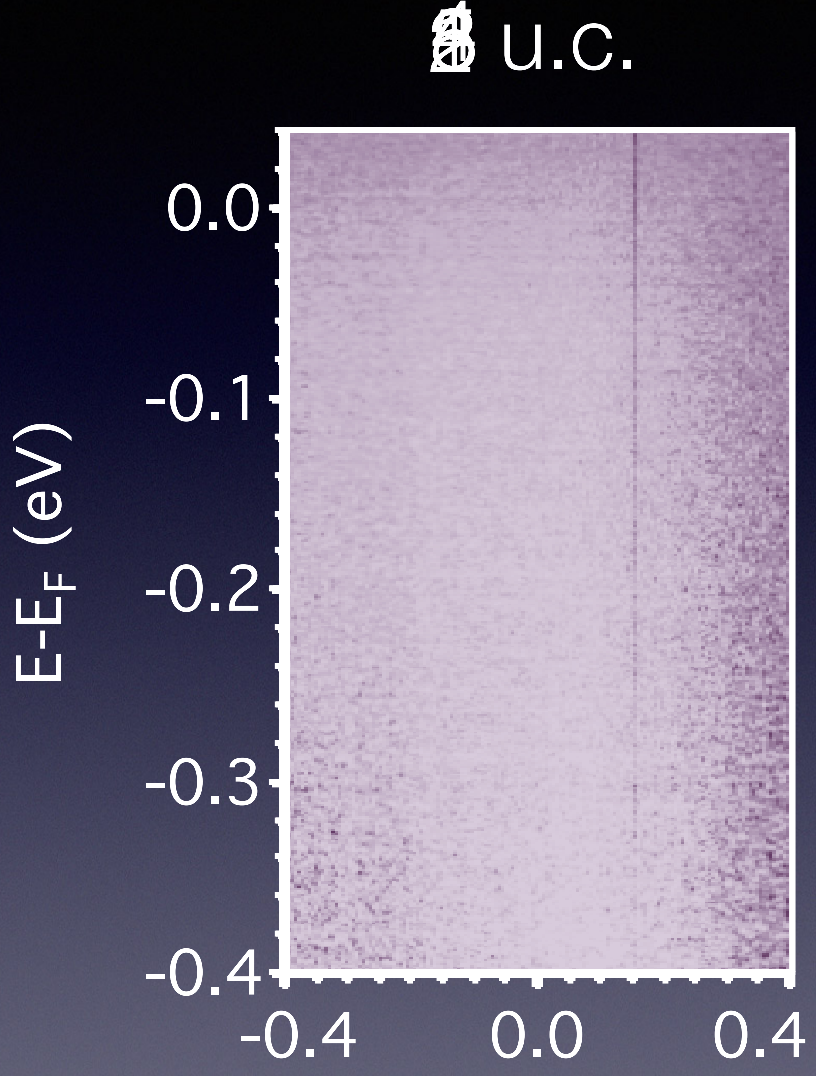
P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)
E.A. Nowadnick, J.P. Ruf et al., *Phys. Rev. B* **92**, 245109 (2015)

