Photoemission and the electronic structure of magnetic oxides

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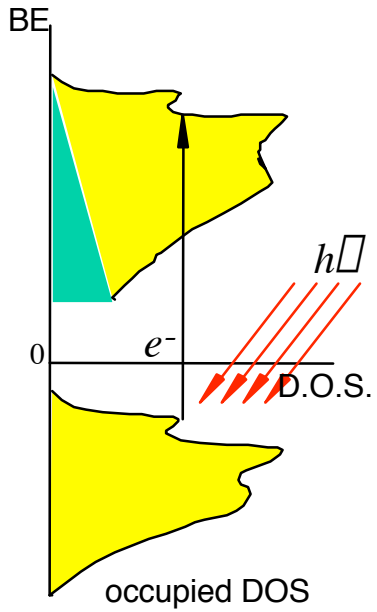
Einstein's Photoelectric effect
XPS, UPS, ARPES

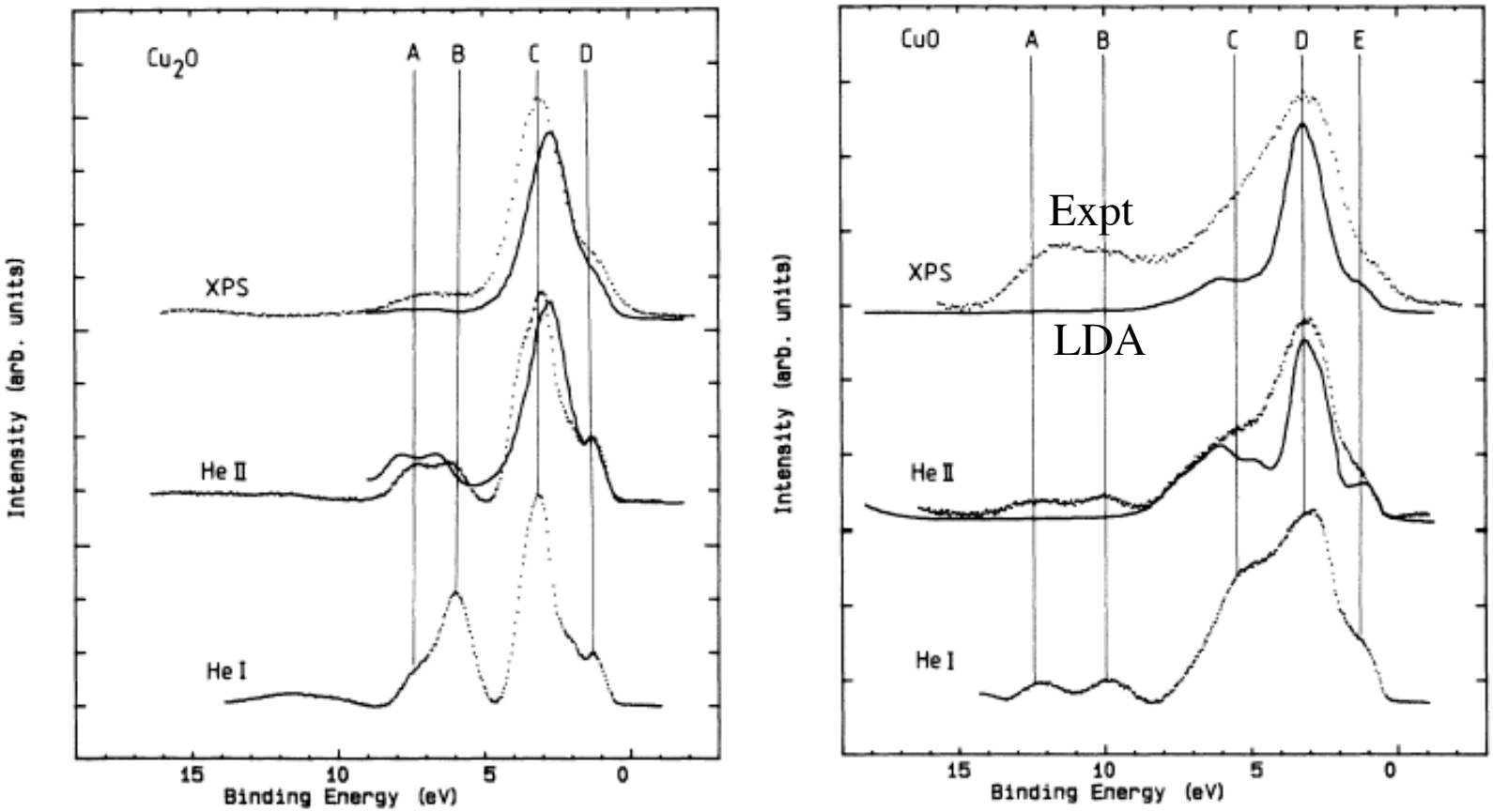


PHYSICAL REVIEW B VOLUME 38, NUMBER 16 1 DECEMBER 1988

Electronic structure of Cu_2O and CuO

J. Ghijsen, L. H. Tjeng, J. van Elp, H. Eskes,
J. Westerink, and G. A. Sawatzky
Department of Solid State and Applied Physics, Material Research Centre, University of Groningen, Nijenborgh 18, NL-9747 AG Groningen, The Netherlands

M. T. Czyzyk
ESM Afdeling, Fakulteit der W & N, Catholic University of Nijmegen, Toernooiveld, NL-6525 ED Nijmegen, The Netherlands
(Received 25 July 1988)



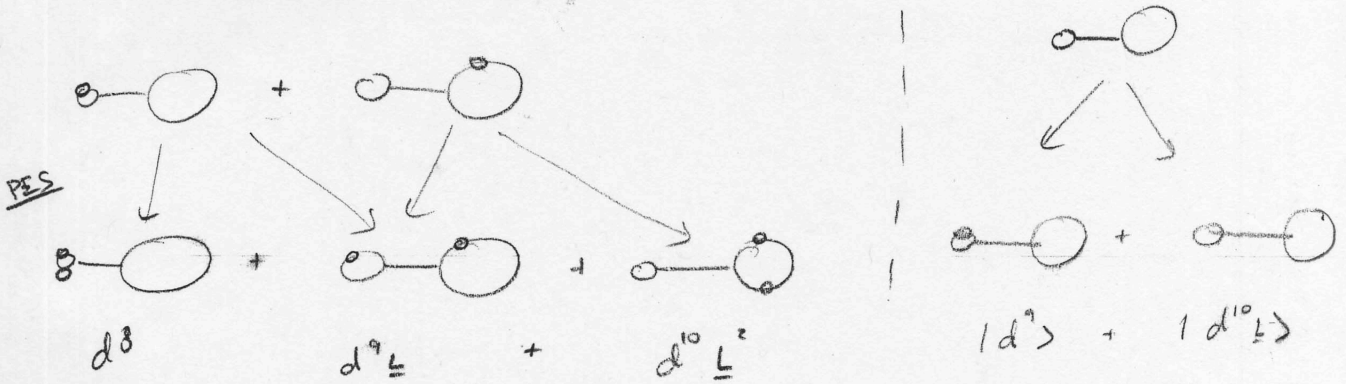
$Cu = 4s^1 3d^{10}$

$Cu^{2+}O^{2-}$ ↔ different by Δ charge transfer energy

$|d^9\rangle + |d^{10}\underline{L}\rangle$

Cu_2O

$|d^{10}\rangle$



costs U_{dd}

costs U_{pp} (small)

core

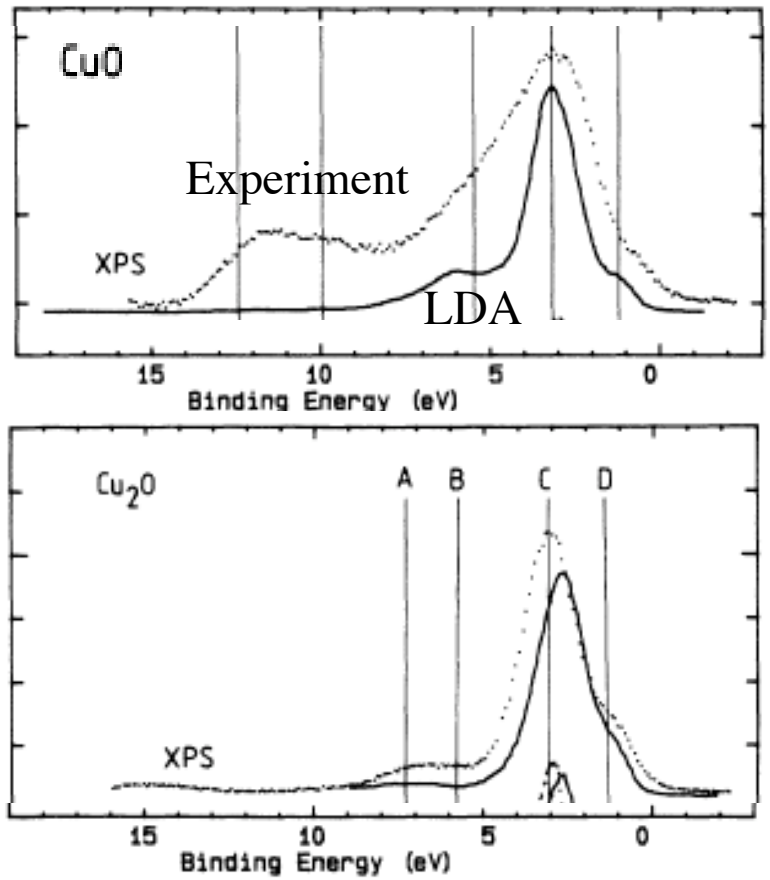
$|d^9\rangle + |d^{10}\underline{L}\rangle$

xPS $|d^9\underline{I}\rangle + |d^{10}\underline{L}\underline{I}\rangle$

↓

costs U_{pd}

Valence Bands



Intensity

Copper 2p core levels

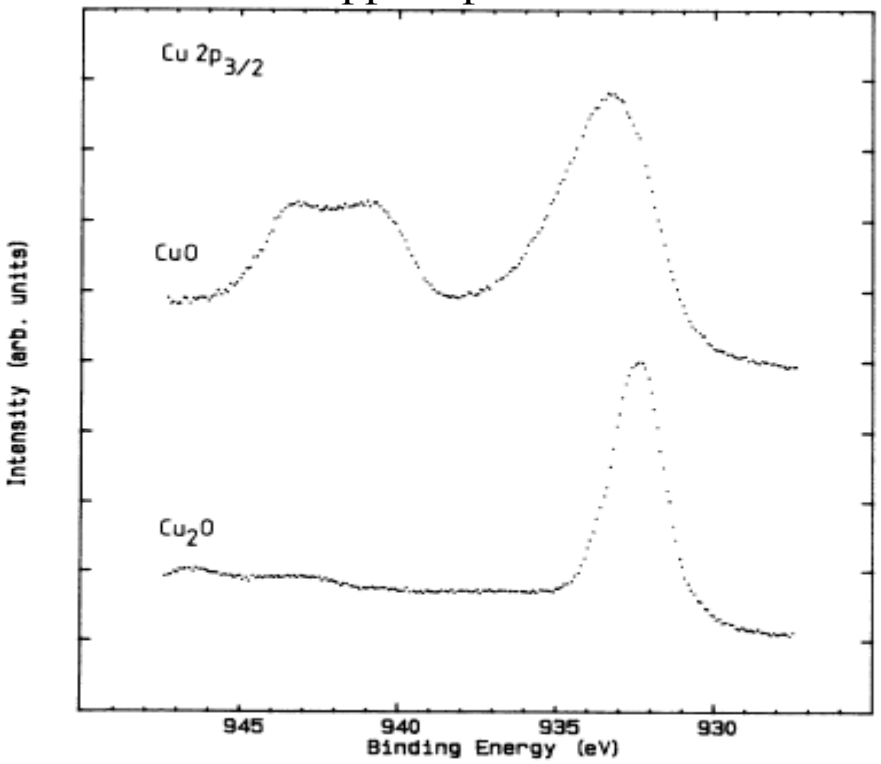
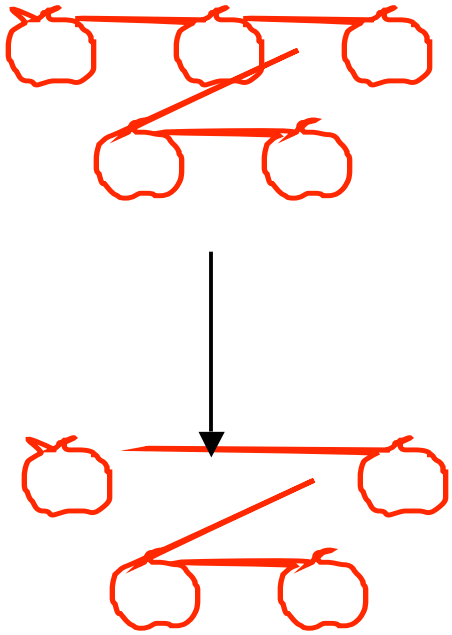


FIG. 2. Cu $2p_{3/2}$ spectra, normalized to the peak height. Bottom, Cu_2O; top, CuO.

