Emergence of Quantum Phases in Novel Materials

VII Edition ICMM-CSIC Postgraduate Course

Ramón Aguado

Leni Bascones

Mª José Calderón

Alberto Cortijo

Instituto de Ciencia de Materiales de Madrid (ICMM-CSIC)

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CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

Emergence

More is different

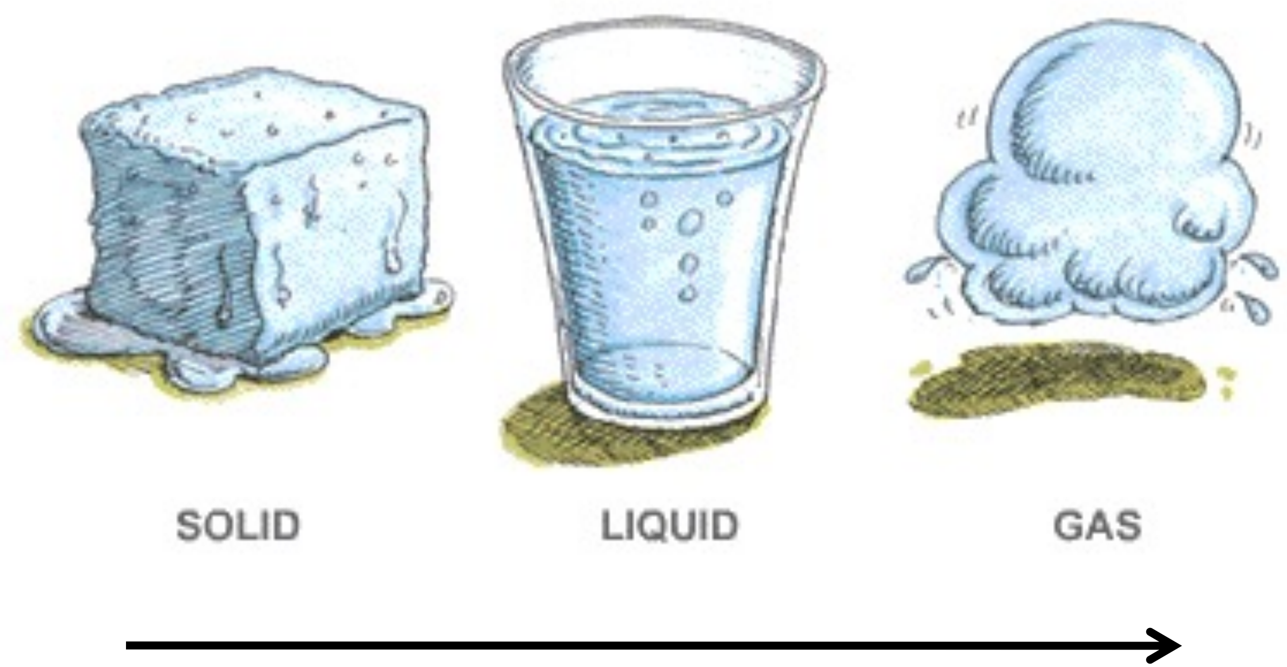
<<The ability to reduce everything to simple fundamental laws does not imply the ability to start from those laws and reconstruct the universe…

At each level of complexity, entirely new properties appear.>>

Anderson
Science 177, 4047 (1972)

New phenomena due to the interactions between particles

Phase transitions



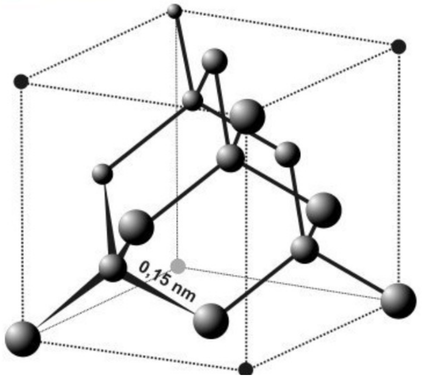
Temperature

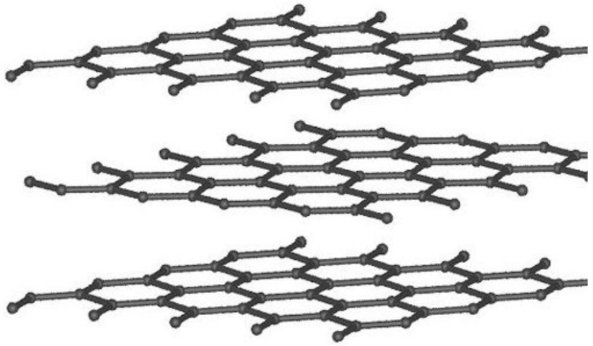
Changes also as a function of pressure …

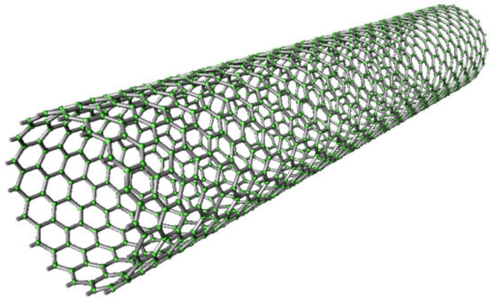
Posibility of detecting but also of controlling the state of the system

Crystalline order in solids

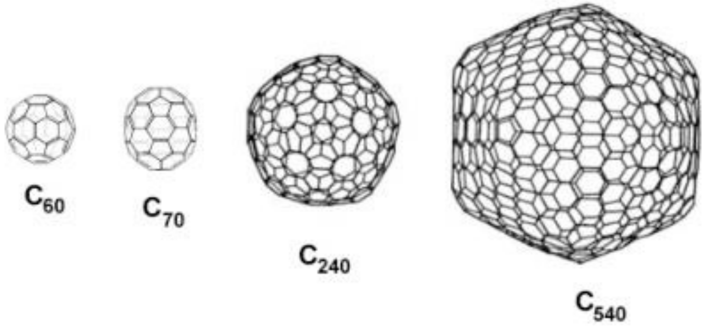
©Itub







Molecules

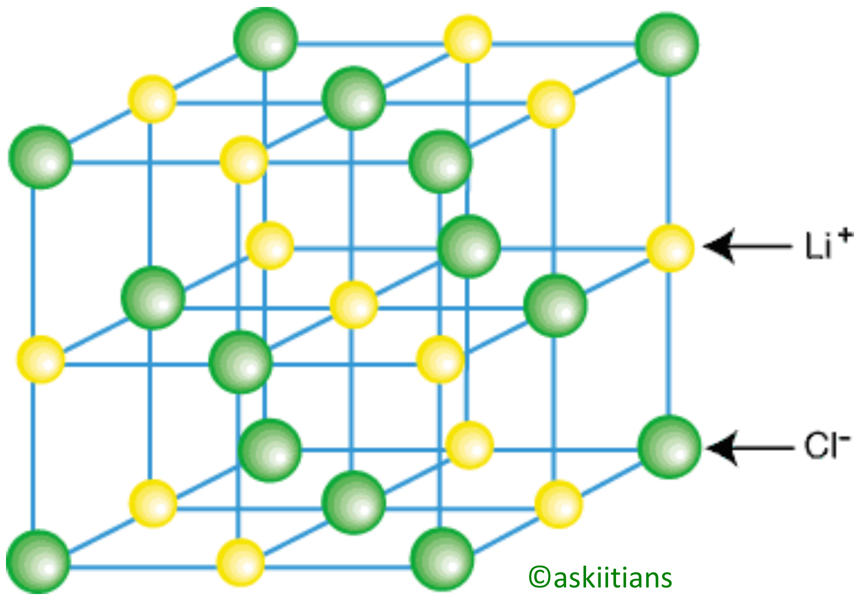


©Fig:web Belén Garrido

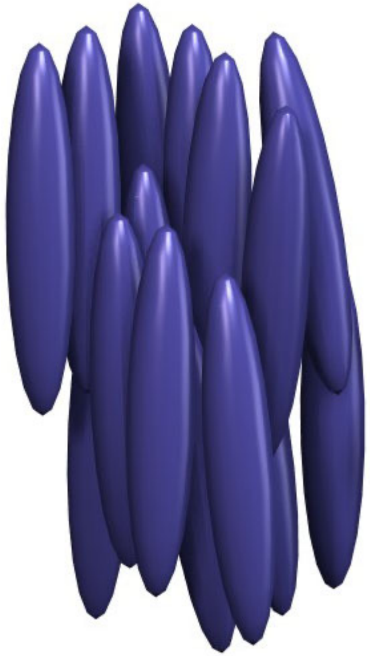
Symmetry breaking (translational symmetry), long-range order: ordered phases

Ordered phases and symmetry breaking

Translational symmetry breaking
(solid)



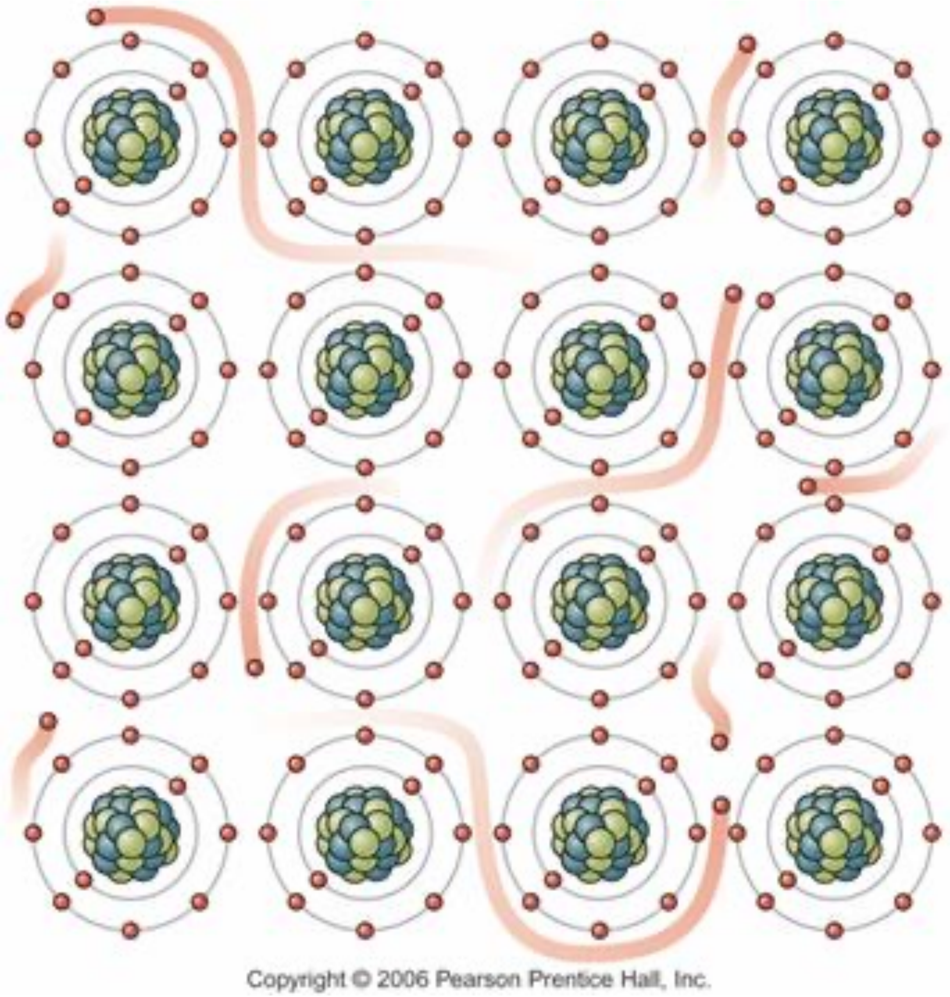
Rotational symmetry breaking
(nematic liquid crystal)



Different symmetries that can be broken

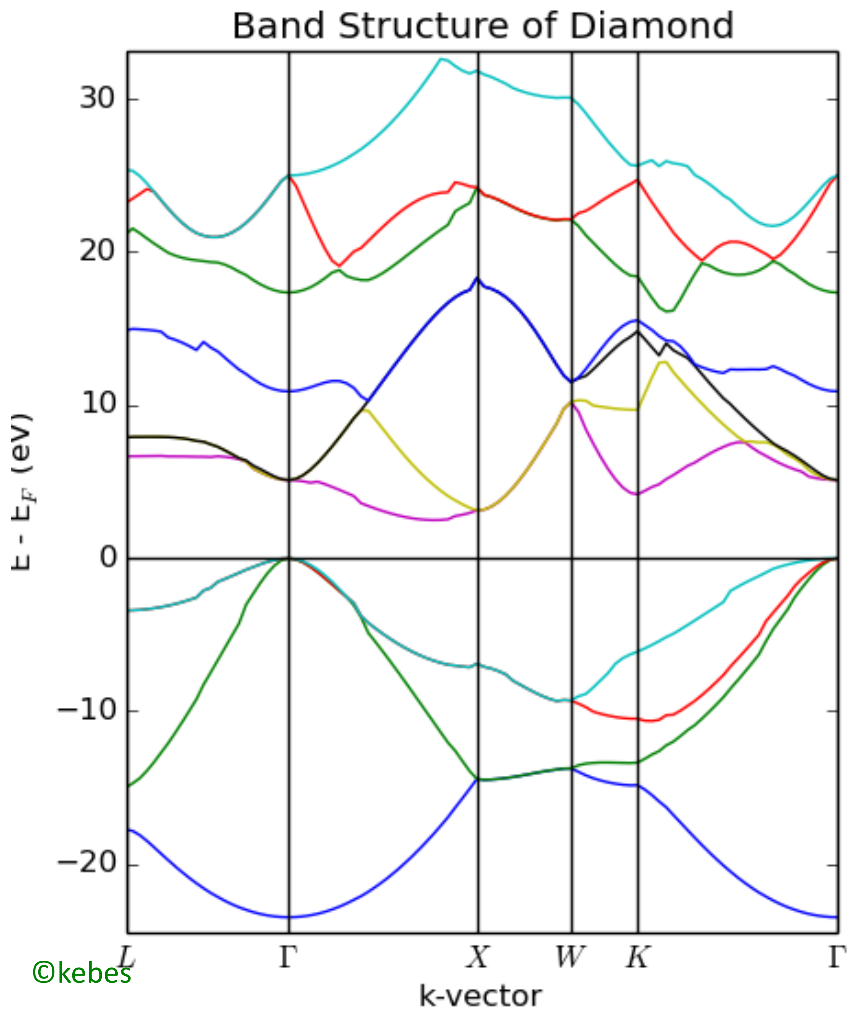
leni.bascones@csic.es

Emergence of quantum phases in novel materials



Electrons interact among them
and with the environment

Band theory: the basis of our undestanding of solids



Bloch states: electronic bands

Band structure E(k)

The electron moves in the average periodic potential of the solid

Density Functional Theory:
Ability to calculate the bands

Band theory: the basis of our undestanding of solids

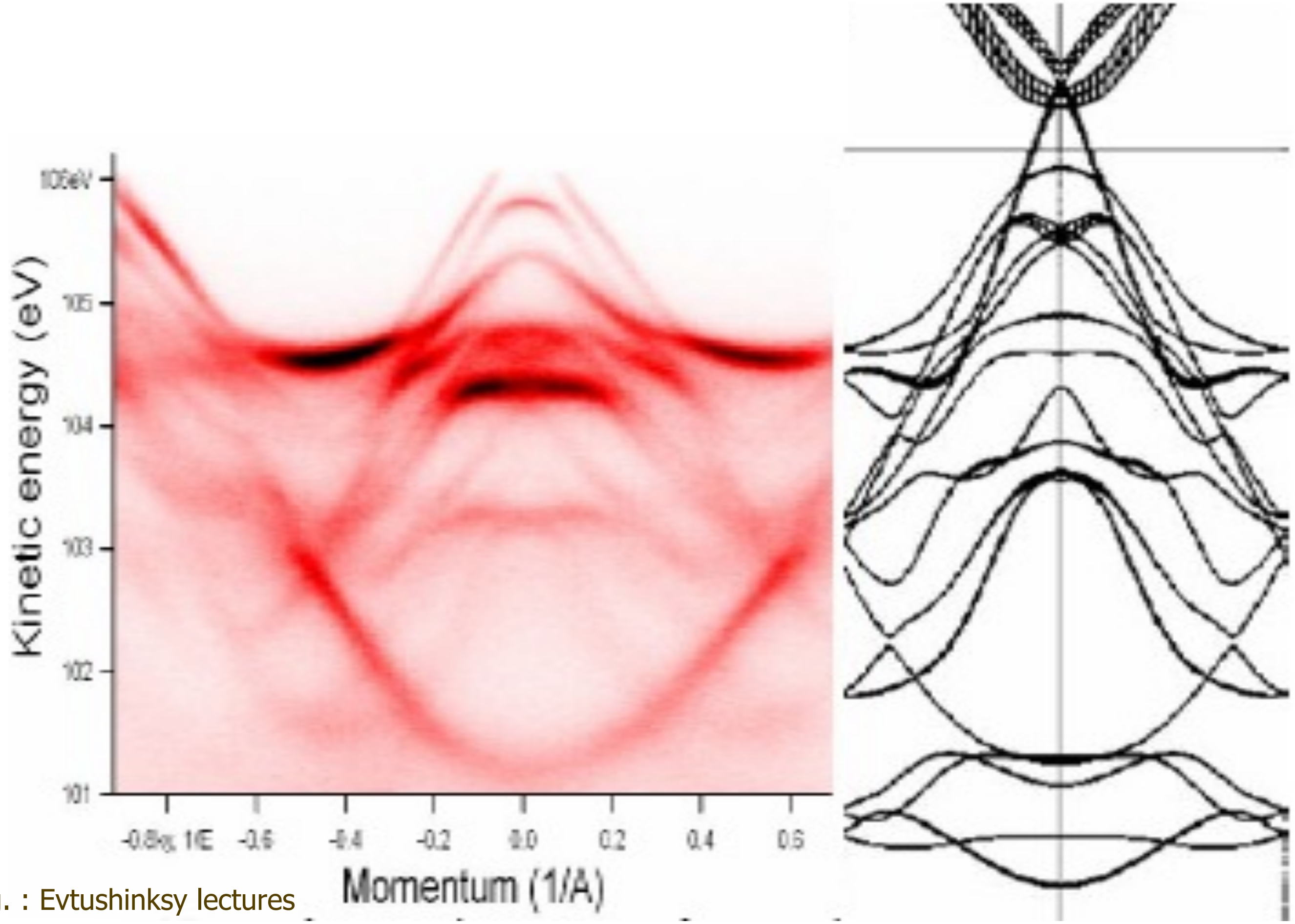
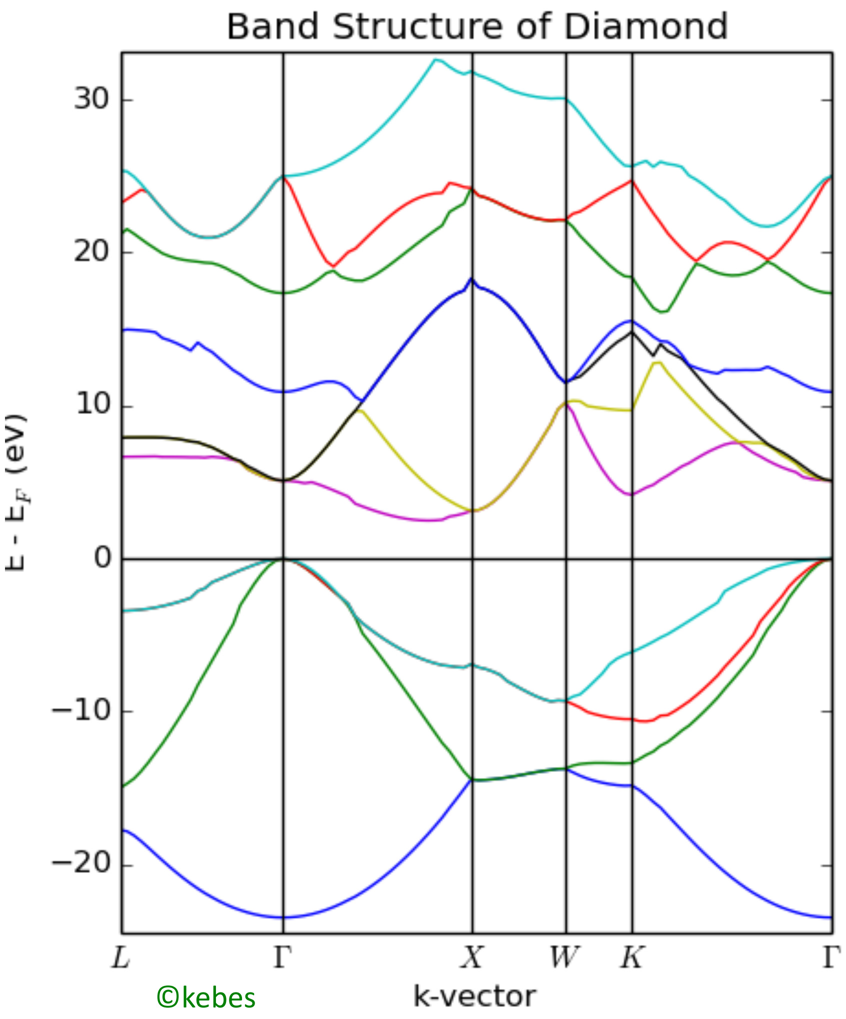


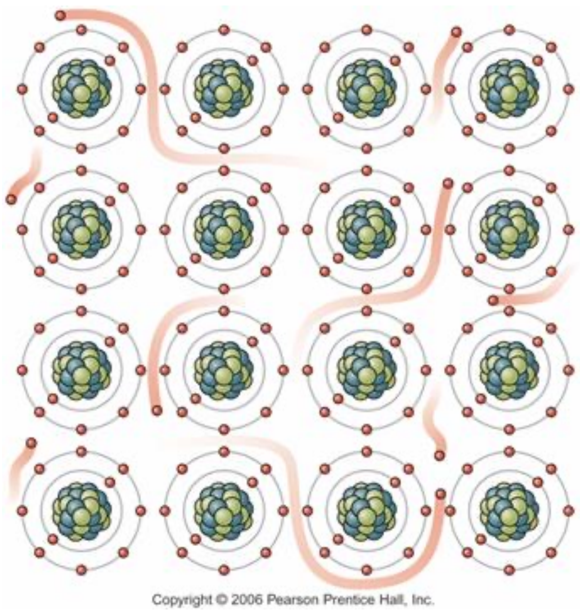
Fig. : Evtushinksy lectures

The bandstructure can be measured Experimentally with probes such as angle resolved photoemission (ARPES) and compared to theoretical predictions

Band theory: an "effective non-interacting" description



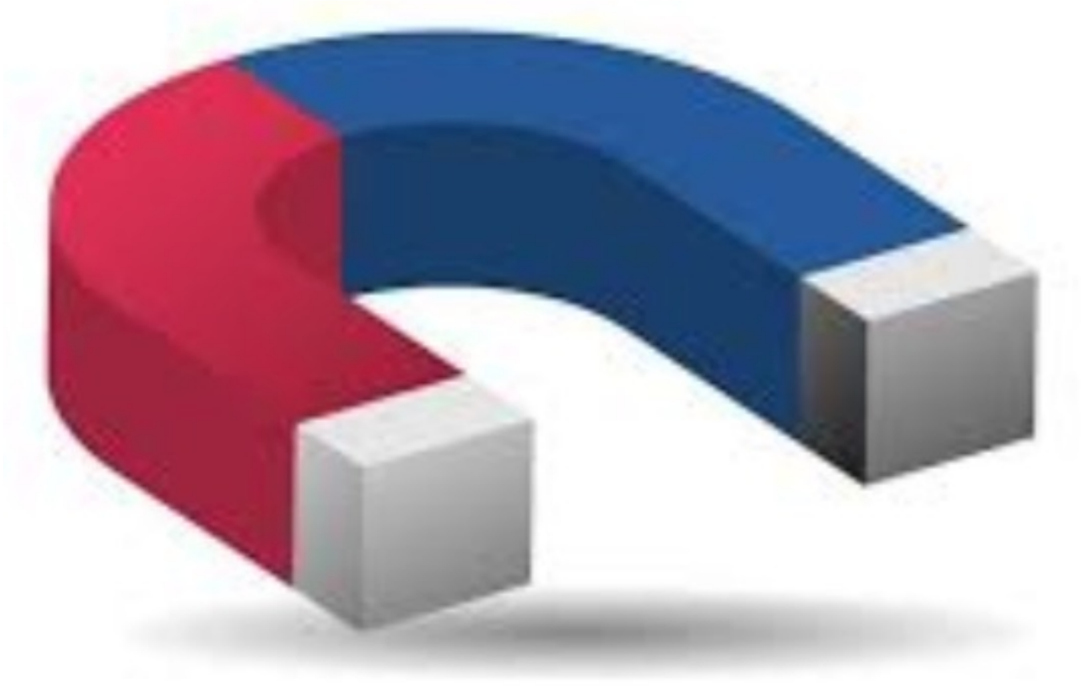
Metals and insulators

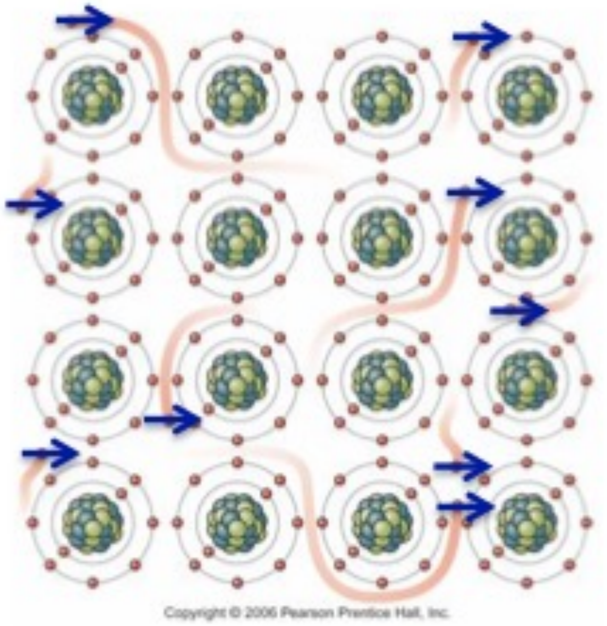


Bandstructure:

Effective "independent" electronic states including in an effective way the symmetry of the lattice and the interactions of the electron with the atomic lattice and with other electrons.