P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

Metal-insulator transition at 2 unit cells

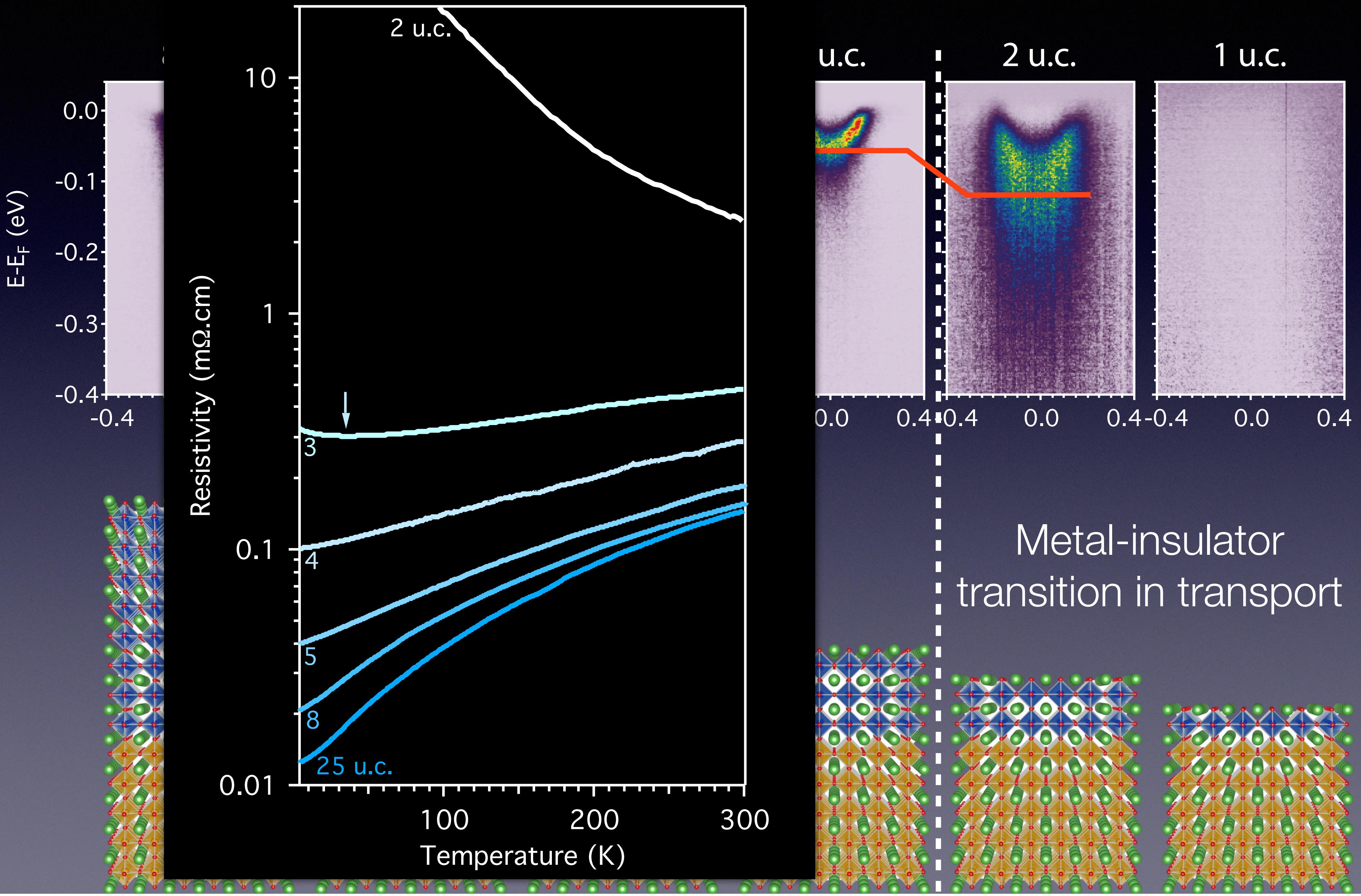






P.D.C. King *et al*, *Nature Nanotechnology* **9**, 443 (2014)

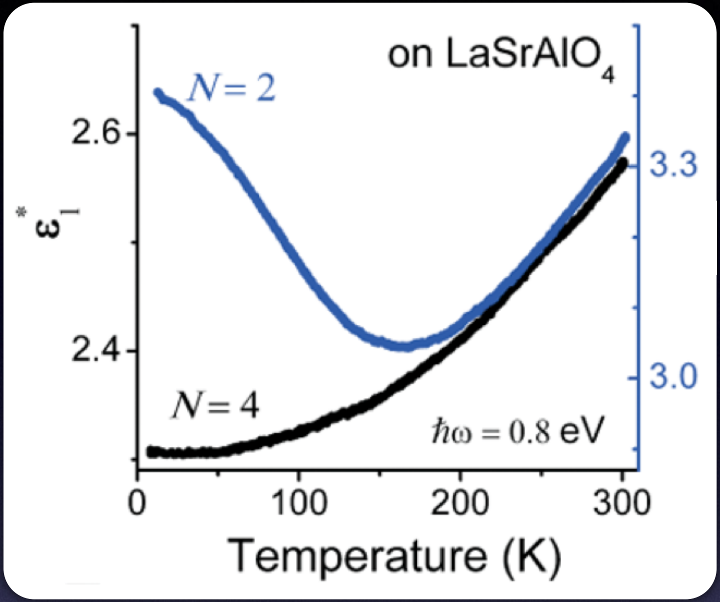
Electronic structure evolution with $LaNiO_3$ thickness