Correlated vs. uncorrelated systems

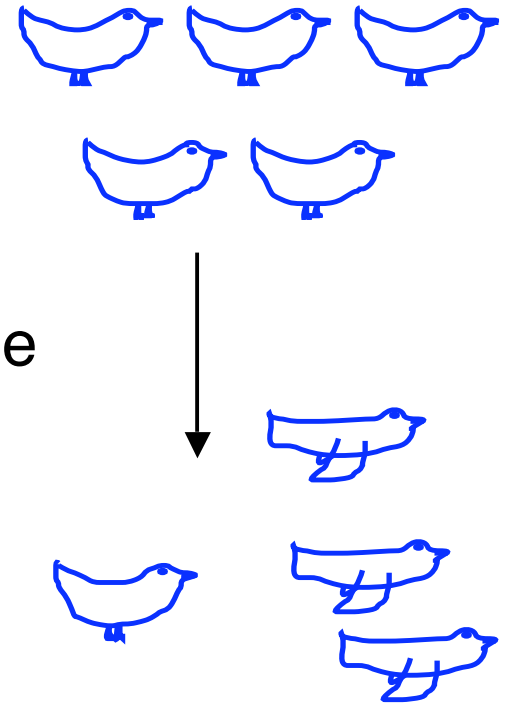
Static



Si, Ge, Cu, etc.
Band theory
Works even though 10^{23} electrons!

remove one

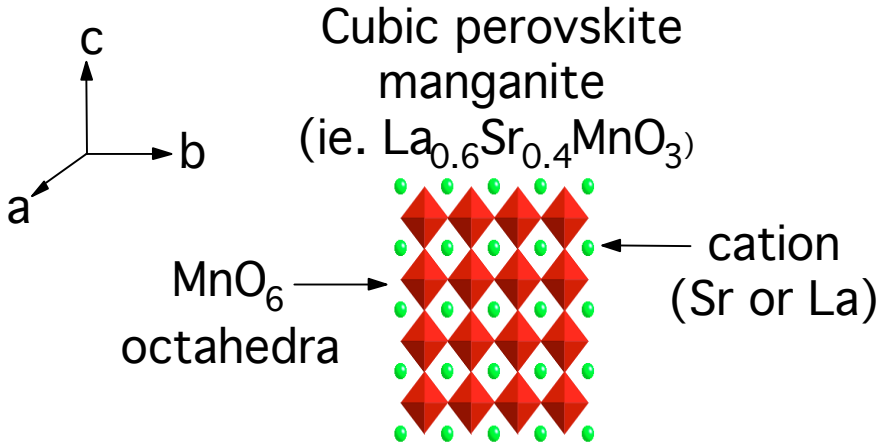
Dynamic



Correlated systems
Many-body theories

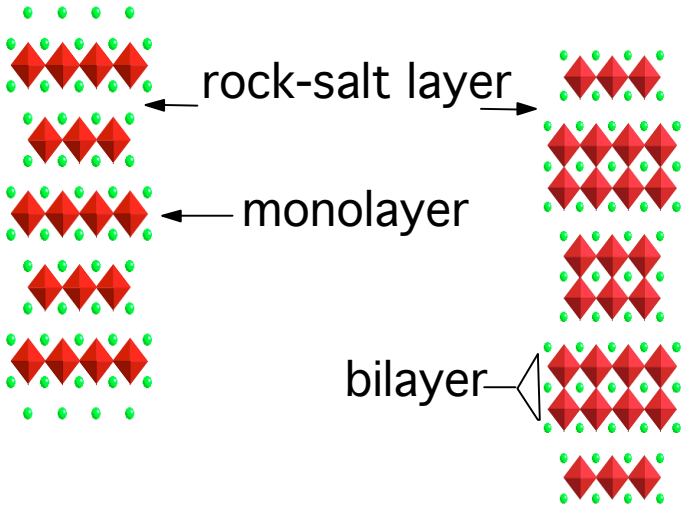
Dynamic interactions --> Novel and rich physics and materials systems.

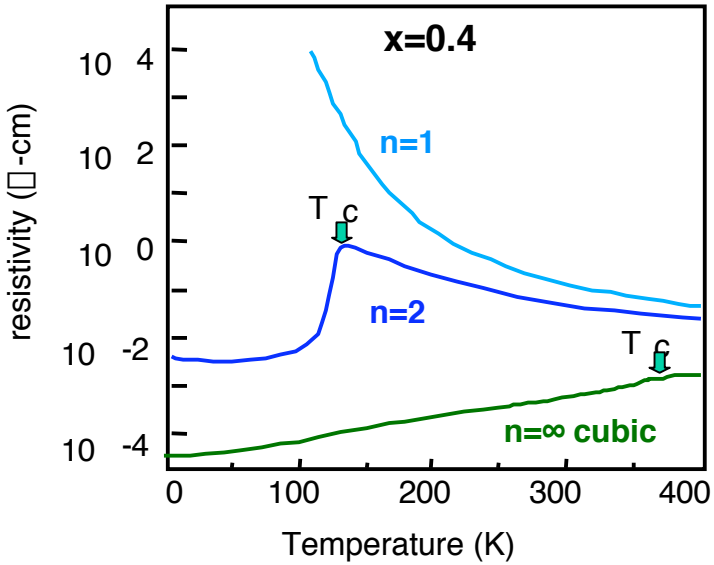
Crystal structure of the manganites



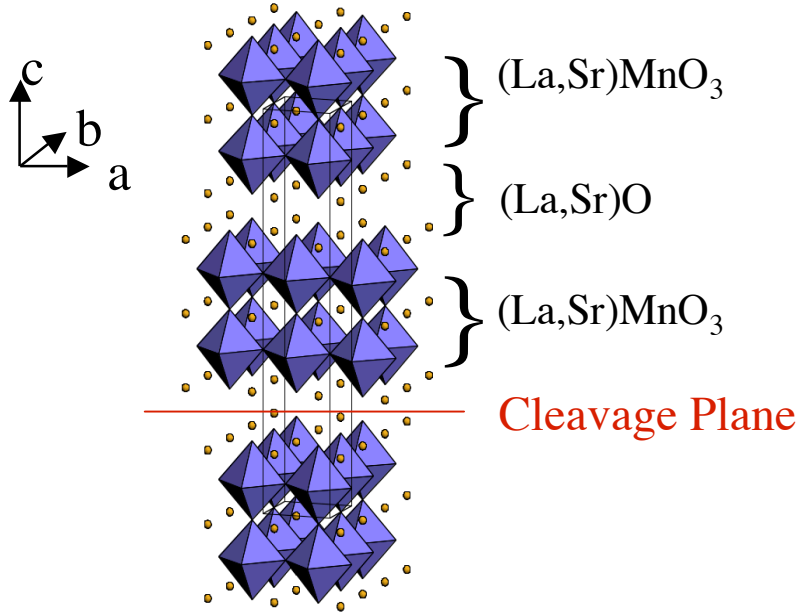
Monolayer perovskite Manganite (i.e. $La_{0.6}Sr_{1.4}Mn_2O_4$)

Bilayer perovskite manganite (ie. $La_{1.2}Sr_{1.8}Mn_2O_7$)

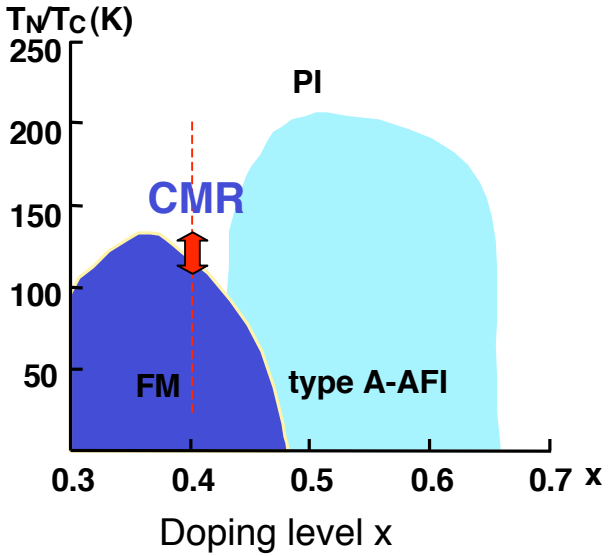




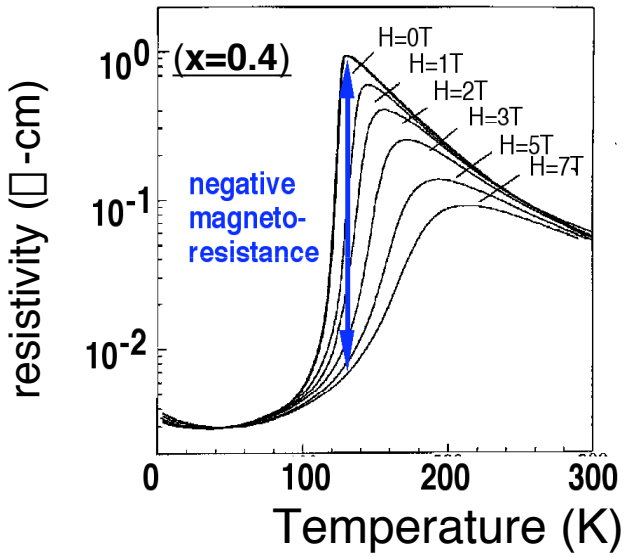
Bilayer manganite - $La_{2-2x}Sr_{1+2x}Mn_2O_7$



C.D. Ling et al.

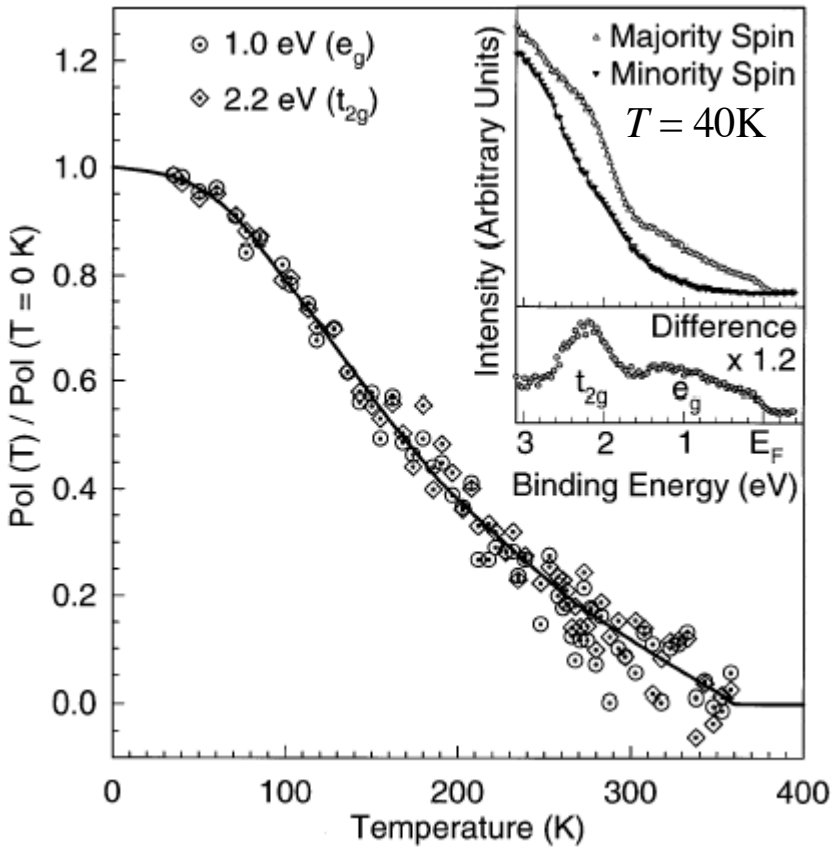


Y. Moritomo et al.



Evidence for half-metallicity

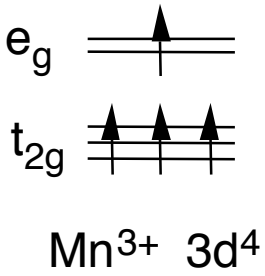
- Strong spin polarization in $La_{0.67}Sr_{0.33}MnO_3$ epi-films:

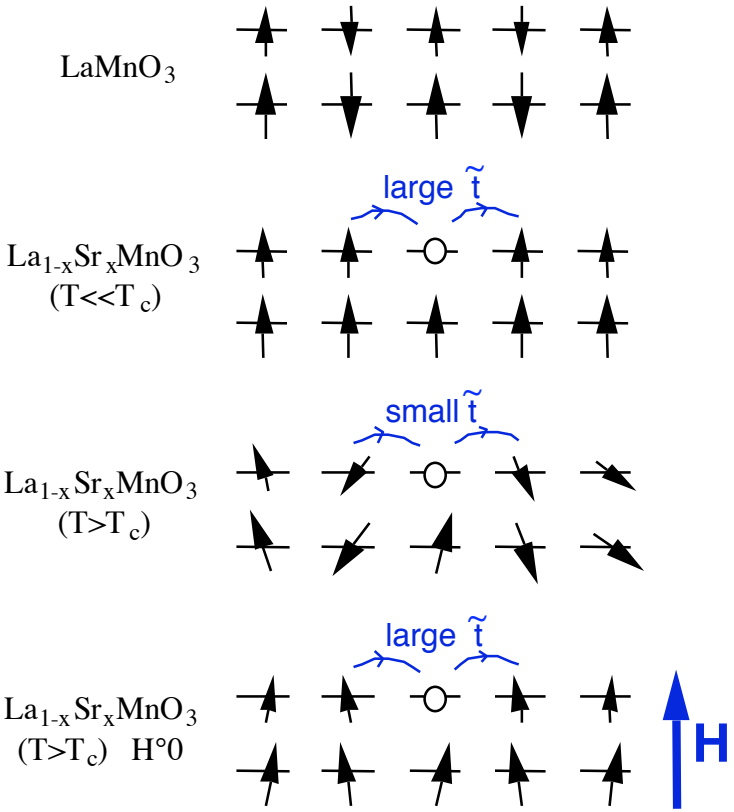


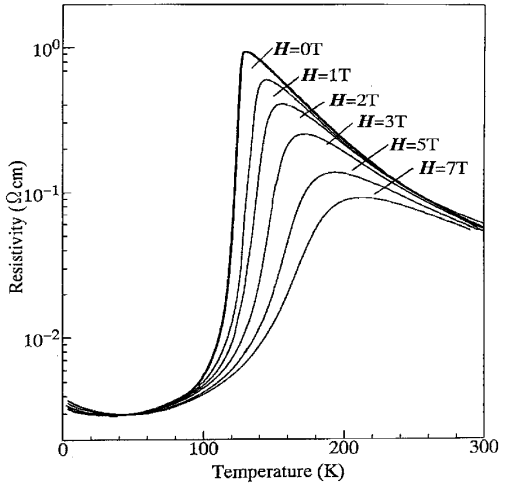
J.H. Park *et al.*, PRL 81, 1953 (1998), Nature 392, 794 (1998).

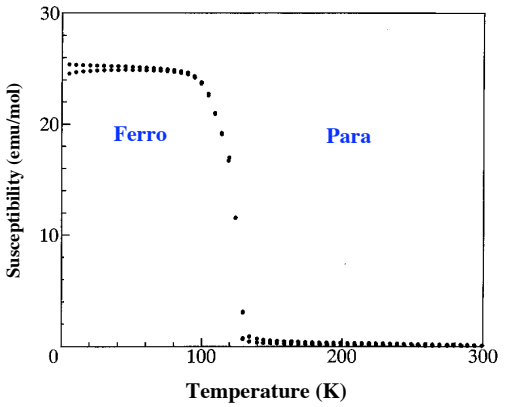
Double Exchange Theory

Kinetic energy gain for ferromagnetic alignment







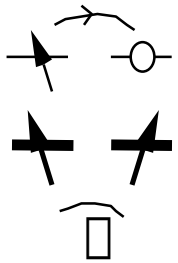


Ferromagnetism and Metallicity go together.

Double Exchange Theory

Kinetic energy gain for ferromagnetic alignment

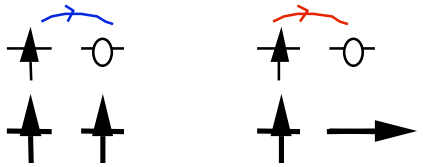
$\tilde{t} = t_o \cos(\theta/2)$



e_g electron
itinerant

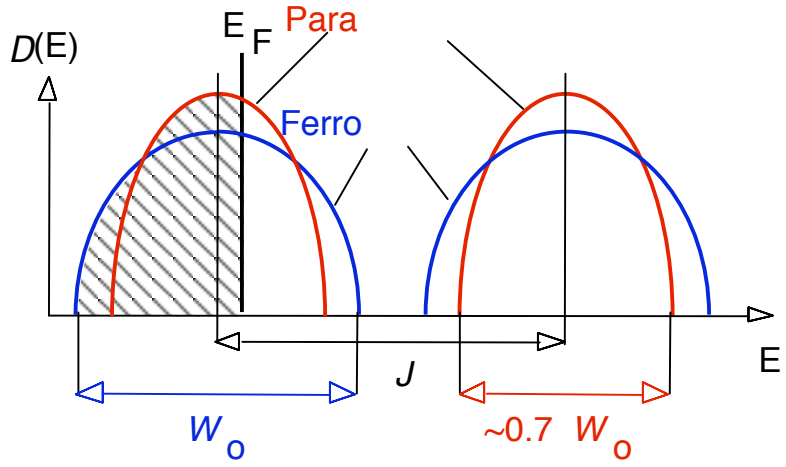
t_{2g} electrons
core-like
S=3/2

Ferro	Para
$\tilde{t} = t_o$	$\tilde{t} \sim .707\ t_o$



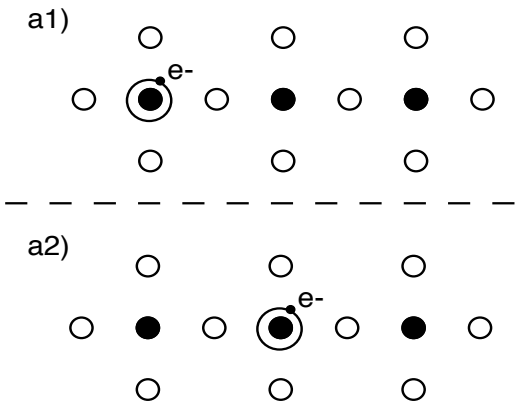
Change in conductivity going across Ferro-Para transition: ~ 30%
Real materials - many orders of magnitude effect! Why?

Change in t --> change in W
Can study with ARPES

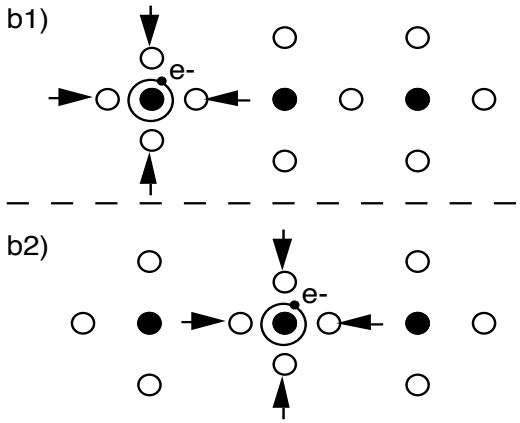


Polarons and conductivity

Transport without polarons

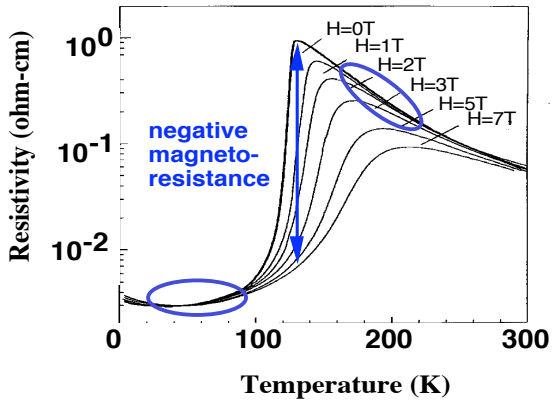


Transport with polarons



To help manganite problem, polaronic effect must be stronger above T_c than below T_c

Feedback effect necessary - Polaronic effect cooperates with Double-exchange and/or other phenomena (charge ordering).



Temperature dependent pseudogaps in CMR Oxides

ARPES experiments

Yi-De Chuang, Zhe Sun, Adam Gromko, Alexei Fedorov,
Fraser Douglas, Max Bunce, D.S.D.
University of Colorado

Samples

T. Kimura, Y. Tokura — *University of Tokyo*
John Mitchell — *Argonne Nat'l Labs*

Thanks to: X.J. Zhou, P. Bogdanov, Z. Hussain ALS Berkeley
K. Altmann, SRC Madison
Support from the DOE and NSF

Y.D. Chuang et al., Science **292**,1509 (2001)

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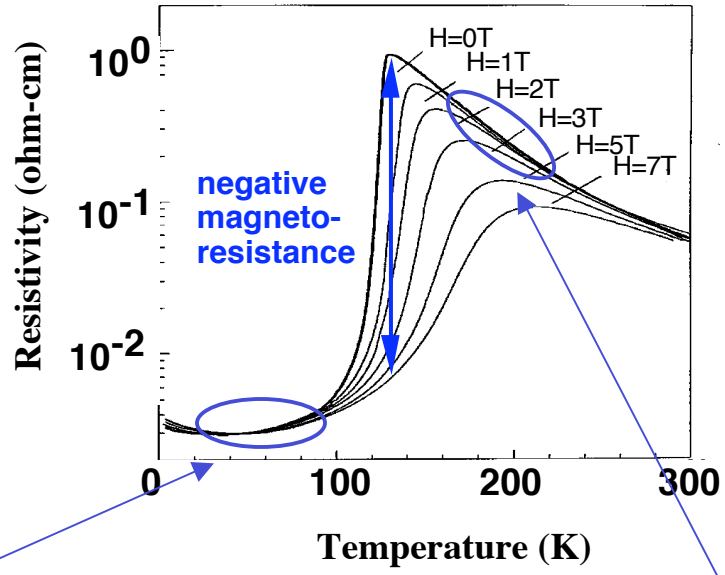
Questions

- How to explain very poor conductivity
  - In the "metallic" state of the manganites?
  - M-I transition into the high T insulating state? (--> CMR)

