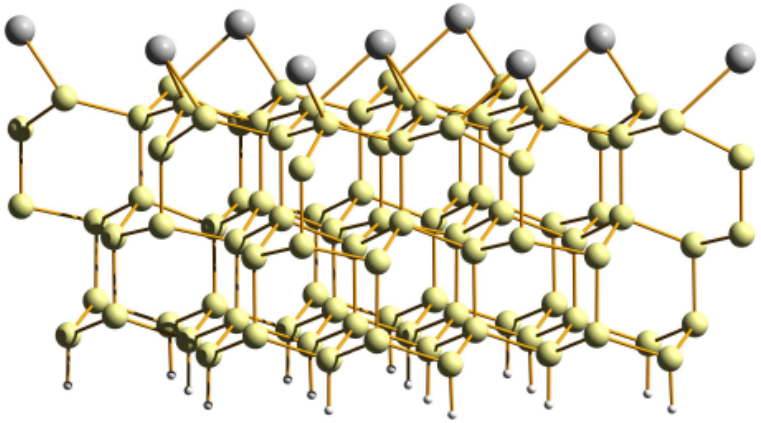
What about U in surface adatom systems?
- Interplay of charge and spin degrees of freedom

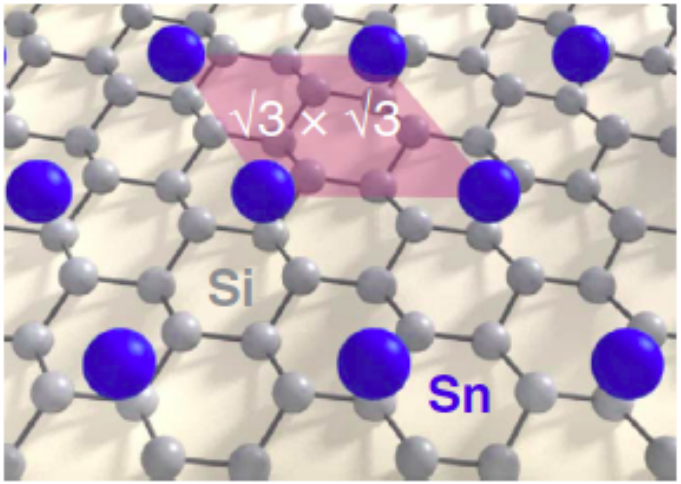
Yusuke Nomura

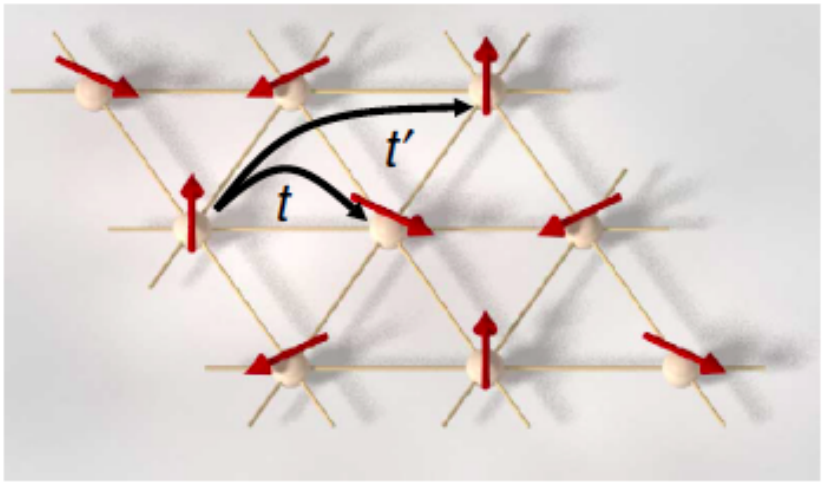
University of Tokyo (from September)
Before: École Polytechnique

Collaborators: P. Hansmann and S. Biermann

X:Si(111) with X=C, Si, Sn, Pb : Structure



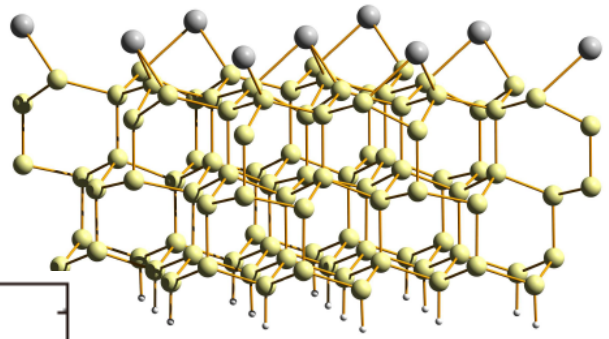


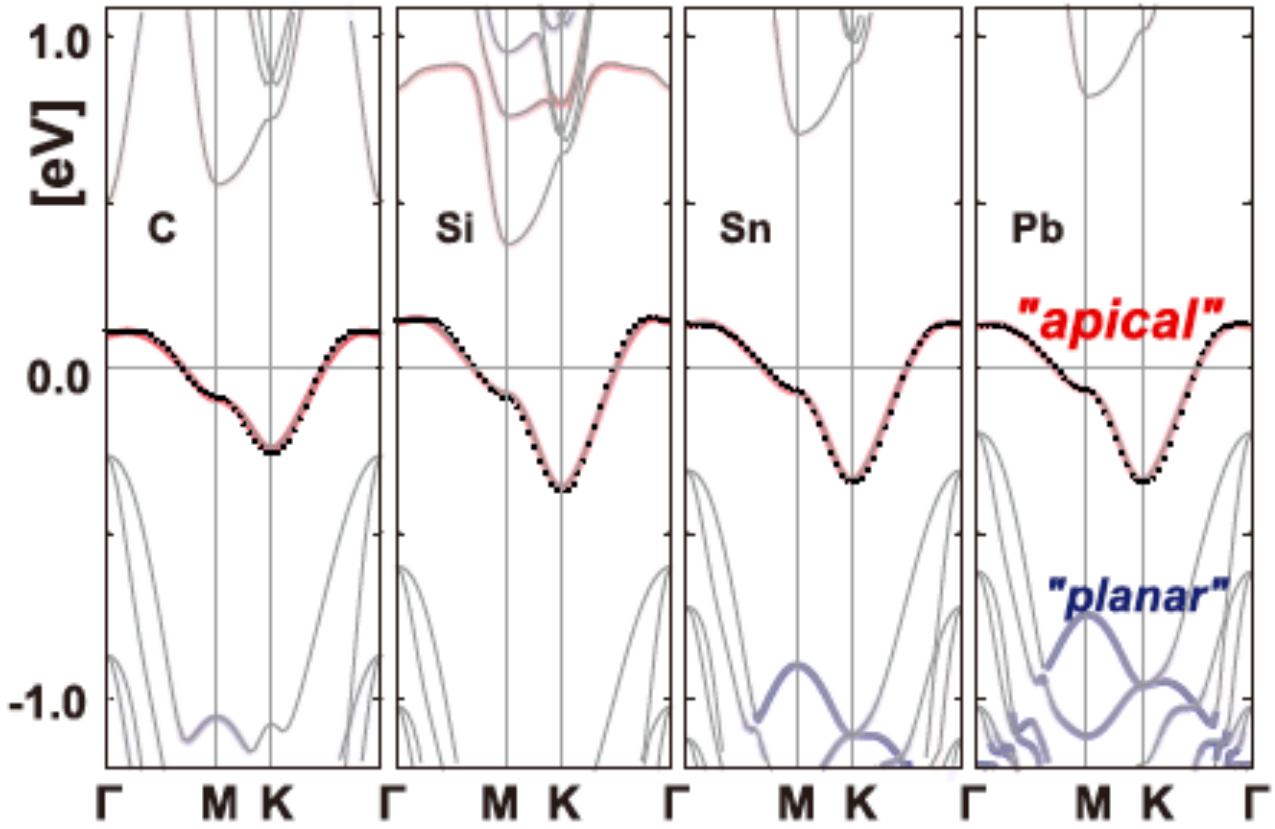


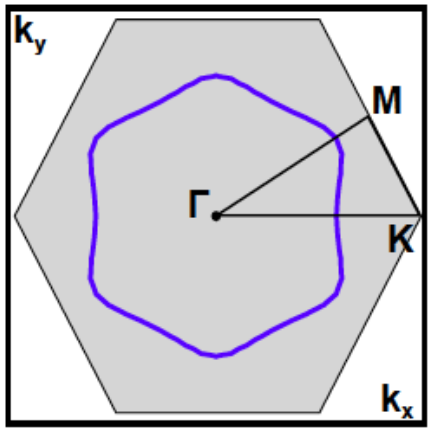
G. Li *et al.*, Nat Commun **4**, 1620 (2013)