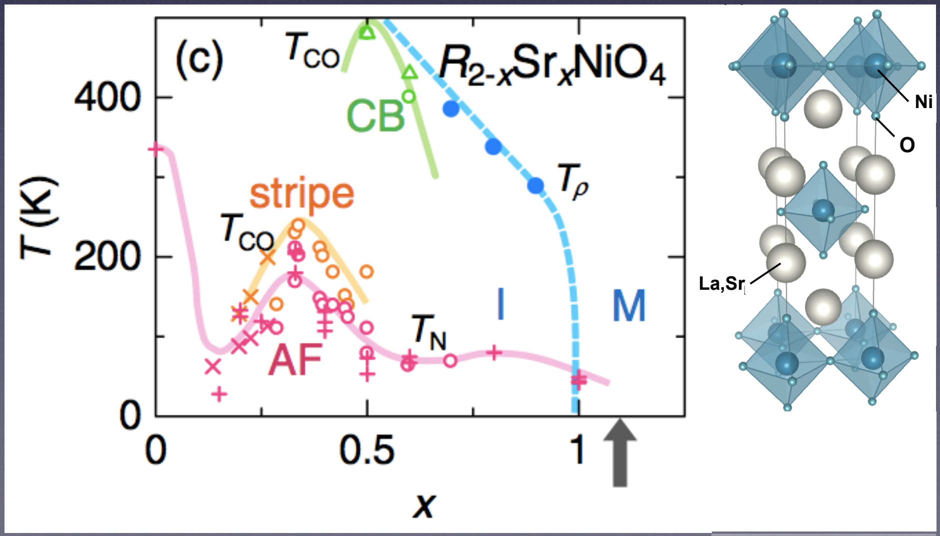
Metal-insulator transition in transport

ordering tendencies in low-dimensional nickelates

Experimental

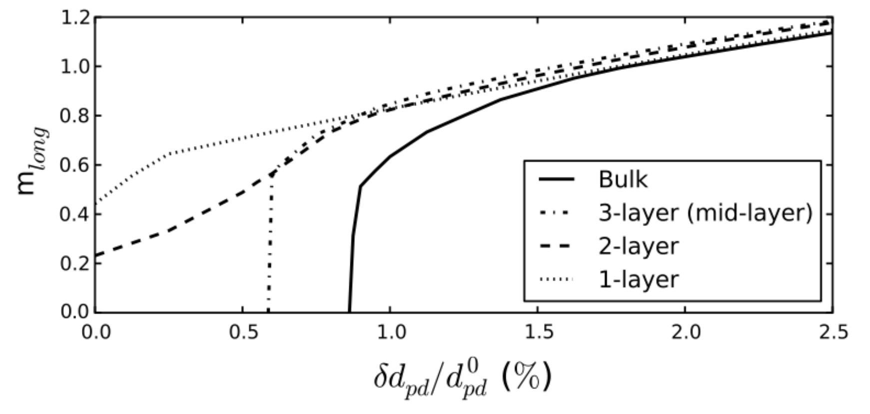


Boris *et al.*, Science 332 (2011) 937

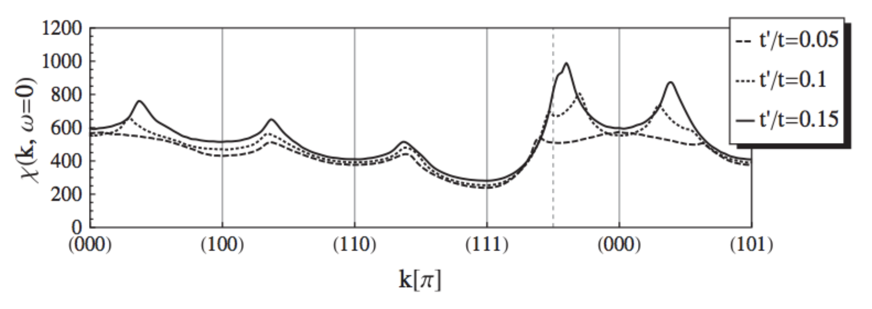


M. Uchida et al., *Phys. Rev. Lett.* **106**, 027001 (2011)

Theoretical



B. Lau & A.J. Millis, *Phys. Rev. Lett.* **110**, 126404 (2013)



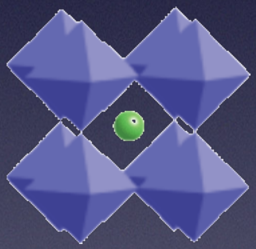
S. Lee, R. Chen & L. Balents, *Phys. Rev. Lett.* **106**, 016405 (2011)

dimensionality plays an important role in layered nickelates

Ruddlesden-Popper Series : $La_{n+1}Ni_nO_{3n+1}$

$n = \infty$

$LaNiO_3$

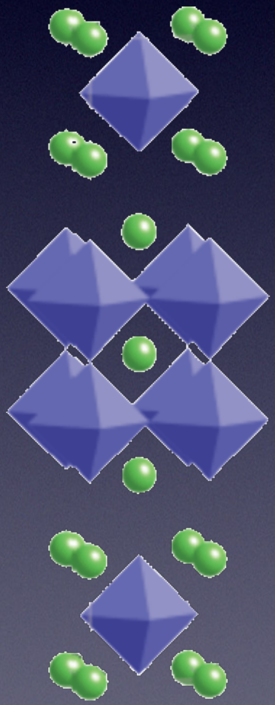


paramagnetic,
correlated metal

• • • • • • • •

$n = 2$

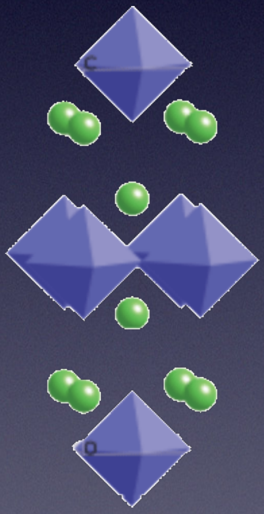
$La_3Ni_2O_7$



paramagnetic
“bad metal”