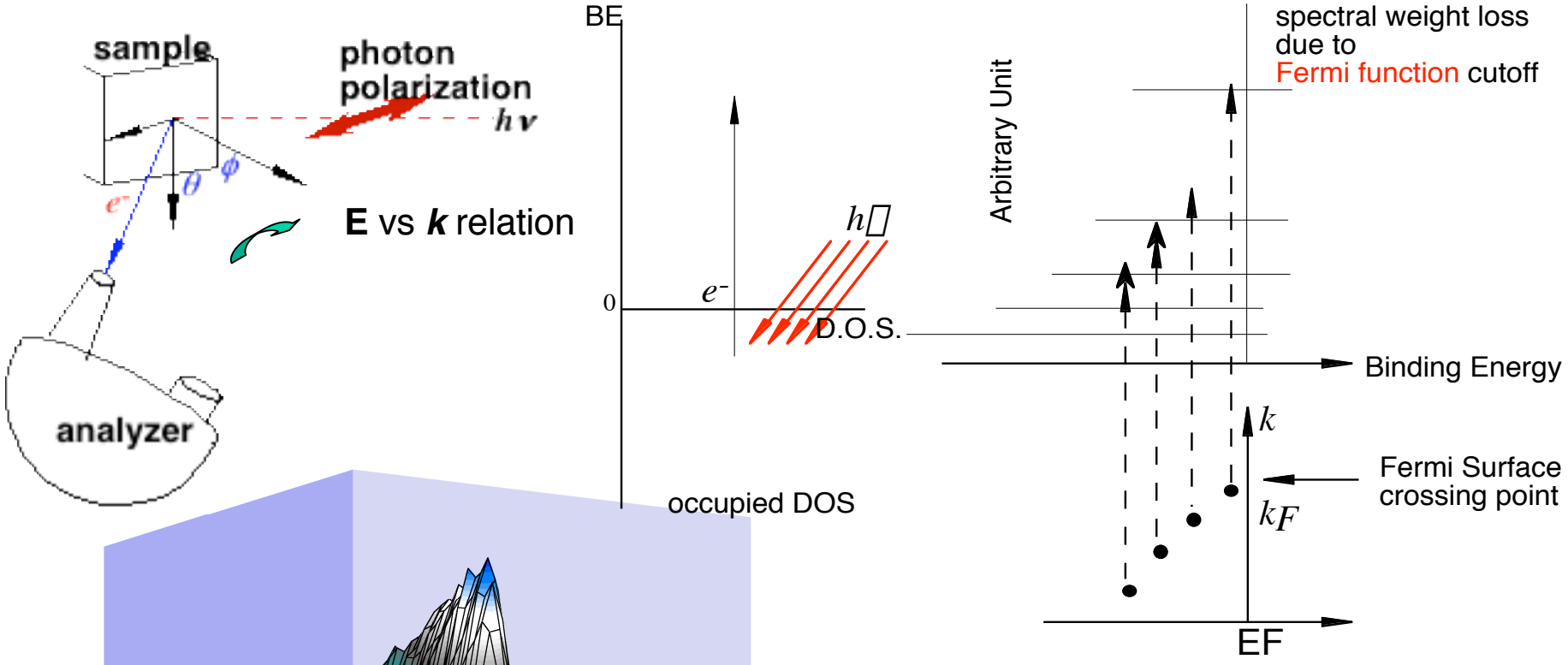


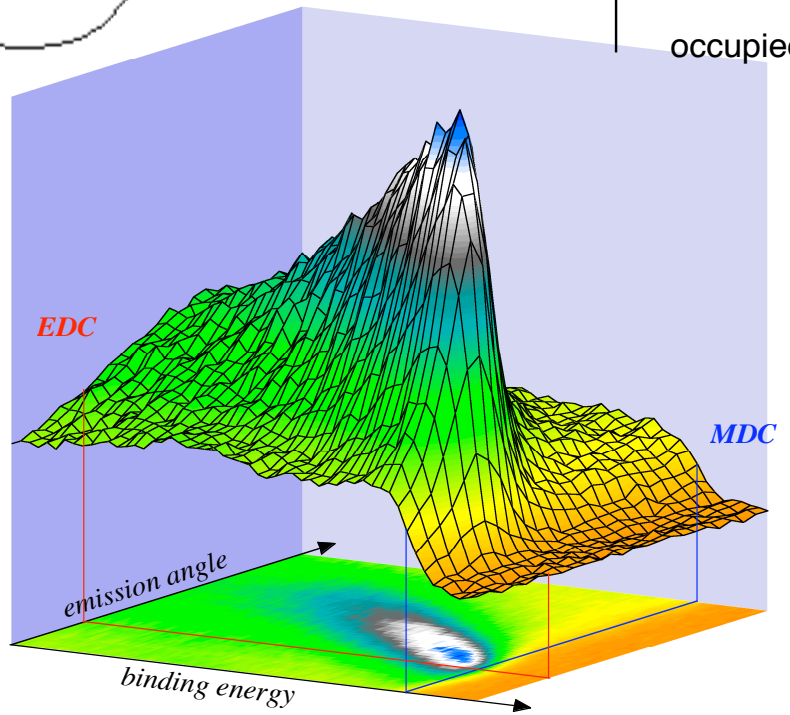
Very poor metal

Insulator

- Possible mechanisms.
  - •Polarons? Large scattering? Charge/orbital ordering?
  - •Temperature dependent pseudogap (includes some of above).

Angle Resolved Photoemission (ARPES)



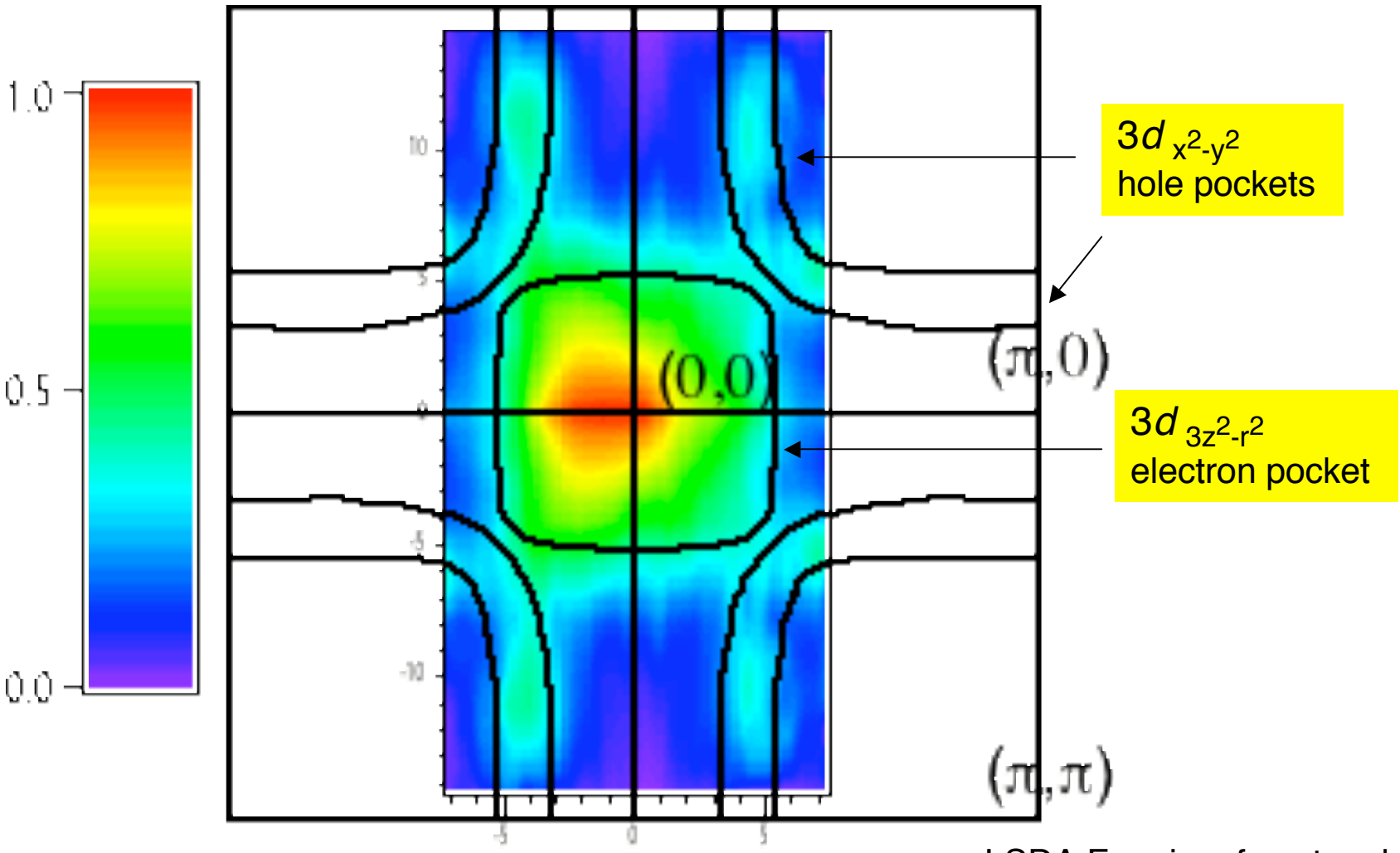


EDC: Energy Distribution Curve (ARPES intensity versus binding energy)

MDC: Momentum Distribution Curve (ARPES intensity versus momentum)

ARPES on $La_{1.2}Sr_{1.8}Mn_2O_7$ (n=2, x=0.4) (Low T Ferromagnetic state)

Integrated spectral weight over (0.2eV,-0.2eV) window



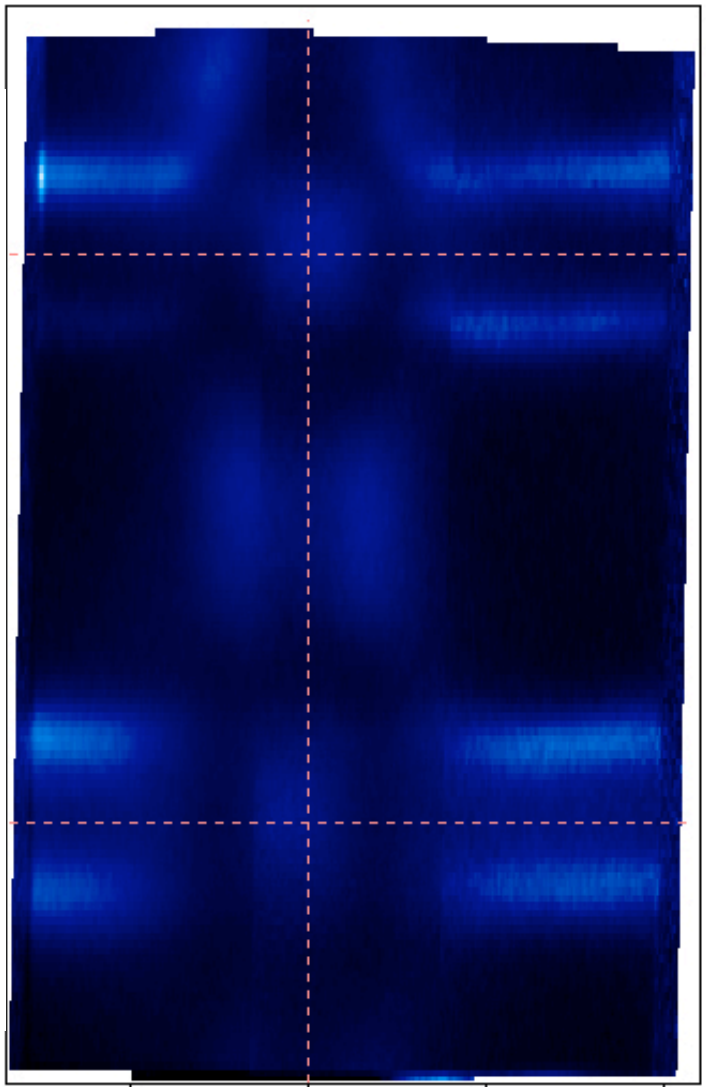
$3d_{x^2-y^2}$ hole pockets

$3d_{3z^2-r^2}$ electron pocket

LSDA Fermi surface topology
(*N. Hamada* unpublished)

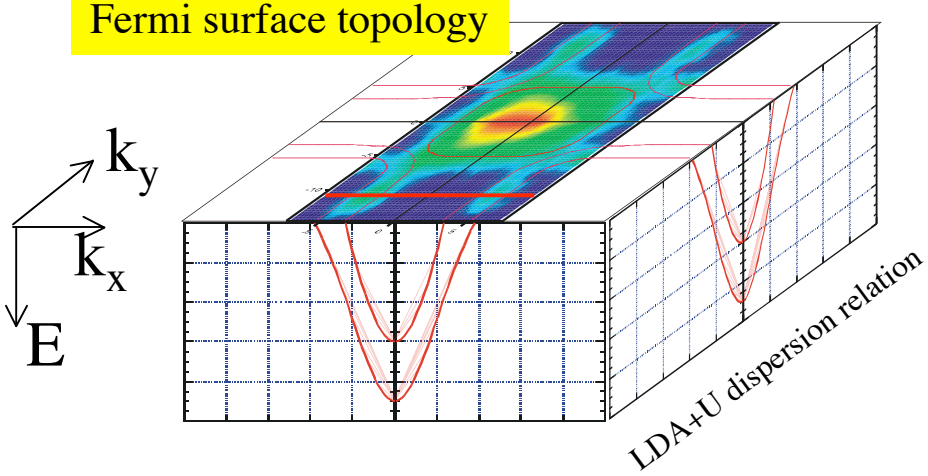
Y.D. Chuang et al., Science **292**, 1509 (2001)

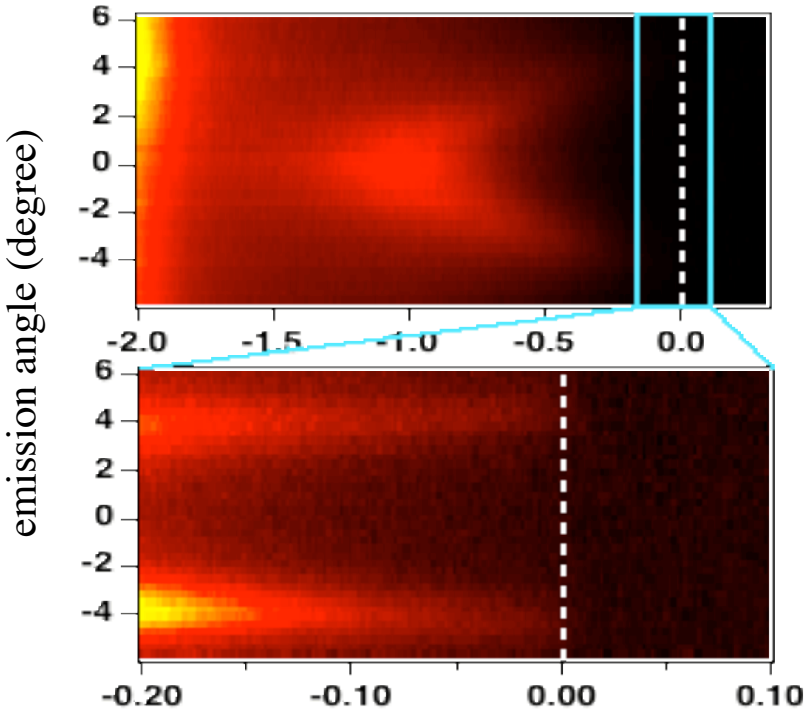
Angle-scanned (unsymmetrized) Fermi Surface
bilayer manganite x=0.4 (Low T Ferromagnetic state)