- ✓ Si(111) surface is unstable by itself. Surface states are stabilized by adatoms
- ✓ {C, Si, Sn, Pb}: Si(111)
- ✓ Monolayer of adatoms forms triangular lattice

X:Si(111) with X=C, Si, Sn, Pb: Band structure







P. Hansmann et al.,
JPCM **25** 094005 (2013)

- ✓ Well represented by single band (half-filling)
- ✓ Narrow band (W ~ 0.5 eV)

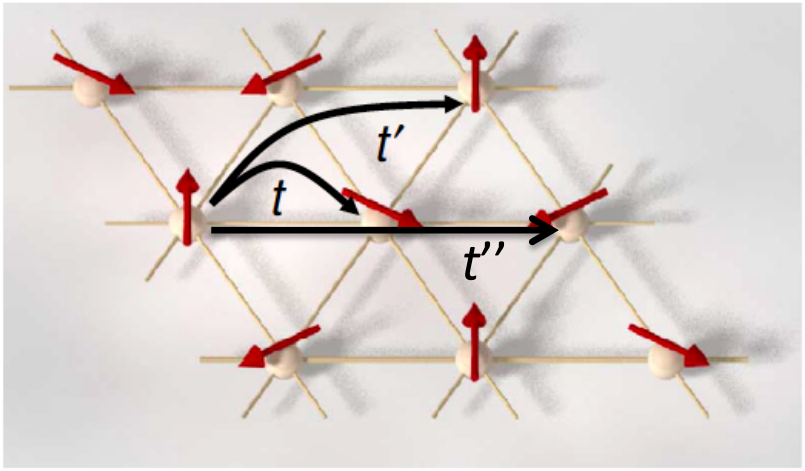
→ Ideal play ground to study correlation effect, geometrical frustration, etc…

E. Tosatti and P.W. Anderson, Jpn. J. Appl. Phys., 381 (1974)

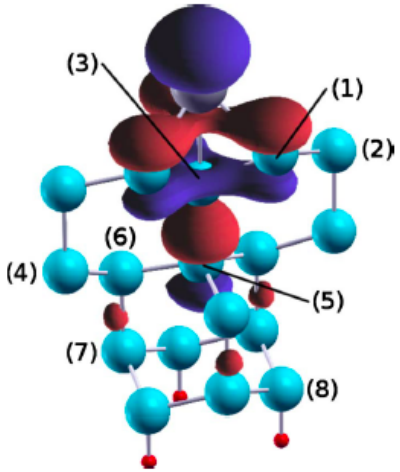
Hopping structure

P. Hansmann et al., JPCM **25** 094005 (2013)
P. Hansmann et al., PRL **110**, 166401 (2013)

		C	Si	Sn	Pb	
Nearest neighbor	t	38.0	50.0	42.0	42.0	[meV]
2nd nearest neighbor	$-t'$	15.0	23.0	20.0	20.0	[meV]
3rd nearest neighbor	t''	0.5	5.0	10.0	10.0	[meV]



➢ Wannier function



S. Schuwalow *et al.*, PRB **82**, 035116 (2010)

✓ Hoppings up to 3rd nearest neighbor sites capture band structure

✓ Noninteracting Fermi surface is deformed because of t'

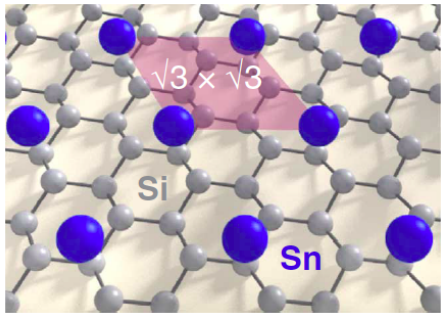
Experimental situation

Pignedoli et al. 2004, Göthelid *et al*. 1995, Uhrberg *et al*. 2000, Morikawa *et al*. 2002, Lobo *et al*. 2003, Modesti *et al*. 2007, Li *et al*. 2013, Horikoshi *et al*. 1999, Slezak et al. 1999, Custance *et al*. 2001, ….

➢ Structure etc…

C: Si(111) – √3× √3 structure (triangular lattice)
Mott

Sn: Si(111) – √3× √3 structure (triangular lattice)
become insulating below ∼ 70 K



Pb: Si(111) – √3× √3 (high-T) ⇔ 3x3 (low T) transition ?
In reality it is more subtle: 1x1 coexist at room temperature
5x√3 structure at low T ?
charge density wave observed at low T

➢ Contradictory results in Sn: Si(111)

- ✓ Core-level spectroscopy sees (mainly) two components => charge order ?
- ✓ ARPES => “backfolding” => interpreted as spin/charge order
- ✓ STM => see no ordering (homogeneous behavior)

Motivation and Outline

Motivation

- Charge vs Spin ?

  → system is close to the crossover between charge- and spin-dominant regime

- Why contradiction in Experiment ?

  → time scale of the probe matters

Outline

- Previous theoretical studies

  Study on Hubbard model (LSDA+U, DMFT, DCA, DF, …) => spin order
  GW+EDMFT study on extended Hubbard model => charge order
  → need a method beyond GW+EDMFT

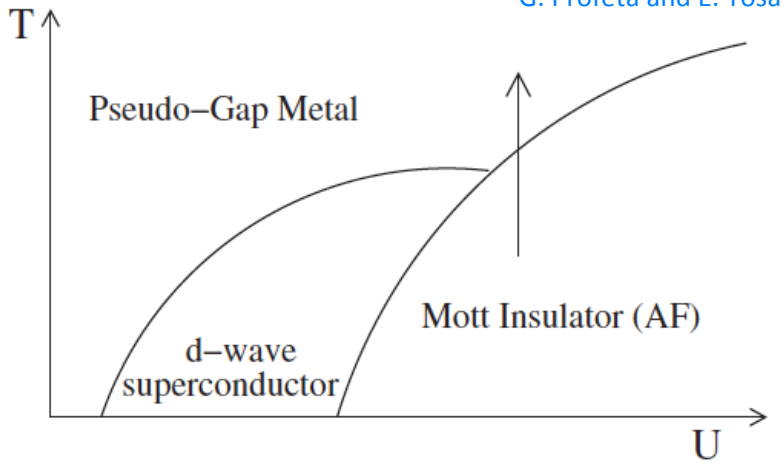
- TRILEX --- a method treating spin and charge on same footing

- TRILEX result (preliminary)

Previous theoretical studies ①

- LSDA+U calculation

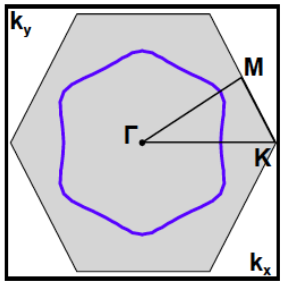
G. Profeta and E. Tosatti, PRL **98**, 086401 (2007))



→ Predict magnetic Mott insulating ground state

- Noncollinear spin-dependent DFT calculation

S. Schuwalow, D. Grieger, and F. Lechermann, PRB **82**, 035116 (2010)



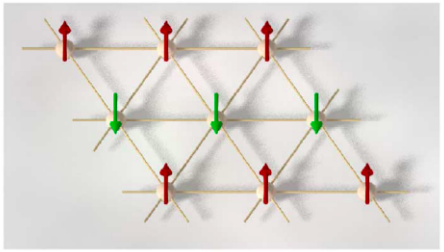
Magnetic ordering	E (meV)	M (μ_B)	M_{Sn} (μ_B)
Ferromagnetic	1.5	0.80	0.031
Collinear ferrimagnetic	–1.6	0.44	0.055
120° noncollinear	–3.6	0.00	0.058

✓ Predict noncollinear magnetic state => 3x3 structure (instability at **q** = K)