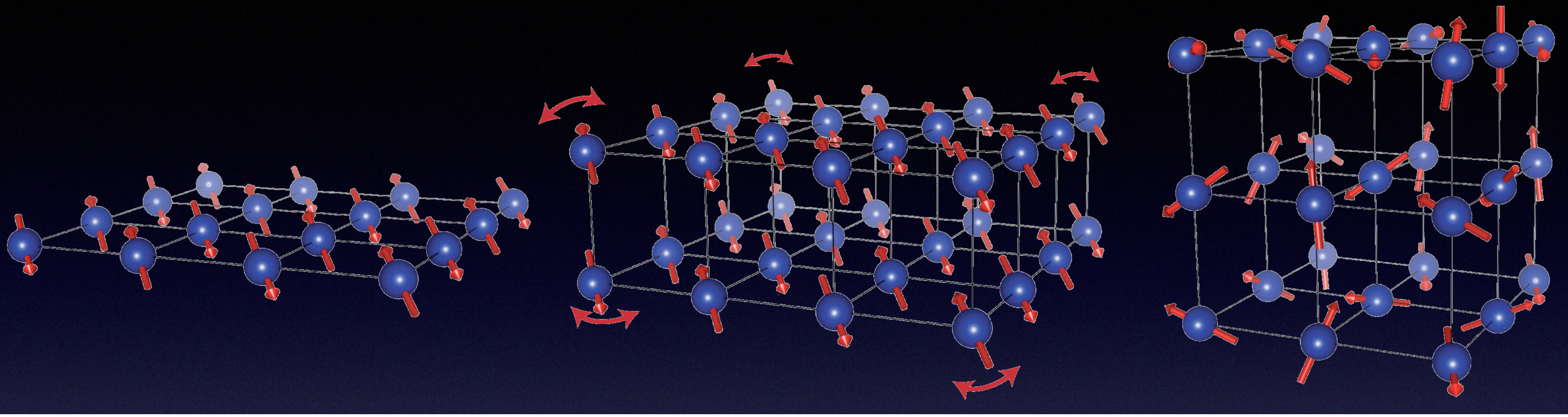
$n = 1$

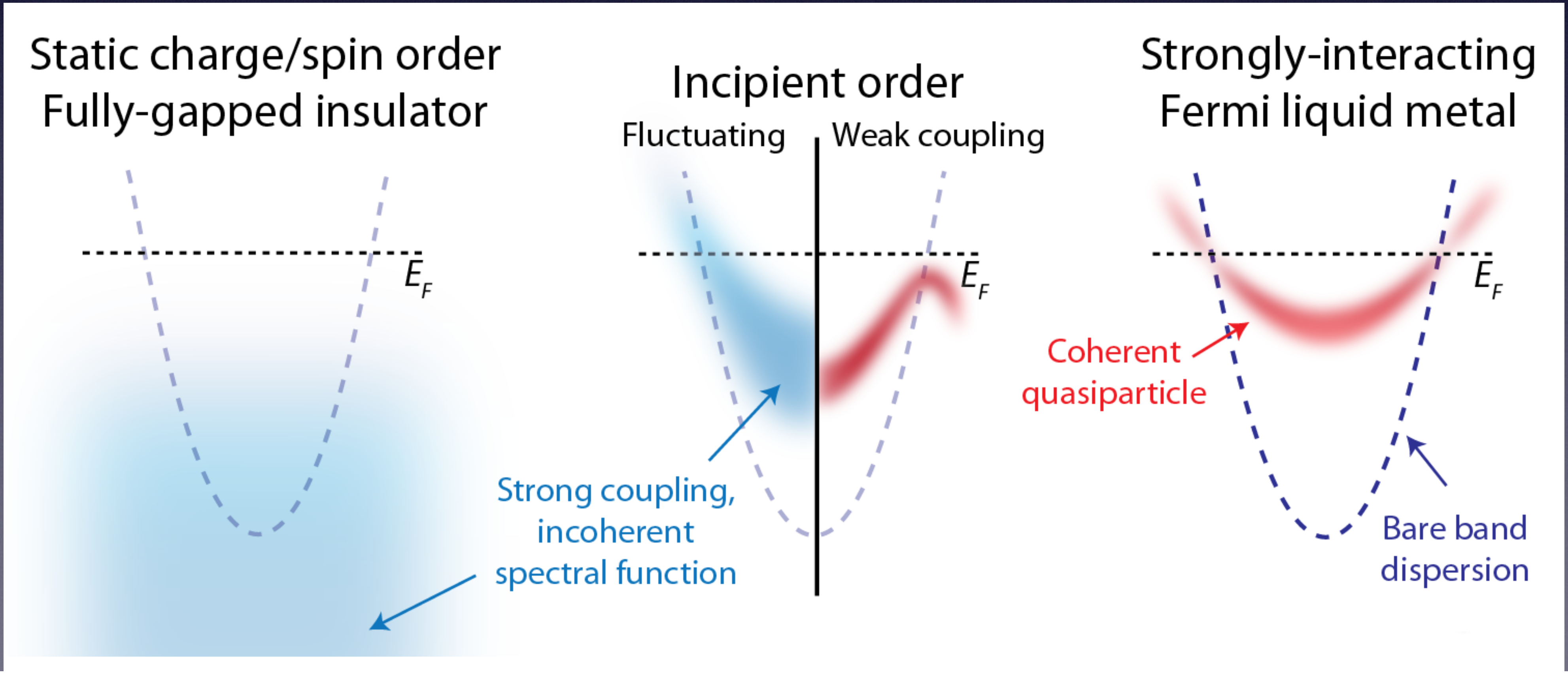
La_2NiO_4



correlated
antiferromagnetic
insulator, insulating
charge / spin stripes

Ordering instability likely onsets below 3 unit cells





Static charge/spin order
Fully-gapped insulator

Incipient order

Fluctuating | Weak coupling

Strongly-interacting
Fermi liquid metal

Strong coupling,
incoherent
spectral function

Coherent
quasiparticle

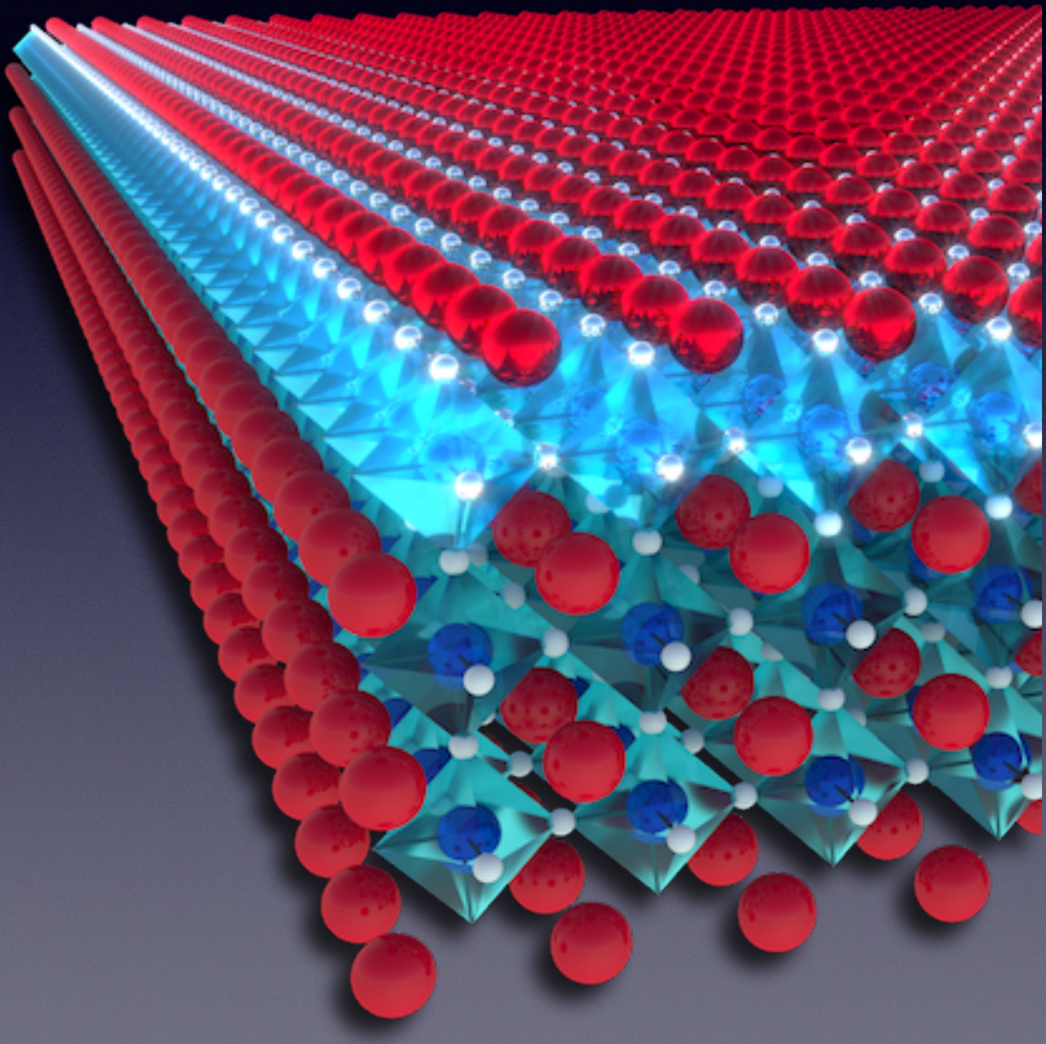
Bare band
dispersion

Scientific Needs from NSLS-II

- can we directly detect static spin/charge ordering stabilized in the ultrathin limit?
- can we observe short-ranged spin fluctuations before static long-ranged order onsets?
- what is the effect of octahedral rotations and/or polar distortions near the film-substrate interface?

Ultrathin

$LaNiO_3$



P.D.C. King *et al*, *Nature Nanotechnology*, 9, 443 (2014)

recent examples in atomically thin interface materials

- quantum materials in the ultrathin limit : atomically thin $LaNiO_3$

- enhanced high-T_c superconductivity in monolayer FeSe grown on $SrTiO_3$

characteristics of superconductors

nature of the superconducting wavefunction

$$\Psi(r_1, s_1; r_2, s_2) = \phi(r_1, r_2)\chi(s_1, s_2) \begin{cases} \phi(r_1, r_2) & \text{spatial part} \\ \chi(s_1, s_2) & \text{spin part} \end{cases}$$

pair wavefunction must be antisymmetric

~ 99% of SC : $\chi(s_1, s_2) \propto (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$