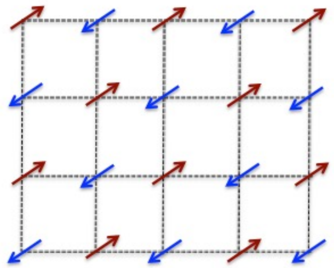
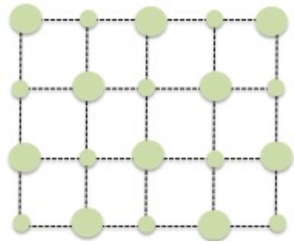
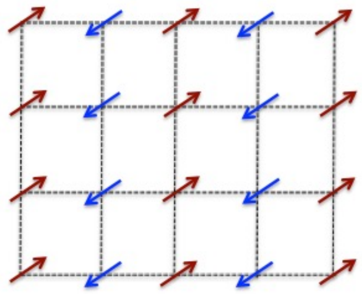
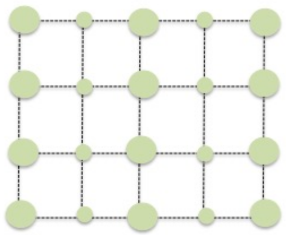
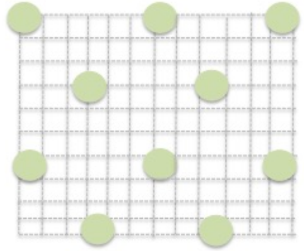
Electronic phases: symmetry breaking





Ferromagnetism:
Spin order
It does not require lattice symmetry breaking

Electronic phases with lattice symmetry breaking

	Spin Order		Charge Order	
Lattice translational symmetry broken (density waves)		Neel antiferromagnet		Charge density wave
Lattice translational and rotational symmetry broken (stripe-like phases)		Stripe antiferromagnet	Charge stripes 	Wigner crystal 
Lattice rotational symmetry broken (nematic states)		Spin nematic		Charge nematic

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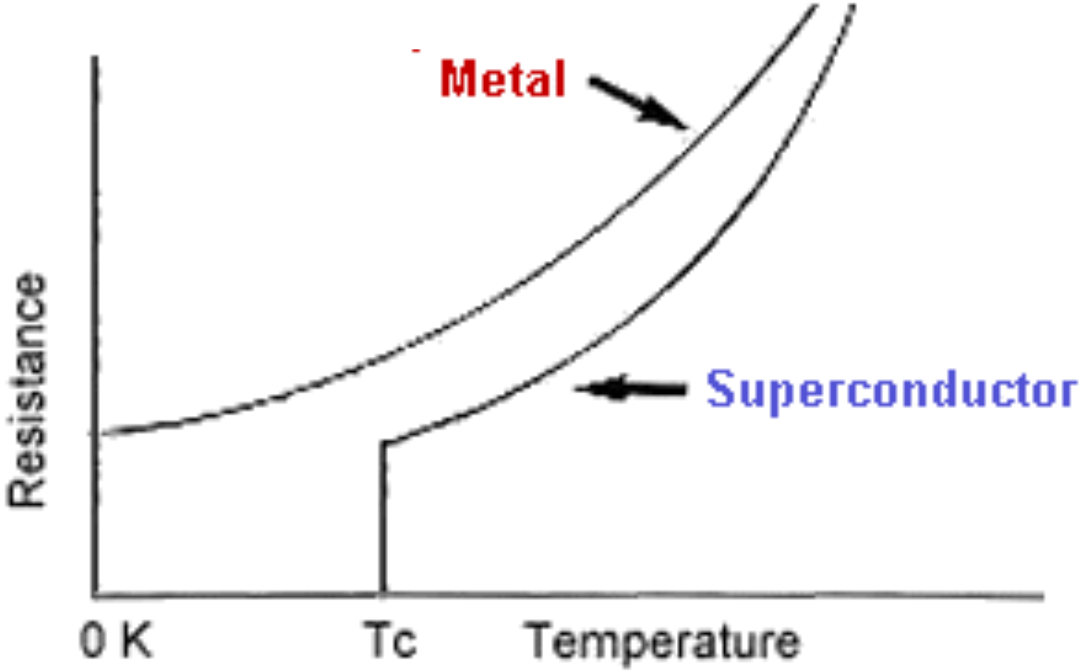
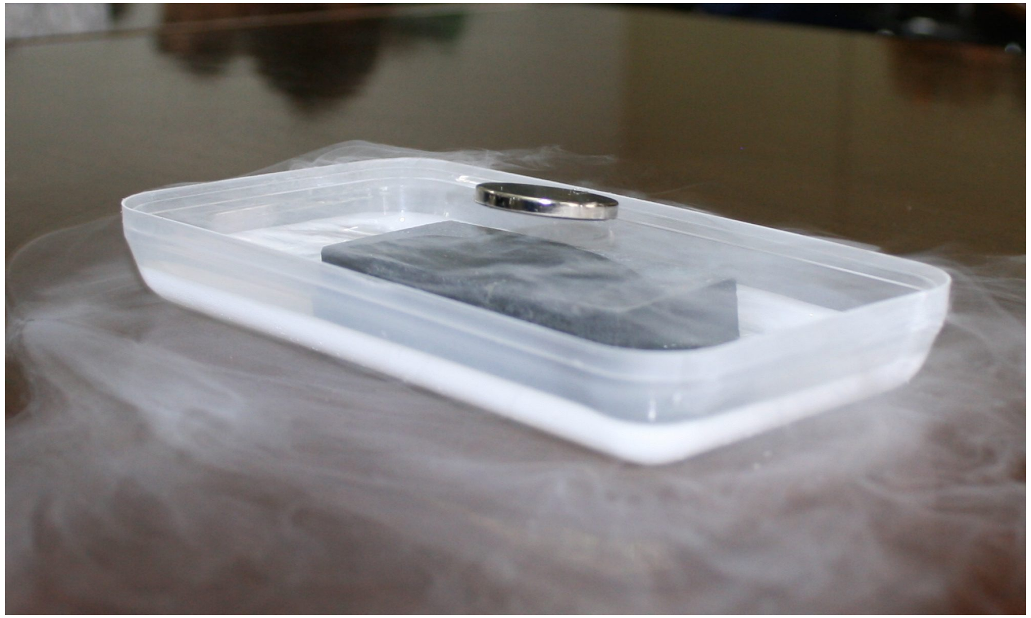
Superconductivity





Breaking of gauge symmetry

Normal vs ordered state

Decreasing Temperature

Normal state

Transition Temperature

Often tunable with doping, pressure, magnetic field or other external knobs

Ordered state

Long-range order
Symmetry breaking
Order parameter

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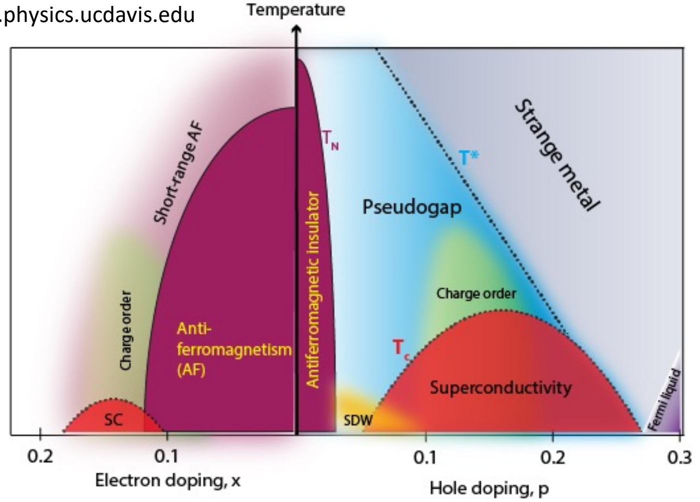
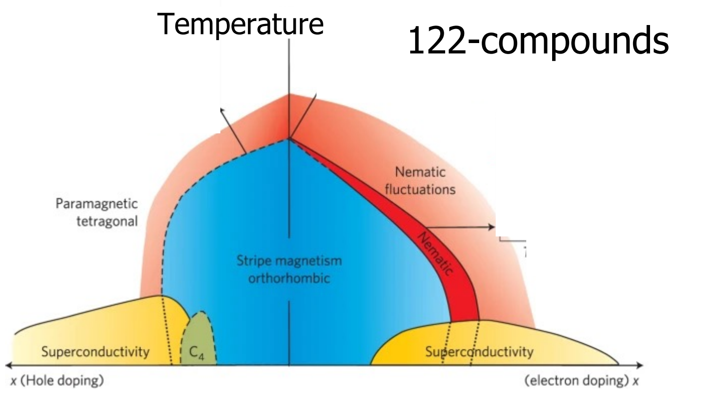
Strongly correlated electron systems: complex phase diagrams

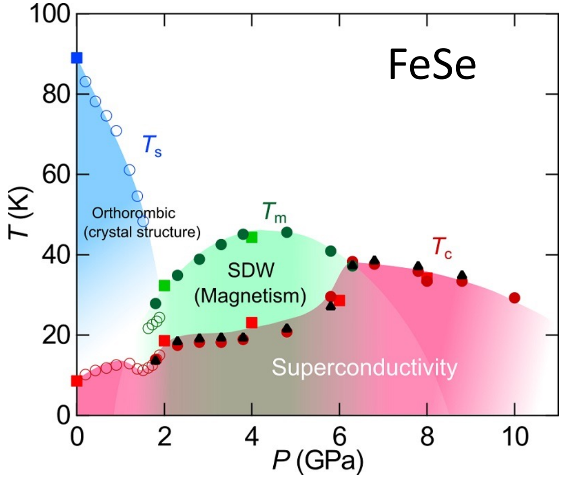
High-Tc superconducting cuprates

Fig: cme.physics.ucdavis.edu

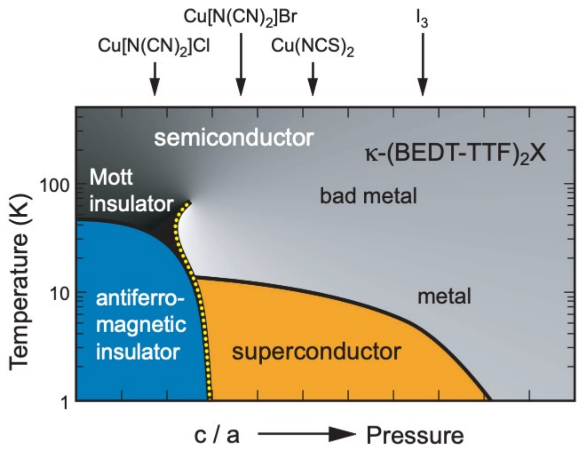


Iron superconductors

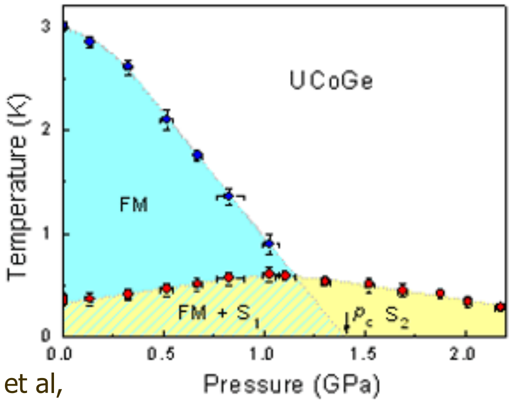




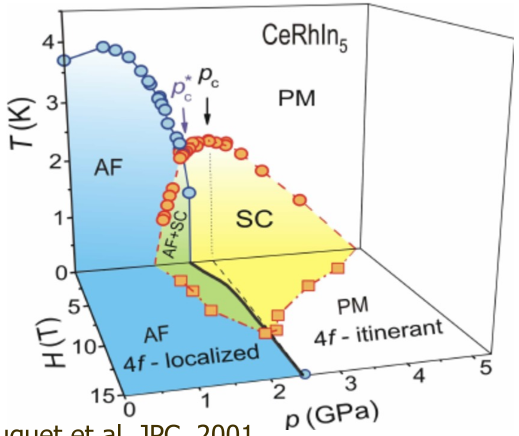
Organic superconductors



Heavy fermion compounds

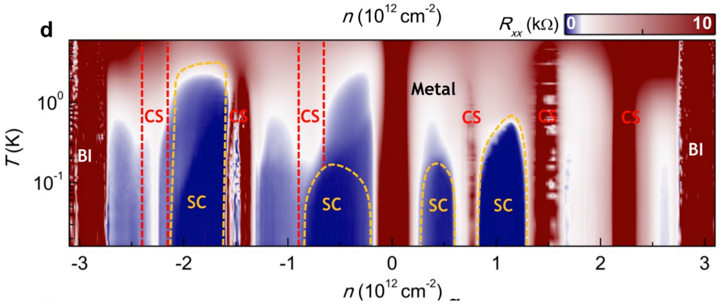


Slooten et al,
PRL 103 , 097003(2009)



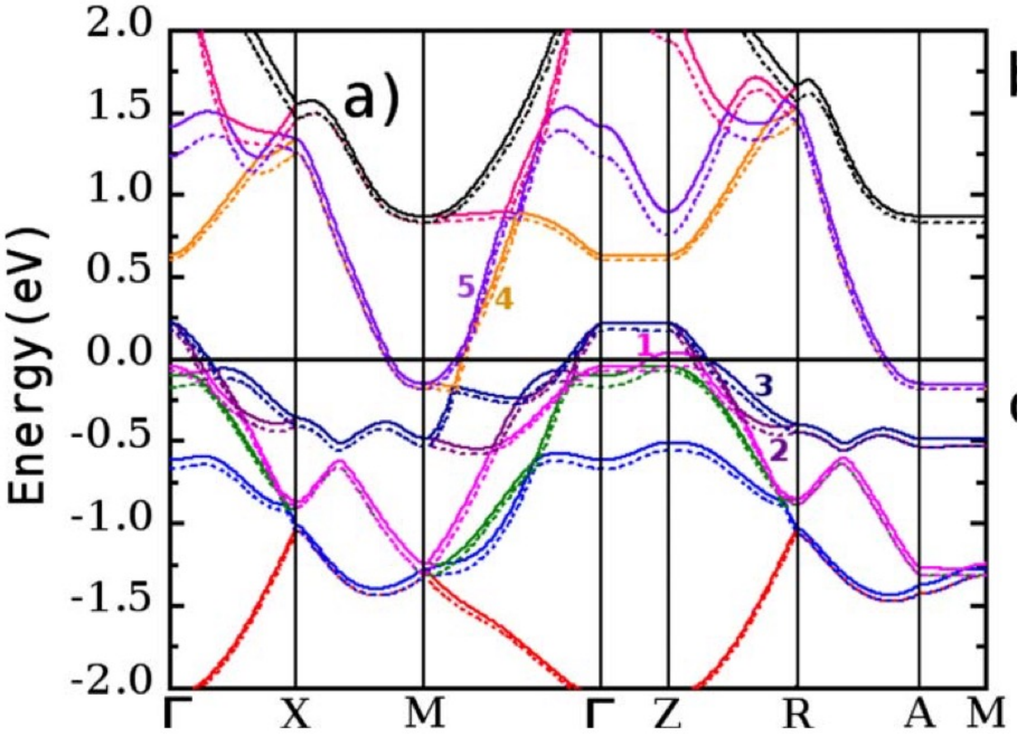
Flouquet et al, JPC, 2001

Twisted bilayer graphene

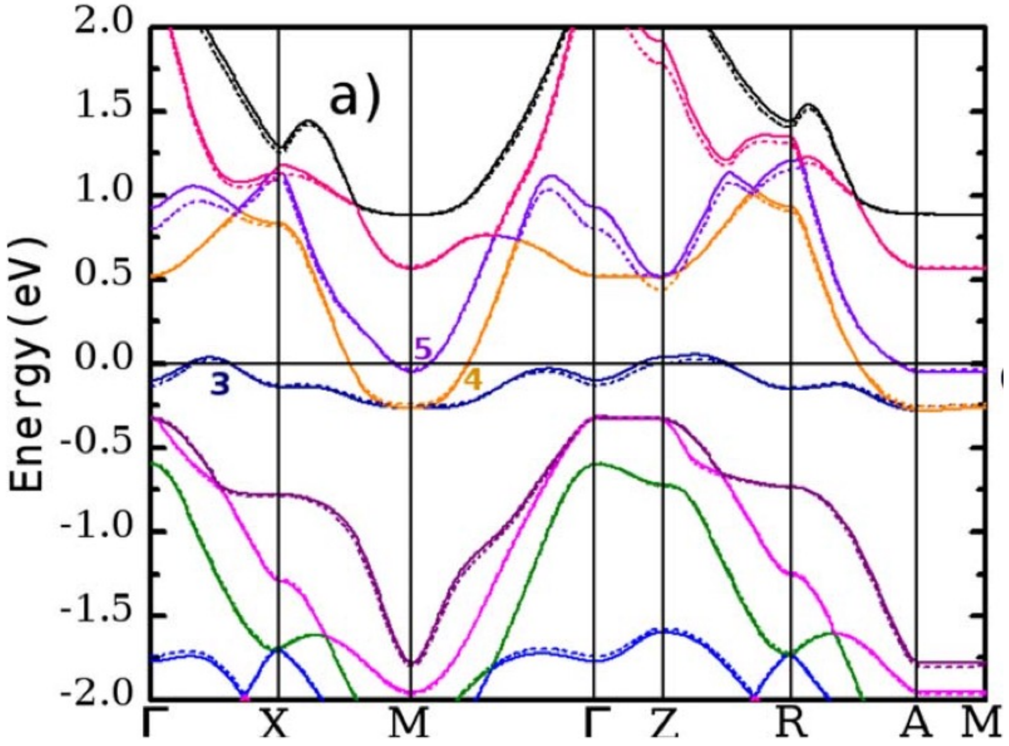


Band structure in normal vs ordered state.

Non-ordered "normal" state



Symmetry breaking:
ordered state at low temperature



Ma & Lu, PRB 78, 033111 (2008)

Band structure E(k)
Is it E(k) always well defined? How good are DFT-like and static mean field descriptions?
Why and when does band theory work?

The "normal" state of strongly correlated systems

When the band picture does not work so well

Weakly correlated system

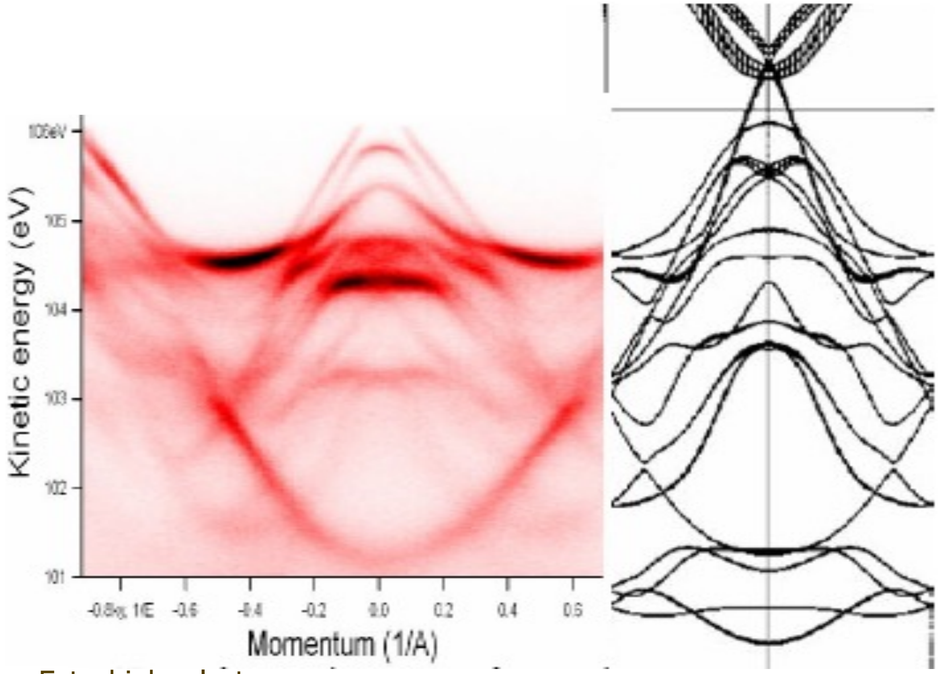


Fig. : Evtushinksy lectures

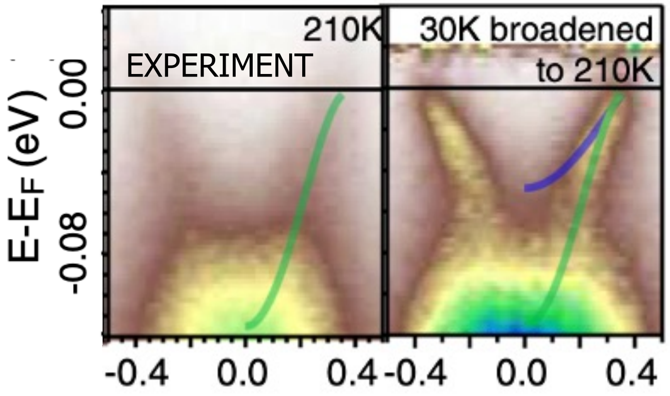
Strongly correlated system

Not just ordered phases and symmetry breaking states

Correlated metals

- Bandwidths which deviate from DFT predictions (larger mass, up to 1000 thousands times larger)
- Blurred spectrum and anomalous temperature behavior

Iron chalcogenide



PRL 110, 067003 (2013)