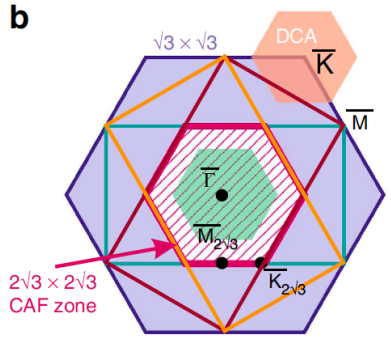
Previous theoretical studies ②

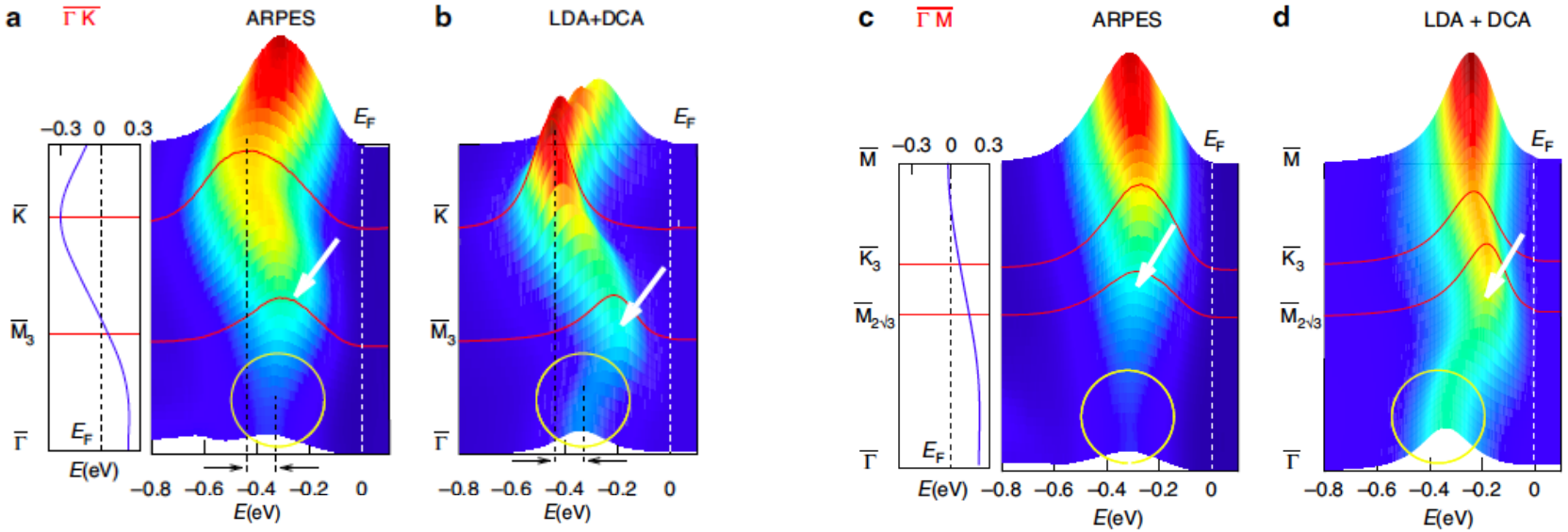
G. Li *et al.*, Nat Commun **4**, 1620 (2013)

➢ LDA + Dynamical cluster approximation on Hubbard model (and comparison with ARPES)



Collinear antiferromagnetism
=> 2√3× 2√3 unit cell
(instability at **q** = M)





→ Shadow band induced by magnetic ordering (collinear AF)

These previous studies consider only local Hubbard interaction…

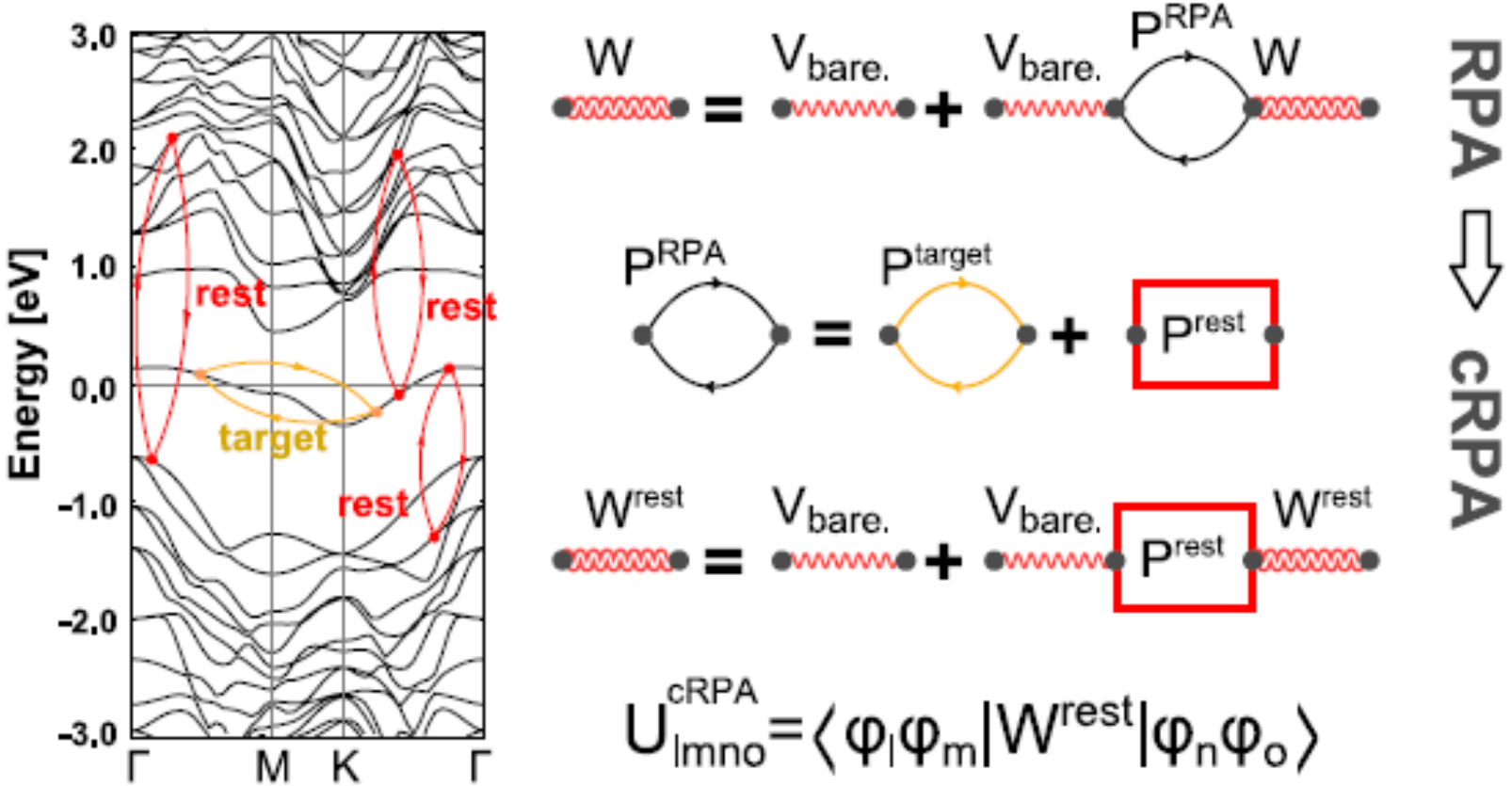
Q. Is the interaction really local ?

A. No. Non-local interaction is substantial.

Realistic Value of Coulomb interaction

- Constrained Random Phase Approximation (cRPA)

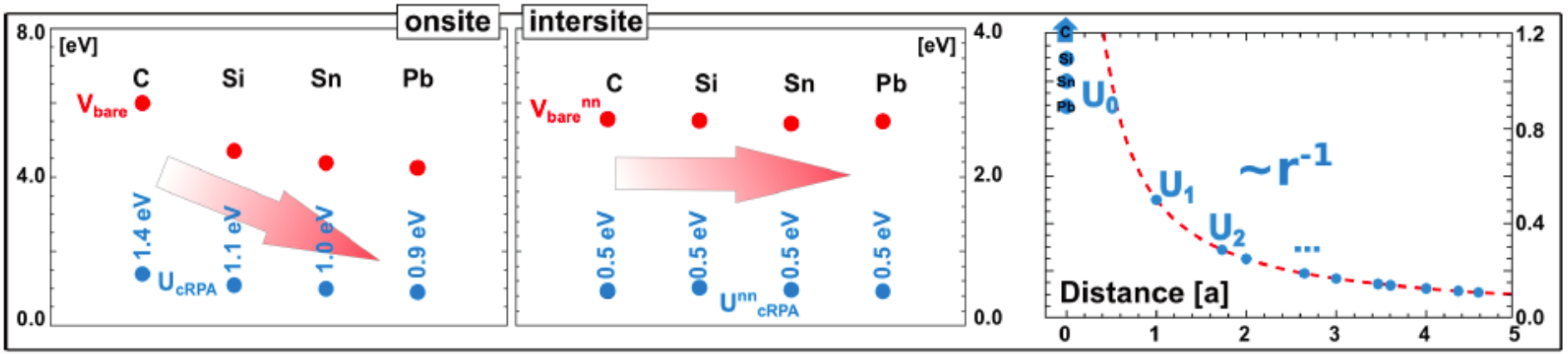
Aryasetiawan, Imada, Georges, Kotliar, Biermann, Lichtenstein, PRB 2004.
[Figure from Hansmann et al., JPCM 2013]