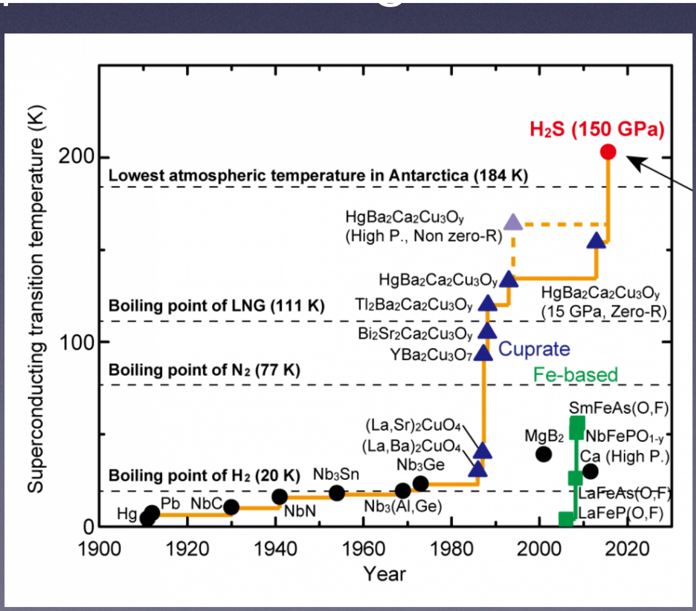
antisymmetric : spin singlet

conventional BCS (Pb, Al, Nb), high Tc cuprates, Fe-SC

very rare : $\chi(s_1, s_2) \propto |\uparrow\uparrow\rangle$

symmetric : spin triplet

superconducting transition temperature (T_c)



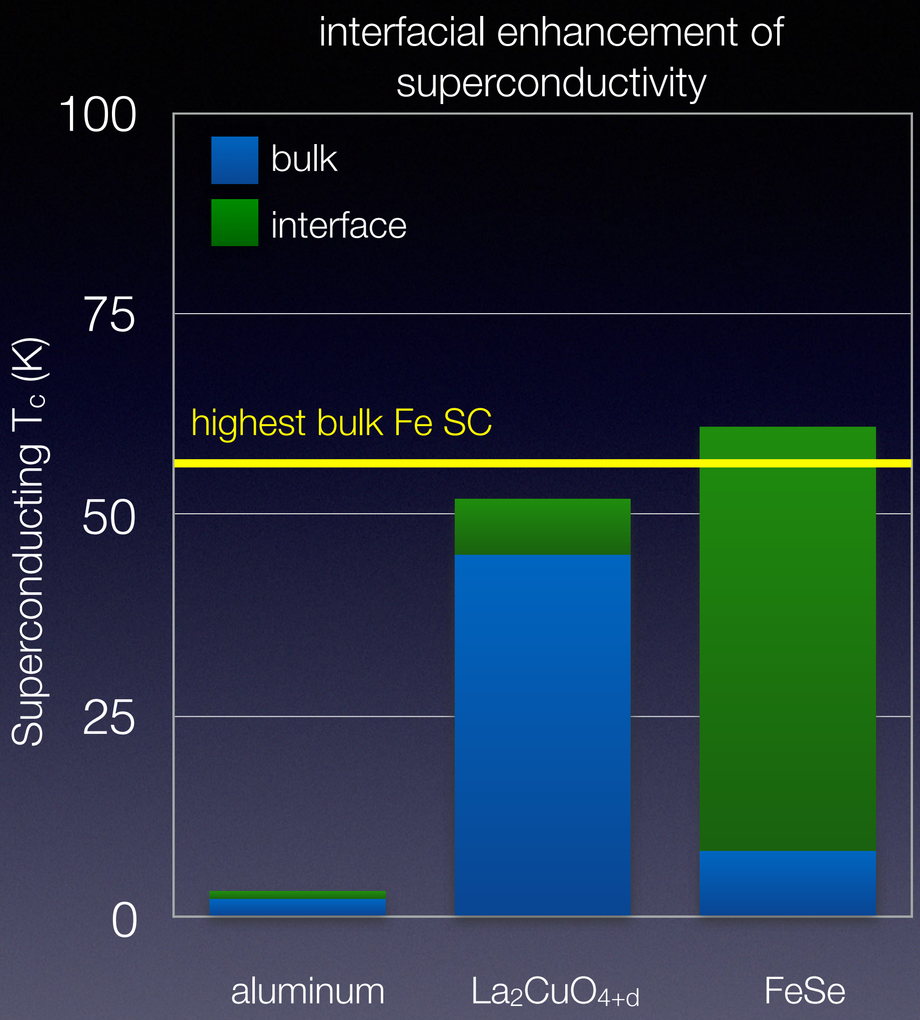
highest T_c known superconductor : H_2S @ 203 K

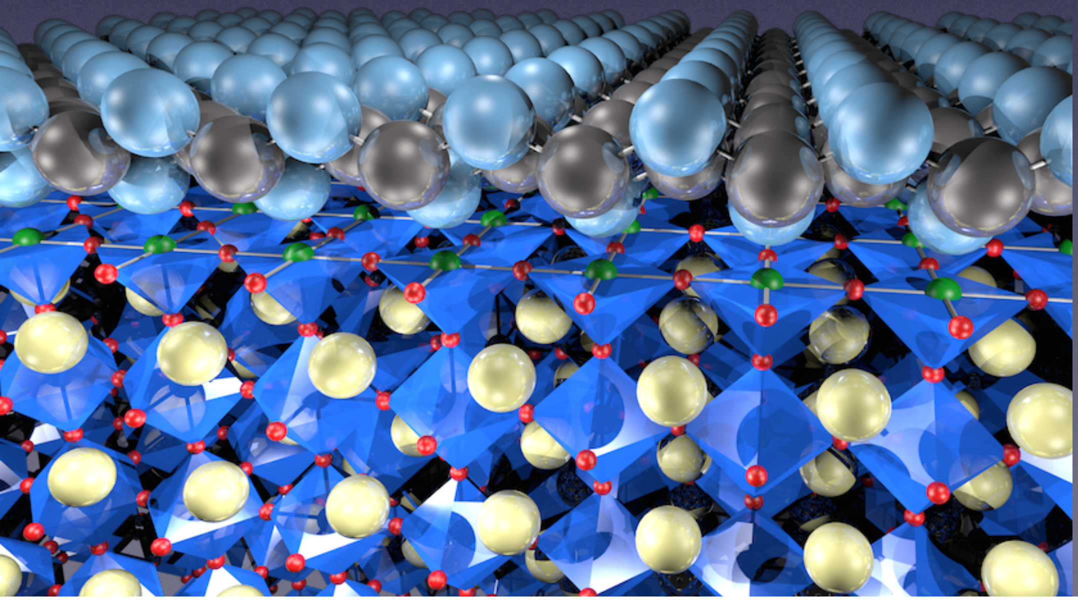
highest T_c ambient SC : $HgBa_2Ca_2Cu_3O_8$ @ 134 K