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Strongly correlated metals

"Normal" state

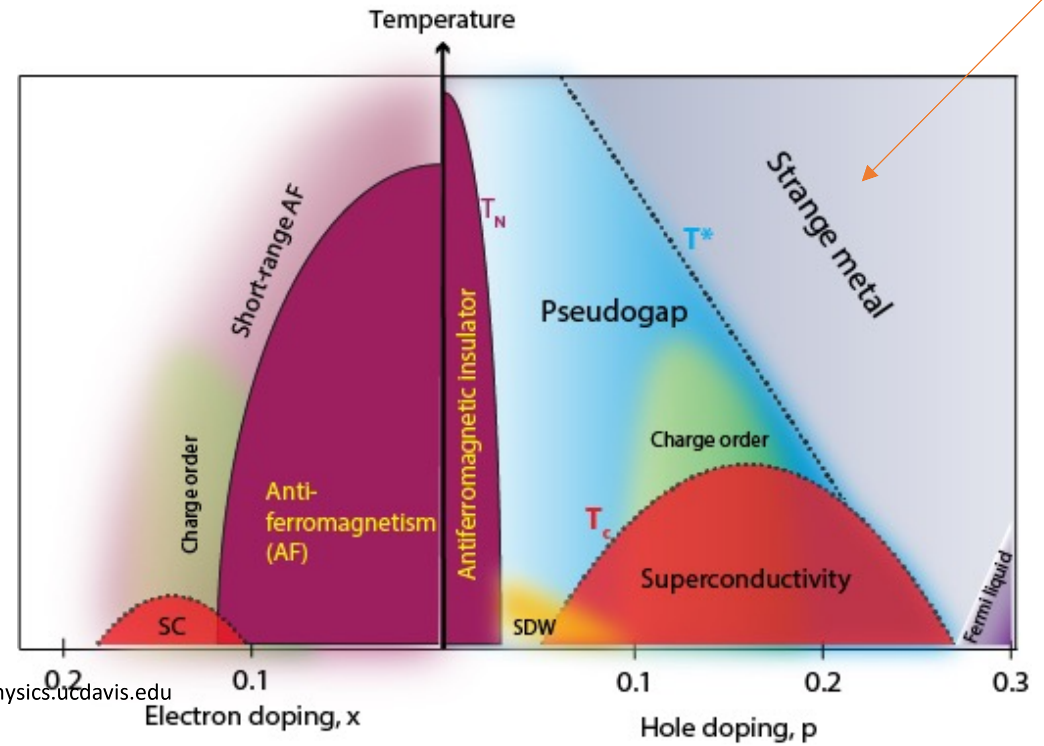
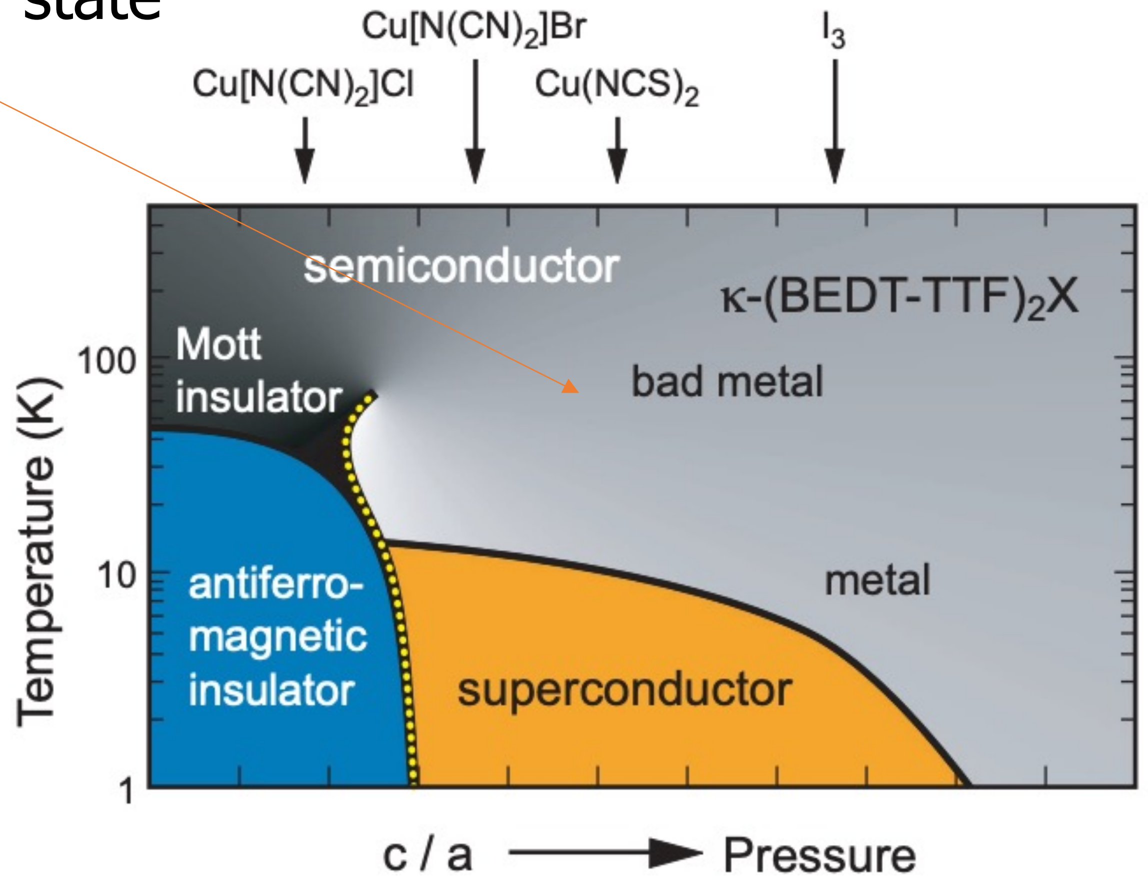


Fig: cme.physics.ucdavis.edu

High-Tc superconducting cuprates



Organic superconductors

Many effects also in the Density of States measured by Scanning Tunnelling microscope (STM) and in other probes

Mott insulator and local moments

Having a metal when we expect an insulator (even without symmetry breaking)

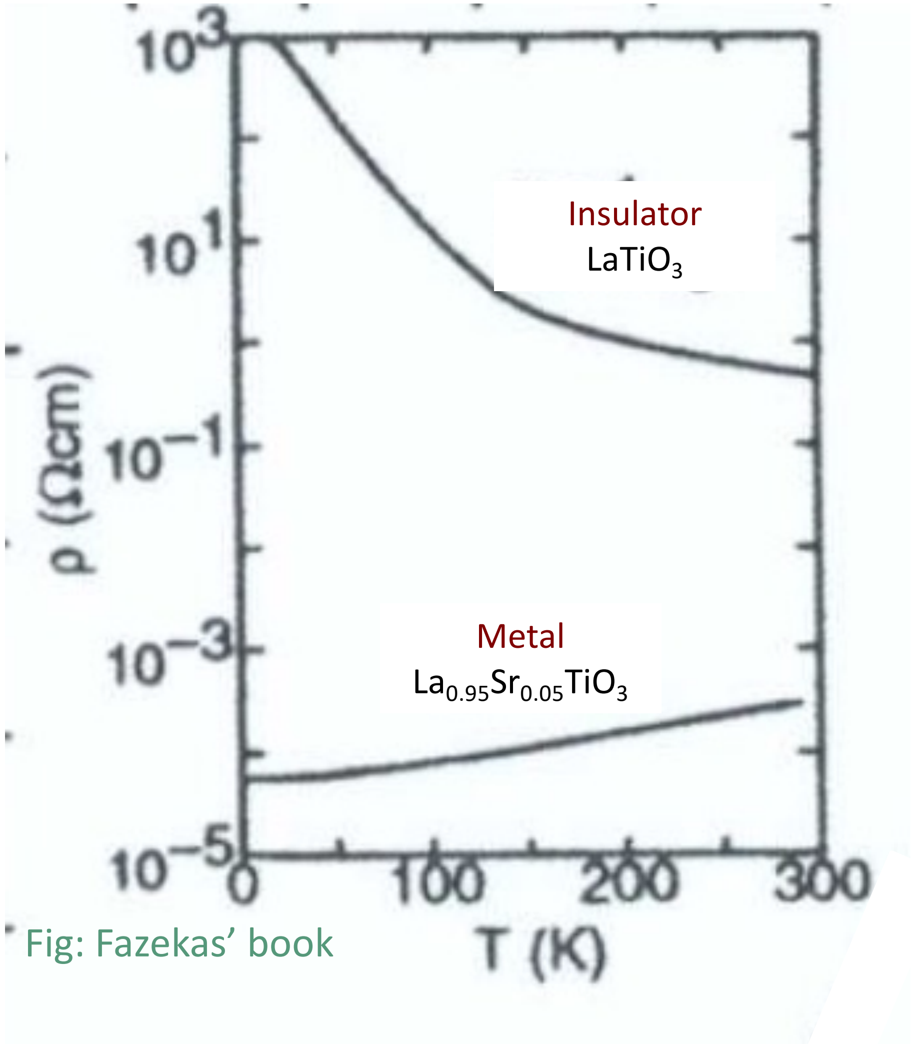
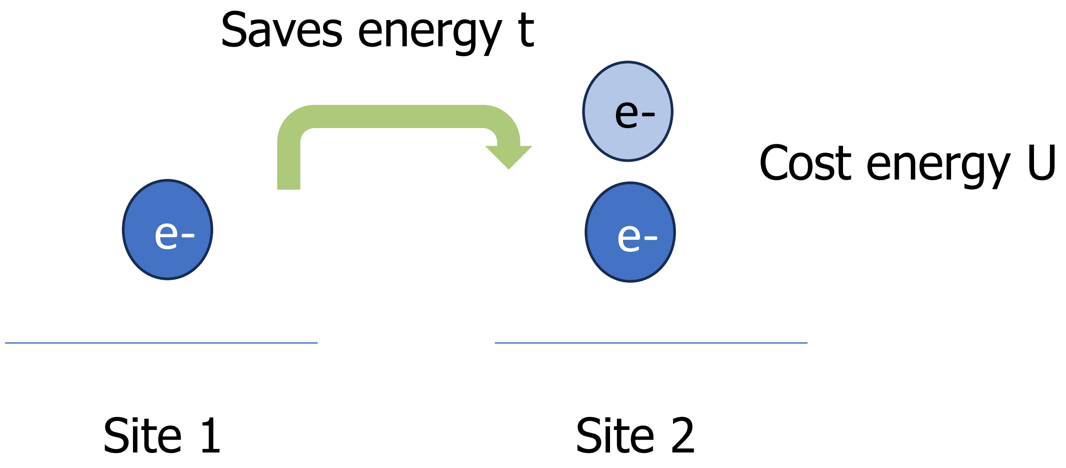


Fig: Fazekas' book



Onsite repulsión U

Hopping energy t

→ Tunnelling suppressed if U>>t

↓

Localization of electrons

↓

Electrons behave as local moments ↗

Local moments and itinerant electrons

Weakly correlated system

Itinerant electrons

Momentum-space

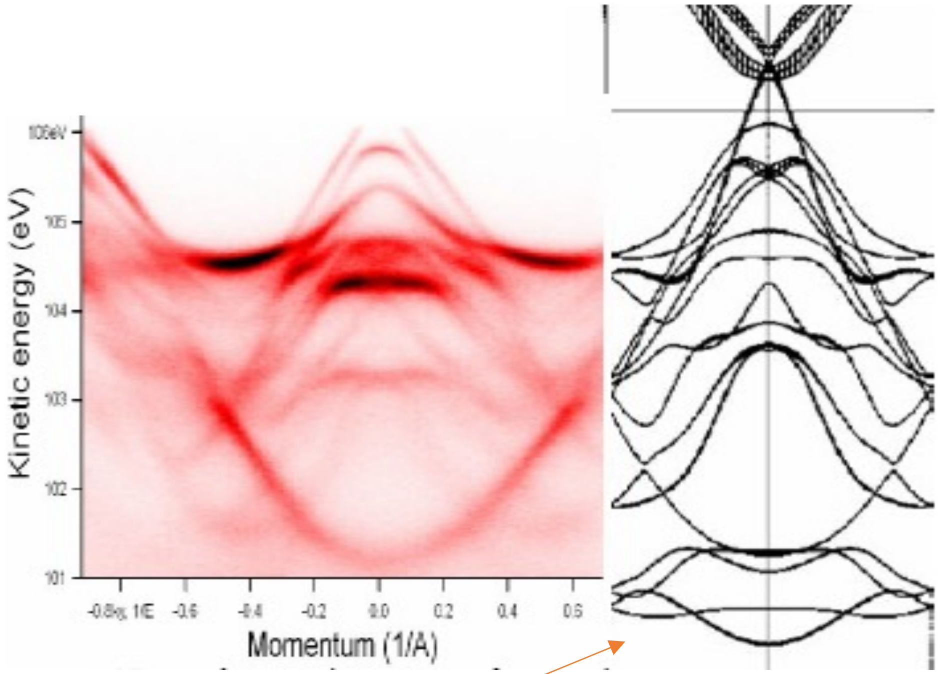


Fig. : Evtushinksy lectures

Band description

Strongly correlated system

Local moments

Real-space

Site 1 Site 2

Charge fluctuations are suppressed

Heisenberg like models ….

Strong modification of the spectrum & not always a binary choice:
Strongly correlated (non-ordered) metals

When different types of electrons play a role

Itinerant electrons

less correlated (larger kinetic energy
or smaller interactions)

+

Local electrons: local moments

More correlated (smaller kinetic energy
or larger interactions)

Kondo effect

Screening of a local moment
by a bath of itinerant electrons
(macroscopic measurable effects)

Heavy fermions

A lattice of local electrons
coupled through tunneling to
Itinerant electrons

Also: Hund's physics in multi-orbital systems ...

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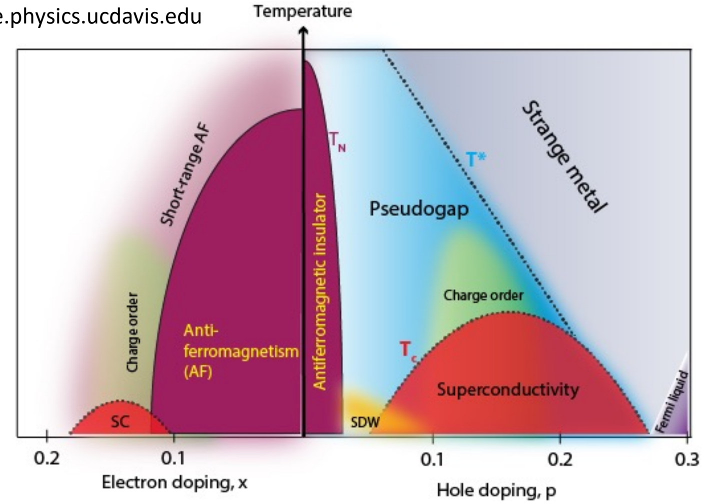
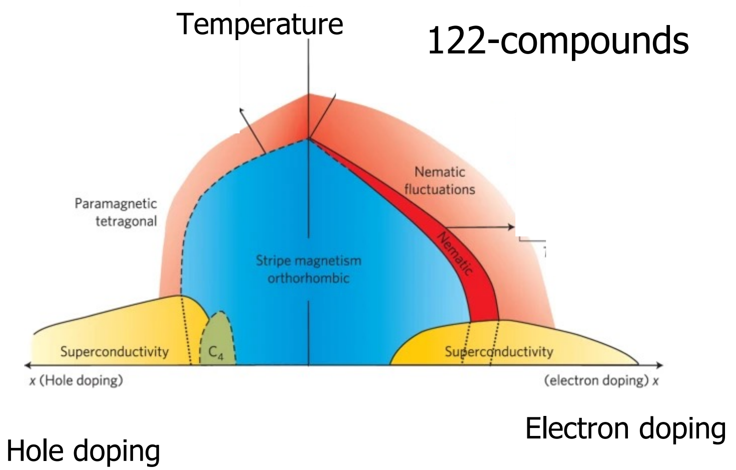
Superconductivity in strongly correlated electron systems

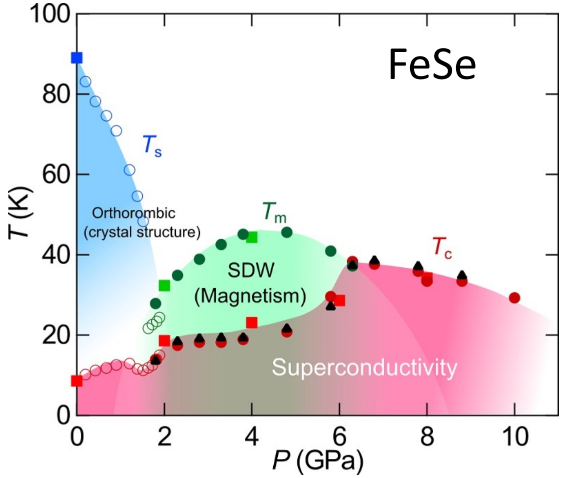
High-Tc superconducting cuprates

Fig: cme.physics.ucdavis.edu

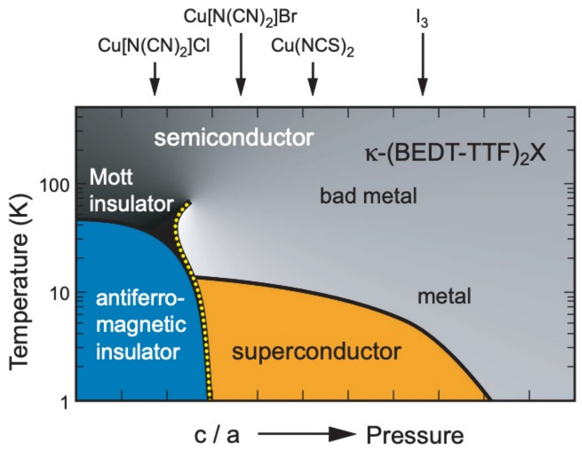


Iron superconductors

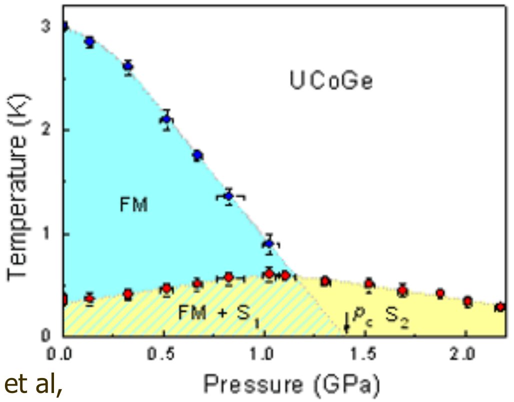




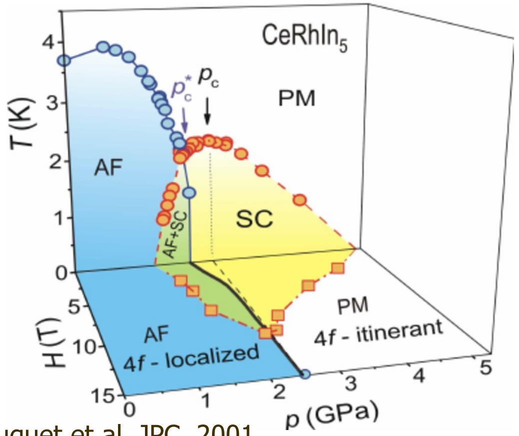
Organic superconductors



Heavy fermion compounds

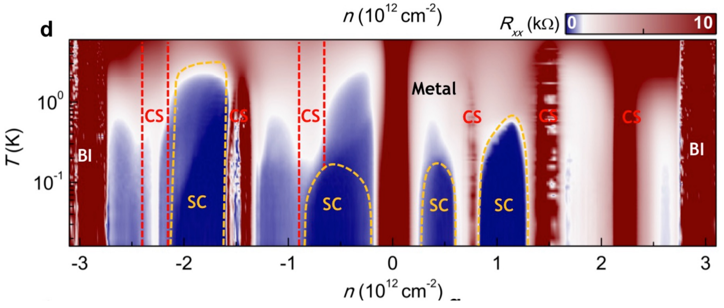


Slooten et al,
PRL 103 , 097003(2009)



Flouquet et al, JPC, 2001

Twisted bilayer graphene



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From a Mott insulator to high-temperature superconductivity

High-Tc superconducting cuprates

Which is the role of Mott (local) physics and strong correlations in high temperature superconductivity?

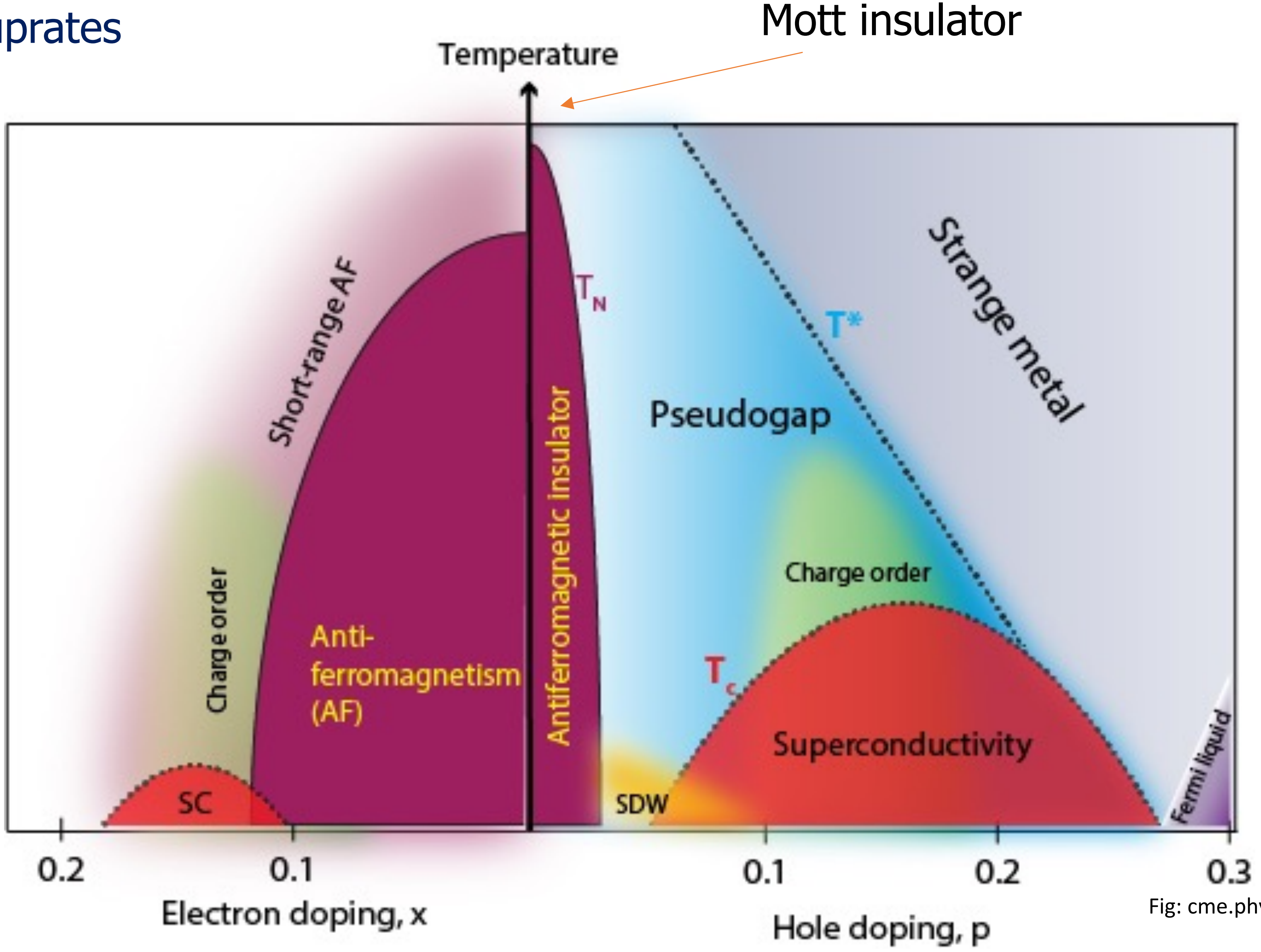

Fig: cme.physics.ucdavis.edu

Quantum materials: lattice and dimensionality

3 dimensions

©Michael Paraskevas

2 dimensions

©Sebastian Dubiel

1 dimension

©Carlos Valenzuela

Interactions are never perturbative in 1 dimension

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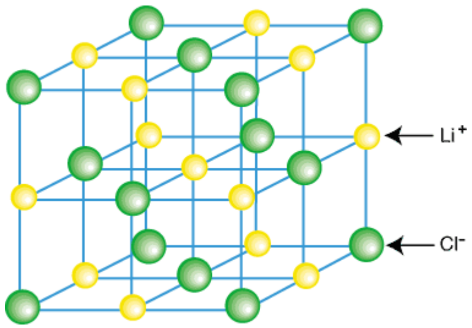
Quantum materials: lattice and dimensionality

