Photo-emission signatures of coherence breakdown in Kondo alloys

Bishal Poudel
LOMA, CNRS, Bordeaux, France
Supervisors : Sébastien Burdin and Gertrud Zwicknagl
In collaboration with Claudine Lacroix

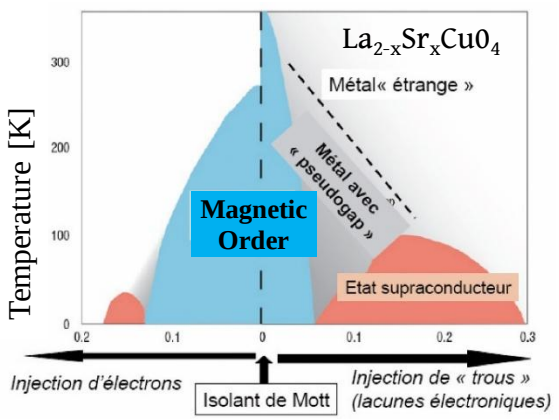
GDR MEETICC
April 2, 2021

Outlines :

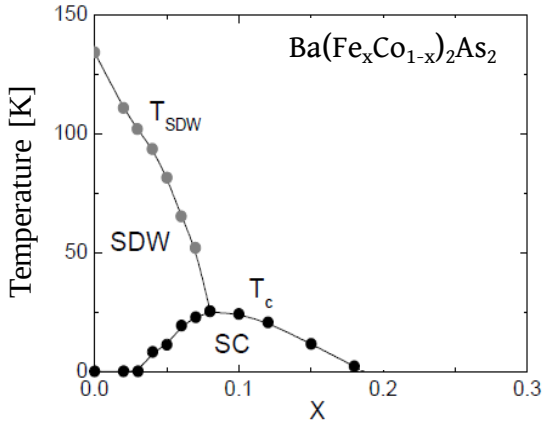
1. **General Motivations**
2. **Introduction to Kondo alloys**
3. **Results and predictions**
   - ARPES signatures
   - Kondo alloys phase diagrams
4. **Conclusions**