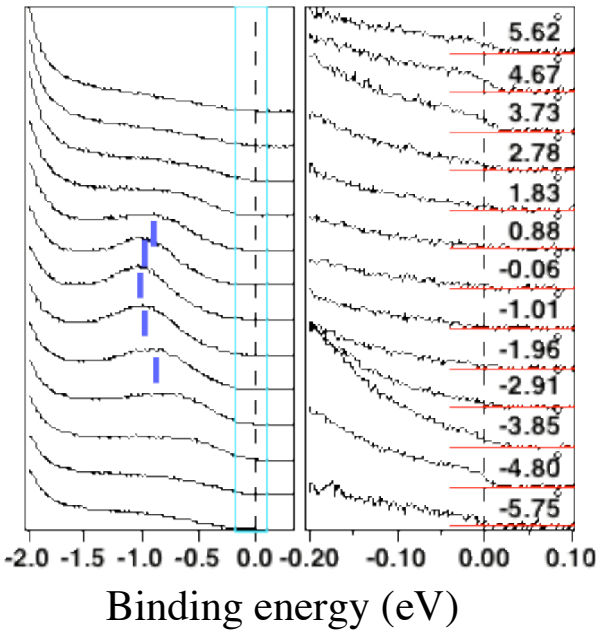
ARPES on bilayer x=0.4 samples - Low T (20K) ferro state

Fermi surface topology

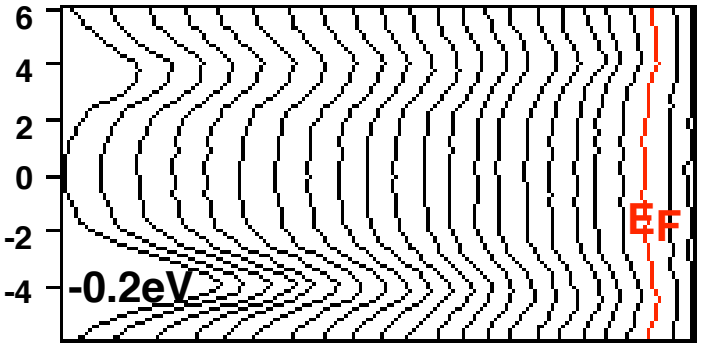




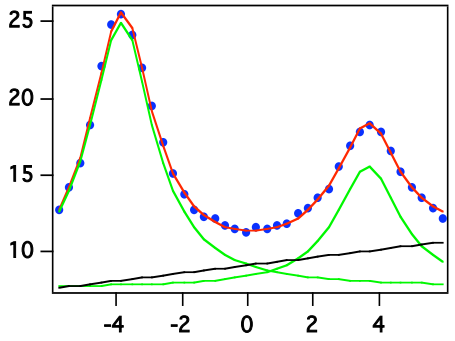
EDCs



MDCs



Double Lorentzian fit to MDCs



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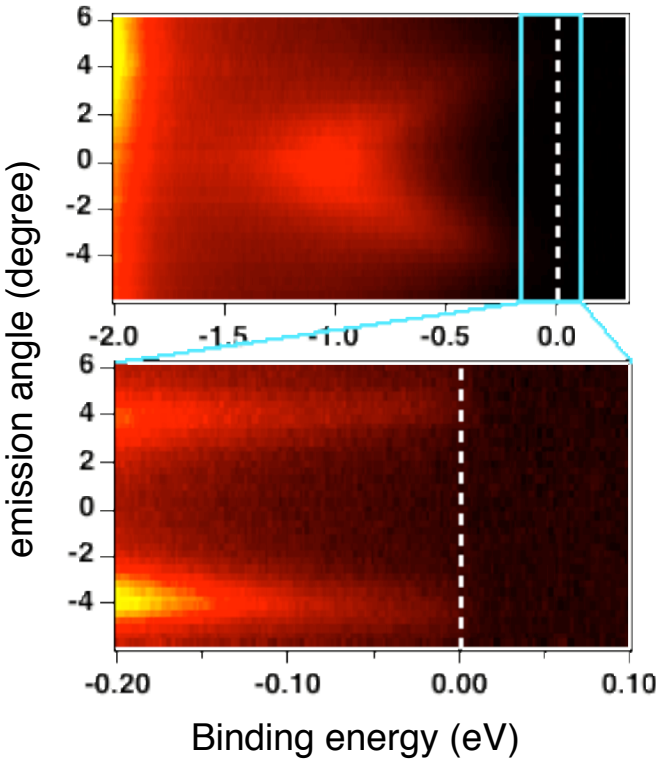

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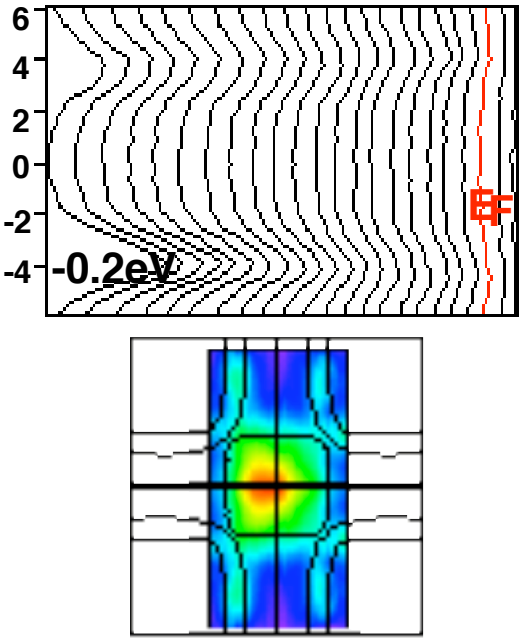

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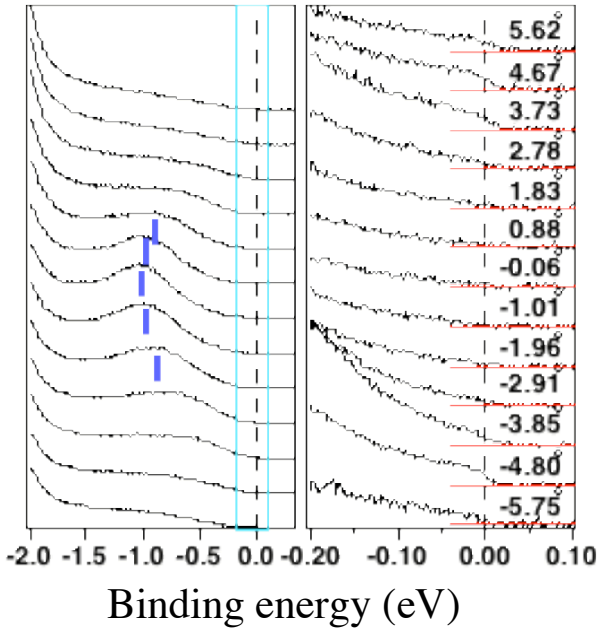
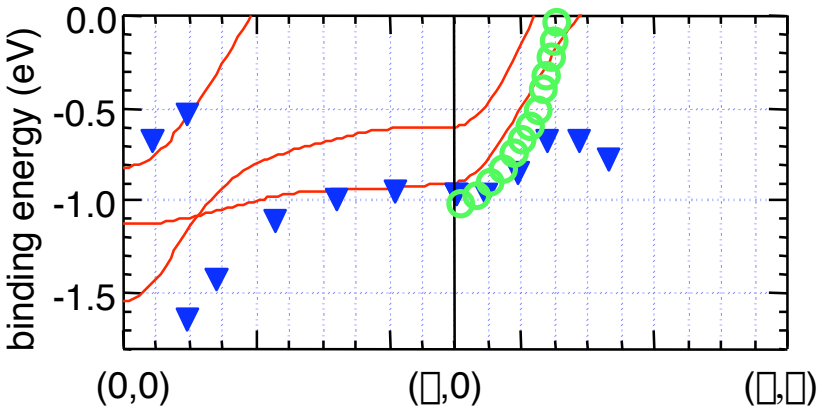
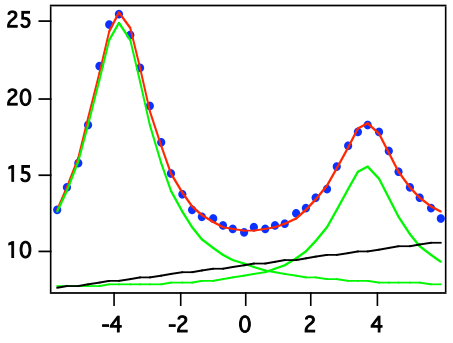




LDA dispersion
MDC centroid
EDC centroid

Double Lorentzian fit to MDCs



Double Exchange Theory

Kinetic energy gain for ferromagnetic alignment

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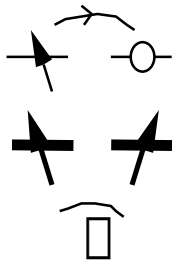

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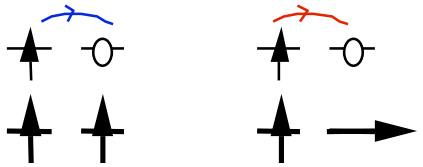

Change in conductivity going across Ferro-Para transition: ~ 30%
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Change in t --> change in W
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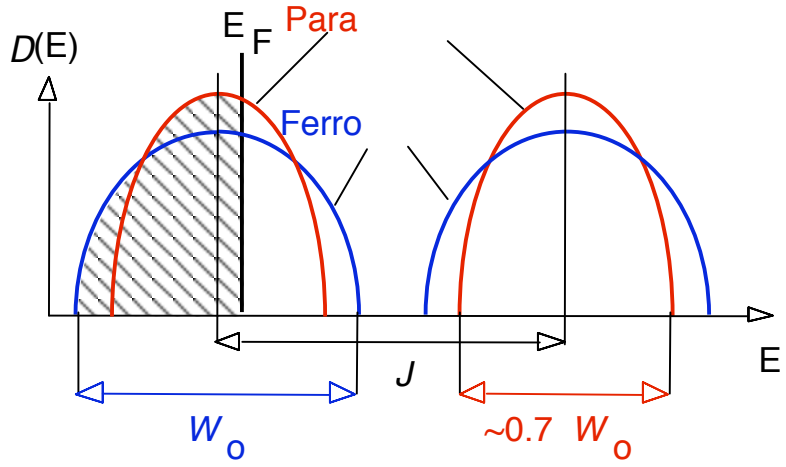
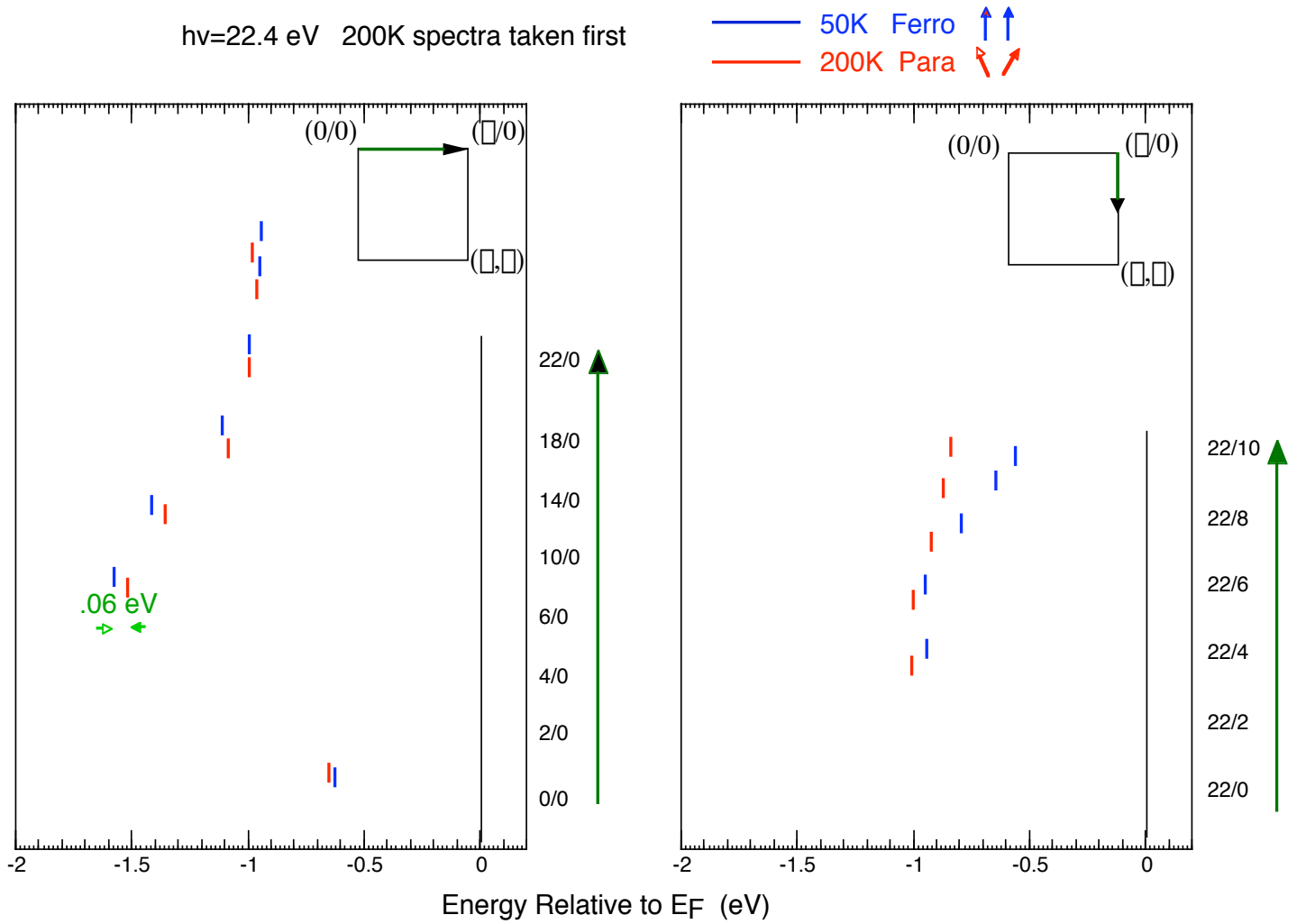