3 dimensions



©askiitians

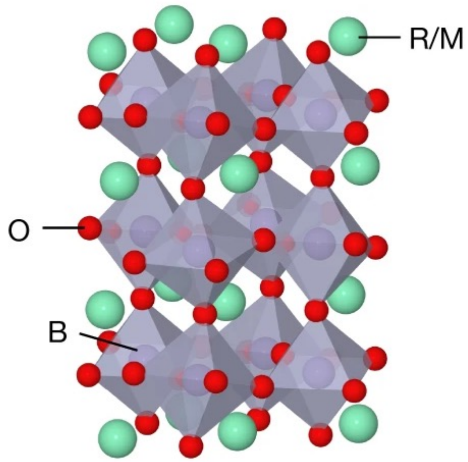
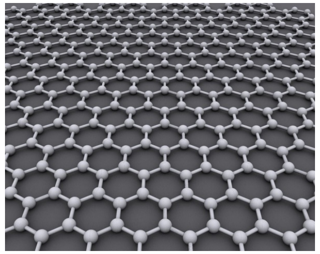


Fig: Guzmán-Verri et al, Nature 576, 429 (2019)

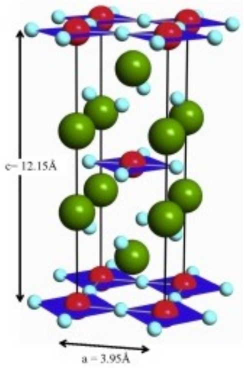
ABO_3 perovskite

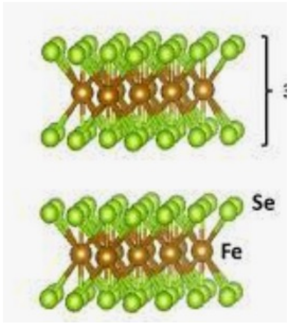
2 dimensions



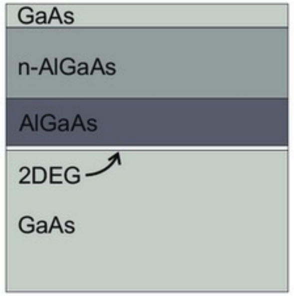
1 or
few layers

Layered quasi-2d materials

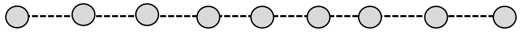




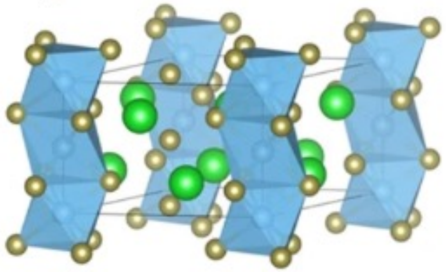
Interfaces

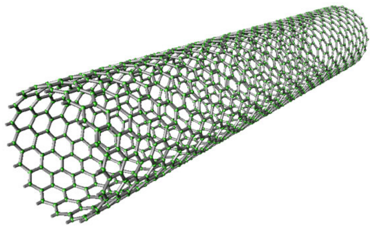


1 dimension



Quasi-1d materials





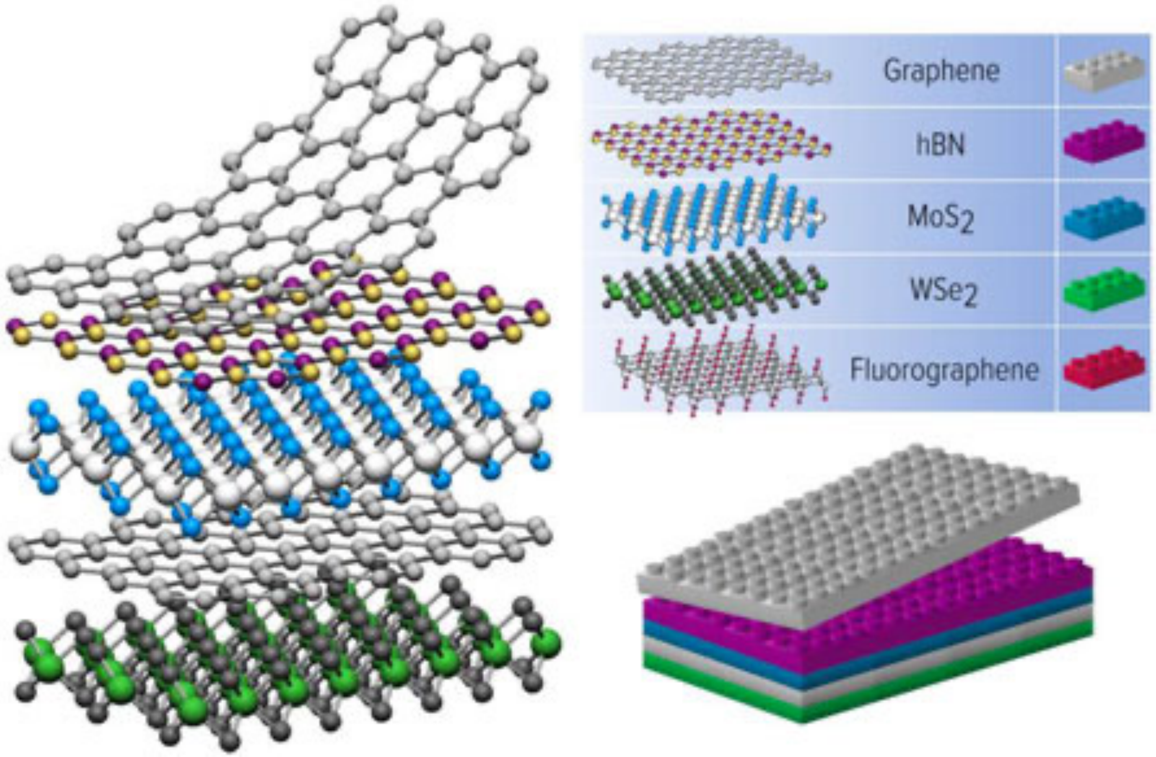
Interactions are never perturbative
in 1 dimension

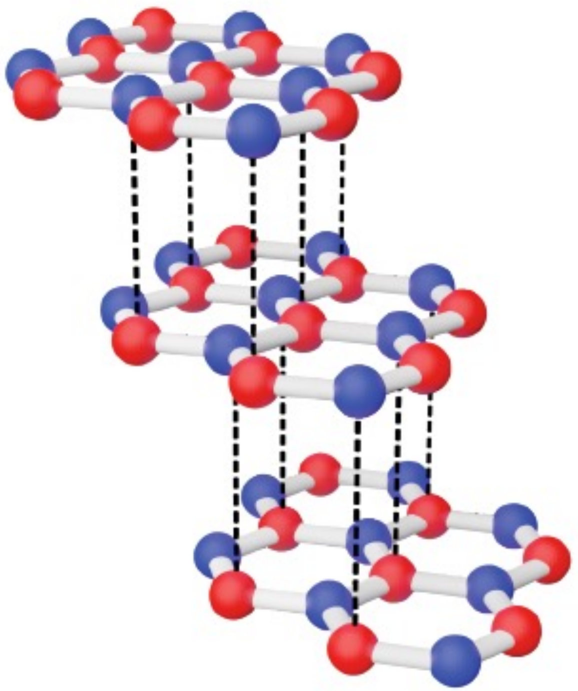
leni.bascones@csic.es

Engineering 2d materials and heterostructures

Stacking different 2d materials

Multilayers with specific stacking

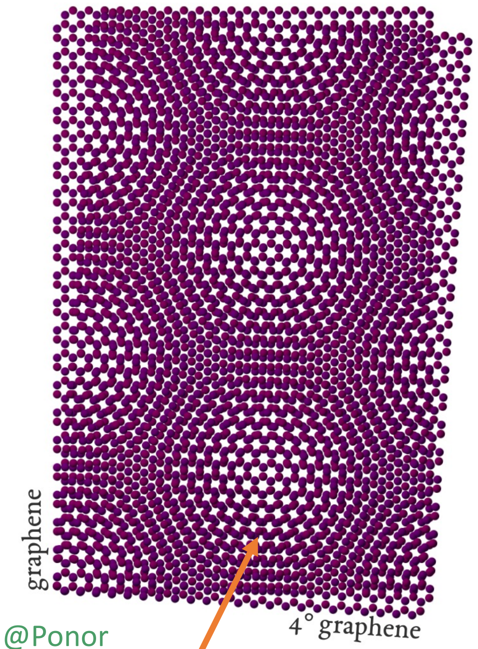




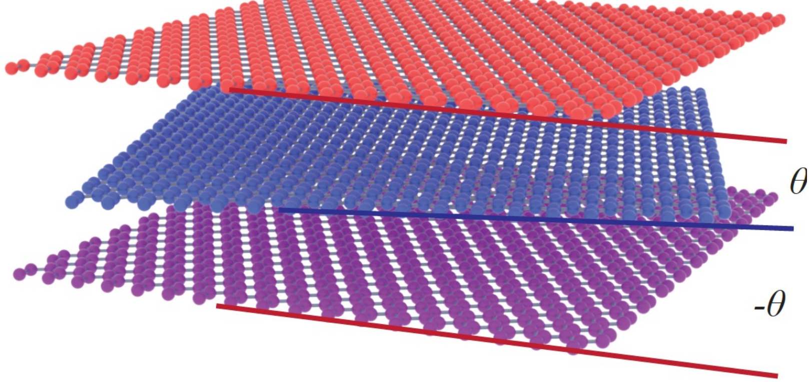
Geim & Grigorieva,
Nature 499, 419 (2013)

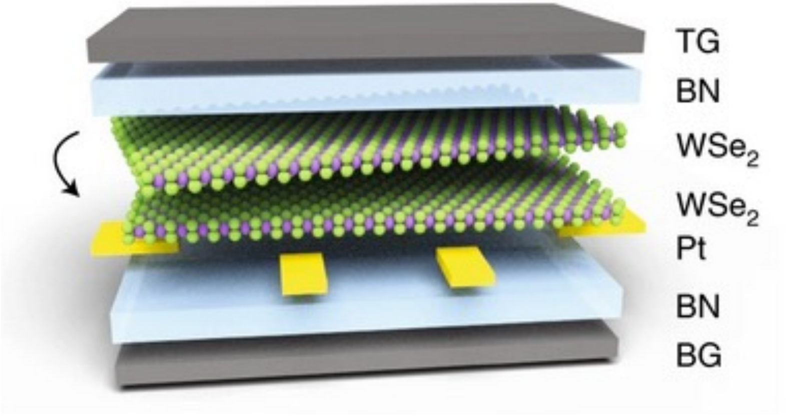
Semimetal, insulating,
metal, ferromagnetic …

Engineering 2d moiré heterostructures. Tunability



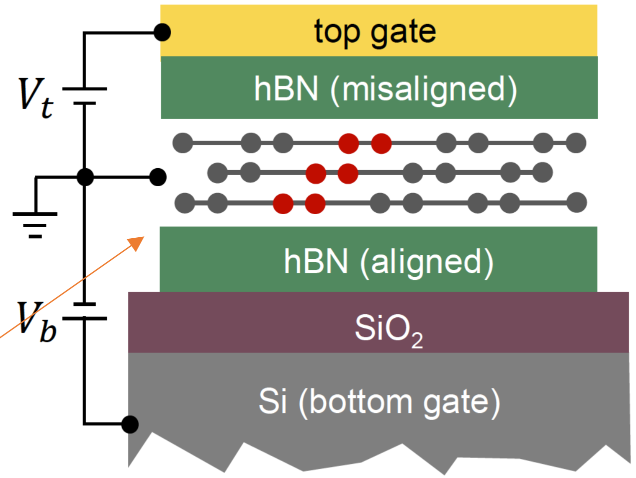
@Ponor





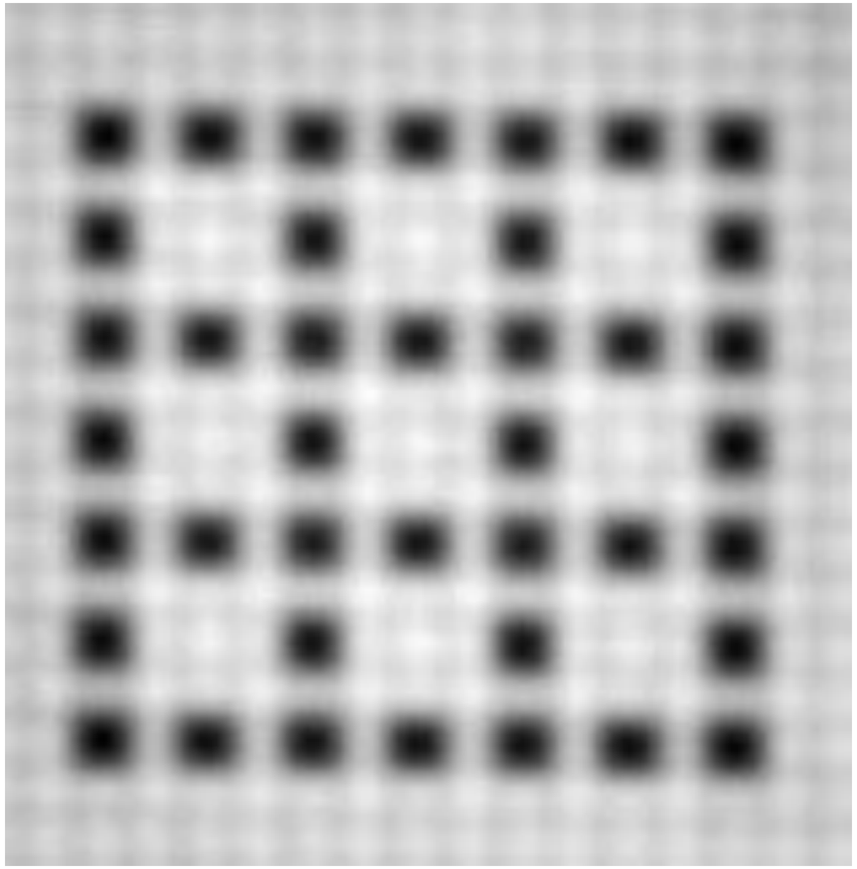
Moiré pattern with
a much larger unit cell

Variety of twisting arrangements with equal or different layers: graphene, dichalcogenides, magnetic 2D materials …



Moiré also through slight mismatch
or through ad-hoc engineered periodic potential

Engineering 2d lattices: atoms on surfaces



Yan & Liljeroth,
Advances in Physics X, 4, 2019

Emergence of Quantum Phases in Novel Materials

Interactions between many particles (electrons)

↓

Correlated Quantum States
in many different and novel materials

↙ ↘

Symmetry breaking states ⟷ Formation of local moments

Tuning parameters for correlated states

Interaction energy vs Kinetic energy

- ❑ Filling of electronic bands
- ❑ Electronic bands details
- ❑ Dimensionality
- ❑ Crystalline lattice
- ❑ Screening
- ❑ External knobs (pressure, …)
- ❑ Interaction with lattice
- ❑ Other (spin-orbit, …)

Active electrons for correlated states

- ❑ Ion= nucleus + core electrons
- ❑ **Focus on electrons close to Fermi level**

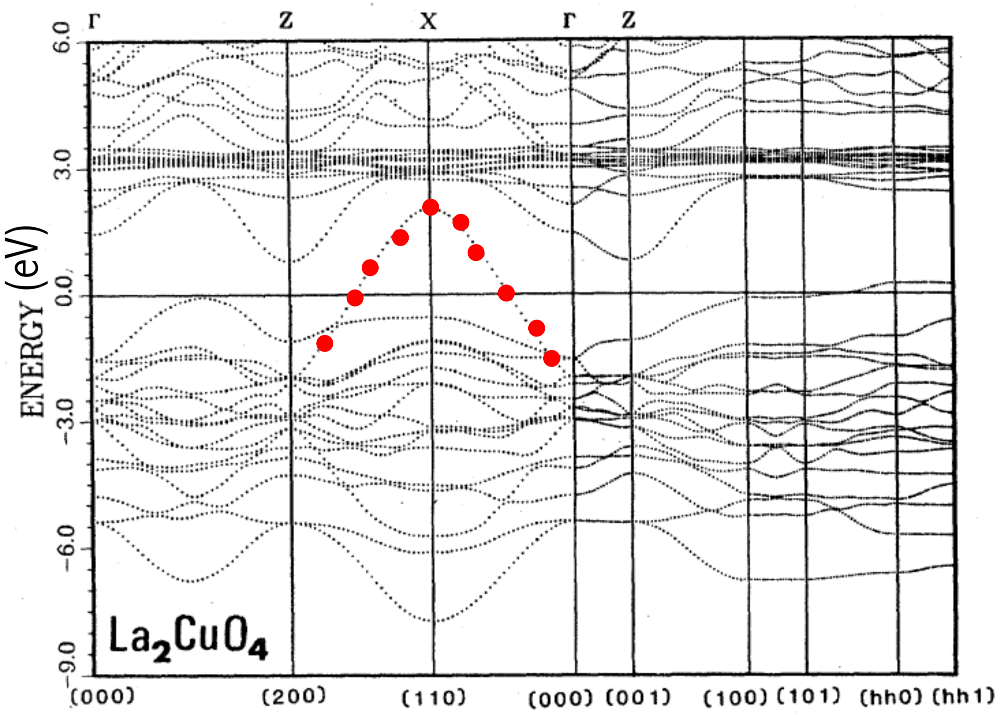
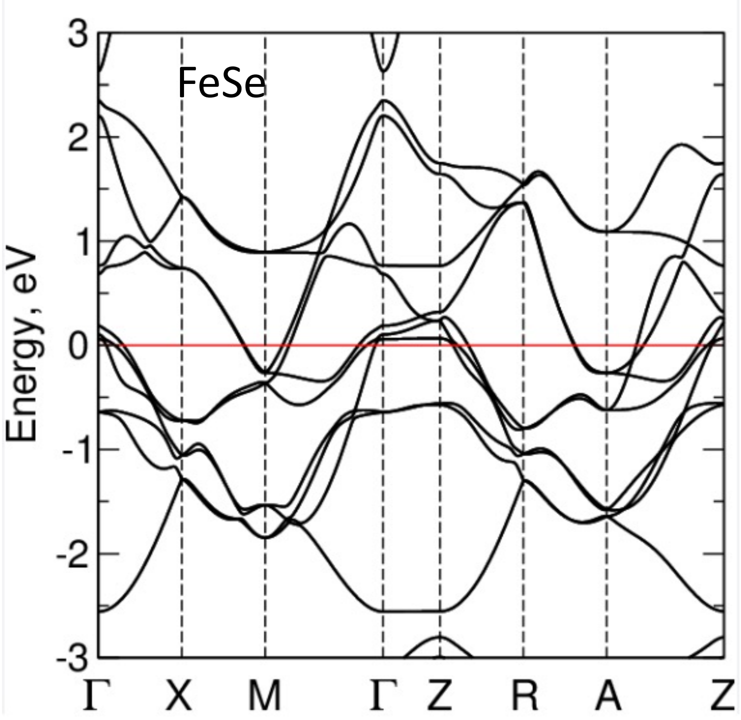
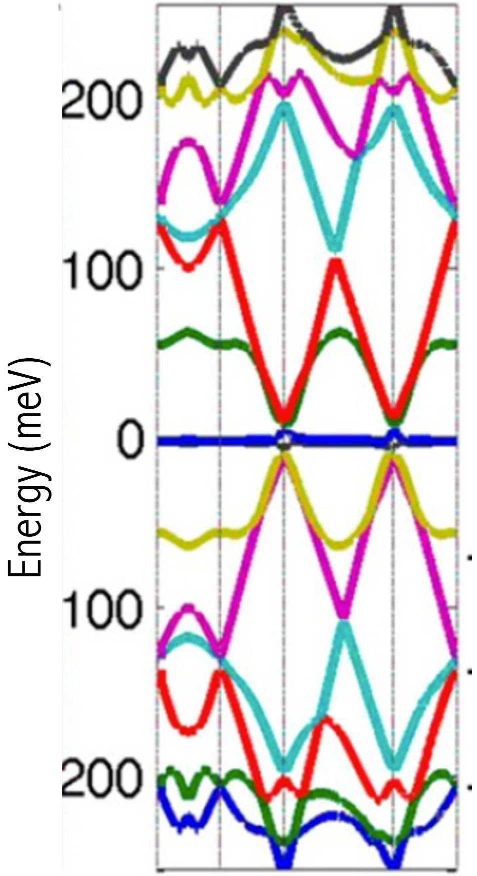


Fig: Pickett, RMP 61, 433 (1989)



Grechnev et al, Ukran. J. Phys 57, 171 (2012)



TBG $\theta = 1.05°$

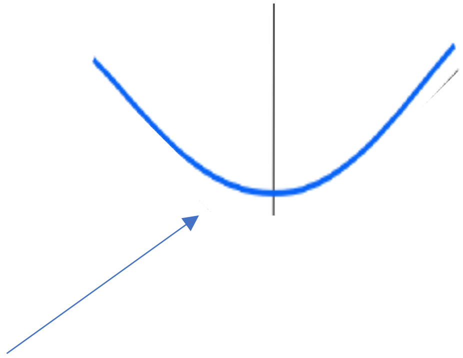
Bistritzer & MacDonald,
PNAS 108, 12233 (2011),

Kinetic energy: lattice versus continuum

Free space: Parabolic band

- Isotropic
- No length scale
- Momentum conserved

Kinetic energy = $\frac{k^2}{2m}$



$m^{-1} = |\partial^2\varepsilon/\partial k^2|$

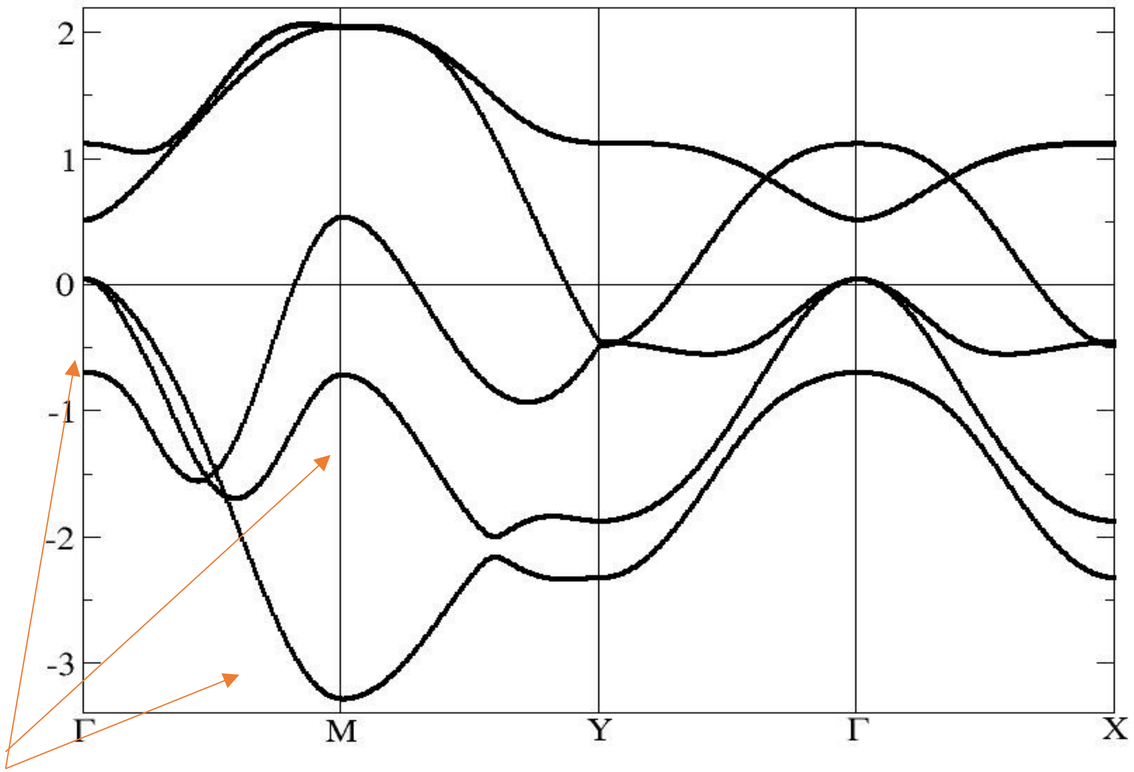
Inverse of second derivative of the energy gives the mass of the electron

Kinetic energy: lattice versus continuum

In a solid:

Periodic potential of the ionic lattice

- Length scale: the lattice constant **a**
- Momentum conserved modulus **2π/a**
- In general, anisotropic potential
- Energy bands filled up to ε_F. Fermi surface