Examples of quantum phase transitions induced by interactions

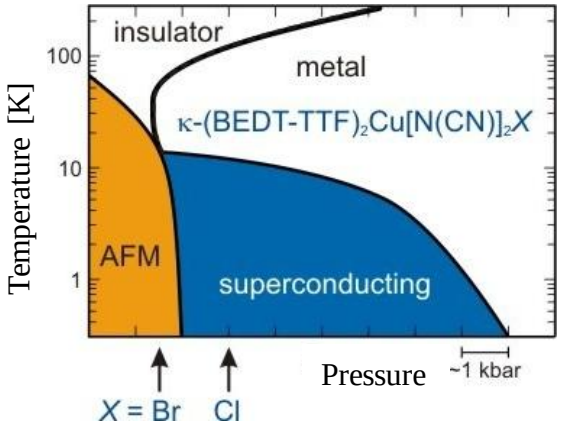
Cuprates



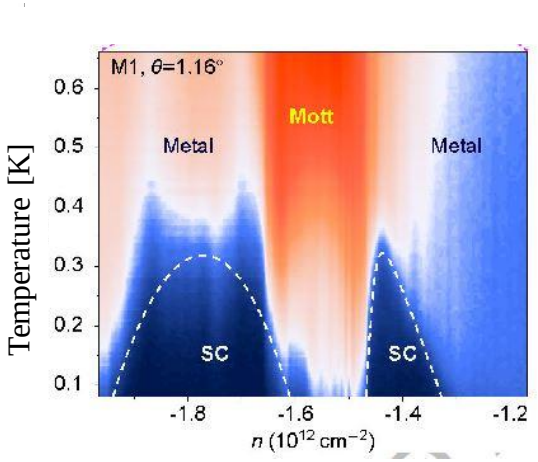
Pnictides



Organics

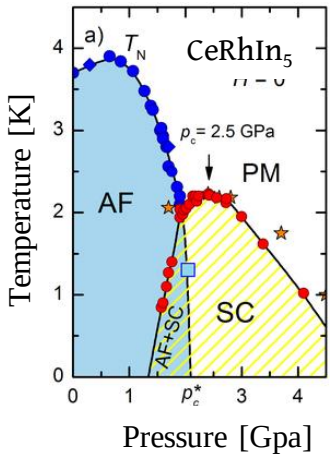


Graphene superlattices

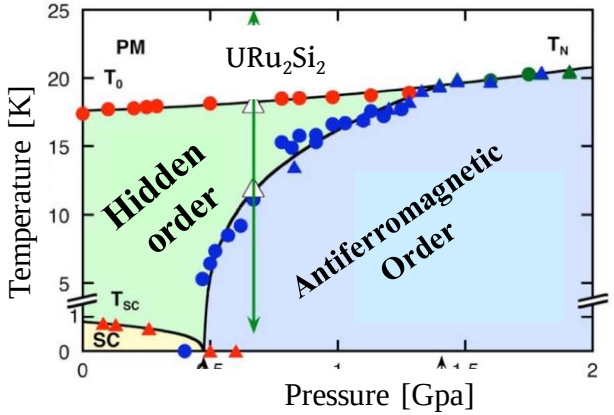


Doping

Rare earths

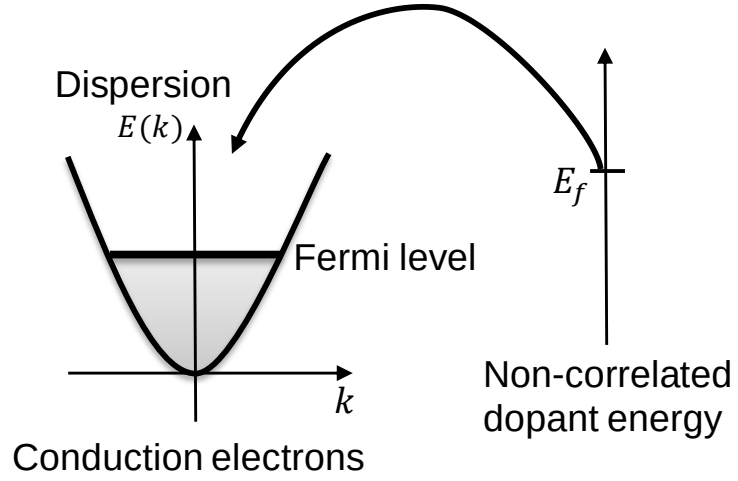


Actinides



Doping with *f*-orbitals in Ce or Yb based compounds

Charge reservoir model



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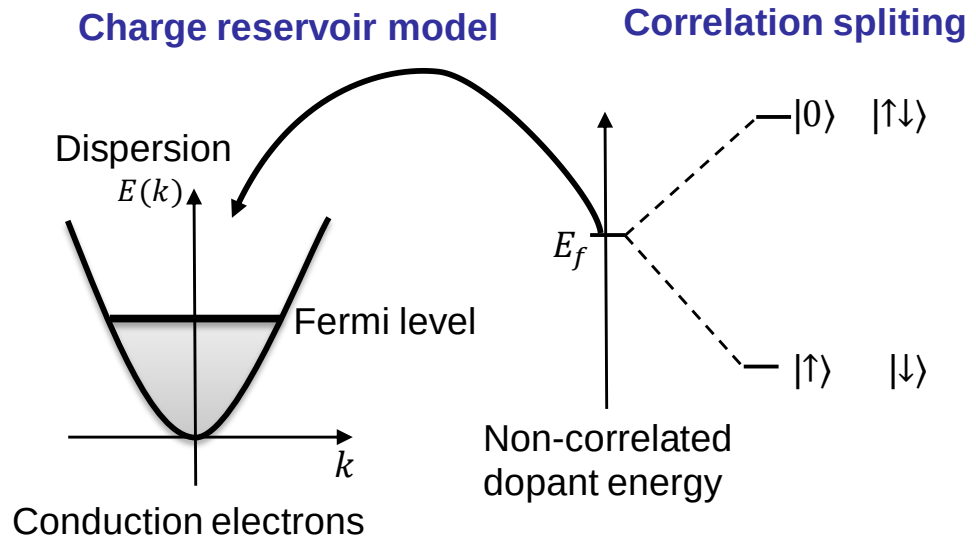

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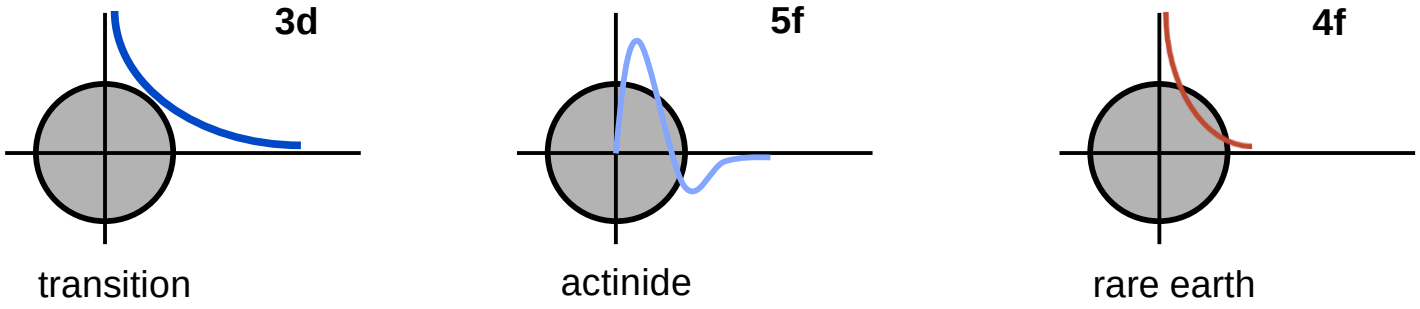
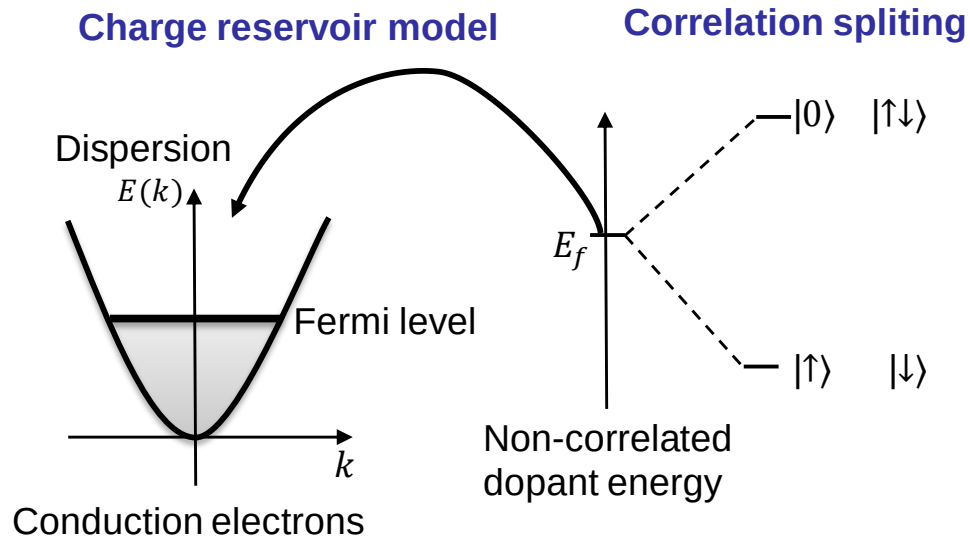
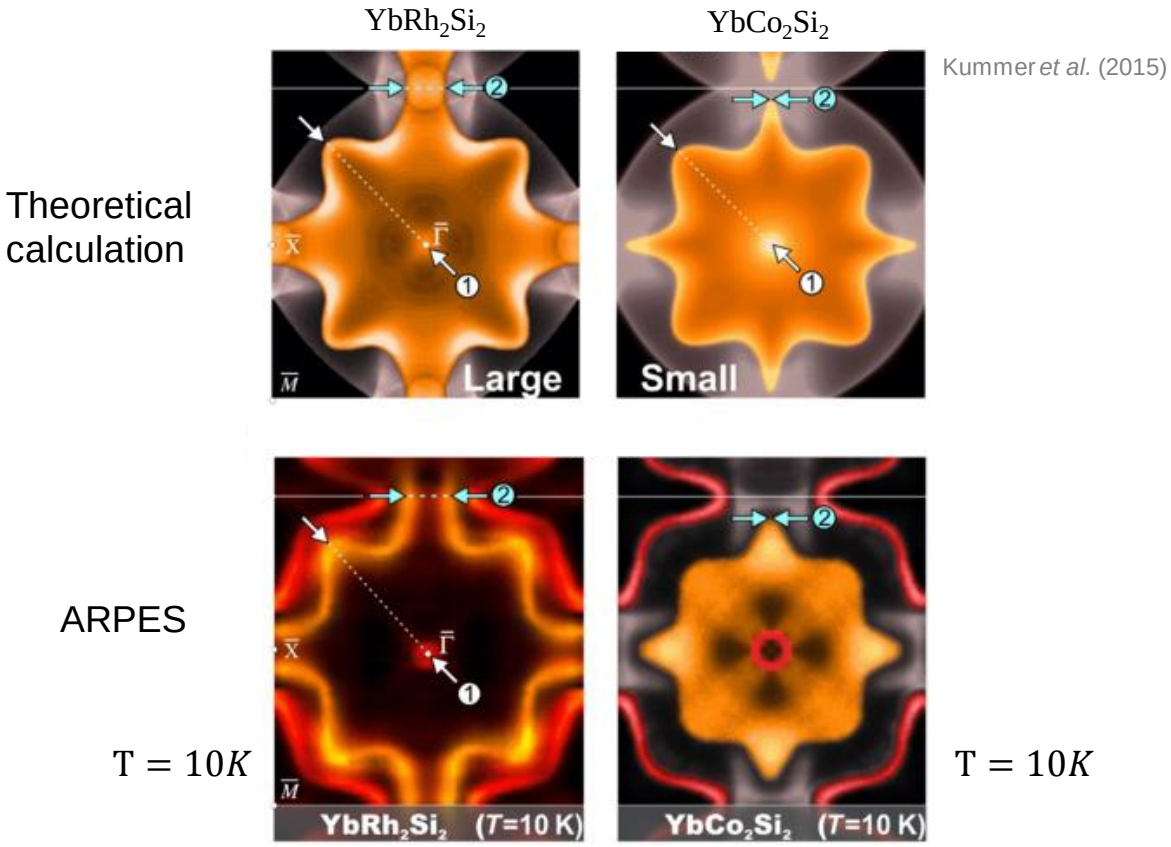