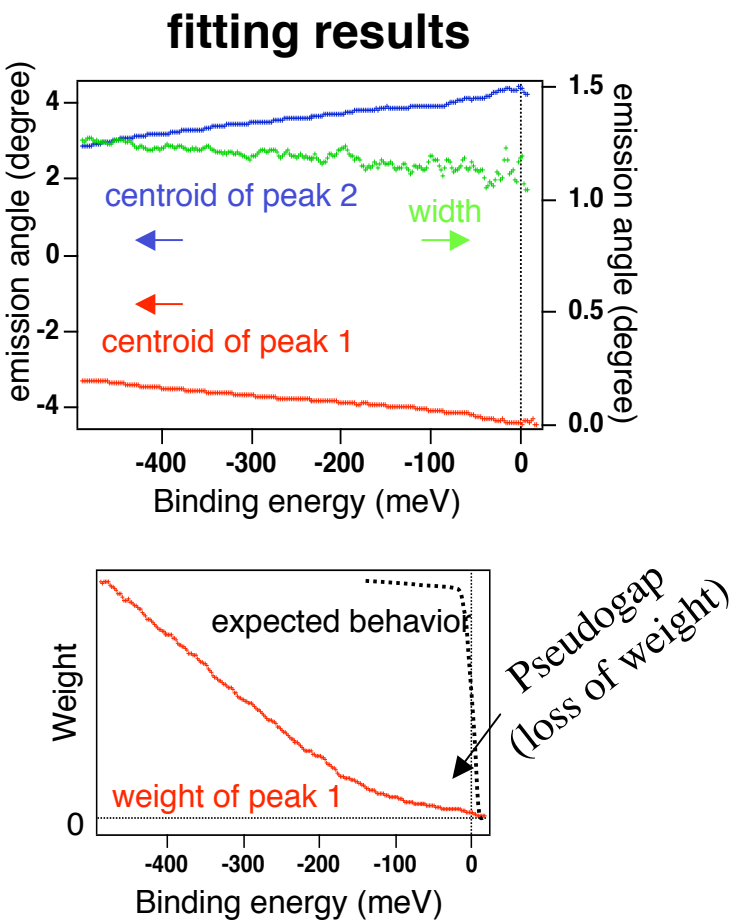
Temperature dependence of $(LaSr)Mn_2O_7$ x=.4 $T_c \sim 130K$



Bandwidth change : .06 eV/1.5 eV = 4%. Much less than the DE prediction of 30%.
==> DE relevant but not key effect.

T. Saitoh et al., PRB (2000)

Extraction of the key transport parameters - low T ferrometal state



1)Fermi velocity (from band dispersion near E_F)
$v_F \sim 0.038\ c$

2)Band mass (fitting dispersion with parabola)
$m \sim 0.27\ m_e$

3)Number of carriers (measurement of the FS volume)
$n \sim 3.4*10^{21}$ holes/cm^3

4)Momentum width (HWHM)
$\Delta k \sim 1.2^o \sim 0.09\ \pi/a \sim 0.07$ A^{-1} (original)

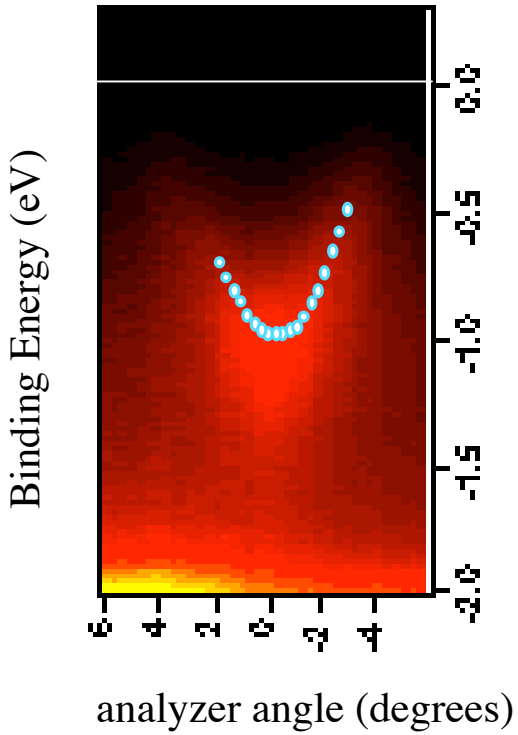
4b)Mean free path (lower limit)
$\Lambda = 1/\Delta k \sim 14$ A ~7 times the Mn-O bond length

4c) Mean free time between scattering (lower limit)
$\tau = \Lambda / v_F \sim 1.24$ fs

$\rho_{ARPES} = 1/\sigma = m^* / n e^2 \tau \sim 1.3*10^{-4}\ \Omega$-cm (Drude)
Real value $\rho_0 \sim 2*10^{-3}\ \Omega$-cm (more resistive)

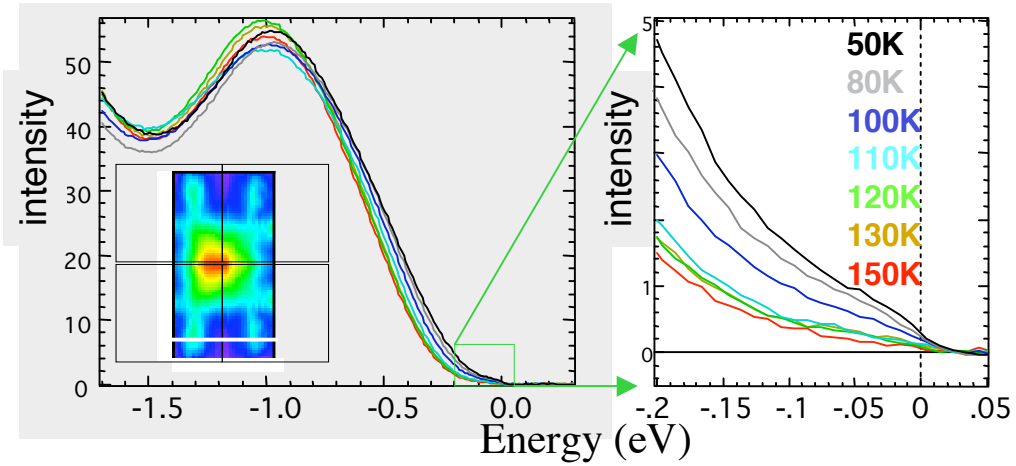
Key ingredient not considered in the Drude calculation: **pseudogap.** Presence of the pseudogap can effectively remove carriers from conduction process thus increasing the resistivity

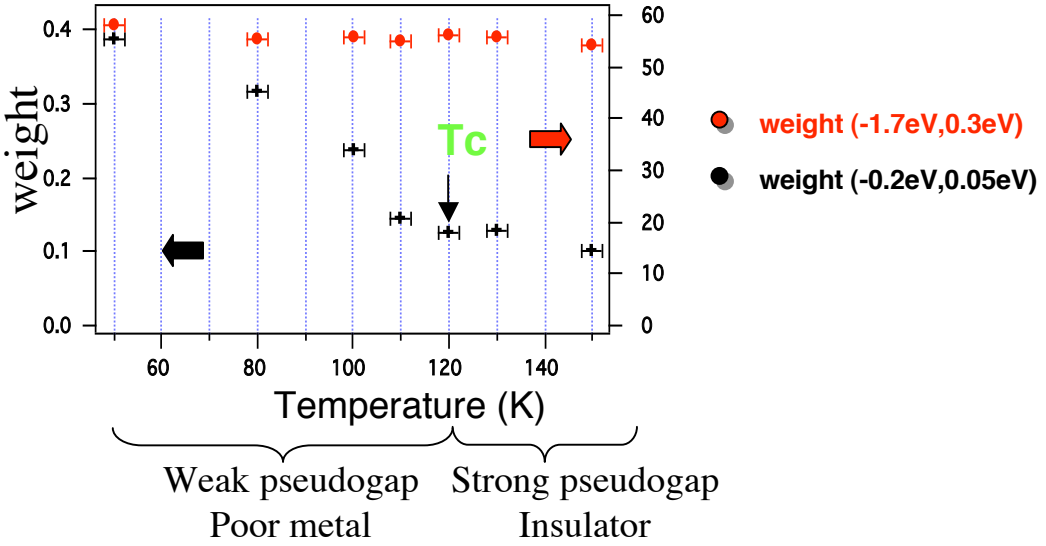
Temperature dependence of the pseudogap



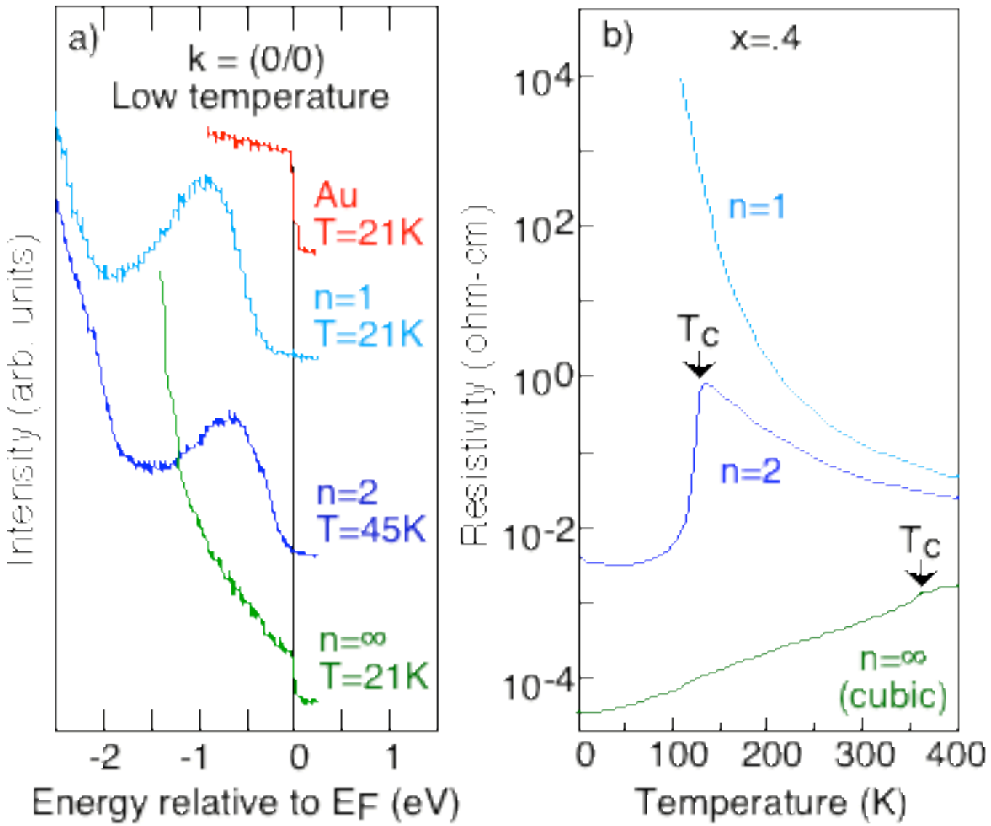
Pseudogap correlates with and may drive the M-I transition.