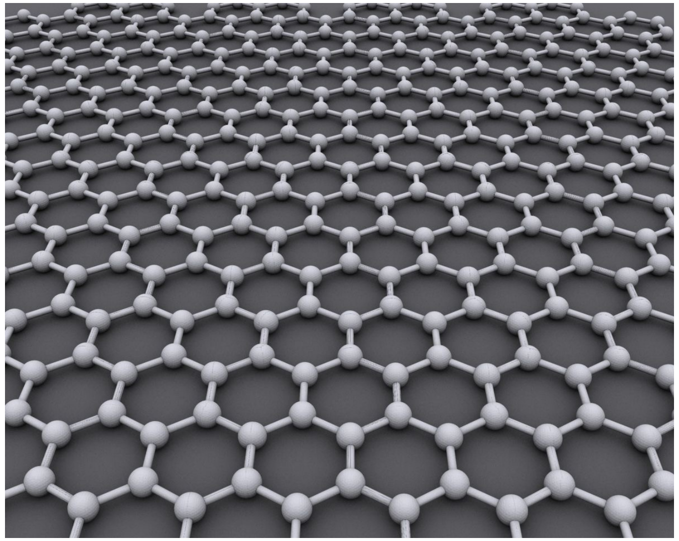
Approximated to parabolic band very close to the bottom/top

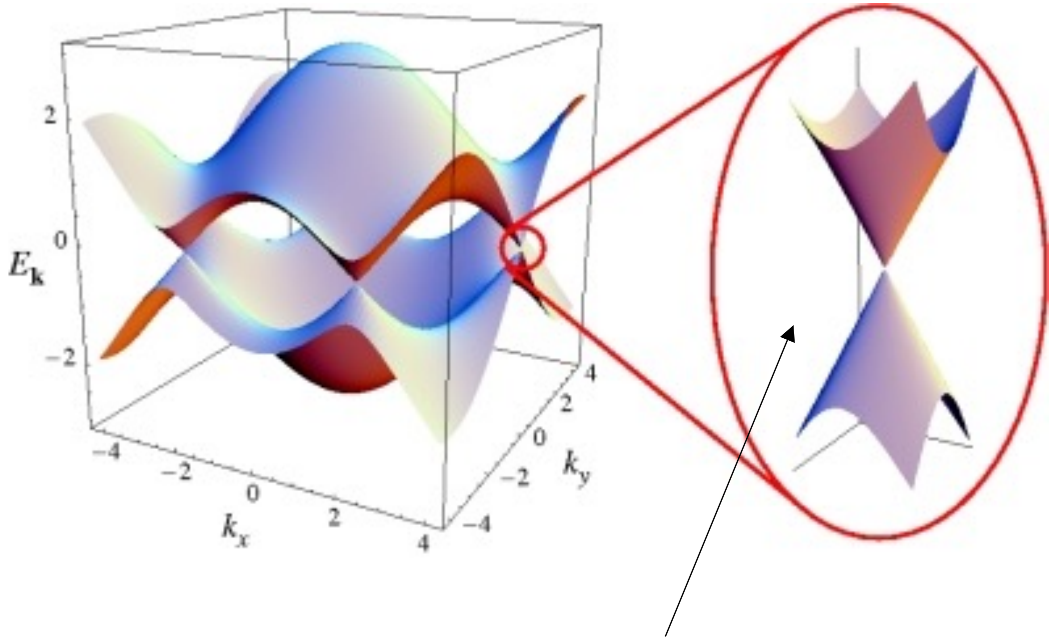
Fig: Calderón et al, PRB, 80, 094531 (2009)

$$m^{-1} = |\partial^2\varepsilon/\partial k^2|$$

Effective mass

Kinetic energy: massless fermions





Linear Dirac dispersion

$$m^{-1} = |\partial^2\varepsilon/\partial k^2| = 0$$

Effective mass vanishes

massless fermions

Kinetic energy: lattice versus continuum

In a solid:

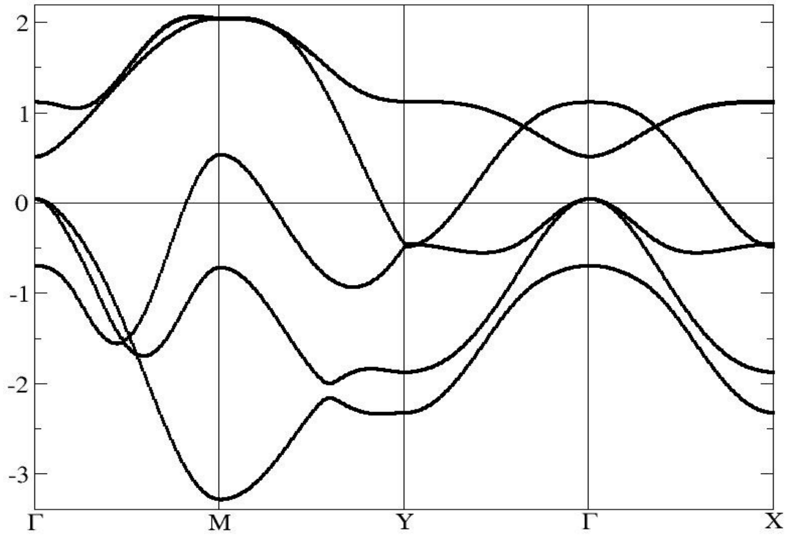
Periodic potential of the ionic lattice

- Length scale: the lattice constant **a**
- Momentum conserved modulus **2π/a**
- In general, anisotropic potential
- Energy bands filled up to ε_F. Fermi surface

Free space:

- Isotropic
- No length scale
- Momentum conserved

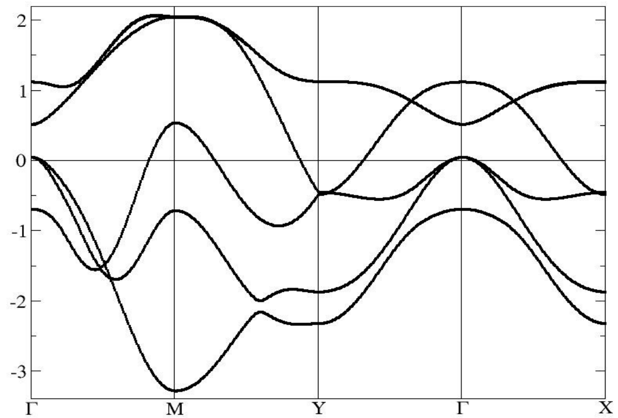
$$\frac{k^2}{2m}$$



Kinetic energy and bandwidth

Kinetic energy: $\int_{-\infty}^{\varepsilon_F} \varepsilon \, N(\varepsilon) \, d\varepsilon$

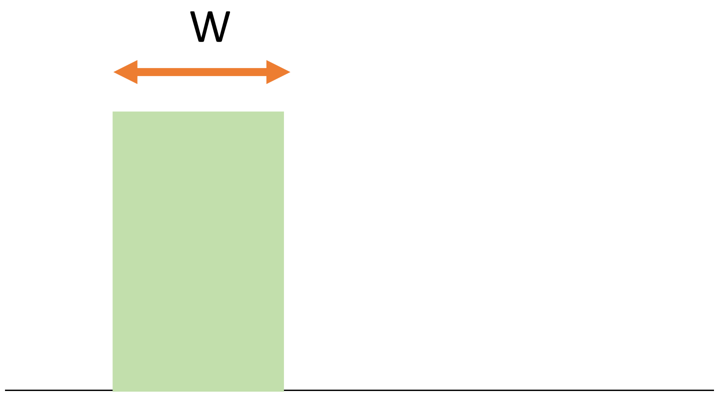
N(ε): density of states

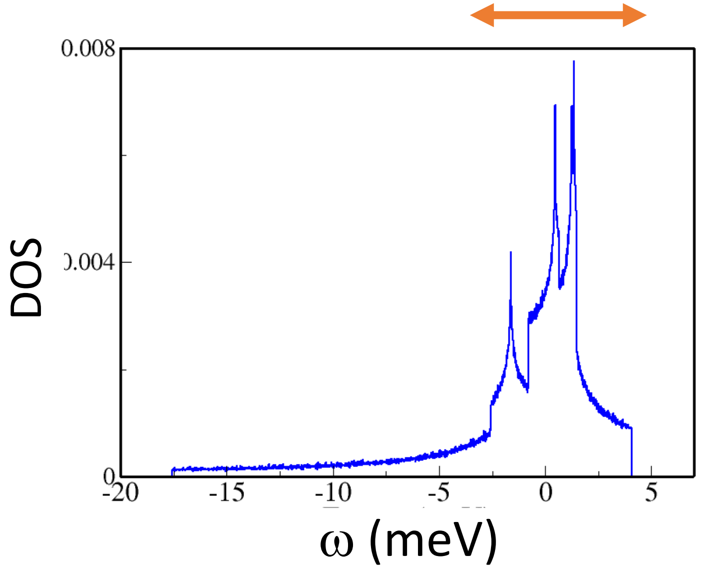


N(ε): constant in [-W/2, W/2], zero otherwise → Kinetic energy ∝ W

W: bandwidth

Flat bands very sensitive to interactions





leni.bascones@csic.es

Kinetic energy and bandwidth

Kinetic energy: $\int_{-\infty}^{\varepsilon_F} \varepsilon\, N(\varepsilon)\, d\varepsilon$

$N(\varepsilon)$: density of states

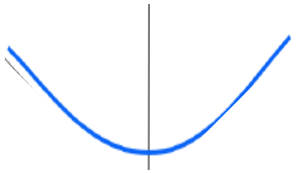
$N(\varepsilon)$: constant in $[-W/2, W/2]$, zero otherwise → Kinetic energy $\propto$ W

W: bandwidth

Flat bands very sensitive to interactions

Larger mass → Narrower band → Smaller bandwidth W → **Smaller Kinetic energy**

$\frac{k^2}{2m}$



Careful! Not applicable to Dirac dispersion

Kinetic energy and bandwidth

Kinetic energy ~Bandwidth W

W ~ eV in most materials

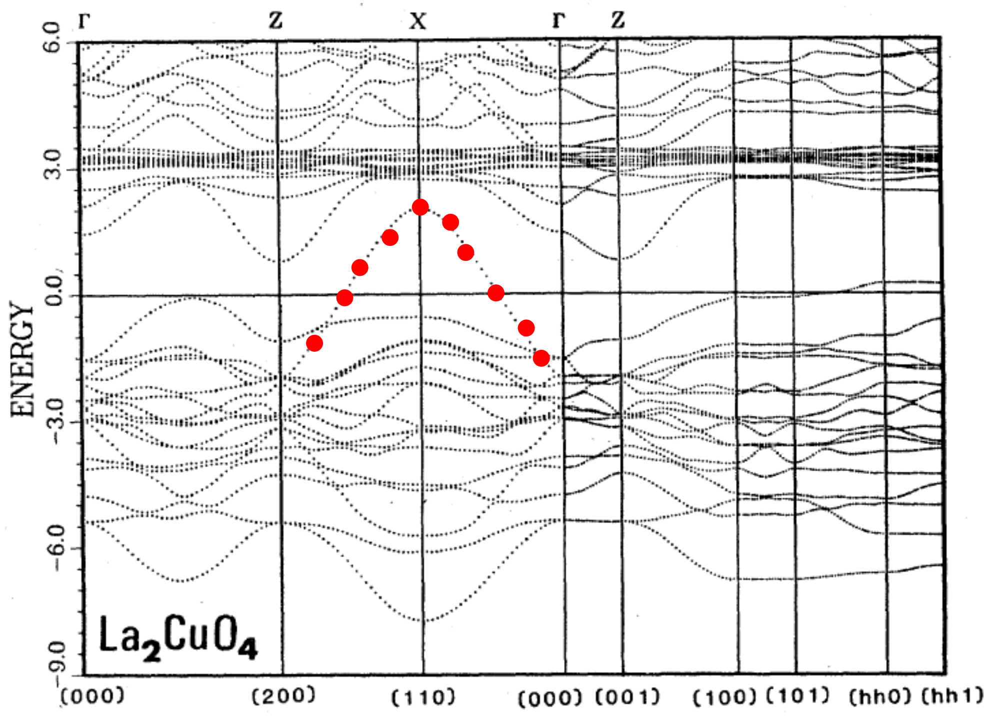
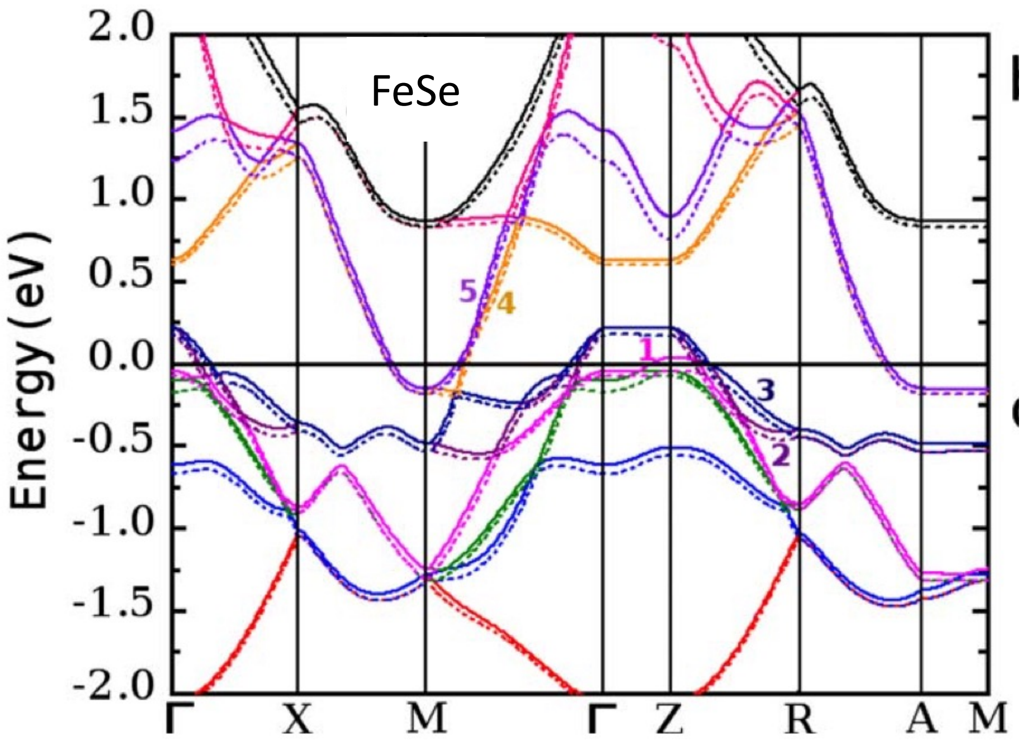
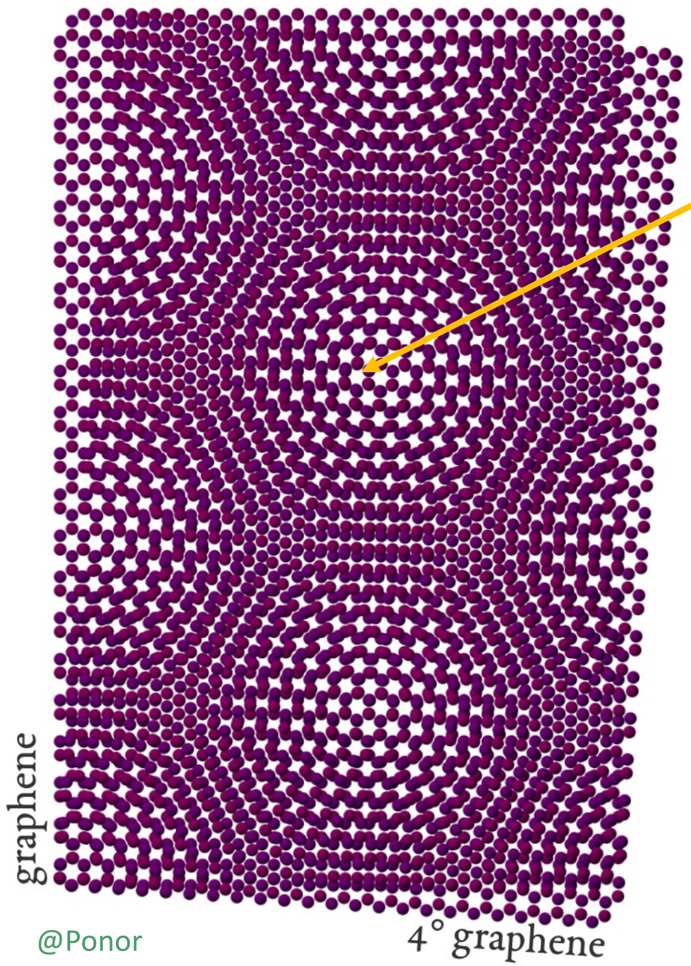


Fig: Pickett, RMP 61, 433 (1989)

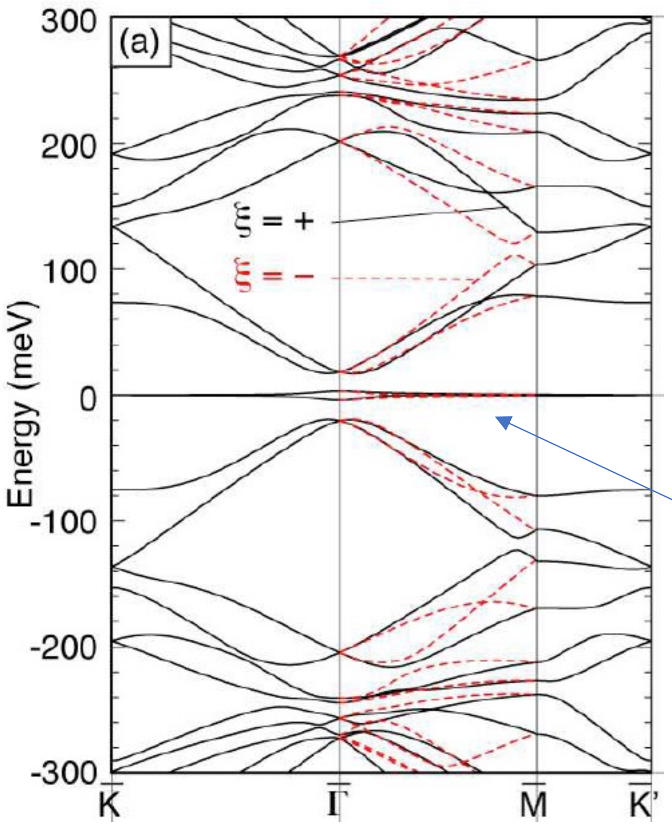


Ma & Lu, PRB 78, 033111 (2008)

Moiré heterostructures: flat bands



Moiré unit cell ~ 10 nm lattice constant



Very narrow bands for certain twist angles (bandwidth 1-10 meV)

Very sensitive to interactions

Strength of interactions

- Ratio between Interaction and Kinetic energies U/W

Kinetic energy ~Bandwidth W

W ~ eV in most materials

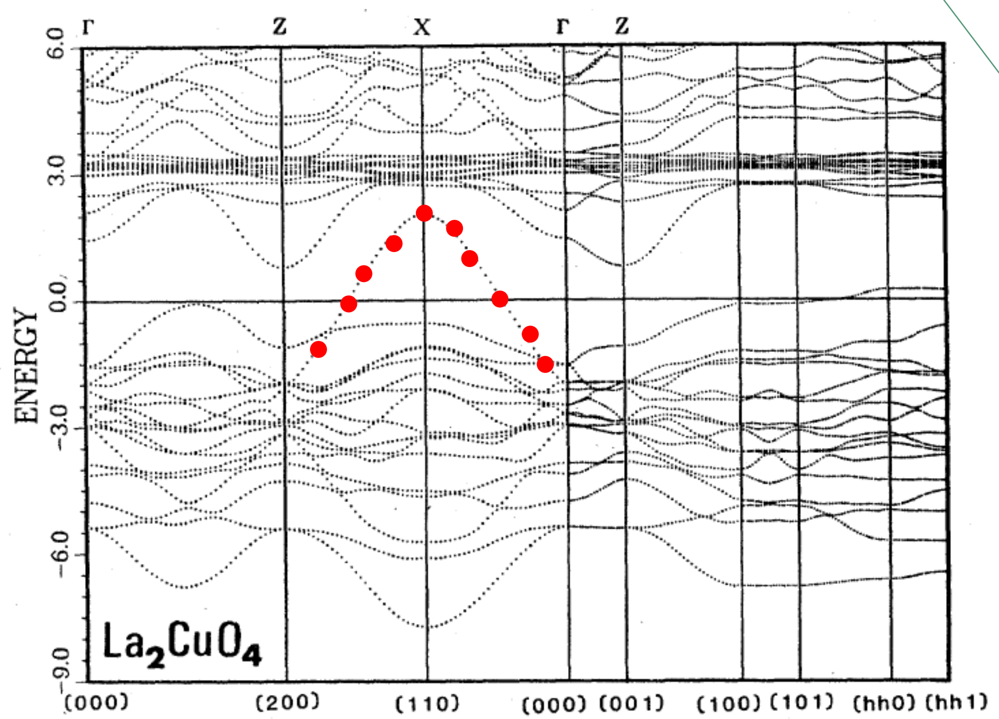


Fig: Pickett, RMP 61, 433 (1989)

Interaction energy:

$$U \sim \left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{1}{\varepsilon_r r_0} = 14.37\ \text{eV Å}\ \frac{1}{\varepsilon_r r_0}$$

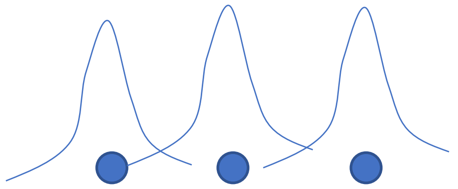
r_0 ~several Å in most materials

Similar energy scales

U/W not small

Strength of interactions

Bands: Tight binding model with atomic orbitals as a basis



Larger spread of wave function
(less localized orbitals, larger r_0)

Larger overlap between orbitals
in neighbouring sites
(larger kinetic energy & bandwidth)

Onsite repulsion
Two electrons in an atom less likely
to be found very close
(smaller effective interaction)

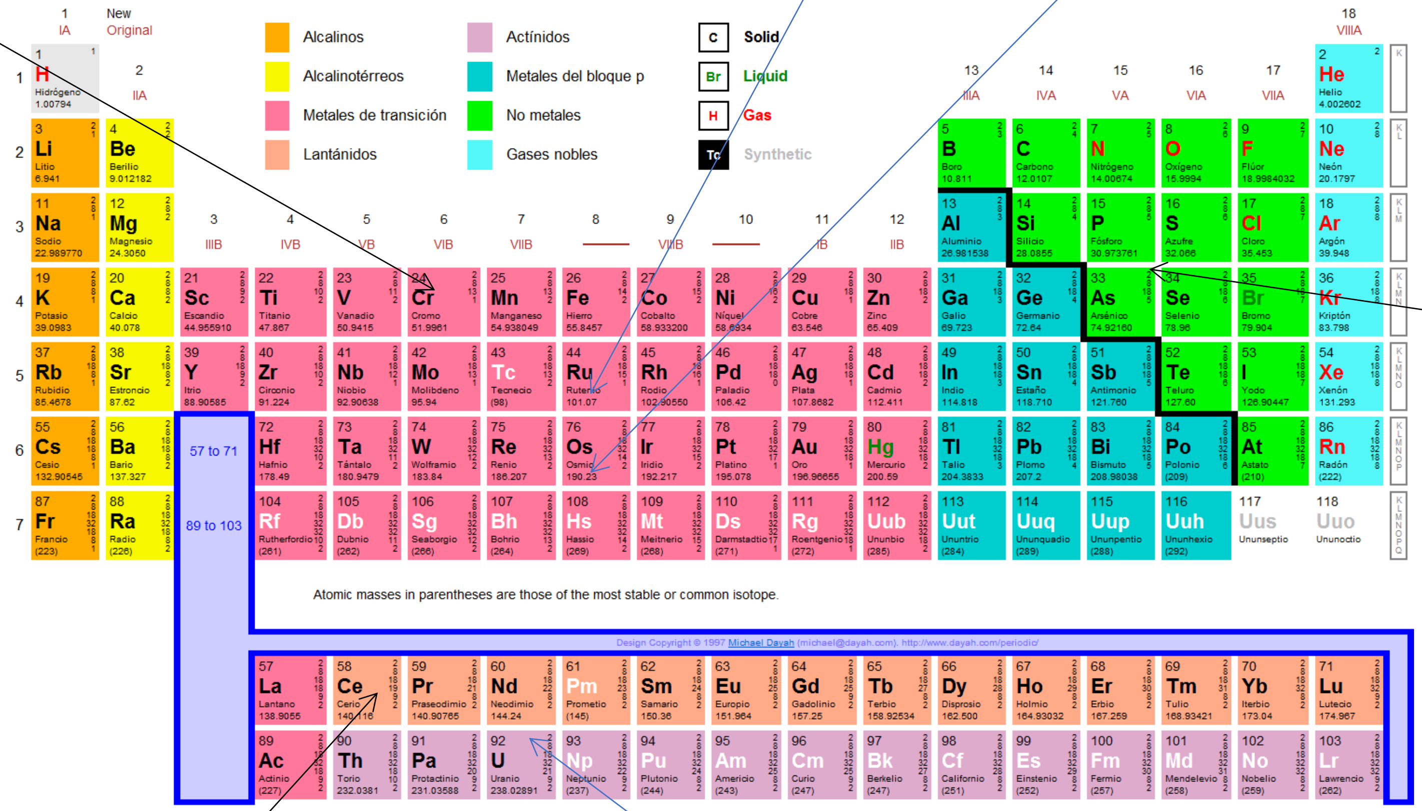
$$U \sim 14.37 \text{ eV} \frac{1}{\varepsilon_r \, r_0}$$

The type of atomic orbital matters

Strength of interactions.