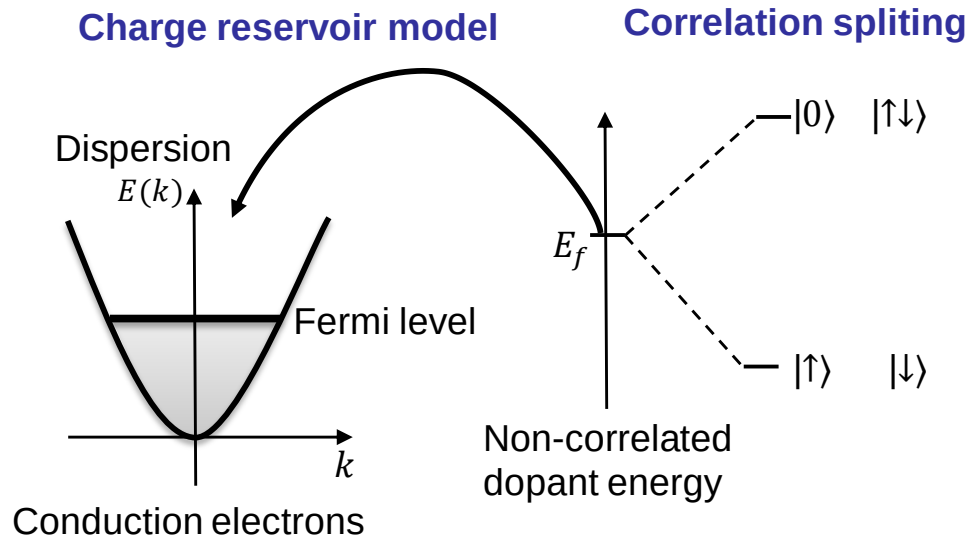
- **f-electrons orbitals are a priori localized and their valences are fixed**
- **But they interact with sea of conduction electrons**

Doping with *f*-orbitals in Ce or Yb based compounds



Doping from *f*-electrons
Enlarged Fermi surface

No doping from *f*-electrons
Small Fermi surface



- **one rare-earth atom per site**
- **Yb valency: $4f^{13}$**

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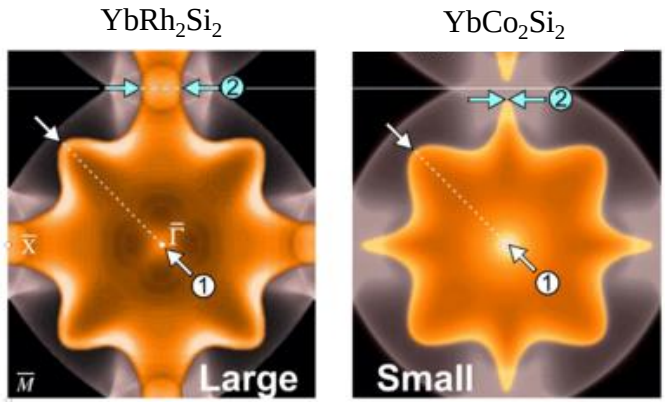
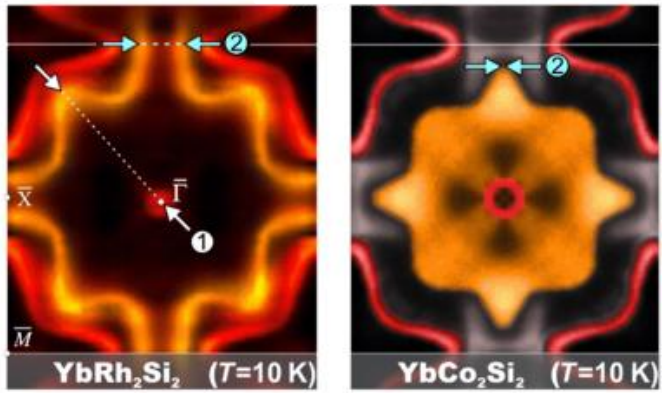


Kummer *et al.* (2015)

Theoretical calculation

ARPES



$T_{Kondo} = 25K < T = 10K$

$T = 10K > T_{Kondo} = 1K$

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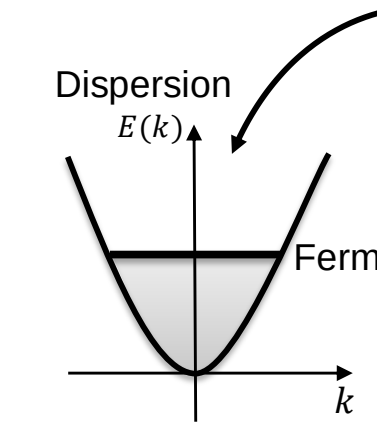
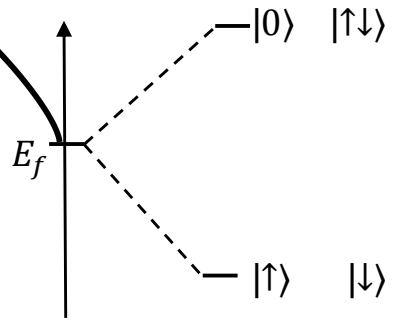