Extended Hubbard Model from cRPA

P. Hansmann *et al.*, JPCM **25** 094005 (2013)
P. Hansmann *et al.*, PRL **110**, 166401 (2013)

		C	Si	Sn	Pb
Hubbard	U_0 (eV)	1.4	1.1	1.0	0.9
Nearest Neighbor	U_1 (eV)	0.5	0.5	0.5	0.5
Long range tail	U_n	U_1/r_a	U_1/r_a	U_1/r_a	U_1/r_a



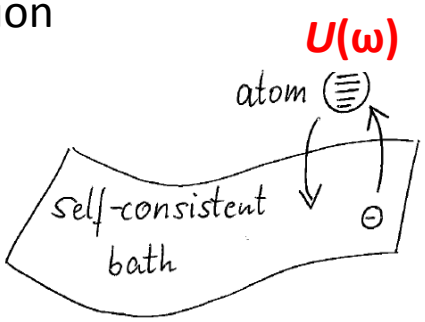
- $U_0 \sim 1$ eV > W (bandwidth) ~ 0.5 eV) → strongly correlated
- $U_1 / U_0 \sim 0.4$-0.5 → Coulomb interaction is highly nonlocal

GW+EDMFT study

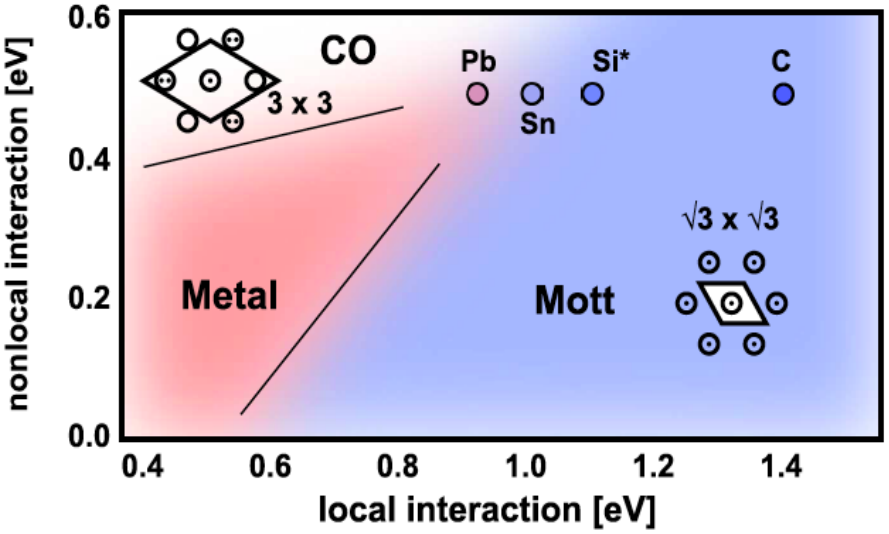
P. Hansmann *et al.*, PRL **110**, 166401 (2013)

- GW + EDMFT : Local correlation from DMFT + Nonlocal correlation from GW
  + Dynamical screening effect from non-local interaction

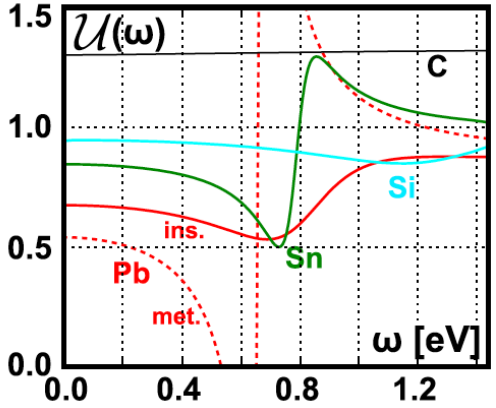
  - ✓ Determine bath and dynamical Hubbard $U(\omega)$ self-consistently
  - ✓ Suitable for studying charge instability
    (whereas spin channel not included)



- Schematic Phase diagram



- Structure of dynamical Hubbard U

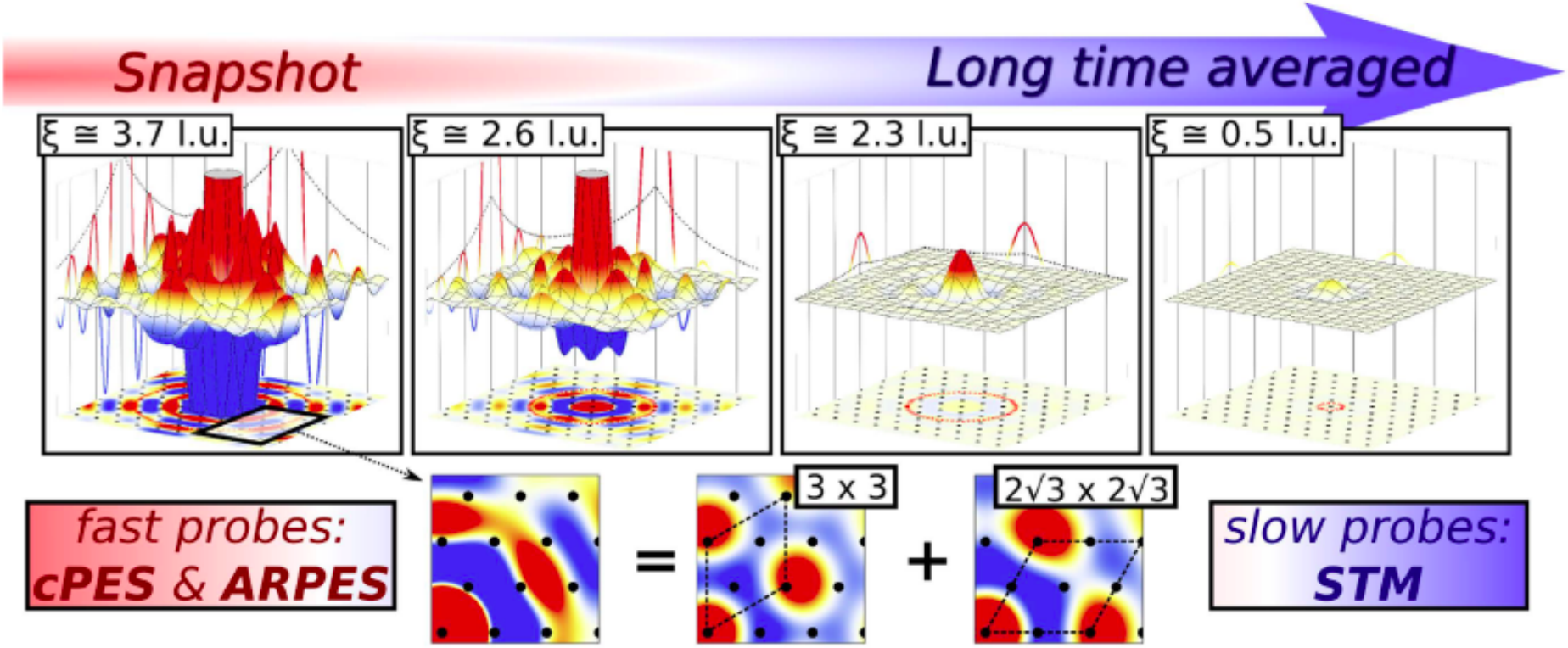


✓ Non-local Interaction induces dynamical screening of Hubbard U and charge instability

GW+EMFT result: charge-charge correlation function

Time-scale of the probe matters!