3d: competition between kinetic energy & interaction

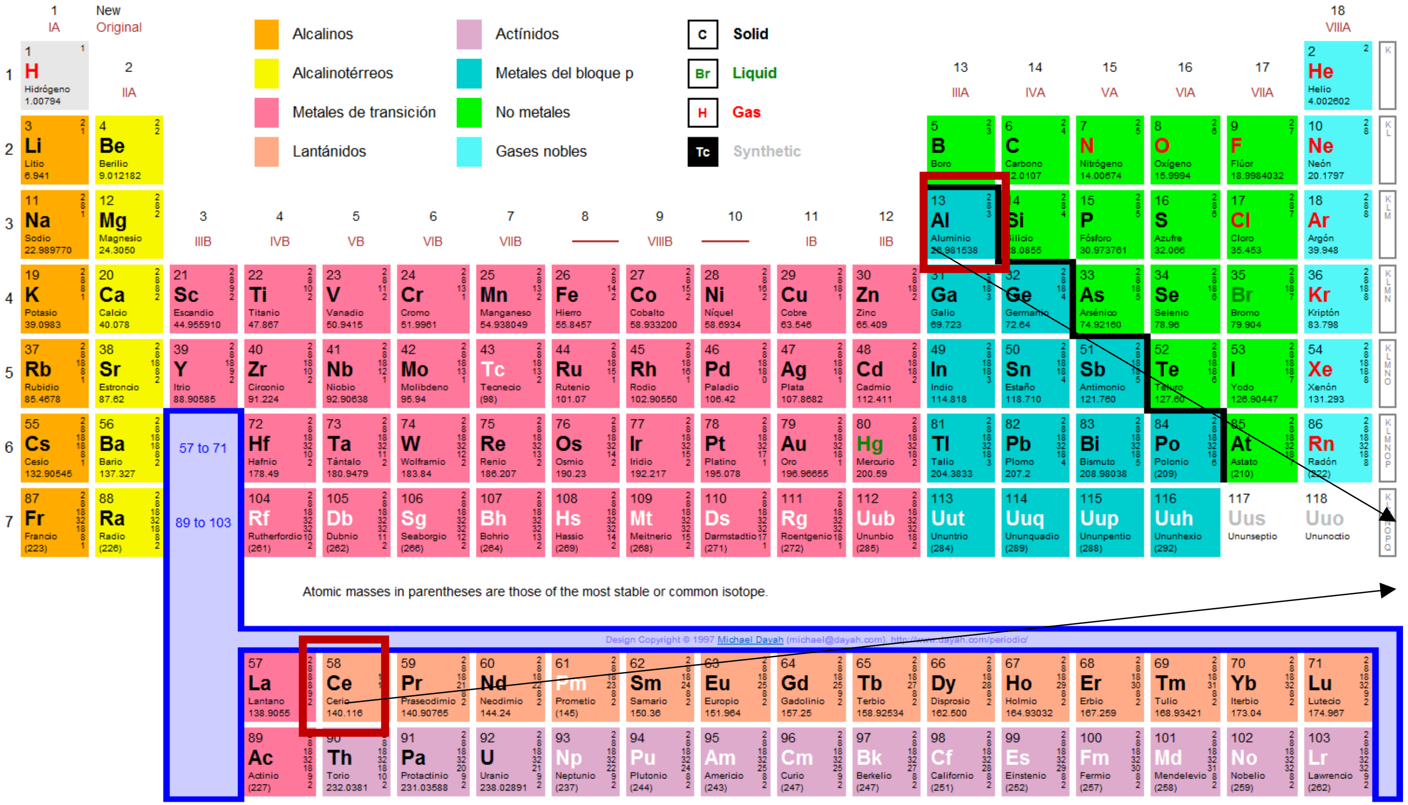
Interaction strength decreases in 4d & overall in 5d due to larger extension of the orbital



s & p electrons kinetic energy more important

4f overall and 5f orbitals are quite localized on a site. Very small kinetic energy. Interactions win

Strength of interactions.



$CeAl_3$

Itinerant + local electrons

Heavy fermion compound

Effective models

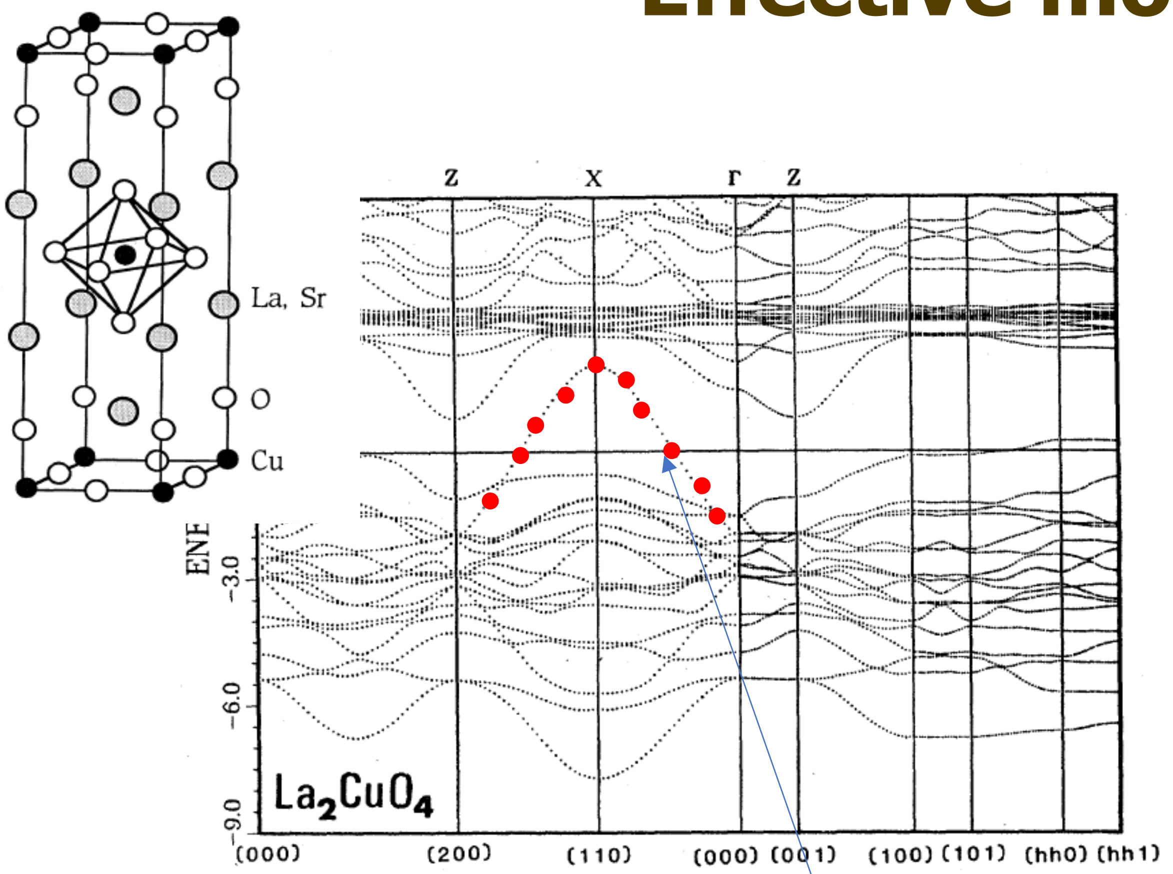


Fig: Pickett, RMP 61, 433 (1989)

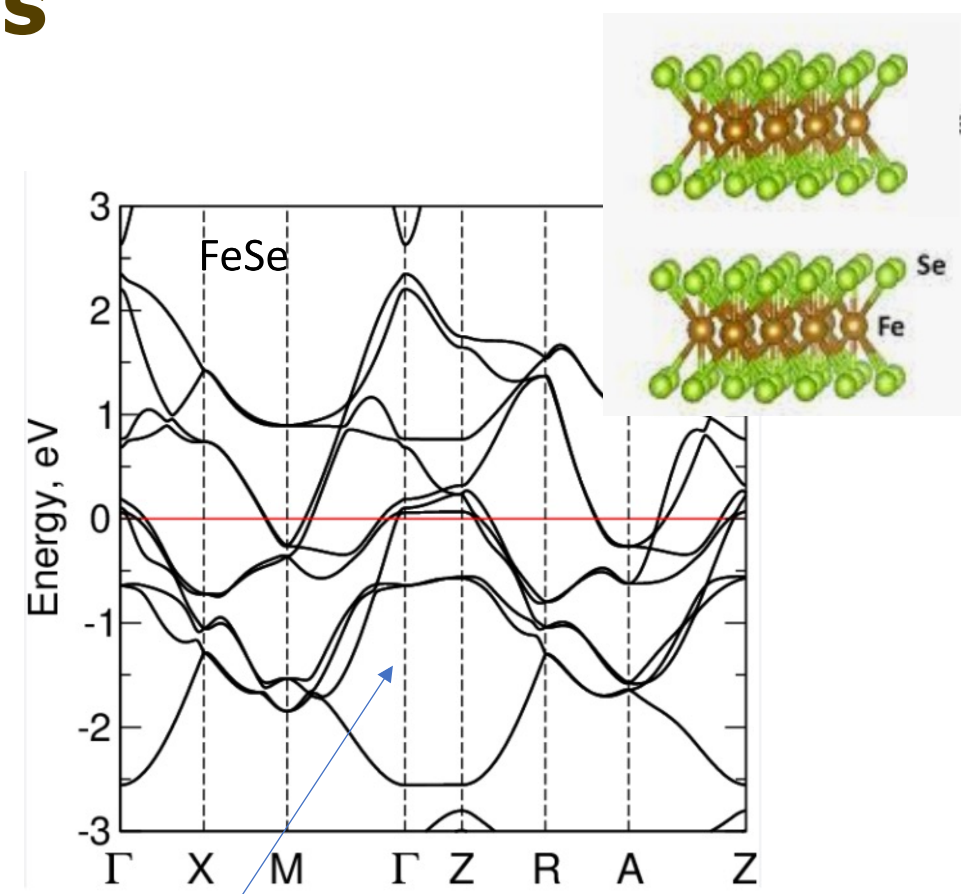


Fig: Grechnev et al, Ukran. J. Phys 57, 171 (2012)

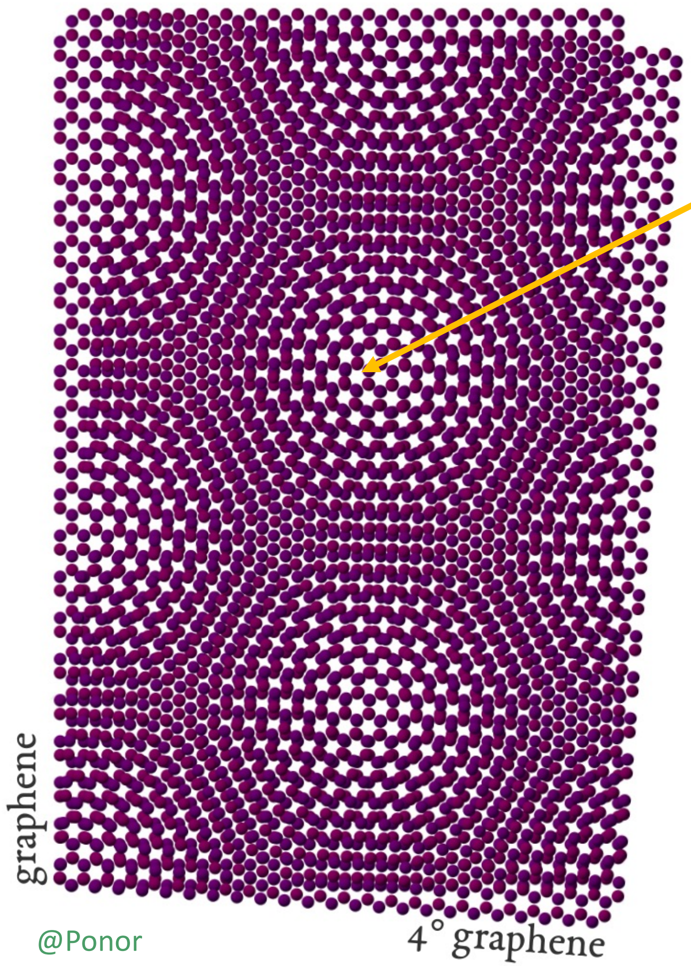
Effective model for Cu electrons close to the Fermi level

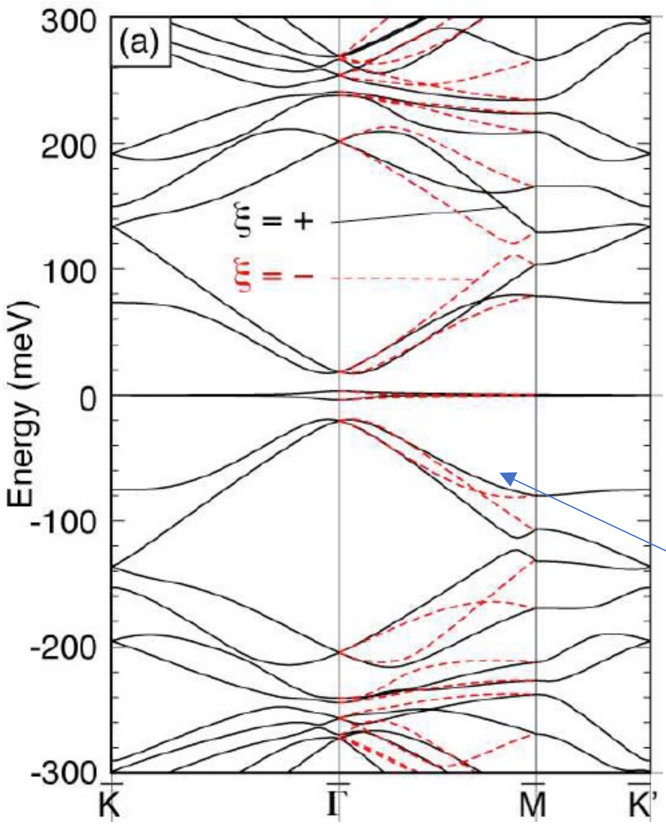
Effective model for Fe electrons close to the Fermi level

Moiré heterostructures

Moiré unit cell ~ 10 nm lattice constant

U ~ 30 meV → r_0 several nm





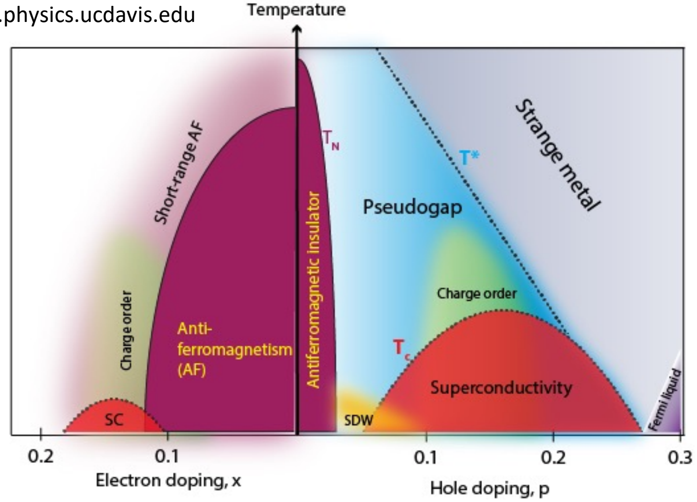
Very narrow bands for certain twist angles (bandwidth 1-10 meV)

Energy scales of the system → Temperature scale of correlated states

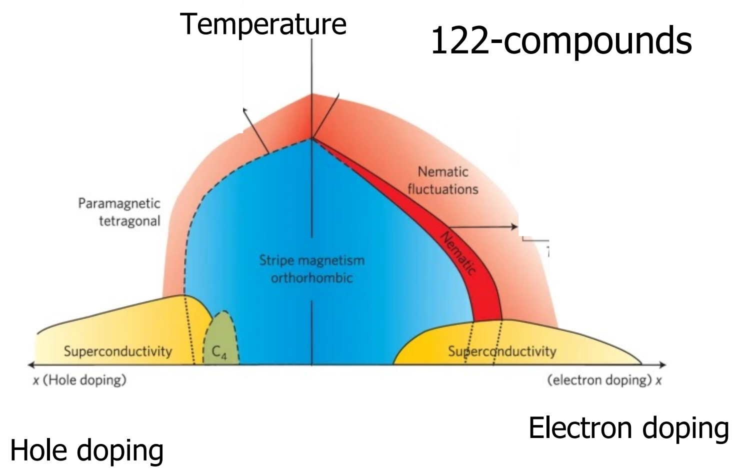
Superconductivity in strongly correlated electron systems

High-Tc superconducting cuprates

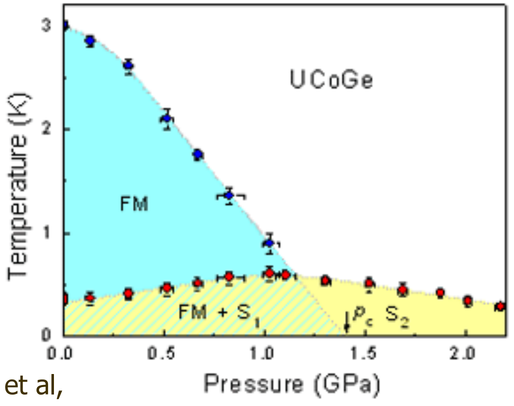
Fig: cme.physics.ucdavis.edu



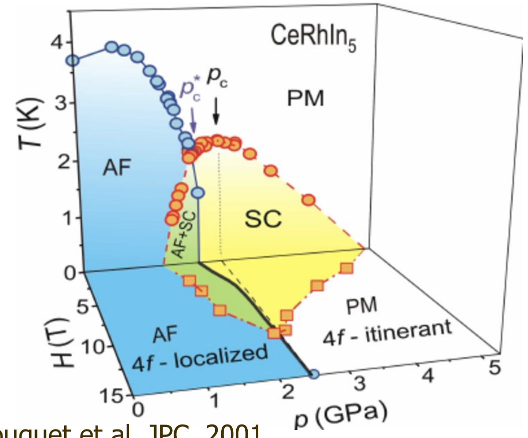
High-Tc iron based superconductors



Heavy fermion compounds

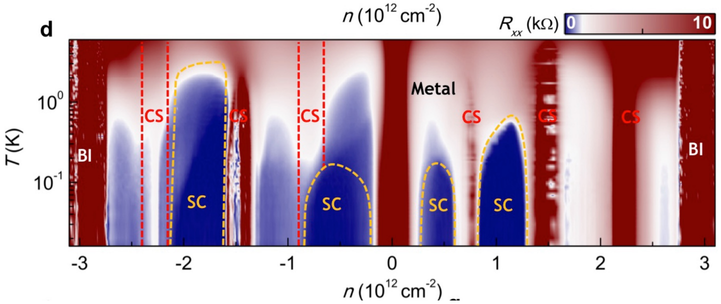


Slooten et al,
PRL 103 , 097003(2009)



Flouquet et al, JPC, 2001

Twisted bilayer graphene



Critical temperatures not small
with respect to the relevant energy scales

leni.bascones@csic.es

Metals and insulators

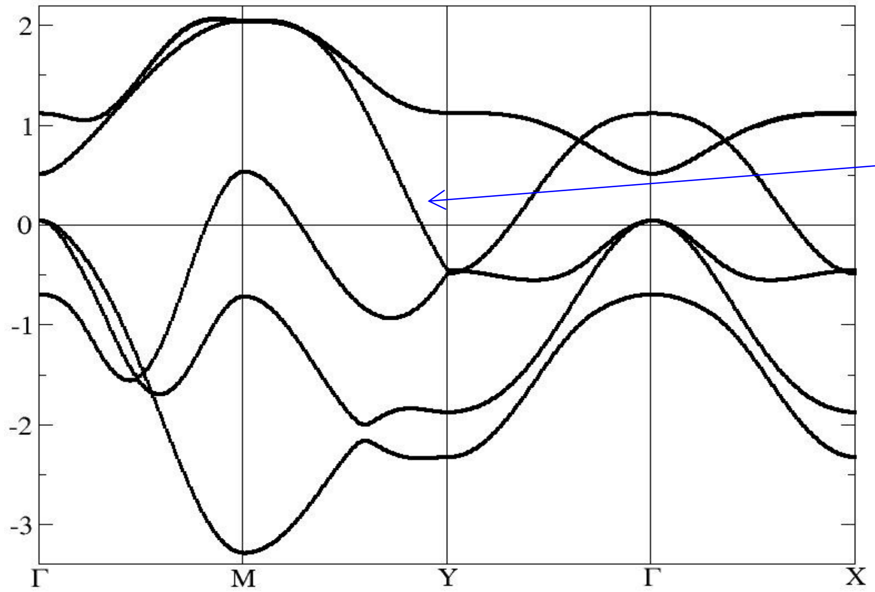


Fig: Calderón et al, PRB, 80, 094531 (2009)

Metallicity
in clean systems
Bands crossing the Fermi level
(finite Density of States (DOS) at E_F)

Insulating behaviour
in clean systems
Bands below
Fermi level filled

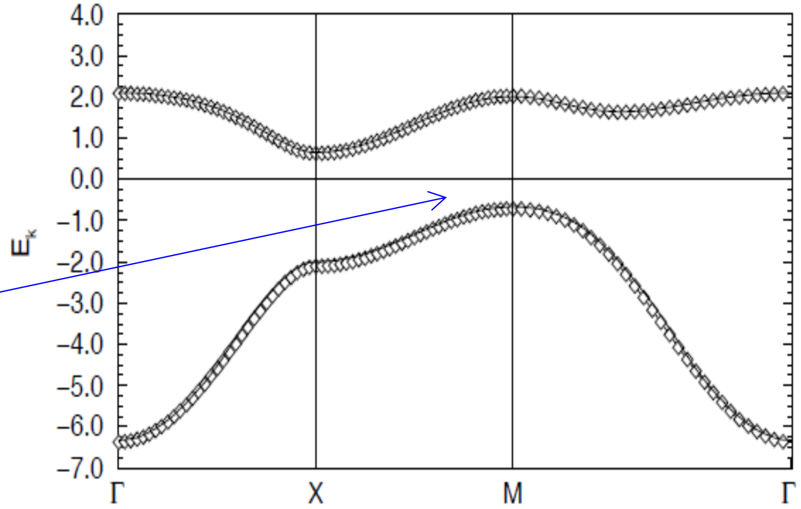


Fig: Hess & Serene, PRB 59, 15167 (1999)