Energy spectra averaged across the entire slice



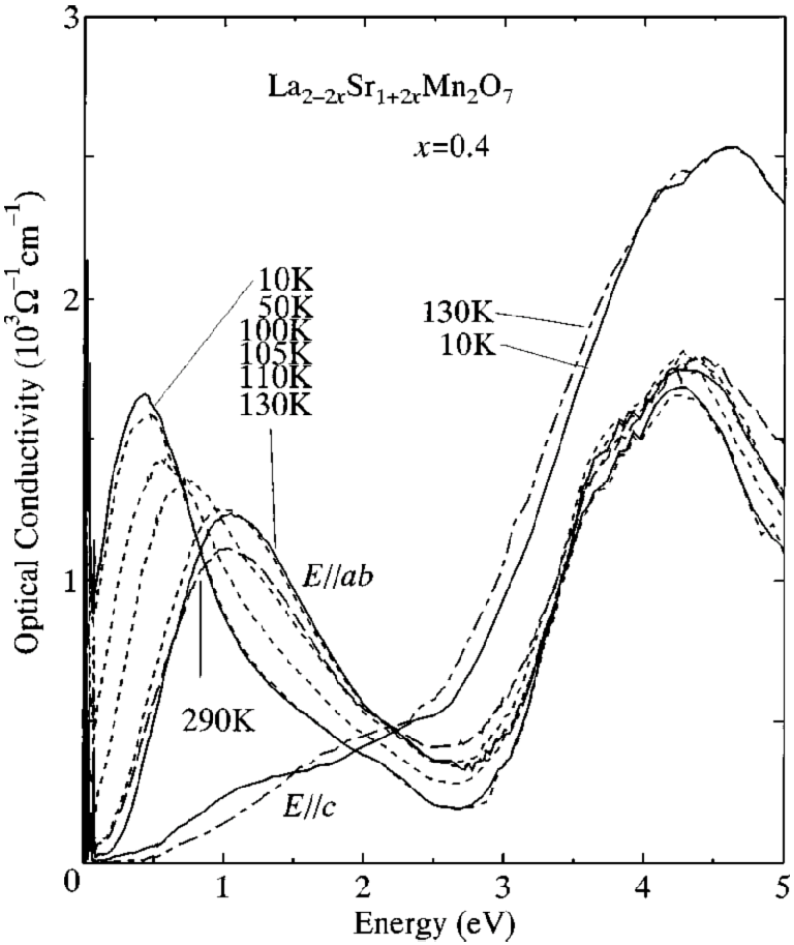


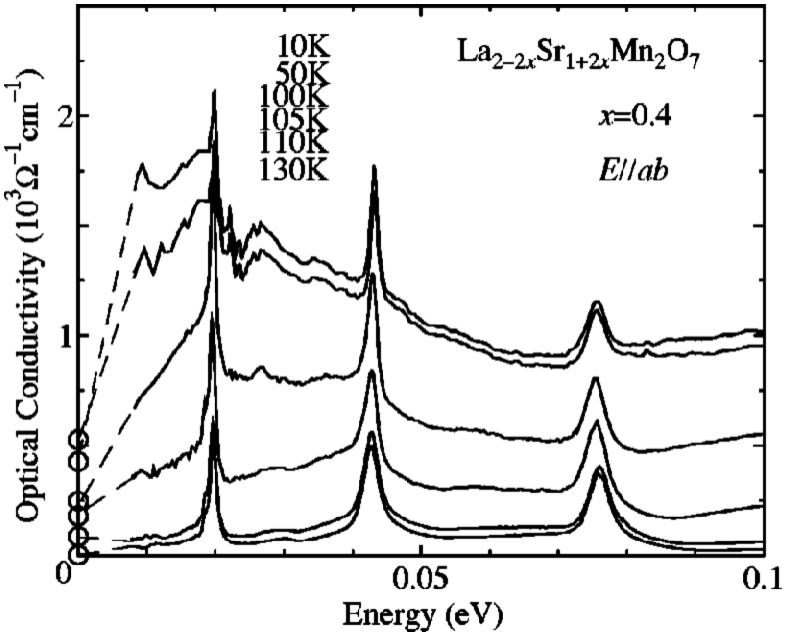
Dimensionality dependence of near-E_F weight



The pseudogap affects all families of the manganites - some very strongly (single layer) and some less so (pseudocubic). Resistivity varies in kind.

Optical Conductivity of layered manganite

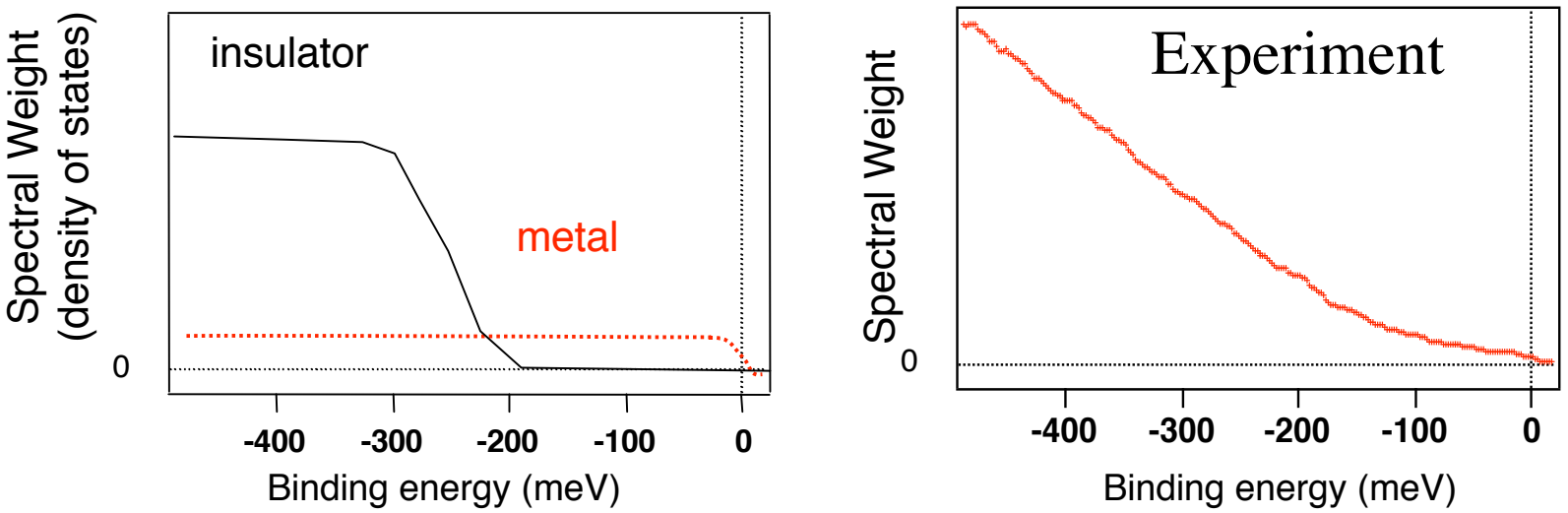




- Spectral weight transfer with temperature over large energy scale
- Gapped low energy spectral weight (no Drude peak)

T. Ishikawa et al., Phys. Rev B 57, R8079 (1998)

Loss of near-E_F weight (pseudogap)
Simple superposition of metallic and insulating regimes?



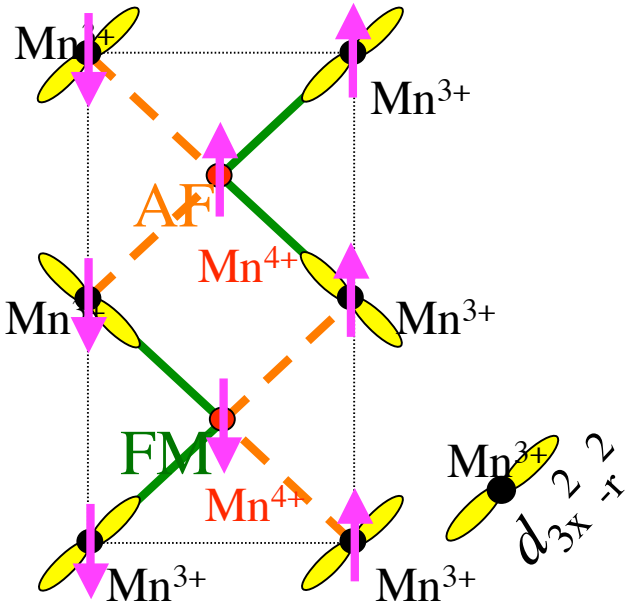
(Need many many types of regions, all with different size gaps)
- May consider fluctuations in space and time to get this.

Also, FS volume matches total # carriers expected by chemical doping (no regions with extra # holes, regions with fewer # holes).

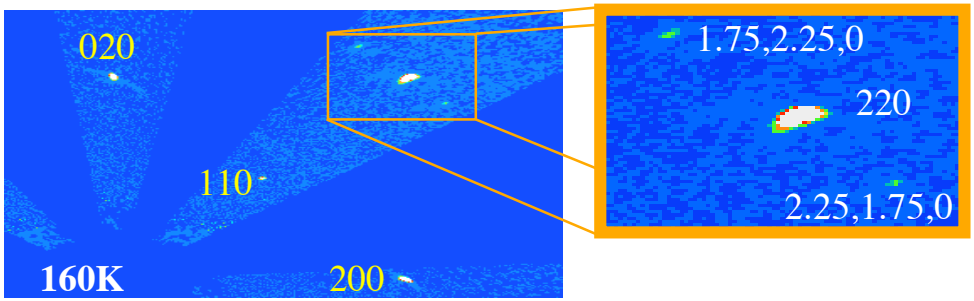
Real-space Charge Density Waves (CDW's) and CDW gaps

Example: $LaSr_2Mn_2O_7$ (x=0.5) commensurately doped insulator

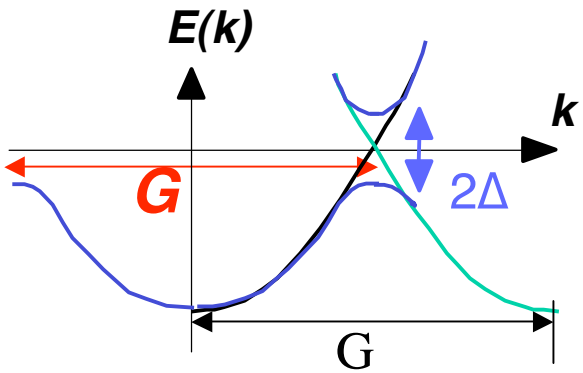
Commensurate/static "CE" ordering



Extra periodicity induced by CDW observable in diffraction experiments as weak superlattice spots at (1/4,1/4,0).



D.Argyriou et al., Argonne/APS

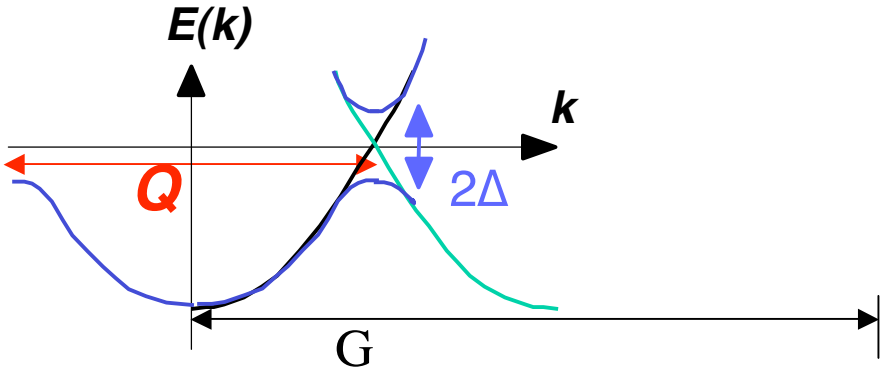


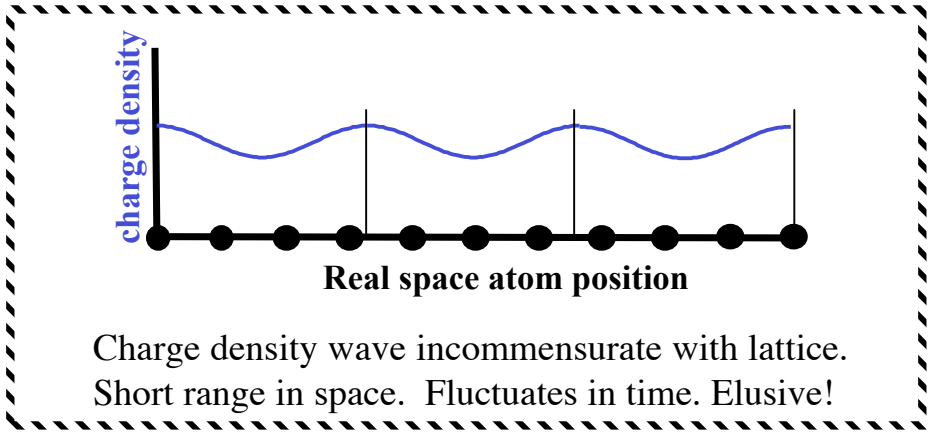
System gains energy when gap is centered at E_F (commensurate doping levels).

k-space driven CDW's and Fermi Surface nesting

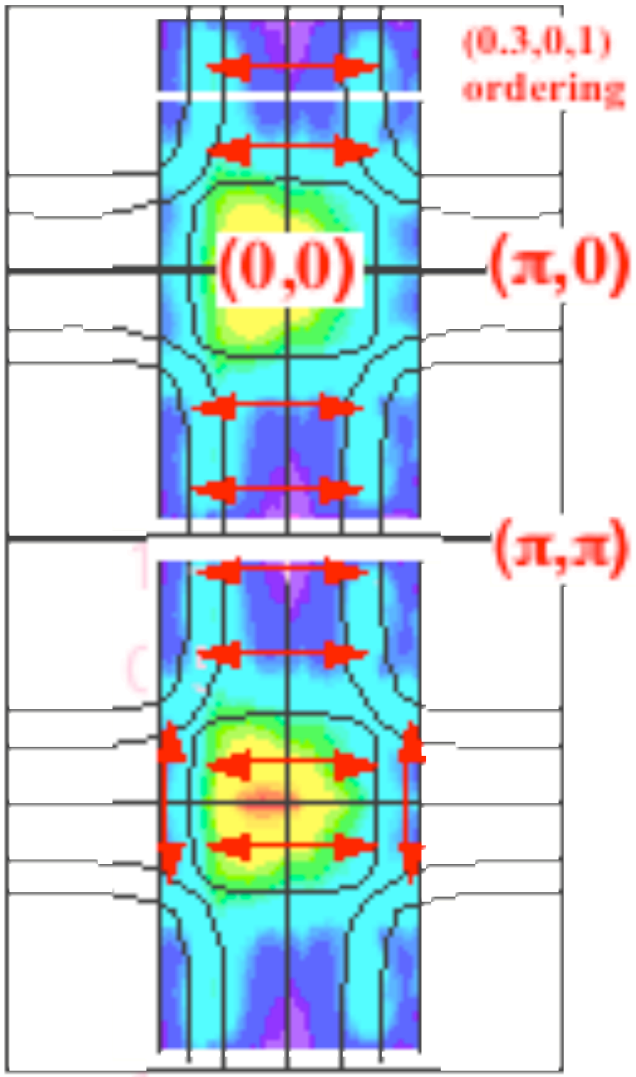
We can guarantee that CDW gap is centered at E_F in a k-space driven CDW --> gain extra energy.