Conduction electrons

Correlation spliting

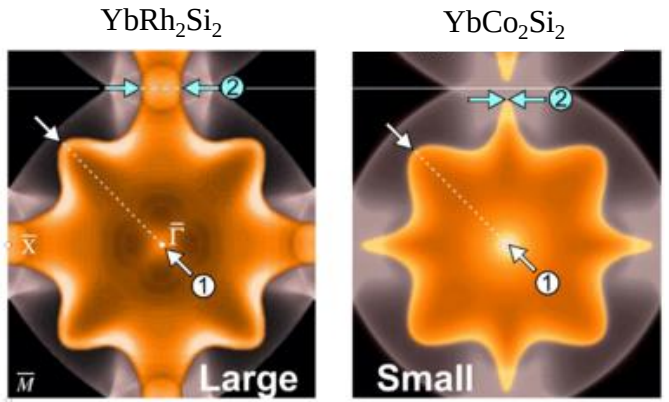


Non-correlated dopant energy

- **one rare-earth atom per site**
- **Yb valency: $4f^{13}$**

Kondo resonance: singlet formation between *f*-electron and conduction electrons

Doping with *f*-orbitals in Ce or Yb based compounds



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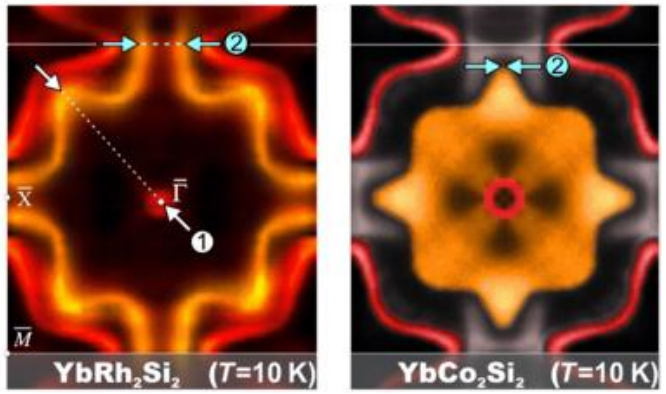

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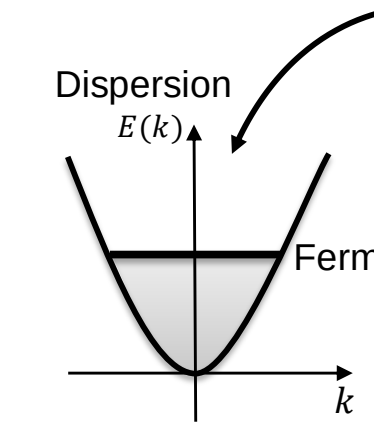

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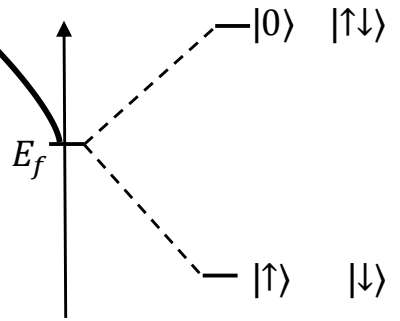


- **one rare-earth atom per site**
- **Yb valency: $4f^{13}$**

Question : what happens when the rare-earth atom is diluted to x atom per site ?

Kondo resonance: singlet formation between *f*-electron and conduction electrons

Kondo alloy model

Kondo Hamiltonian: $H = H_{\mathrm{e-cond}} + J_K \sum_i \vec{S}_i \cdot \vec{\sigma}_i$

Conduction electrons
electronic bandwidth W
electronic filling n_c
"free" density of states $\rho_0 \propto 1/W$

Quantum spins 1/2
Kondo Interaction (antiferromagnetic) between impurities and electrons
concentration x per site

3 relevant parameters: $\boldsymbol{n_c}, \boldsymbol{x}$, and $\boldsymbol{J_K/W}$

Energy scales emerge at low temperature:

- *The Kondo temperature* $\boldsymbol{T_K}$ *(crossover)* characterizing the formation of local singlets

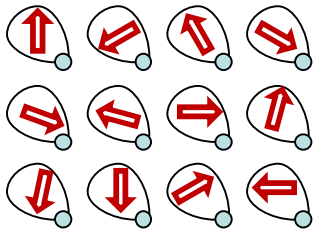
$T_K \propto \exp\left(-1/J_K\rho_0\right)$

Numerical method : adaptation of the DMFT

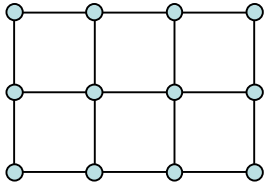
- *General scheme* A.Georges *et al.*, RMP'96

Model defined on a lattice → **Local model: one effective site**

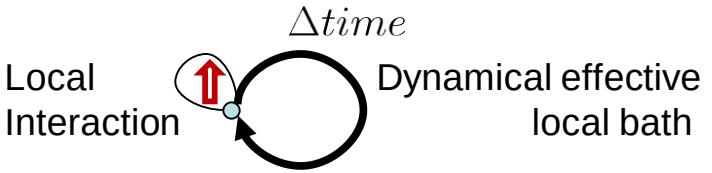
Local interactions & Interactions with neighbors



Kondo Interaction

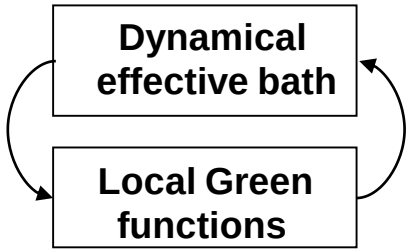


conduction e⁻



System of equations to solve:

Resolution of the effective single site model



Self-consistant DMFT relation

Local action

$$S_{loc} = \sum_{\sigma} \int_0^{\beta} d\tau \int_0^{\beta} d\tau'\, c_{0\sigma}^{\dagger}(\tau)\Delta(\tau-\tau')c_{0\sigma}(\tau') + J_K \int_0^{\beta} d\tau\, \overrightarrow{S_0}(\tau).\overrightarrow{\sigma_0}(\tau)$$

Local Green's function

$$G_{loc}(\omega) = \sum_{\mathbf{k}} \frac{1}{\omega - \varepsilon_{\mathbf{k}} - \Delta(\omega) - 1/G_{loc}(\omega)}$$