typical time scale: fs

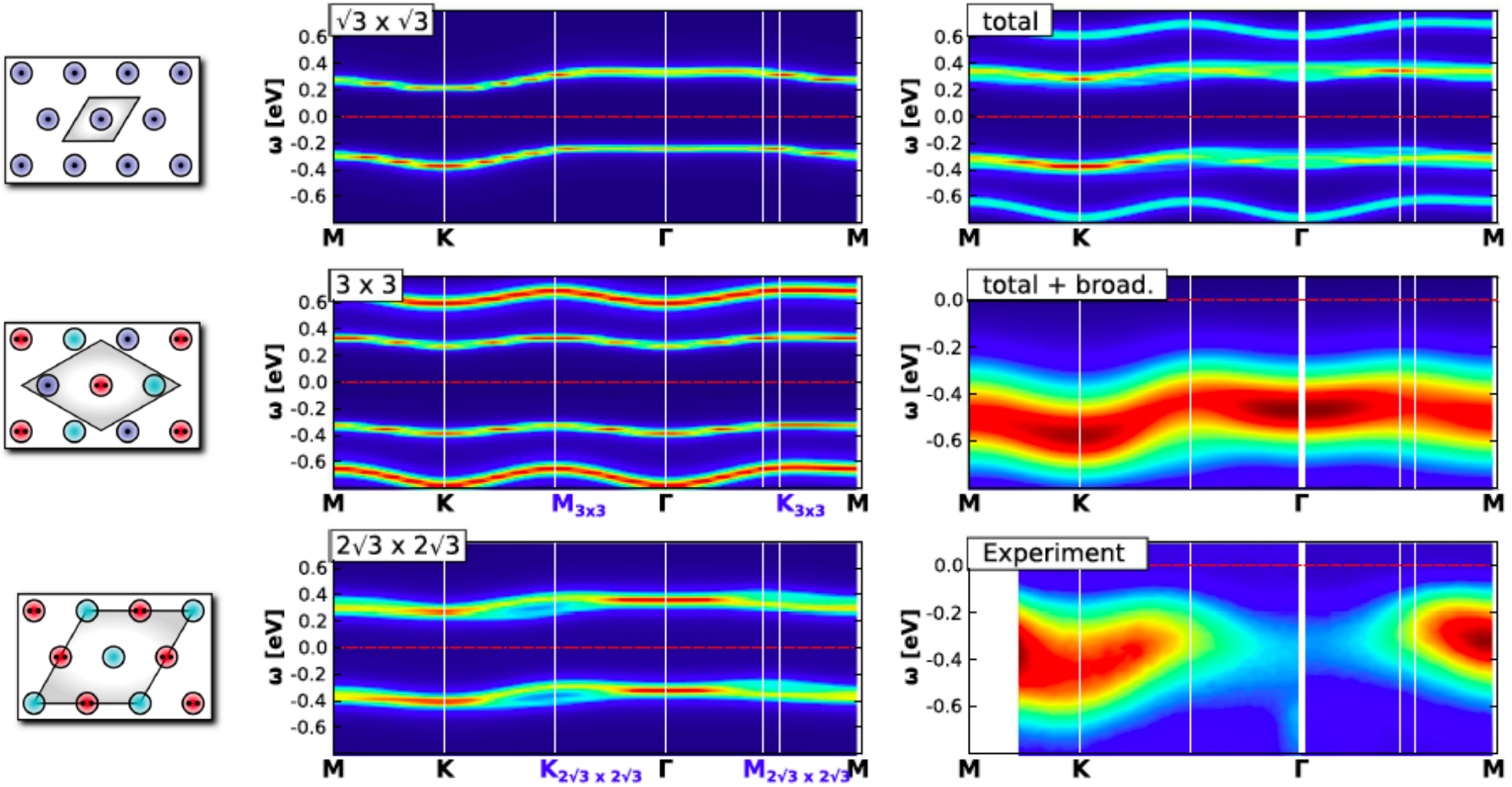


P. Hansmann *et al.*, Scientific Reports 6, 19728 (2016)

- ✓ **STM** (see no order): spatial resolution but temporal average
- ✓ **ARPES/core-level spectroscopy** (suggest ordering): snapshot-like measurement, spatial average

ARPES understood by superposition of different components

P. Hansmann *et al.*, Scientific Reports 6, 19728 (2016)



Q. Charge vs Spin ?

- ✓ Studies on Hubbard model (LSDA+U, DMFT, DCA, DF, …): spin
- ✓ GW+EDMFT on extended Hubbard model: charge
- ✓ Need techniques beyond GW + EDMFT → TRILEX

A. In an interesting regime

Close to the crossover between charge- and spin-dominant regimes

TRILEX approximation

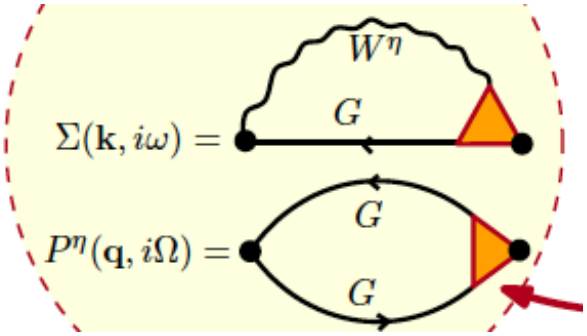
- Trilex → correlation induced by long-range fluctuation + Mott physics (local correlation)

T. Ayral and O. Parcollet, PRB **92**, 115109 (2015); PRB **93**, 235124 (2016)

✓ Exact form of self-energy and polarization (single-band)

$$\Sigma(\mathbf{k}, i\omega) = -\sum_{\substack{\mathbf{q},i\Omega,\\ \eta=\mathrm{ch,sp}}} m_\eta \lambda^\eta G_{\substack{\mathbf{q}+\mathbf{k},\\ i\omega+i\Omega}} W^\eta_{\mathbf{q},i\Omega} \Lambda^\eta_{\substack{\mathbf{k},\mathbf{q},\\ i\omega,i\Omega}}$$

$$P^\eta(\mathbf{q}, i\Omega) = 2\sum_{\mathbf{k},i\omega} \lambda^\eta G_{\substack{\mathbf{q}+\mathbf{k},\\ i\omega+i\Omega}} G_{\mathbf{k},i\omega} \Lambda^\eta_{\substack{\mathbf{k},\mathbf{q},\\ i\omega,i\Omega}}$$



✓ TRILEX (triply irreducible local expansion)

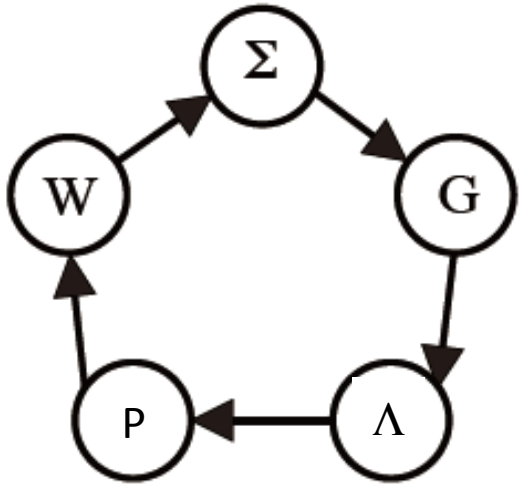
$$\Lambda^\eta(\mathbf{q}, \mathbf{k}, i\omega, i\Omega) \approx \Lambda^\eta_{\mathrm{imp}}(i\omega, i\Omega)$$

c.f. DMFT (dynamical mean-field theory)

$$\Sigma(\mathbf{k}, i\omega) \approx \Sigma_{\mathrm{imp}}(i\omega)$$

Include both spin and charge channel（η= charge, spin) !