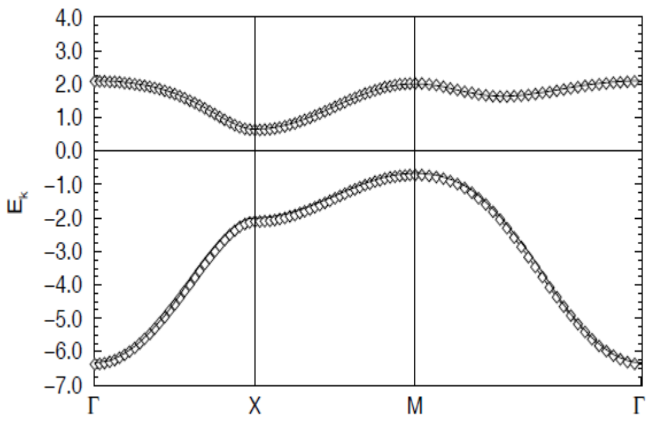
Metals and insulators

Assume a material with spin degeneracy:
Each band can hold 2 electrons per unit cell

Odd number of electrons per unit cell → Metallic

Even number of electrons per unit cell → Insulating

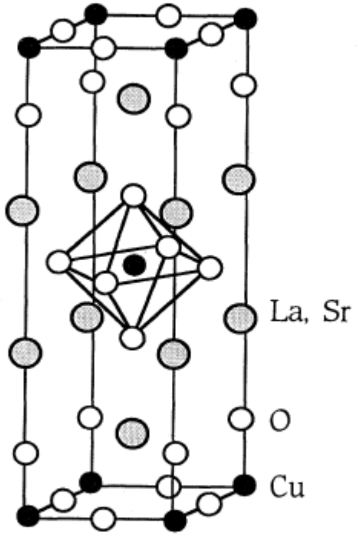
Even number of electrons per unit cell → Metallic (in case of band overlap)



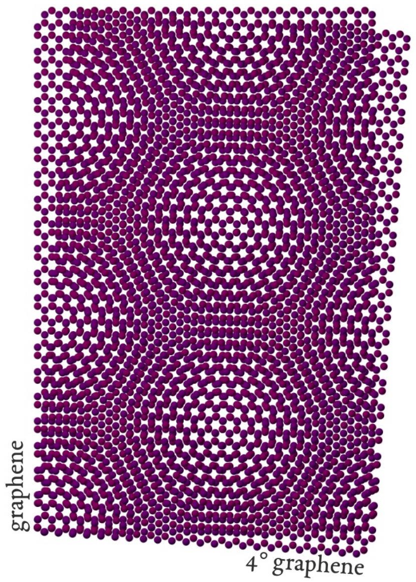
leni.bascones@csic.es

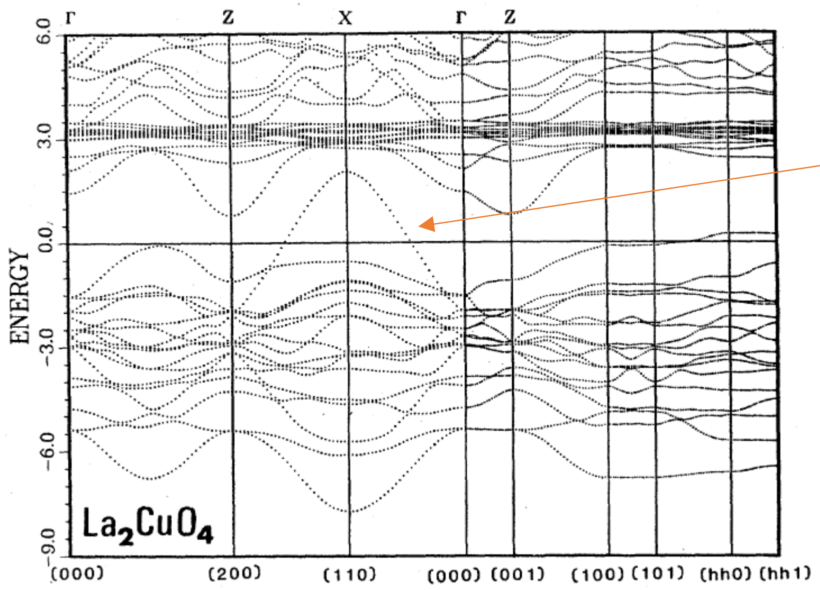
Correlated states and electronic filling

High-Tc
superconducting cuprates

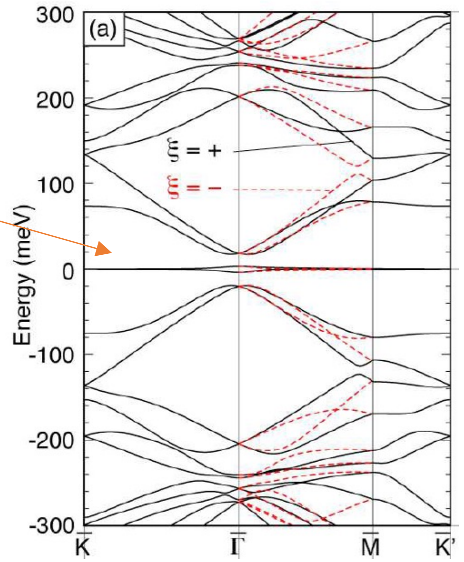


Twisted
bilayer graphene



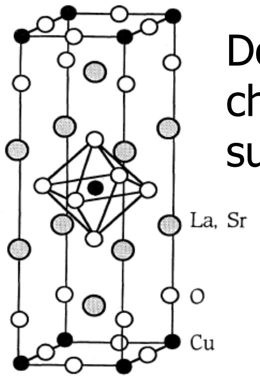


Expected to be metals

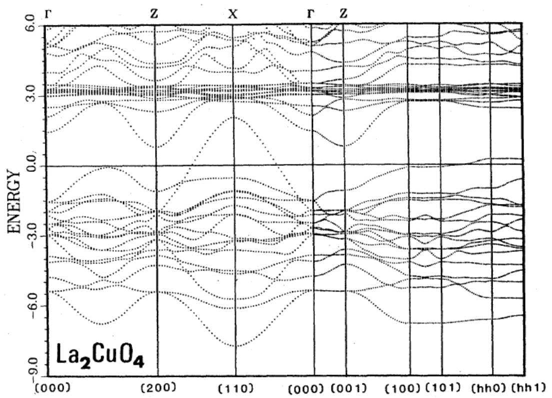


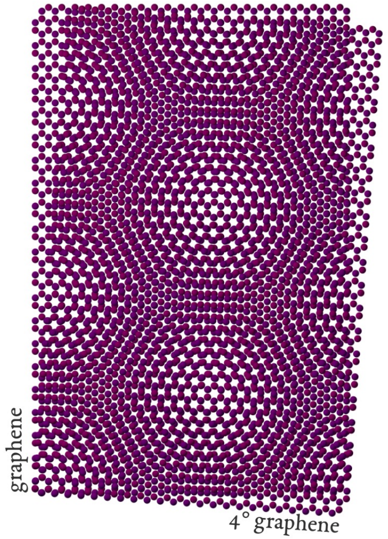
Correlated states and electronic filling

High-Tc superconducting cuprates

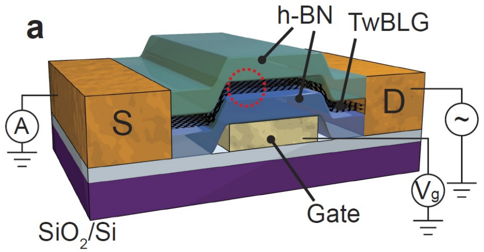


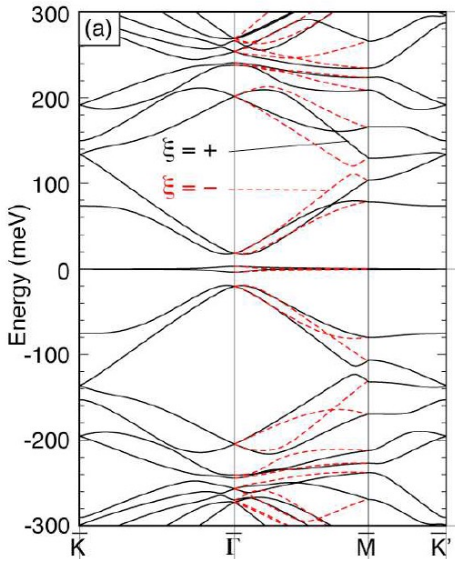
Doping through chemical substitution





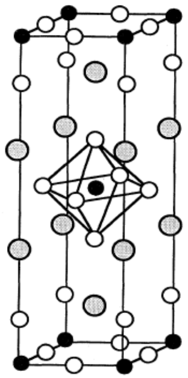
Twisted bilayer graphene



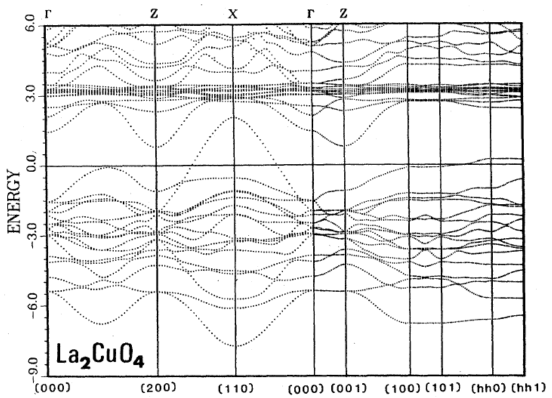


Correlated states and electronic filling

High-Tc superconducting cuprates



Doping through chemical substitution



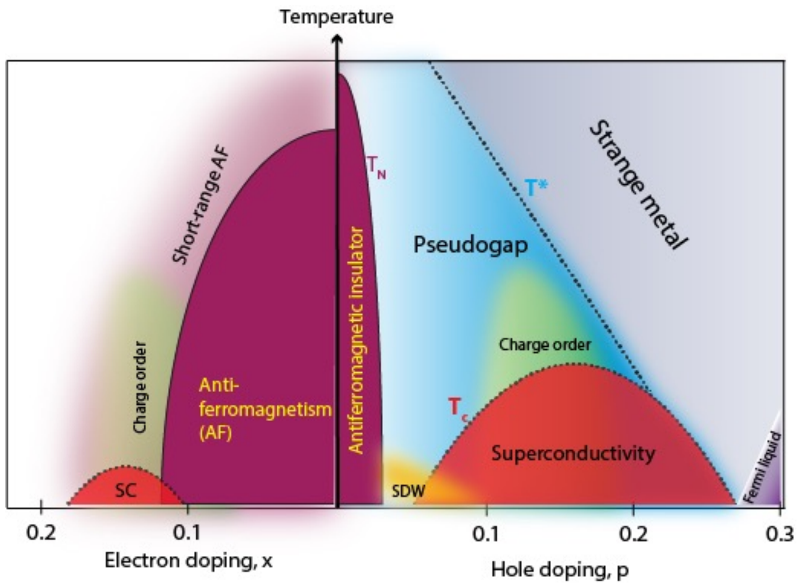
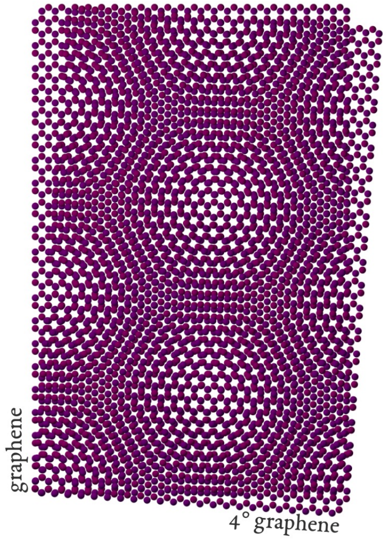
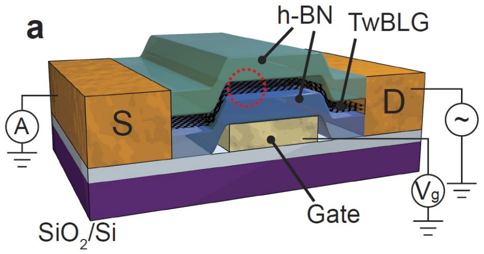
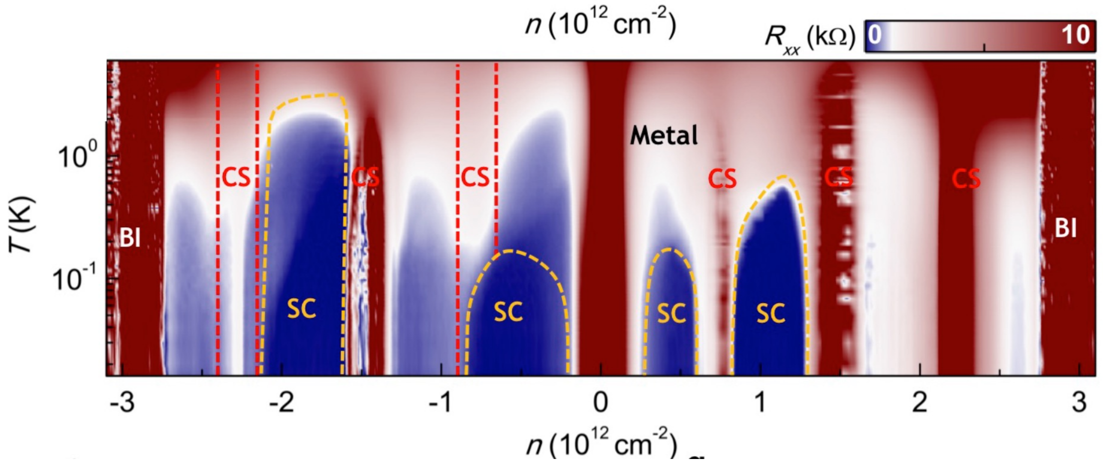


Fig: cme.physics.ucdavis.edu



Twisted bilayer graphene

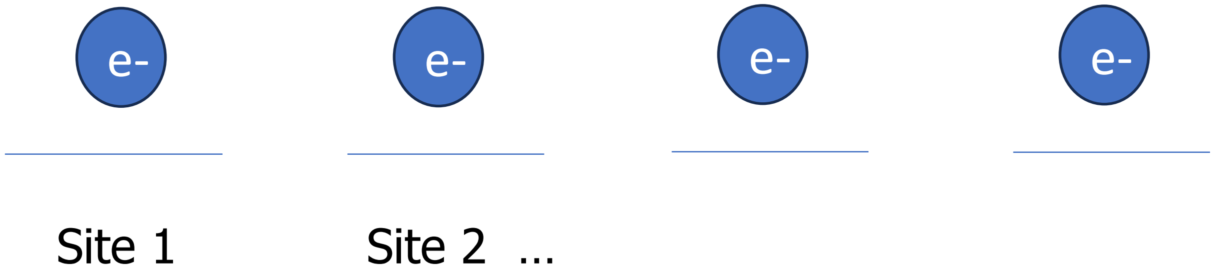




leni.bascones@csic.es

Correlated states and electronic filling

Onsite repulsión U
Hopping energy t



1 electron per site/atom

U>>t Metal insulator transition due to localization of electrons

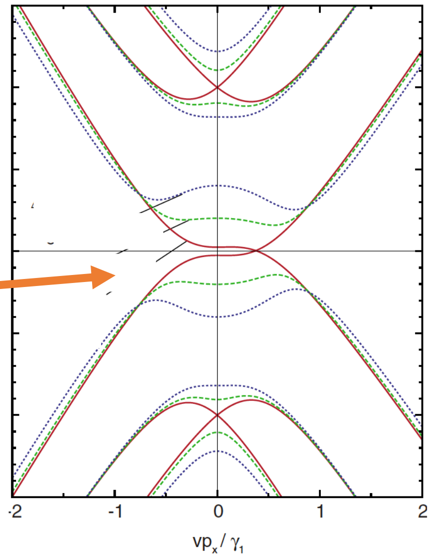
Partial electronic filling per site/atom

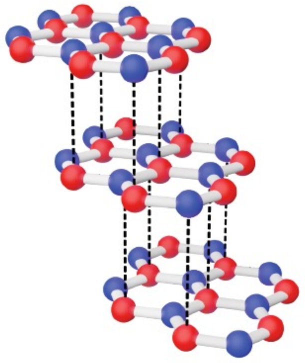
Always metallic (if no symmetry breaking)

But maybe correlated metal!

Band structure "details"

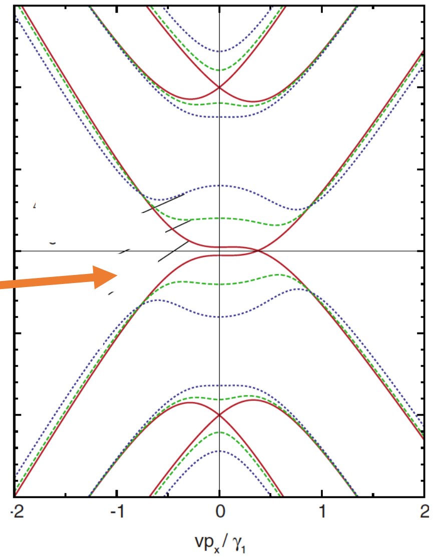
- Dimensionality
- Bandwidth W (flat bands)
- Flat band regions
  (large density of states at E_F)

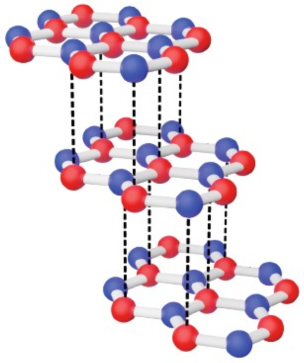




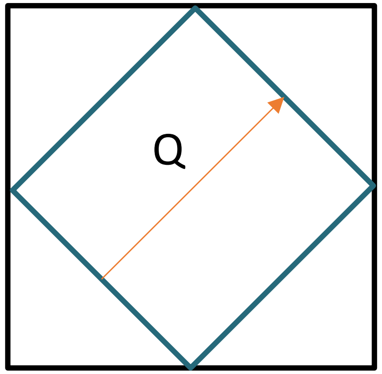
Band structure "details"

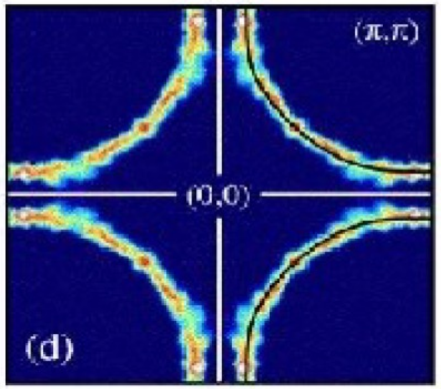
- Dimensionality
- Bandwidth W (flat bands)
- Flat band regions
  (large density of states at E_F)
- Nesting with momentum Q
  (always in 1D)



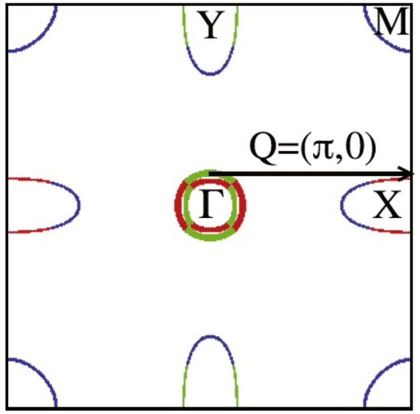


Fermi surfaces in 2D





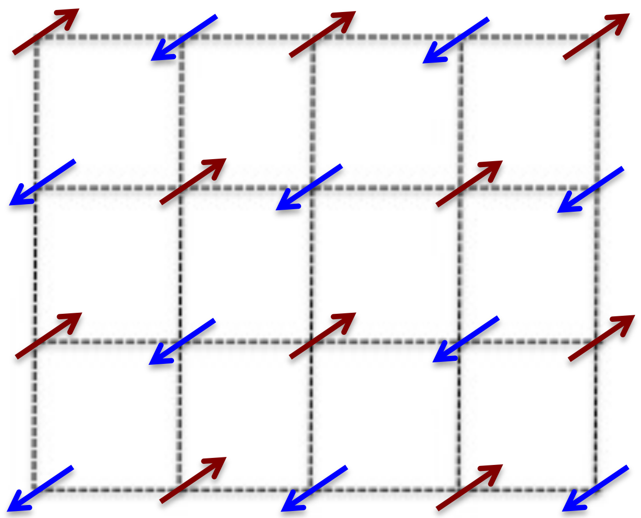
Fermi Surface of Cuprates



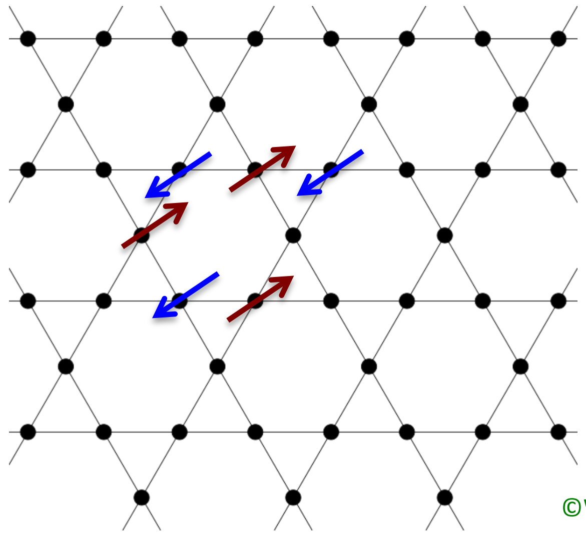
Fermi Surface of Iron superconductors

Dimensionality and crystalline lattice

Bipartite lattice

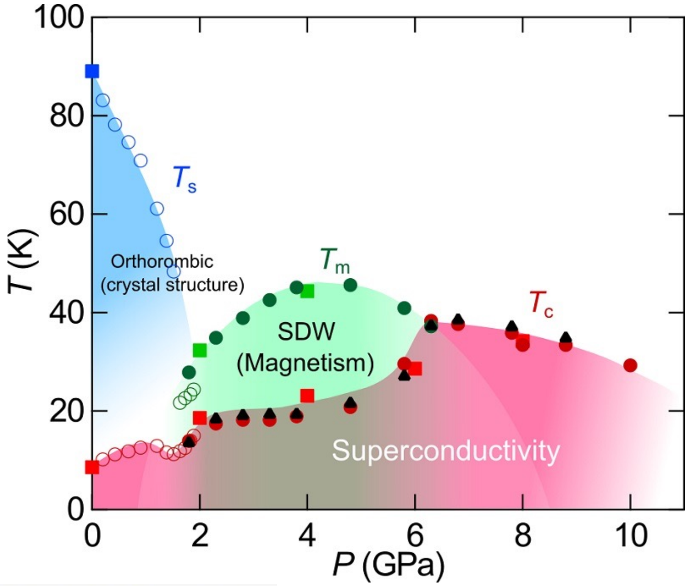


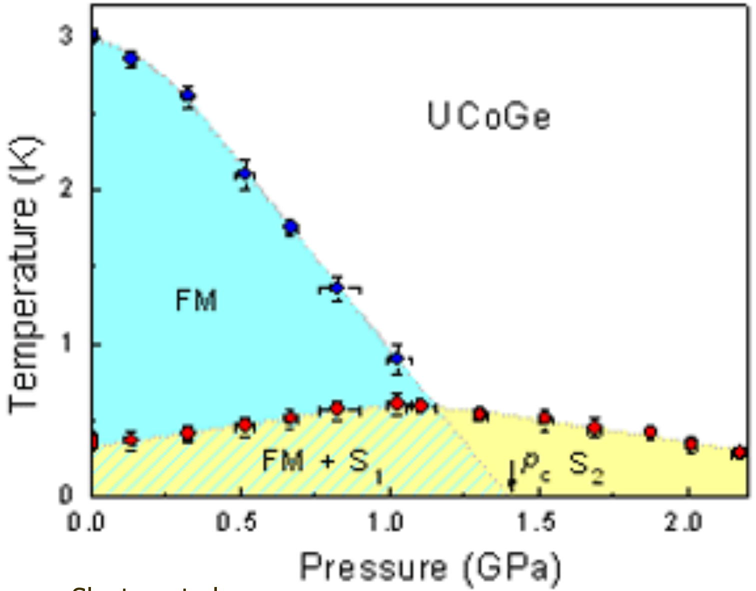
Frustrated lattice



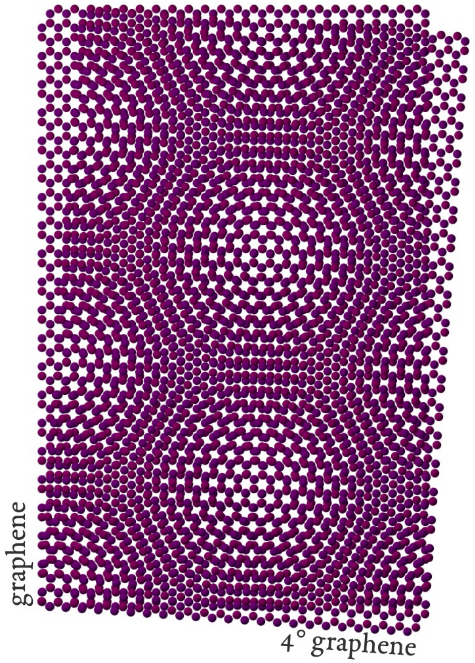
Gran variedad de estados electrónicos correlacionados