ARPES signal

$$A(\mathbf{k}, \omega = 0) = -\frac{1}{\pi} Im\{G(\mathbf{k}, \omega = 0)\}$$

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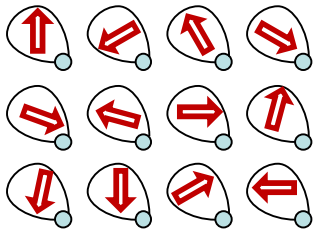

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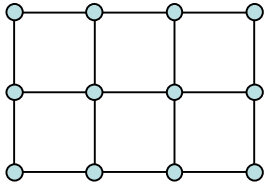

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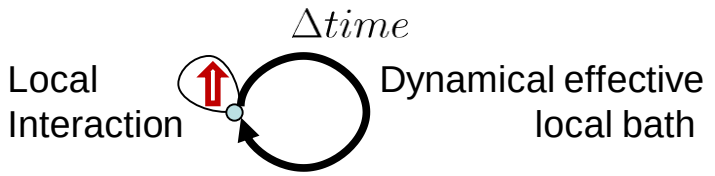

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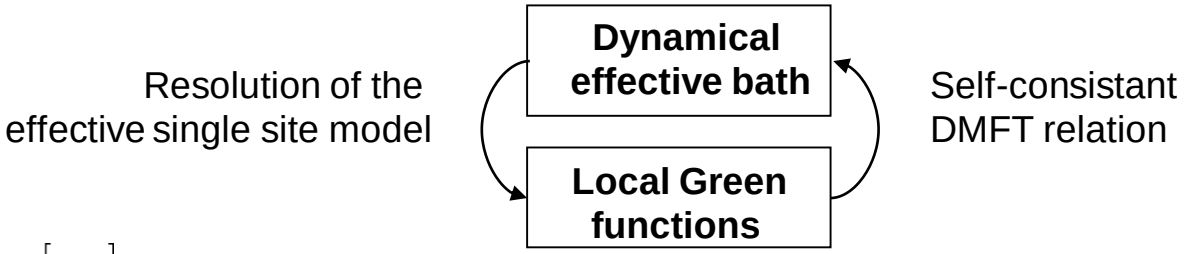
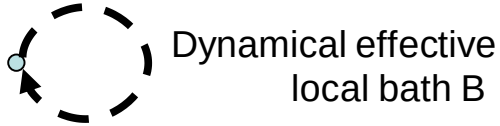


- *Binary Kondo alloys : matrix DMFT* $\mathrm{A}_x\mathrm{B}_{1-x}[\cdots]$

Burdin *et al.*, PRB'07, PRL'13, '19 **Poudel** *et al.* JMMM'20 **Poudel** *et al.* arXiv'21

Kondo effective site (atom A):

effective site atom B:



Locally, the Kondo interaction is treated in the mean-field « slave boson » approximation

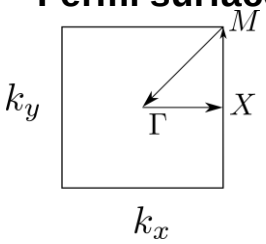
Angle resolved photo-emission spectroscopy

Strong $T_K/W \approx 0.1$:

x : concentration of *f*-electrons atoms
n_c : electronic filling

Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21

Fermi surface

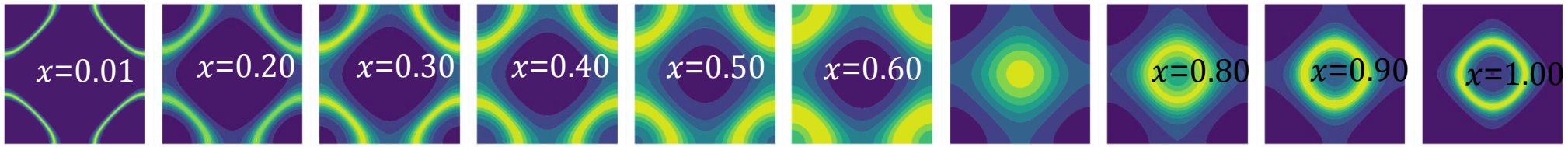


Square lattice
Brillouin zone

Shrinking →

$x = n_c = 0.70$

Enlarging →



Diluted Kondo

***f*-electrons substitution = hole doping**

Dense Kondo

***f*-electrons substitution = electron doping**

A Liftshitz-like transition is observed at $x = n_c$

Angle resolved photo-emission spectroscopy

Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21

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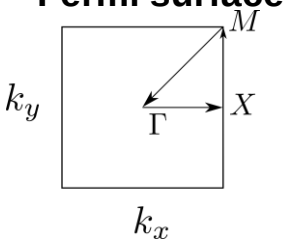

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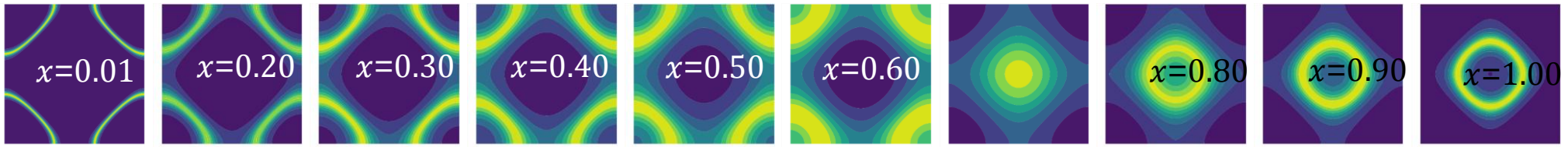

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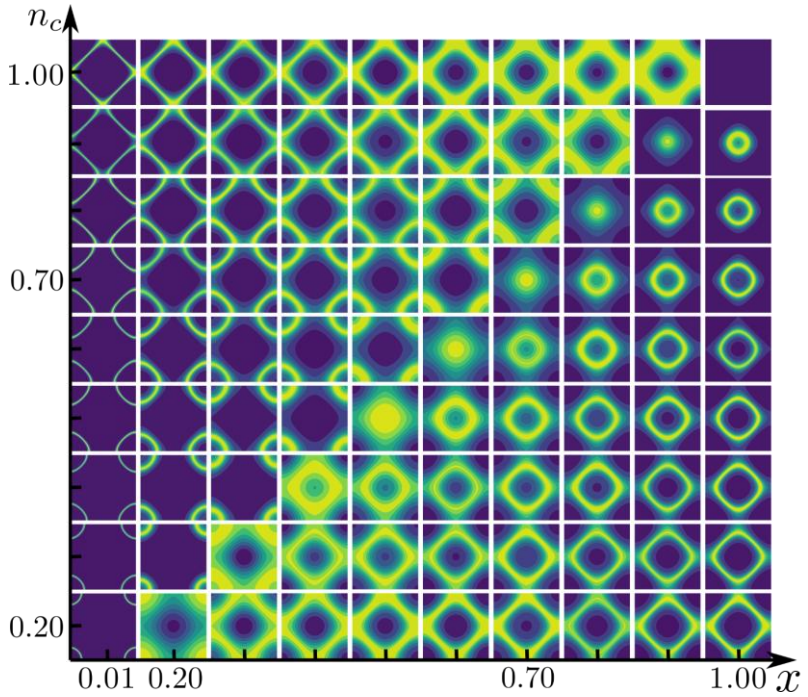