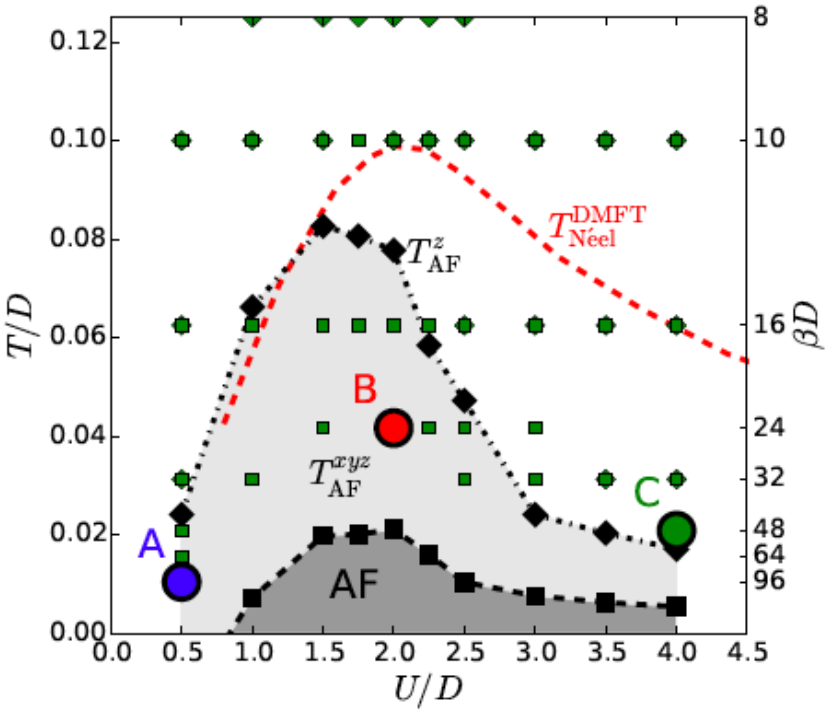
Hedin's equation



Example of Trilex result: structure of Λ in 2D square lattice

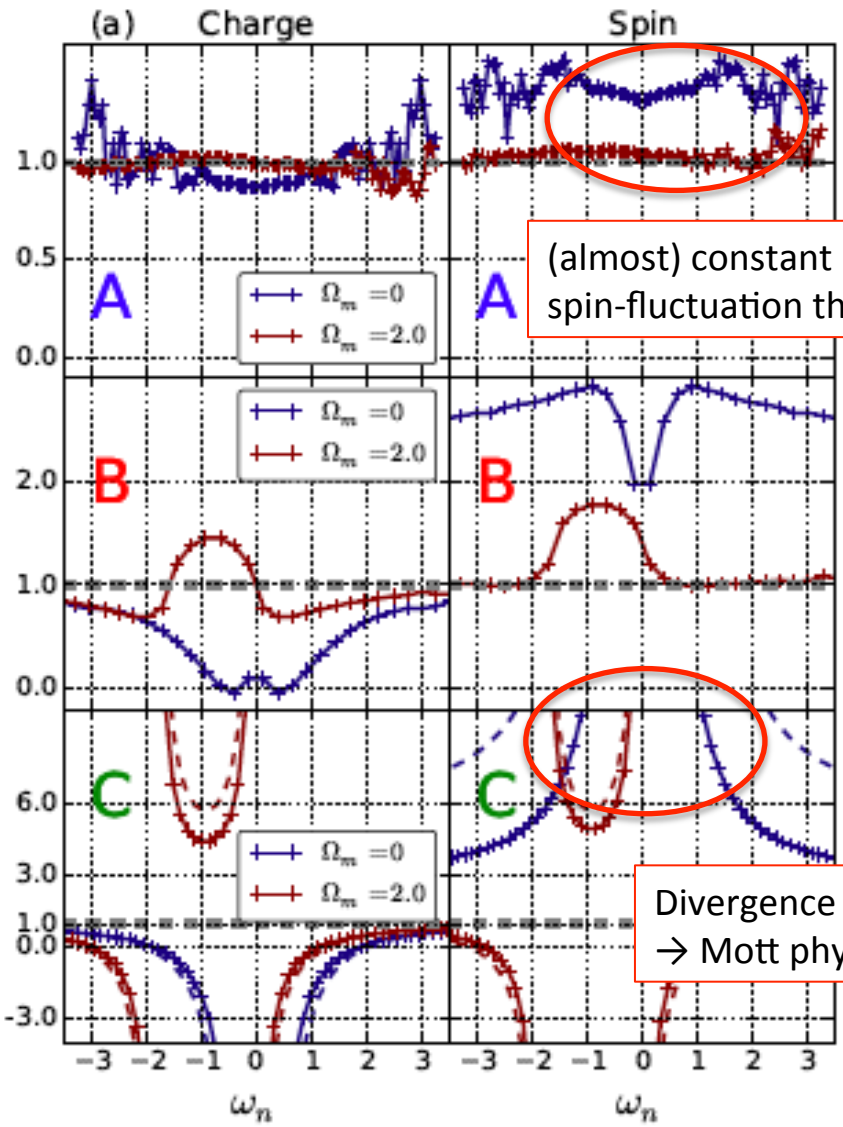
T. Ayral and O. Parcollet, PRB **92**, 115109 (2015); arXiv:1512.06719

➢ Phase diagram (2D square lattice)