If many parts of the Fermi Surface connect (nesting condition) the instability is greatly enhanced.





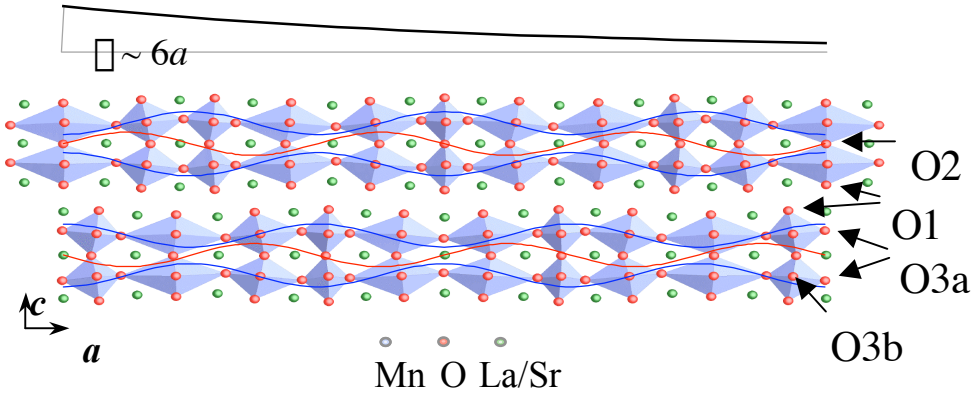
Charge density wave incommensurate with lattice.
Short range in space. Fluctuates in time. Elusive!



Entire measured FS of $La_{1.2}Sr_{1.8}Mn_2O_7$ is gapped due to large parallel segments (nearly perfect nesting).

Real space picture of the k-space driven CDW fluctuations

From an analysis of the intensity of 108 superlattice reflections (only observed in the high temperature paramagnetic state)

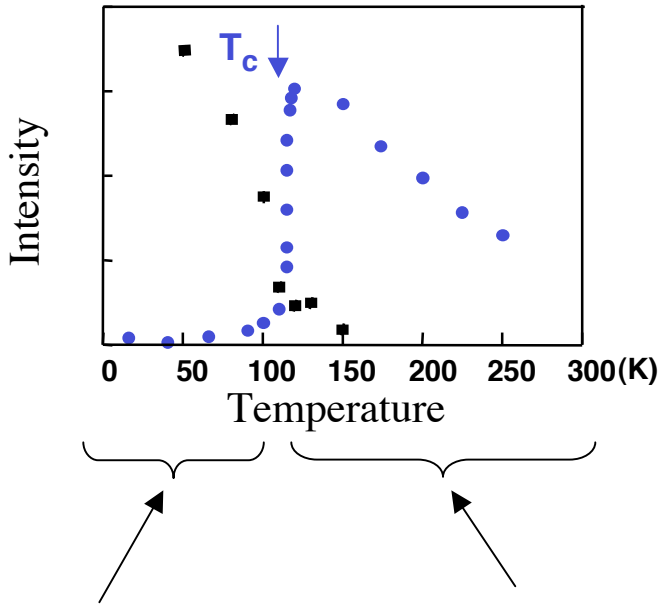


- Fermi-Surface-driven CDW cooperates with the Jahn-Teller effect to distort MnO_6 octahedra. --> increased energy scale of gap.
- Elastic strain mediates the correlations.
- Incommensurability with the lattice --> order is short ranged in real space

B. Campbell et al, (Phys. Rev. B 2001)

Temperature Dependence

ARPES Weight at E_F | Diffraction I(0.3,0,1)

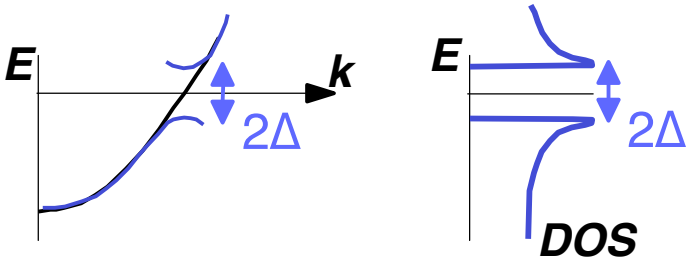


Weak Pseudogap
Poor metal
Fluctuating CDW
(not observed in diffraction!)

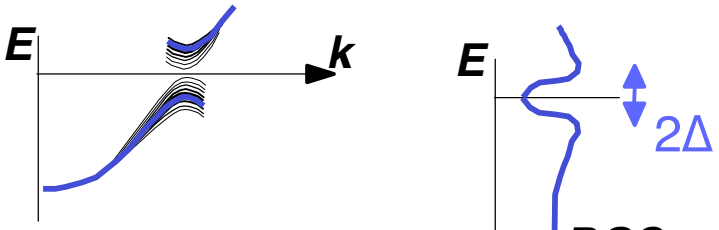
Strong Pseudogap
Insulator
Semi-static CDW

Sharpness of the energy gap

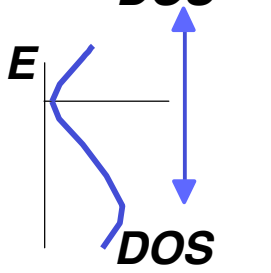
CDW with long range order



With short range order

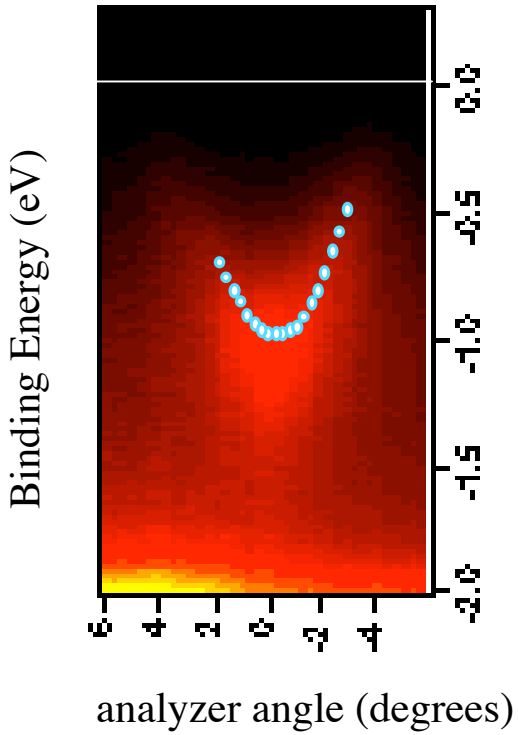


Including polaronic (Jahn-Teller) energy scale

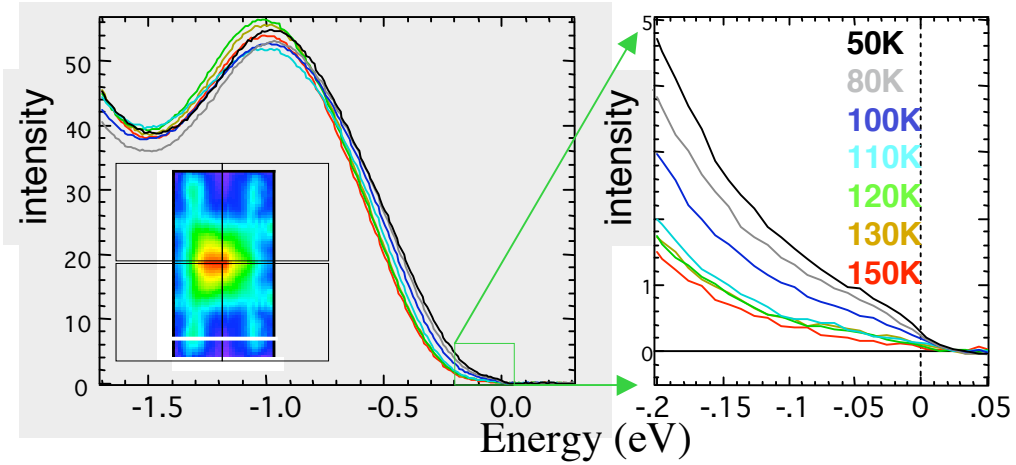


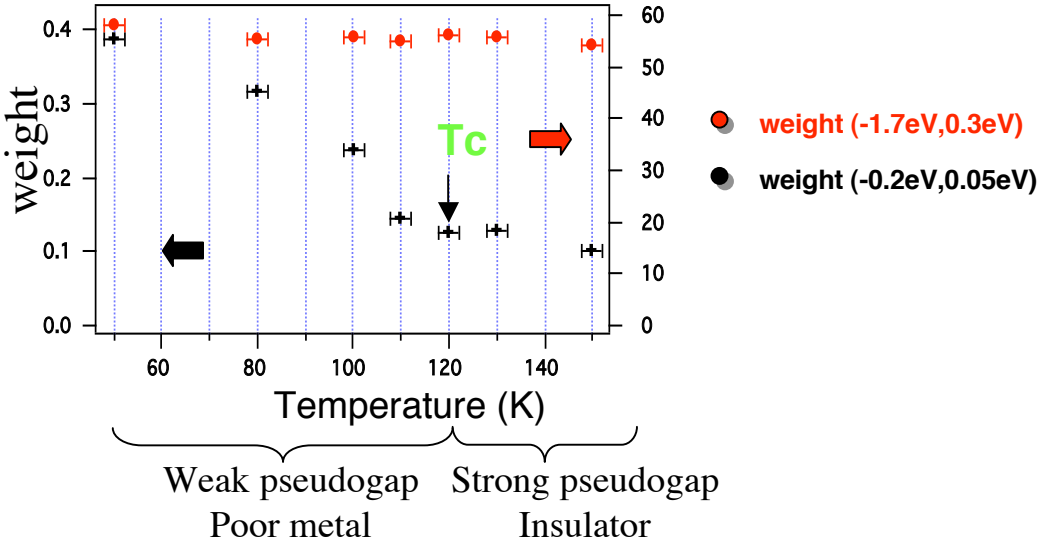
Looks like lower figure even in low T ferromag state!
Polarons and/or charge order at all Temps (with varying strengths).

Temperature dependence of the pseudogap



Energy spectra averaged across the entire slice





Pseudogap correlates with and may drive the M-I transition.

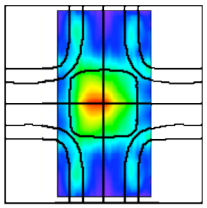
Competition and cooperation

***k*-space**

electronic

ARPES

Fermi surface topology

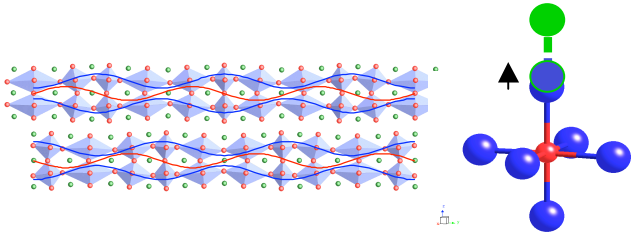


(0.3,0,1) orbital stripe (*k*-space)

CDW (short range)

***r*-space**

XRD,EXAFS



charge/orbital modulation *charge* *orbital*

+

Jahn-Teller distortions of various magnitude *lattice*

pseudogap

CMR effect

Double Exchange *spin*