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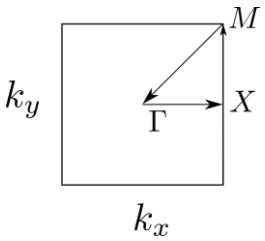
Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21



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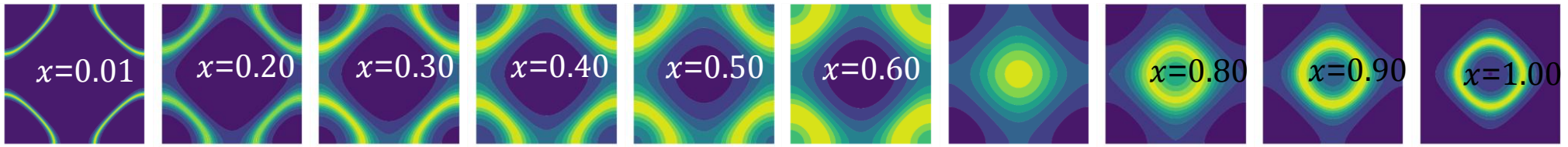

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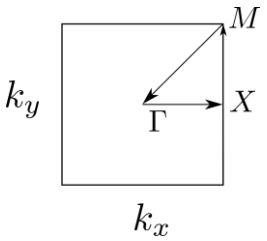

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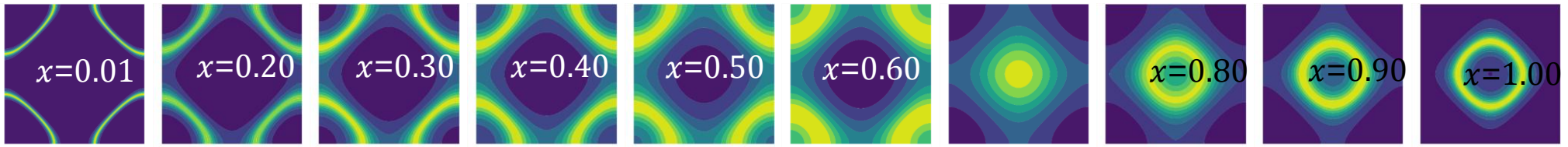

Weak $T_K/W \approx 0.01$:

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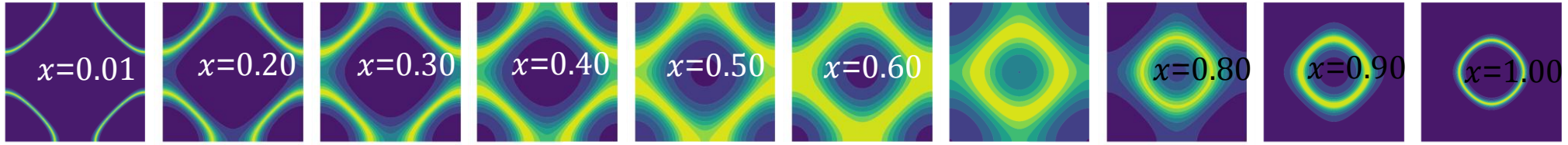

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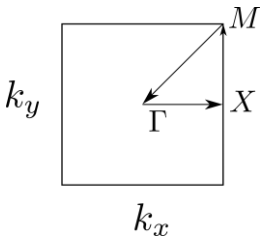

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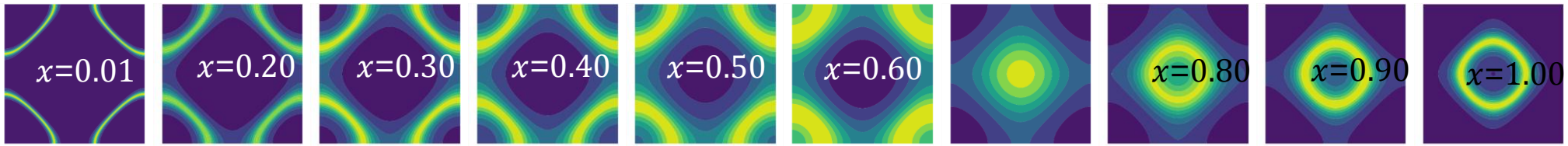
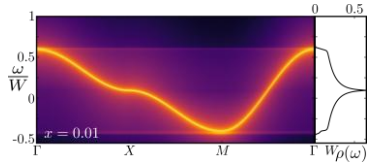
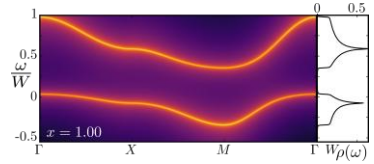




one-branch dispersion



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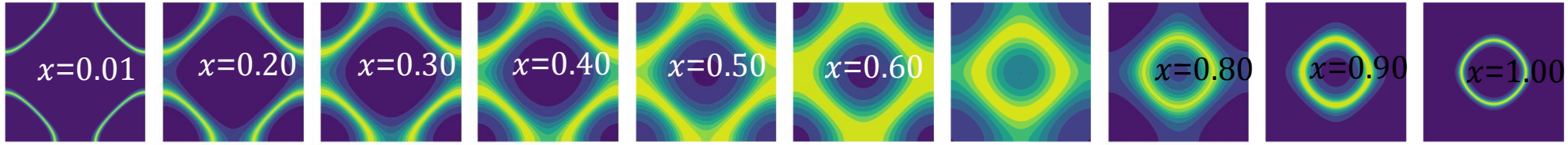
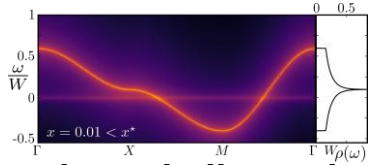
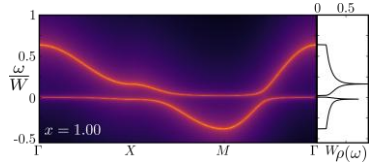