D: half-band width = 4*t*

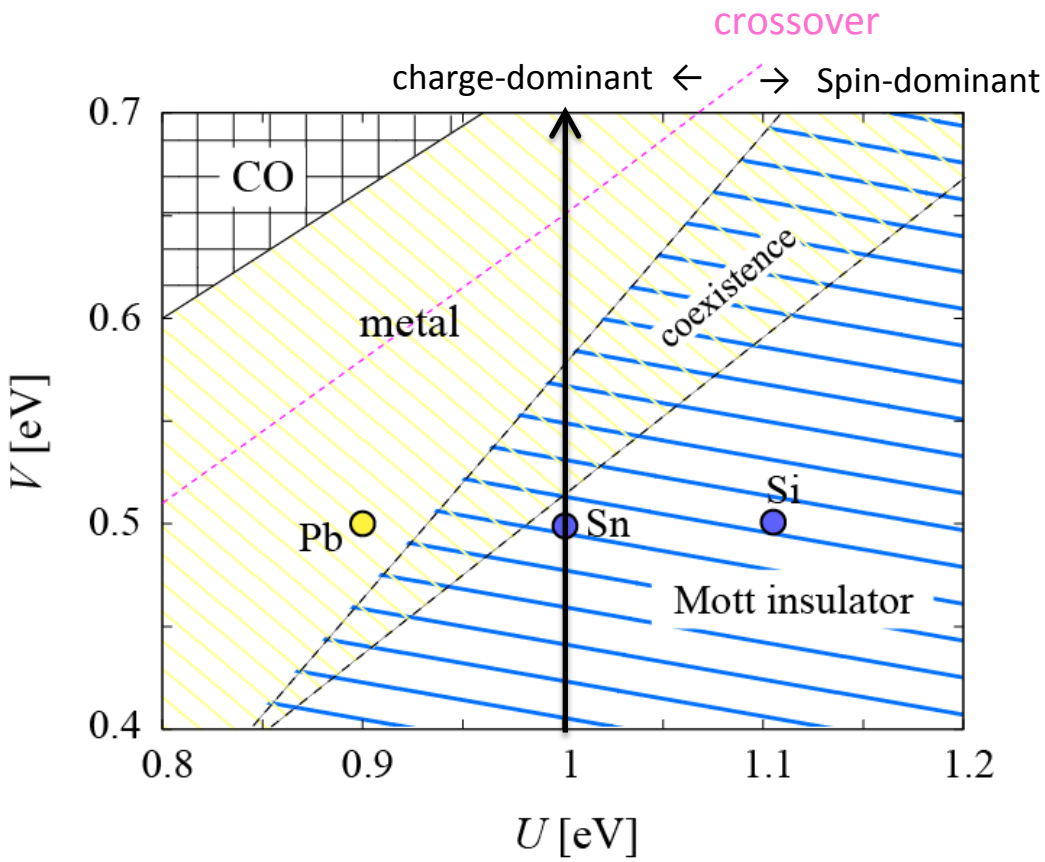
➢ Frequency dependence of Λ

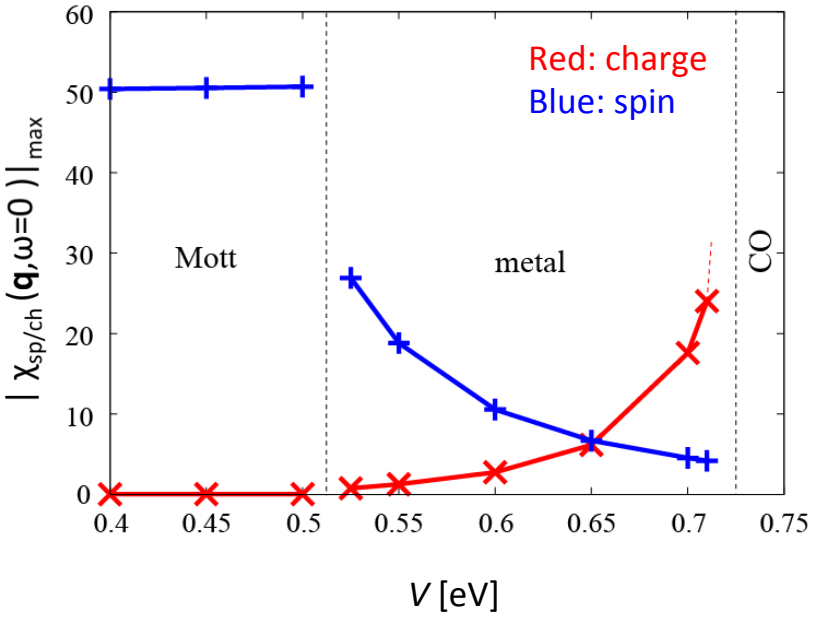


(almost) constant → spin-fluctuation theory

Divergence of Λ → Mott physics

Trilex result: schematic phase diagram at T = 116 K

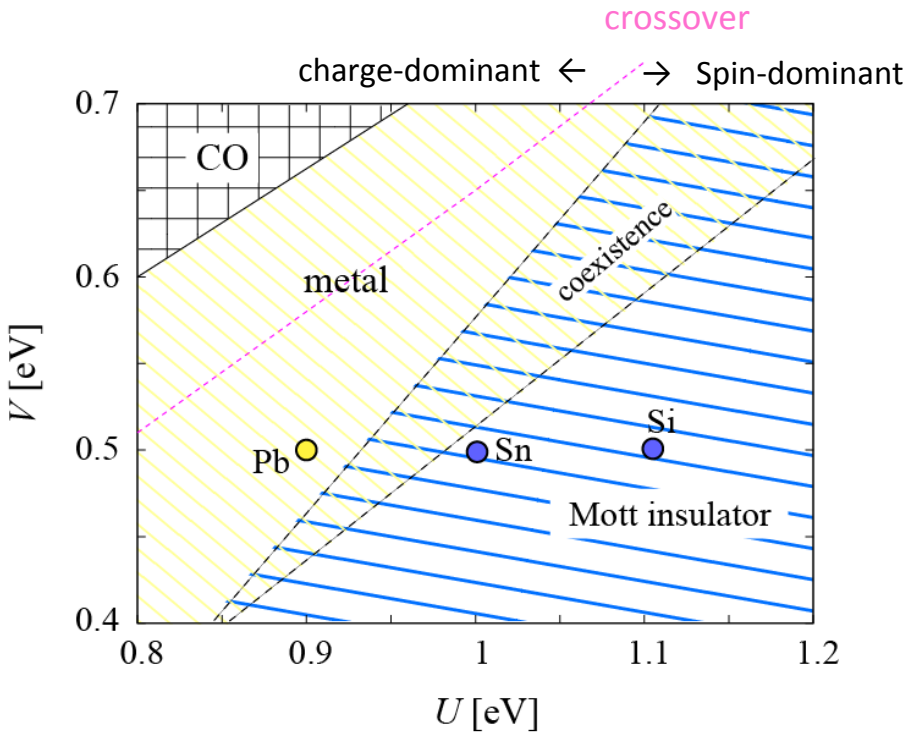




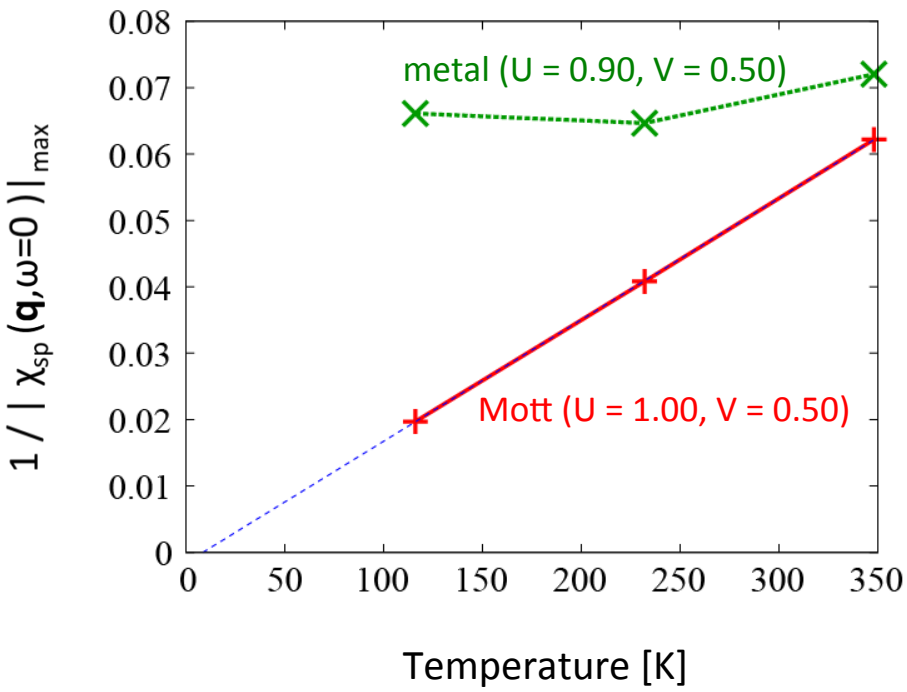
- ✓ crossover between spin-dominant and charge-dominant regimes
- ✓ Real material very close to crossover (within ~10 % parameter difference)

Trilex result: Neel temperature is low

- Phase diagram at 116 K



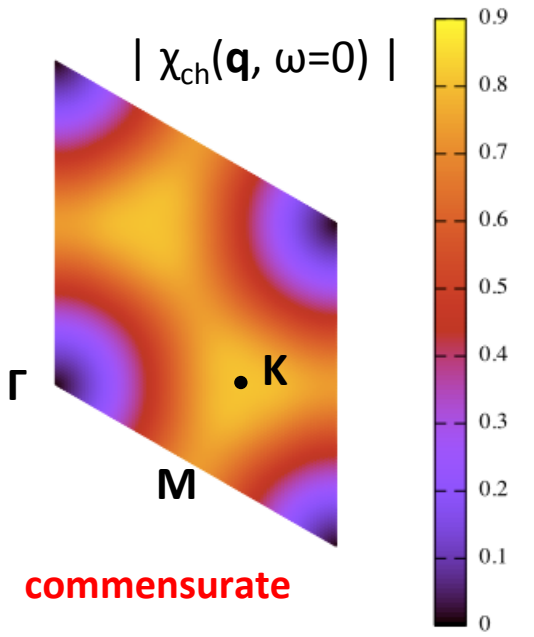
- Inverse spin susceptibility



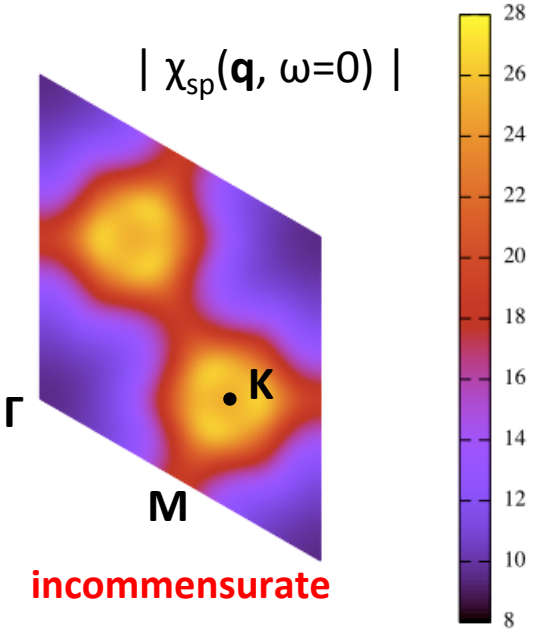
- ✓ No magnetic ordering at 116 K
- ✓ If spin order exists, Neel temperature should be low