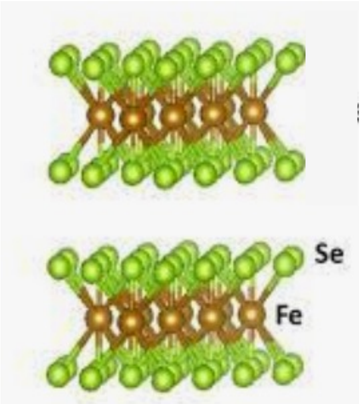
External knobs to control the correlated states: pressure, strain …





Slooten et al,
PRL 103 , 097003(2009)



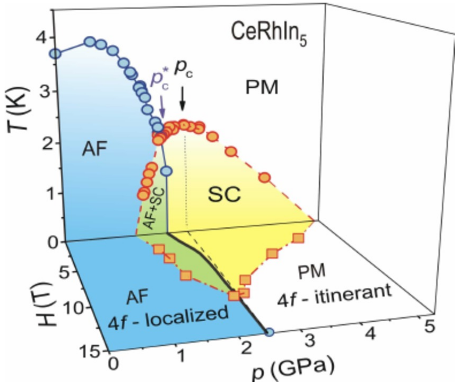


Modification of kinetic energy (lattice constant, Fermi Surface details …)

In a given material certain correlated states appear only under pressure

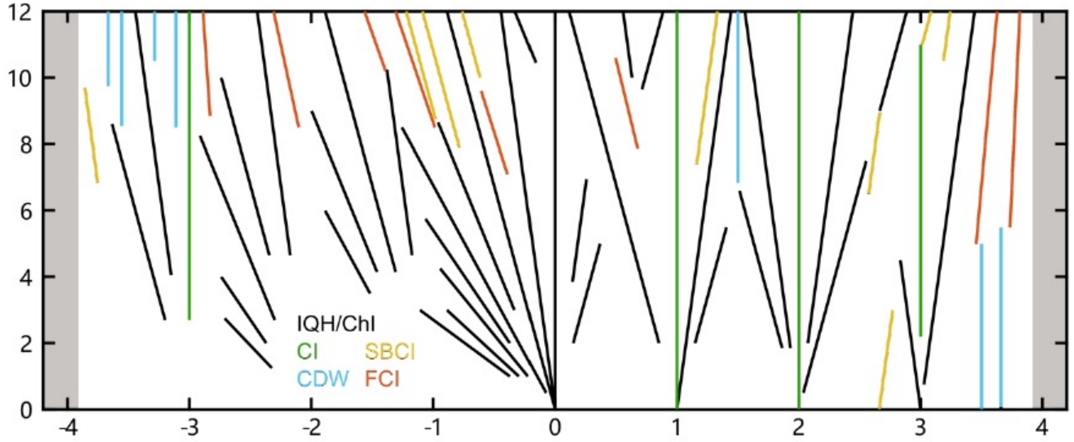
External knobs to control the correlated states: magnetic and electric field…

Magnetic field: spin and orbital effect



Flouquet et al, JPC, 2001

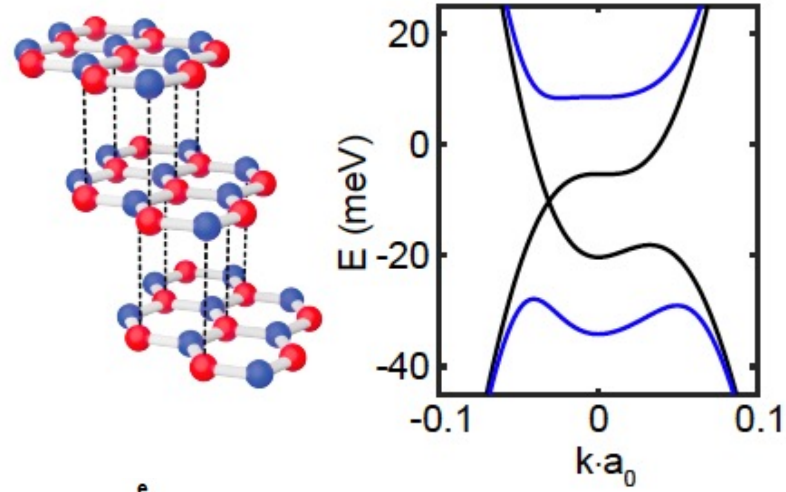
Twisted Bilayer Graphene (compressibility measurements)

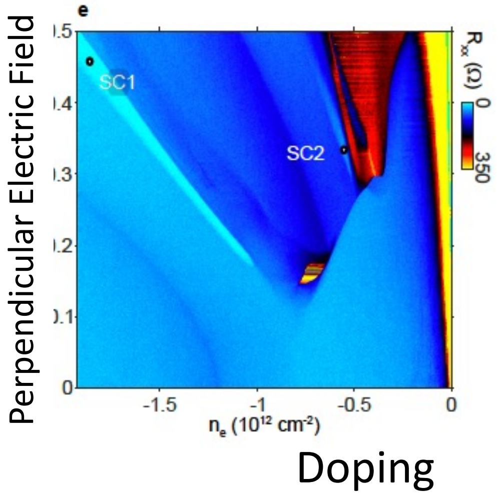


Xie et al, arXiv: 2107.10854

Different types of insulating correlated states

Perpendicular electric field





Zero field

ABC Graphene trilayer

The role of screening: from Coulomb to Hubbard

Coulomb interaction $\left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{1}{\varepsilon_r r_0} = 14.37 \text{ eV Å} \frac{1}{\varepsilon_r r_0}$

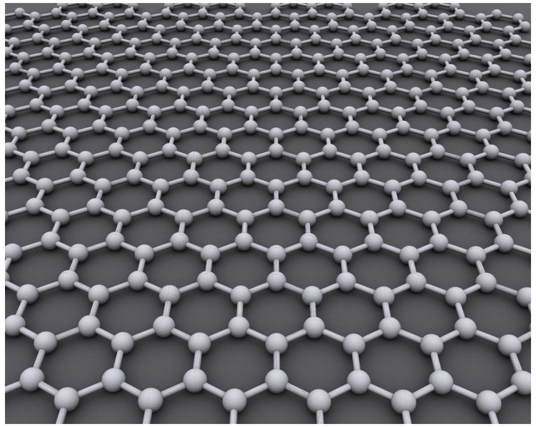
Tuning ε_r and maximum interaction length r_0

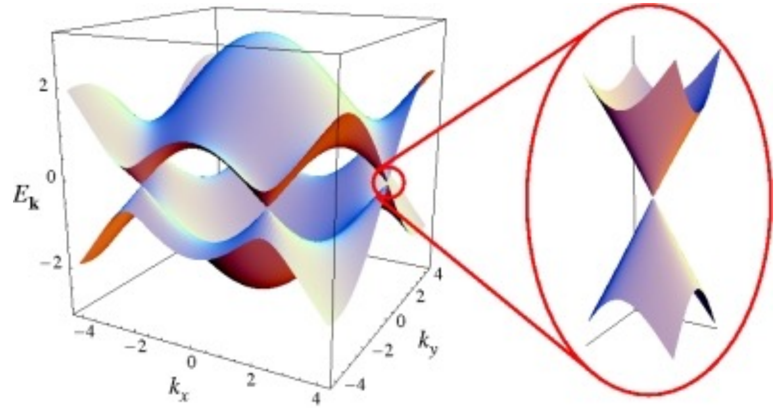
e- e- e- e-

Site 1 Site 2 …

If not screened the electronic interaction decays a 1/r

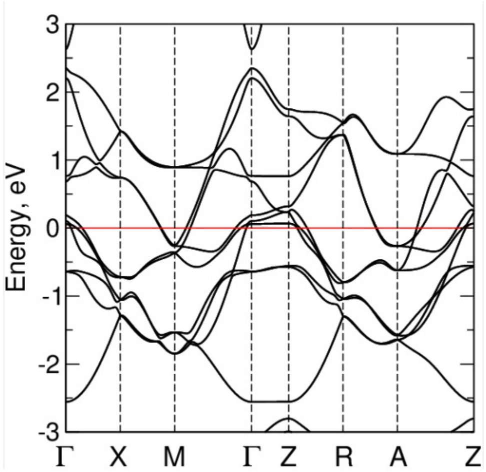
Graphene: 1/r interaction



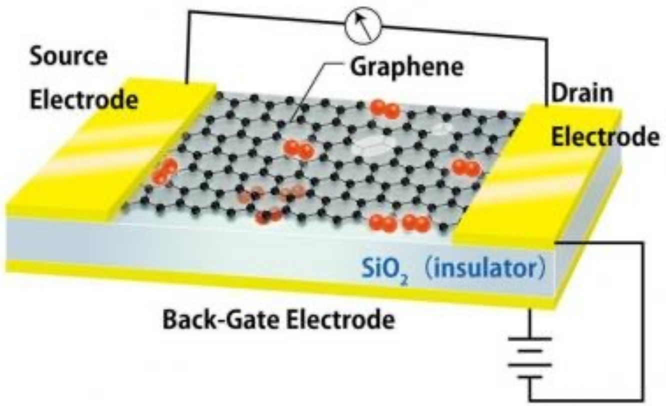


The role of screening: from Coulomb to Hubbard

Screening in metals



Screening with a gate



Coulomb interaction

$$\left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{1}{\varepsilon_r\, r}$$

→

Screened interaction

$$\left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{1}{\varepsilon_r}\frac{\mathrm{Exp}(-r/\lambda_0)}{r}$$

The interaction decreases faster with distance

↓

Tune interaction length

ε_r also determined by screening from deep bands, substrate, gates…:

leni.bascones@csic.es

The role of screening: from Coulomb to Hubbard

$$\left(\frac{e^2}{4\,\pi\,\varepsilon_0}\right)\frac{1}{\varepsilon_r}\,\frac{\mathrm{Exp}(-r/\lambda_0)}{r}$$

$\lambda_0 \sim$ lattice constant

Only onsite repulsión U :

Hubbard model

Site 1 Site 2 …

$\lambda_0 \rightarrow \infty$

Coulomb 1/r

Interaction decays slowly

and extends to all neighbors

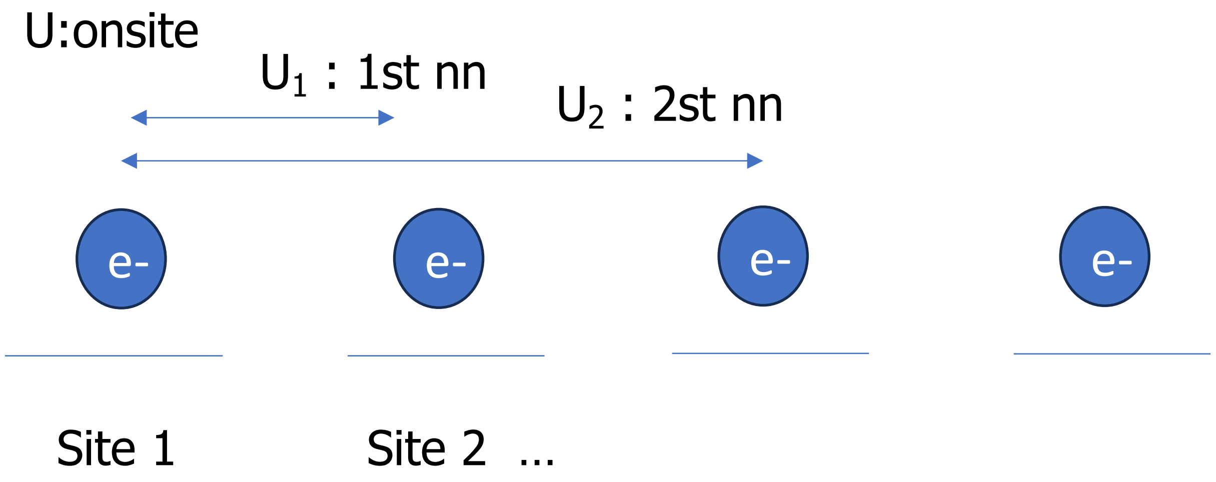
leni.bascones@csic.es

The role of screening: from Coulomb to Hubbard

$$\left(\frac{e^2}{4\pi\varepsilon_0}\right)\frac{1}{\varepsilon_r}\,\frac{\mathrm{Exp}(-r/\lambda_0)}{r}$$

Extended Hubbard model

Interaction up to first or second neighbors



The role of screening: from Mott to Wigner

Onsite repulsión U
Hopping energy t

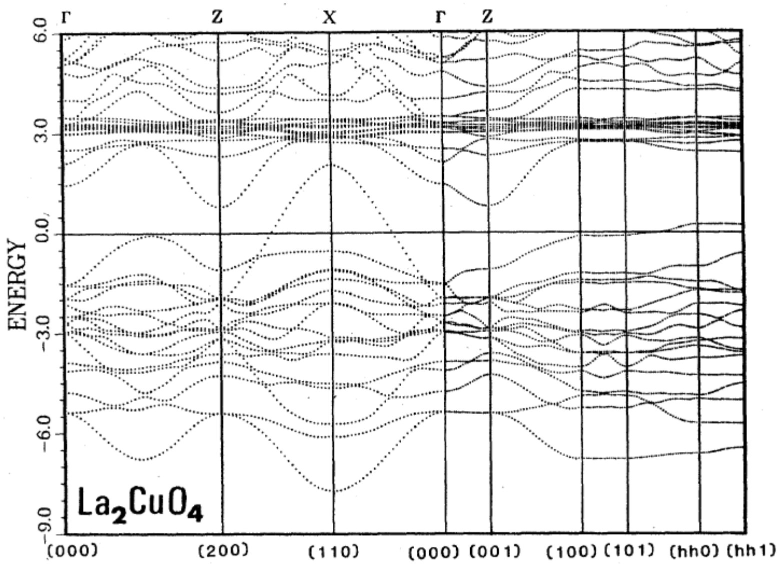
Site 1 Site 2 …

1 electron per site/atom

U>>t Metal insulator transition due to localization of electrons

Mott insulator
(Mott-Hubbard)

Large density of electrons, onsite repulsion & role of lattice



leni.bascones@csic.es

The role of screening: from Mott to Wigner

Assume: 3D, free space (continuum)

Coulomb $\propto \frac{e^2}{r}$

Electronic density $n = N_e / V$

N_e: Number of electrons

V: Volume

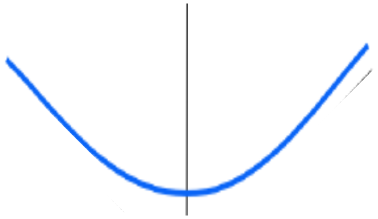
Kinetic energy

$$\int_{-\infty}^{\varepsilon_F} \varepsilon \, N(\varepsilon) \, d\varepsilon \propto n^{2/3}$$

density of states

Interaction energy

$$\propto \langle 1/r \rangle \cong n^{1/3}$$

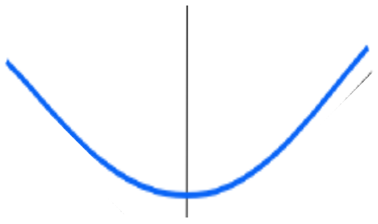


$$\frac{\text{Interaction energy}}{\text{Kinetic energy}} \propto n^{-1/3}$$

← Larger for small density

Important in semiconductors

The role of screening: from Mott to Wigner



$$\frac{\text{Interaction energy}}{\text{Kinetic energy}} \propto n^{-1/3}$$

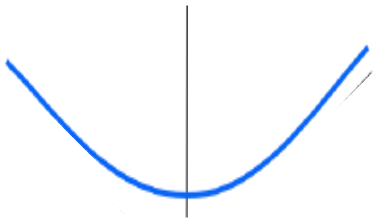
← Larger for small density

$$\propto \frac{e^2}{r}$$

Electrons far apart see each other

Important role of
Pauli principle in correlations

The role of screening: from Mott to Wigner



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Larger for small density

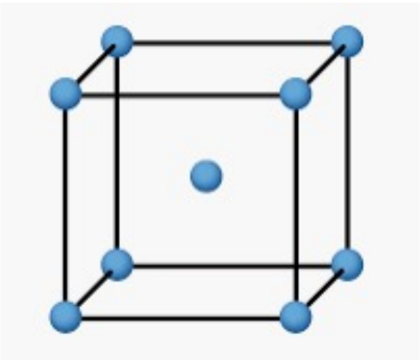
$$\propto \frac{e^2}{r}$$

Electrons far apart see each other

Important role of
Pauli principle in correlations

Wigner crystal

BCC lattice in 3D



Triangular
lattice in 2D

leni.bascones@csic.es

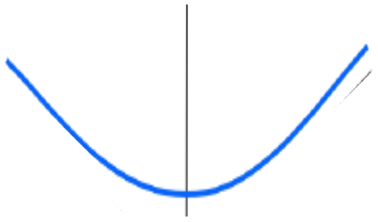
The role of screening: from Mott to Wigner

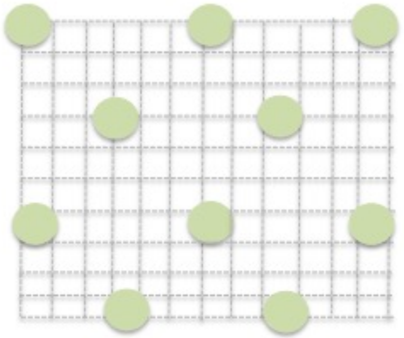
Wigner crystal

Free space (continuum)

Coulomb interaction $\propto \frac{e^2}{r}$

Insulating for small densities (inconmensurate)





Close to the bottom of the band
The lattice less important

Breaks lattice symmetry

Mott insulator

Lattice (unit cell)

Onsite interaction U

Larger effect of interactions
at integer filling of the lattice sites
(conmensurate filling)

Does not break
the lattice symmetry

Onsite repulsion only:
Electrons far apart
do not interact
(Pauli principle still matters)

leni.bascones@csic.es

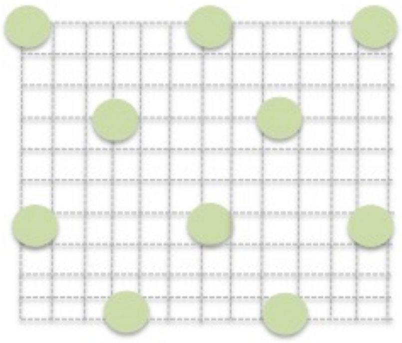
The role of screening: from Mott to Wigner

Wigner crystal

Free space (continuum)

Coulomb interaction $\propto \frac{e^2}{r}$

Insulating for small densities (inconmensurate)



Mott insulator

Lattice (unit cell)

Onsite interaction U

Larger effect of interactions
at integer filling of the lattice sites

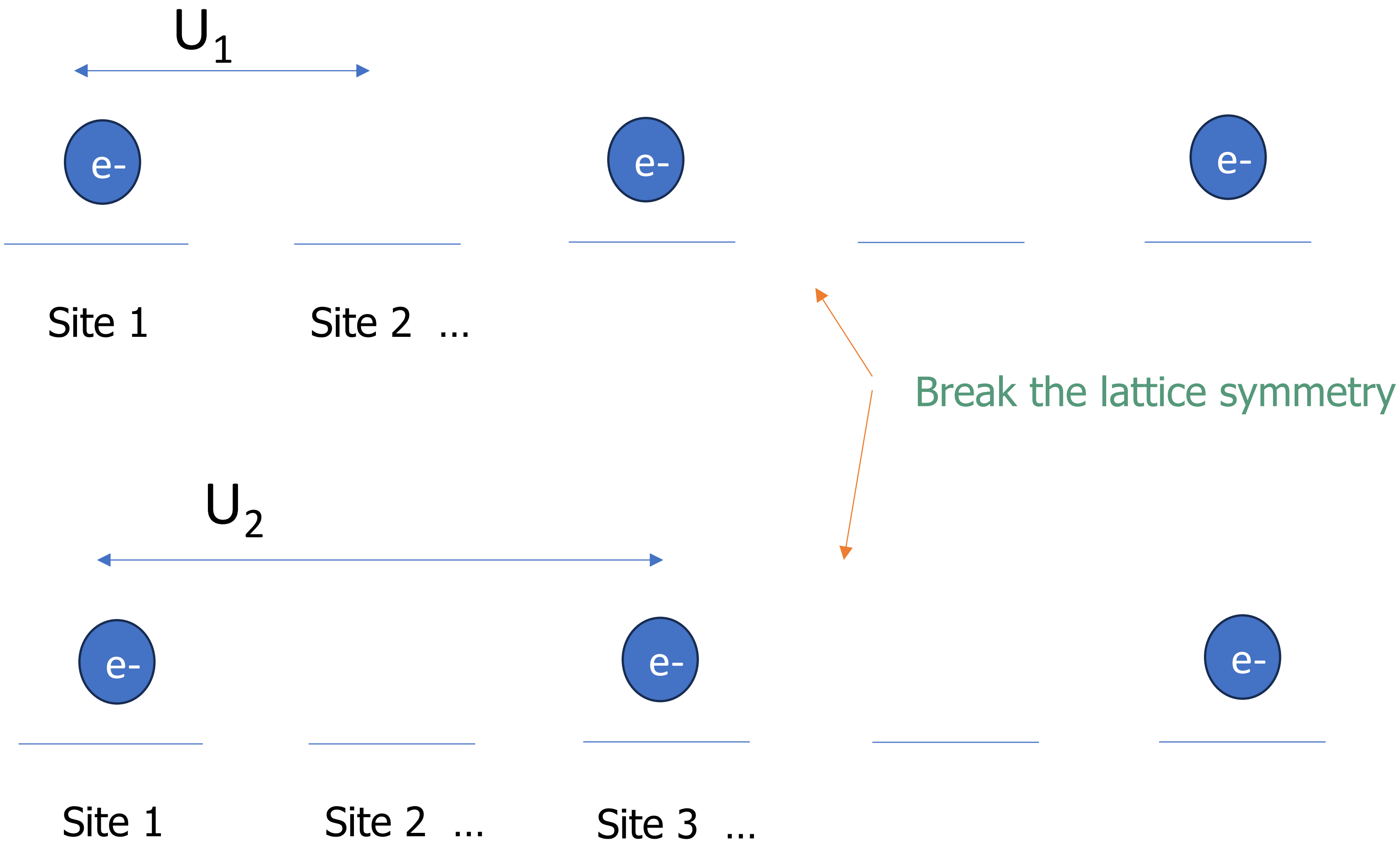
Charge localization (spin not invoked in insulating behavior)

leni.bascones@csic.es

Generalized Wigner or Wigner-Mott

Extended Hubbard: finite range of interaction

Commensurate filling



Repulsion up to
1st nearest neighbors

Repulsion up to
2nd nearest neighbors

leni.bascones@csic.es

Generalized Wigner or Wigner-Mott

Extended Hubbard: finite range of interaction

Commensurate filling

Repulsion up to
1st nearest neighbors

leni.bascones@csic.es

Emergence of Quantum Phases in Novel Materials

Interactions between many particles (electrons)

Correlated Quantum States

Symmetry breaking states

Formation of local moments

leni.bascones@csic.es

Tuning parameters for correlated states

Interaction energy vs Kinetic energy

- ❑ Filling of electronic bands
- ❑ Electronic bands details
- ❑ Dimensionality
- ❑ Crystalline lattice
- ❑ Screening
- ❑ External knobs (pressure, …)
- ❑ Interaction with lattice
- ❑ Other (spin-orbit, …)

40 years of intensive research effort and continuously new surprises appear

Emergence of quantum phases in novel materials

Basic ideas & generic concepts

Different kinds of phenomena
& why they happen

Theoretical description

Many different approaches and
analytical & numerical methods

Experimental signatures
of the quantum states

Materials and platforms
where these quantum phases appear