one-branch dispersion

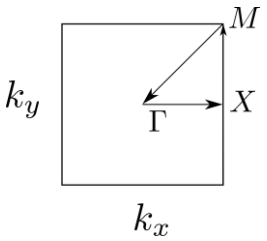


Angle resolved photo-emission spectroscopy

Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21

Strong $T_K/W \approx 0.1$:

x : concentration of f-electrons atoms
n_c : electronic filling



Square lattice
Brillouin zone

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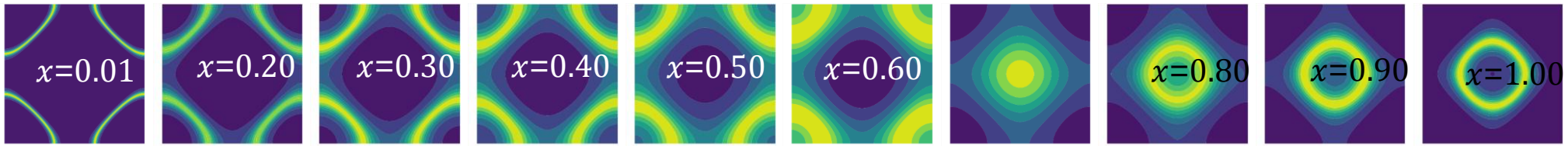
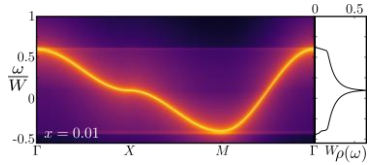
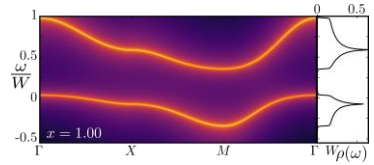




one-branch dispersion



two-branch dispersion
one gap

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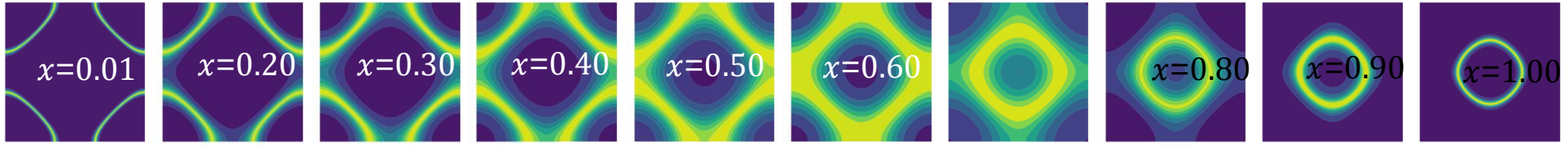

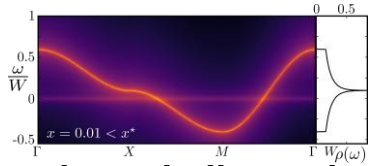

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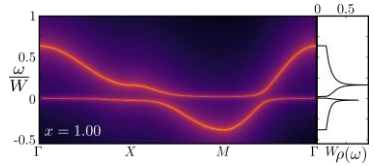

two-branch dispersion
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Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21

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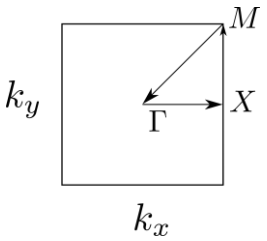

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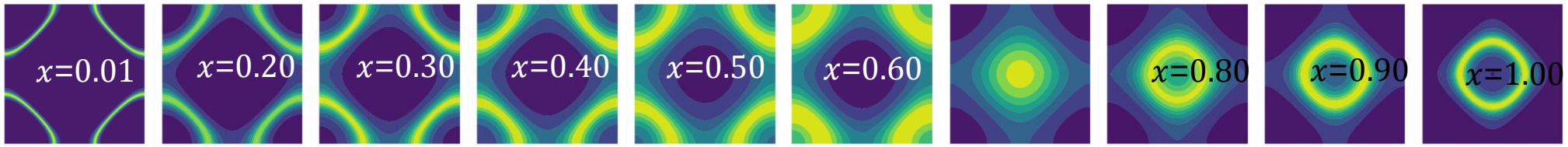

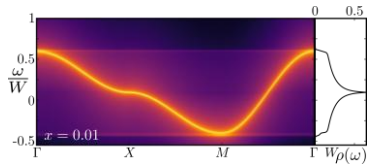

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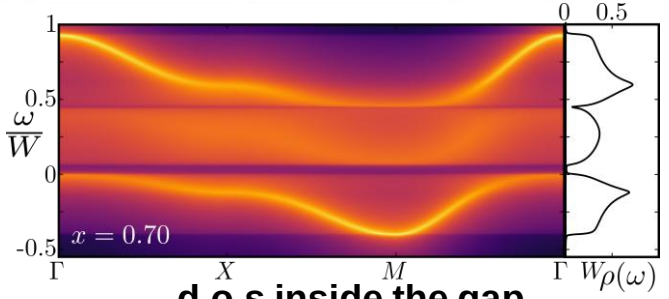
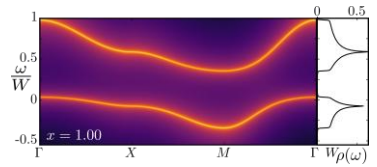


d.o.s inside the gap



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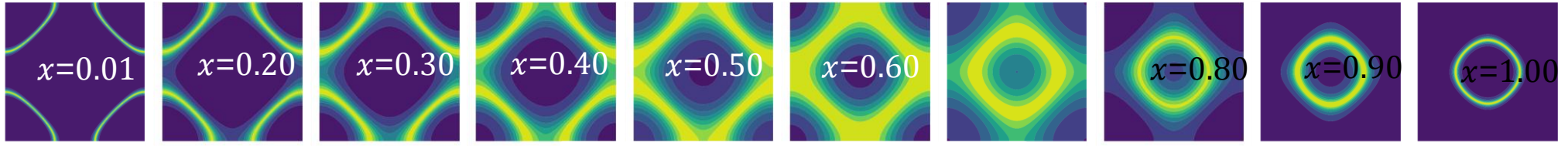