highest T_c Fe-based SC : SmFeAsO ~ 56 K

interfacial enhancement of superconductivity

- idea of interfacial enhancement of SC dates back to Ginzburg (1964)
- small (10%) effects observed in metal films and cuprate thin films
- in bulk, FeSe has a T_c of 8 K, and is structurally the simplest of the Fe-based SCs
- initial measurements of ~ 60-70 K T_c's of FeSe / $SrTiO_3$ performed by STM and ARPES





V. Ginzburg. *Phys. Lett.* **13**, 101 (1964)

M. Strongin *et al*., *Phys. Rev. Lett.* **21**, 1320 (1968)

I. Bozovic *et al*., *Phys. Rev. Lett.* **89**, 107001 (2002)

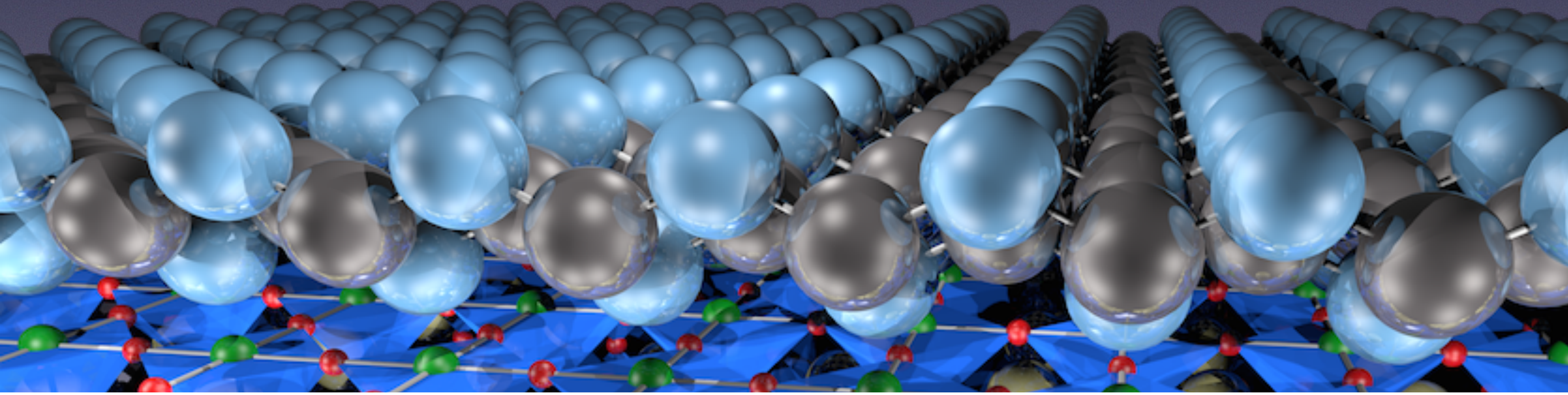
Q.Y. Wang *et al*., *Chin. Phys. Lett.* **29**, 017401 (2012)

central questions about monolayer FeSe / $SrTiO_3$

- what is the mechanism of the interfacial enhancement of T_c?
- how does the T_c measured spectroscopically compare to other probes?
- what happens to the superconductivity as the FeSe film gets thicker?

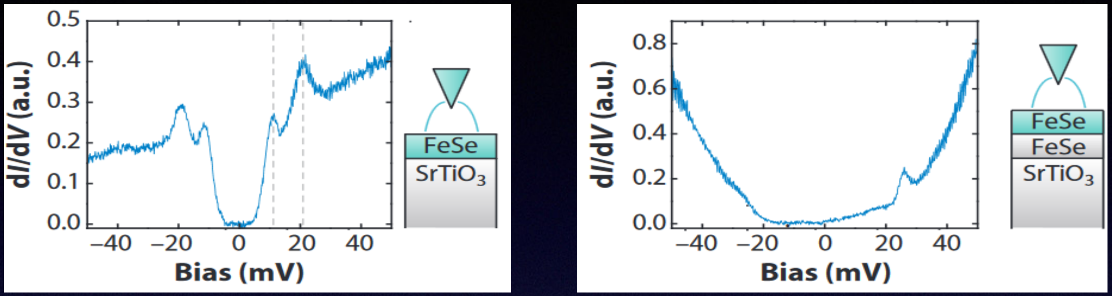
challenges

- superconductivity of FeSe / $SrTiO_3$ monolayer does not survive removal into air (unless samples are capped)
- capped samples appear to exhibit lower T_c's when measured *ex situ* than reported by *in situ* spectroscopic probes
- a given sample is not measured by different techniques; T_c's cannot be compared



what happens as the FeSe film gets thicker?