Momentum Structure of χ in spin-dominant regime

U = 0.90 eV
V = 0.45 eV
T = 116 K
Metal

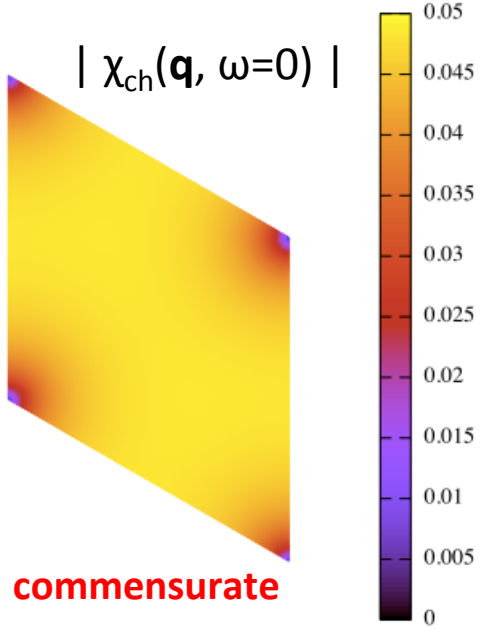


commensurate

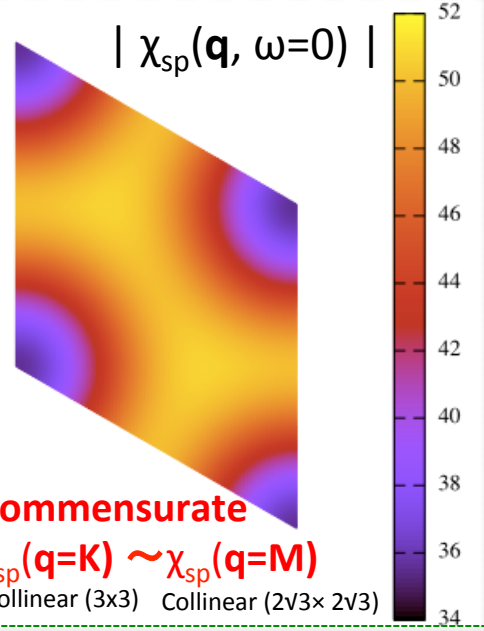


incommensurate

U = 1.00 eV
V = 0.50 eV
T = 116 K
Mott insulator



commensurate

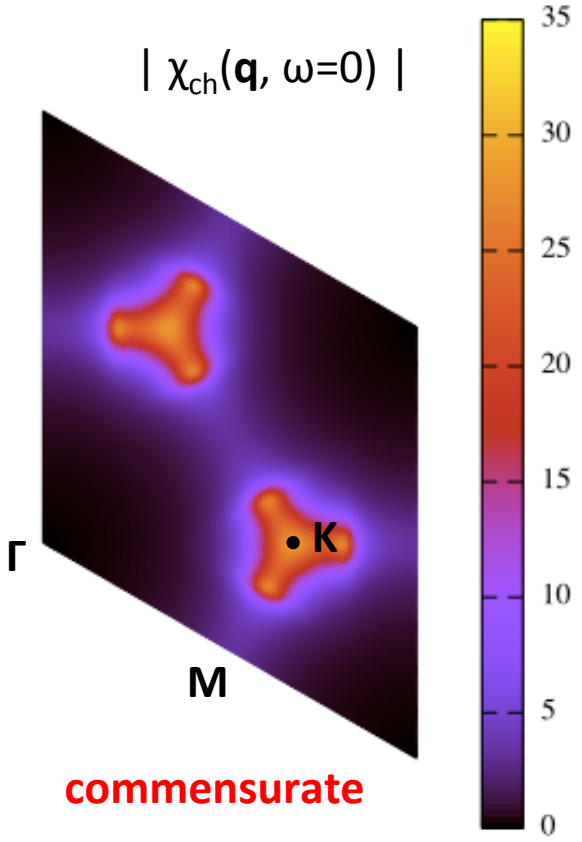


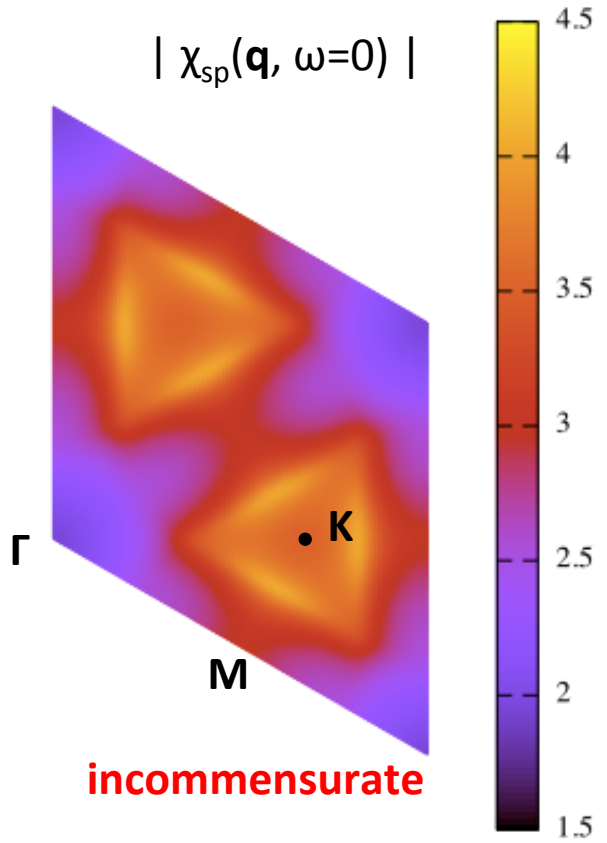
Commensurate
$\chi_{sp}(q=K) \sim \chi_{sp}(q=M)$
Noncollinear (3x3) Collinear (2√3× 2√3)

Consistent with the previous study by G. Li et al. (dual fermion)

Momentum Structure of χ in charge-dominant regime

U = 0.90 eV, V = 0.65 eV, T = 116 K, Metal





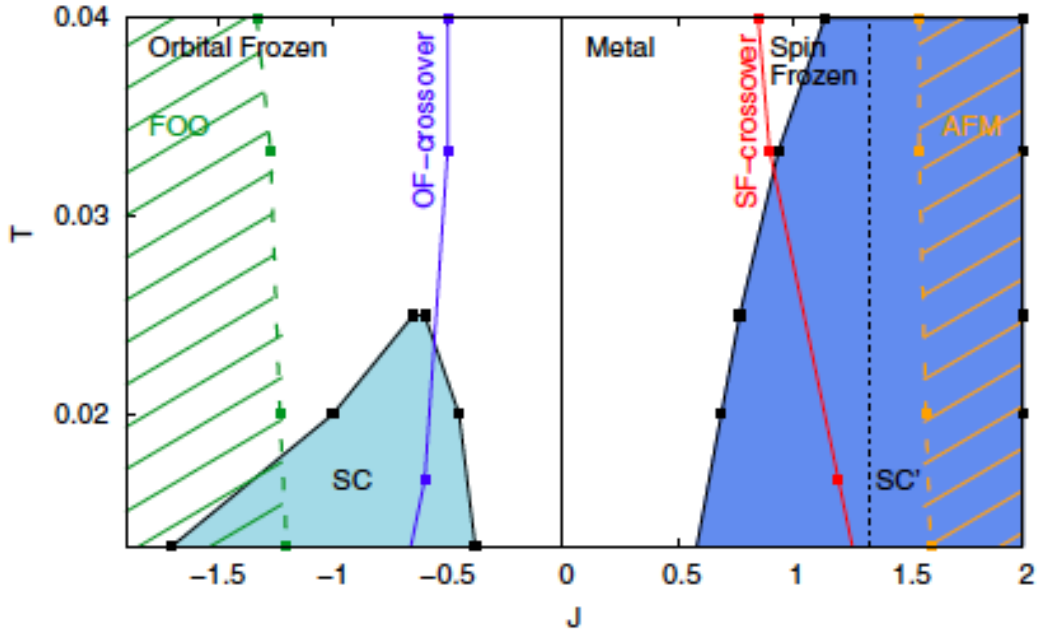
- ✓ | χ_{ch} | is maximum at commensurate wave vector **q** = K
- ✓ But instability at incommensurate wave vector is almost degenerate

Summary

- Surface adatom is an ideal playground to study the interplay between charge and spin
- Coulomb interaction is highly nonlocal
- TRILEX treat charge and spin on same footing
- Existence of non-local interactions push the system into a subtle regime close to crossover between spin and charge dominant regions
- Ordering vector can be incommensurate
- Interpretation of experimental results tricky when timescales of probes and fluctuations coincide

Poster No. ***

Spin/orbital freezing and triplet/singlet superconductivity in two-orbital Hubbard model



In collaboration with

Karim Steiner, Shintaro Hoshino, and Philipp Werner

K. Steiner *et al*., Phys. Rev. B 92, 115123 (2015)

See also related talk by Ryotaro Arita (Friday morning)