in situ probes (ARPES, STM) see no gap for films thicker than 1 UC



1 unit cell FeSe

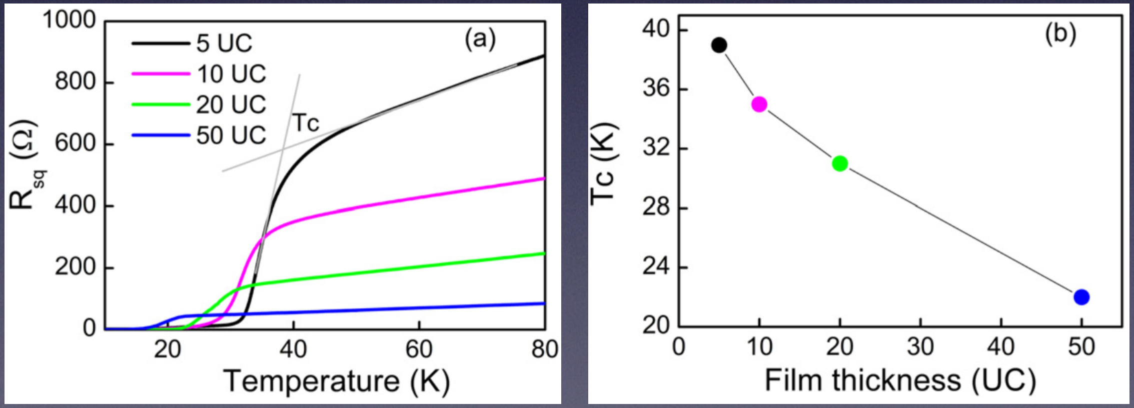
2 unit cells FeSe

Wang et al., *Chin. Phys. Lett.* **29**, 037402 (2012)

- superconducting gap disappears for all films thicker > 1 uc

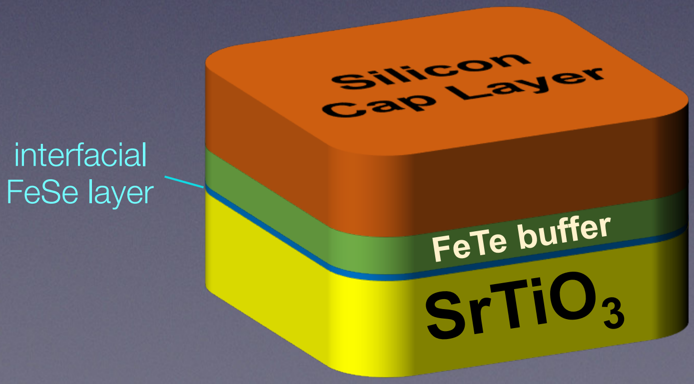
surface-sensitivity cannot see buried interface?

ex situ measurements on capped films



Wang et al., *2D Materials* **2**, 044012 (2015)

- SC observed for thicker capped films, but T_c appears to decrease with film thickness
- capping layers believed to reduce T_c

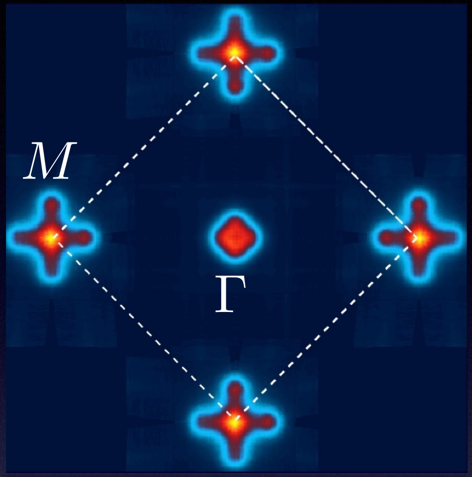


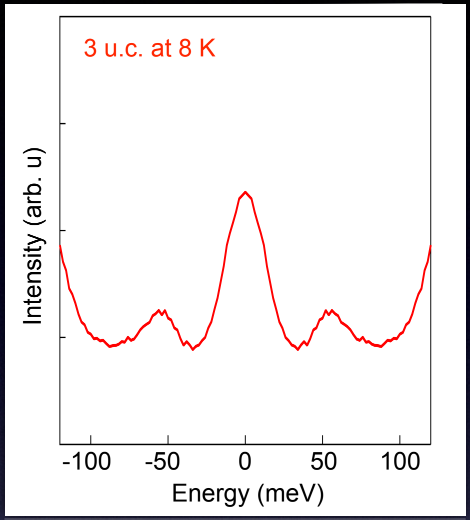
ARPES measurements as a function of film thickness

bulk-like Fermi surface

no detectable SC gap

3 unit cell
FeSe





- ARPES data for 3 unit cell sample nearly identical to bulk FeSe
- No observable SC gap measured down to base temperatures

missing hole pocket at Γ

15 meV gap

gap closes ~ 60 K

1 unit cell
FeSe