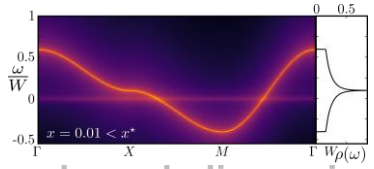

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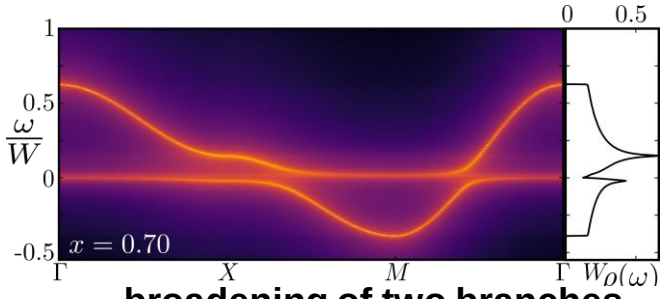
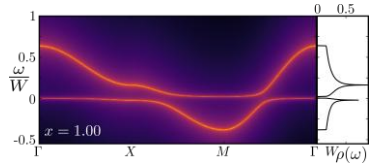


broadening of two branches



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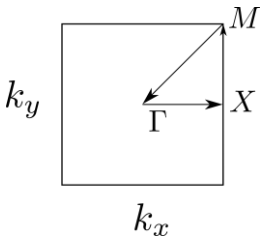
Burdin *et al.* PRL'13 **Poudel** *et al.* arXiv'21



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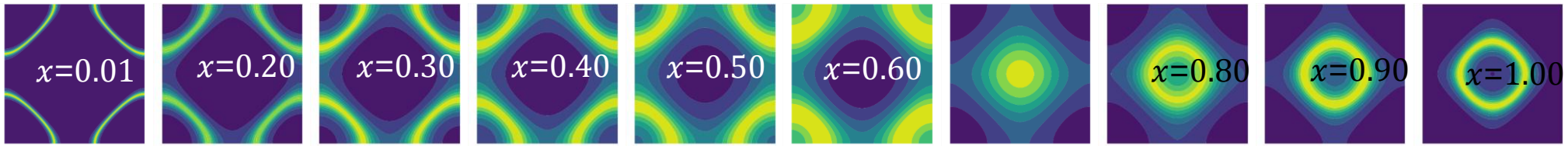
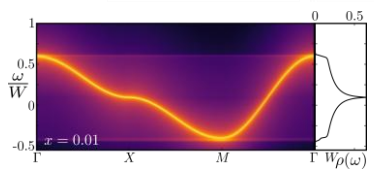
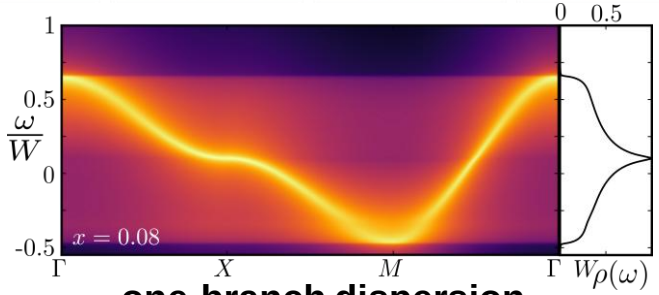




one-branch dispersion



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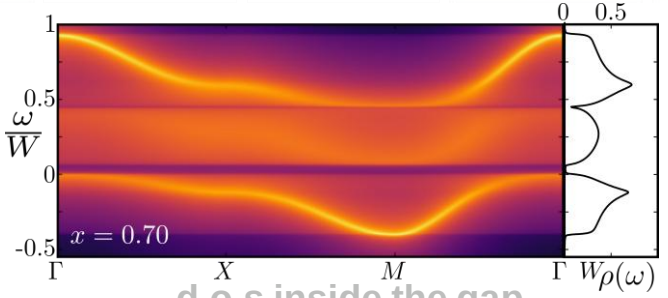
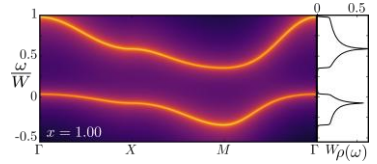


d.o.s inside the gap



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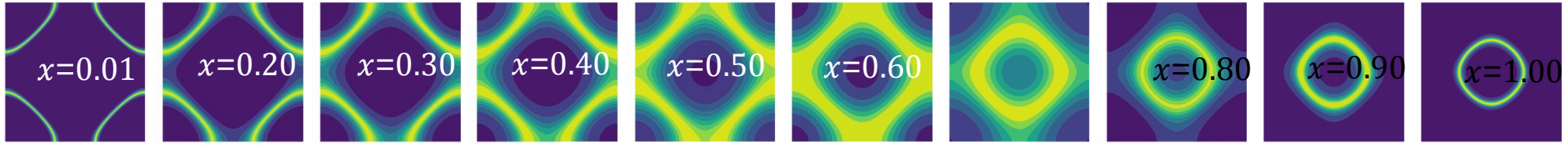

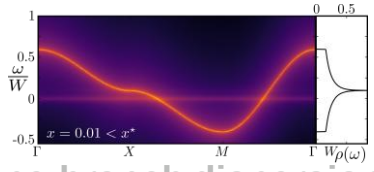

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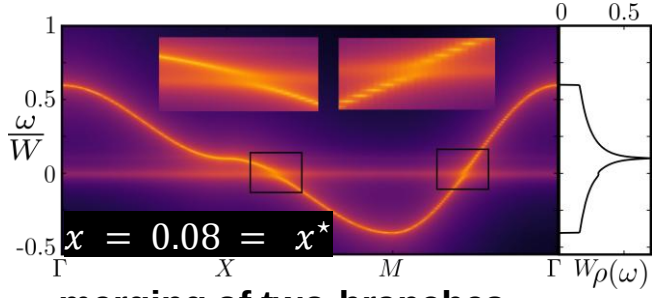
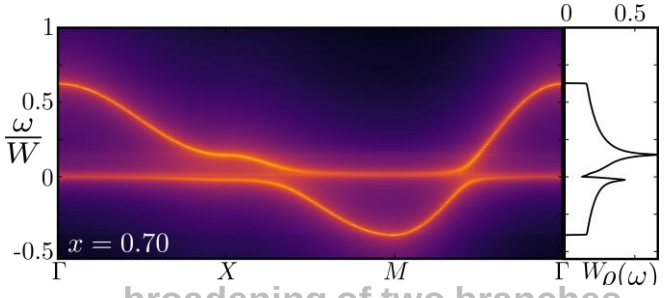
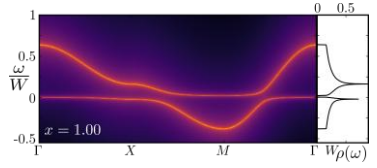


merging of two-branches
vanishing of effective mass $m^\star$



broadening of two branches

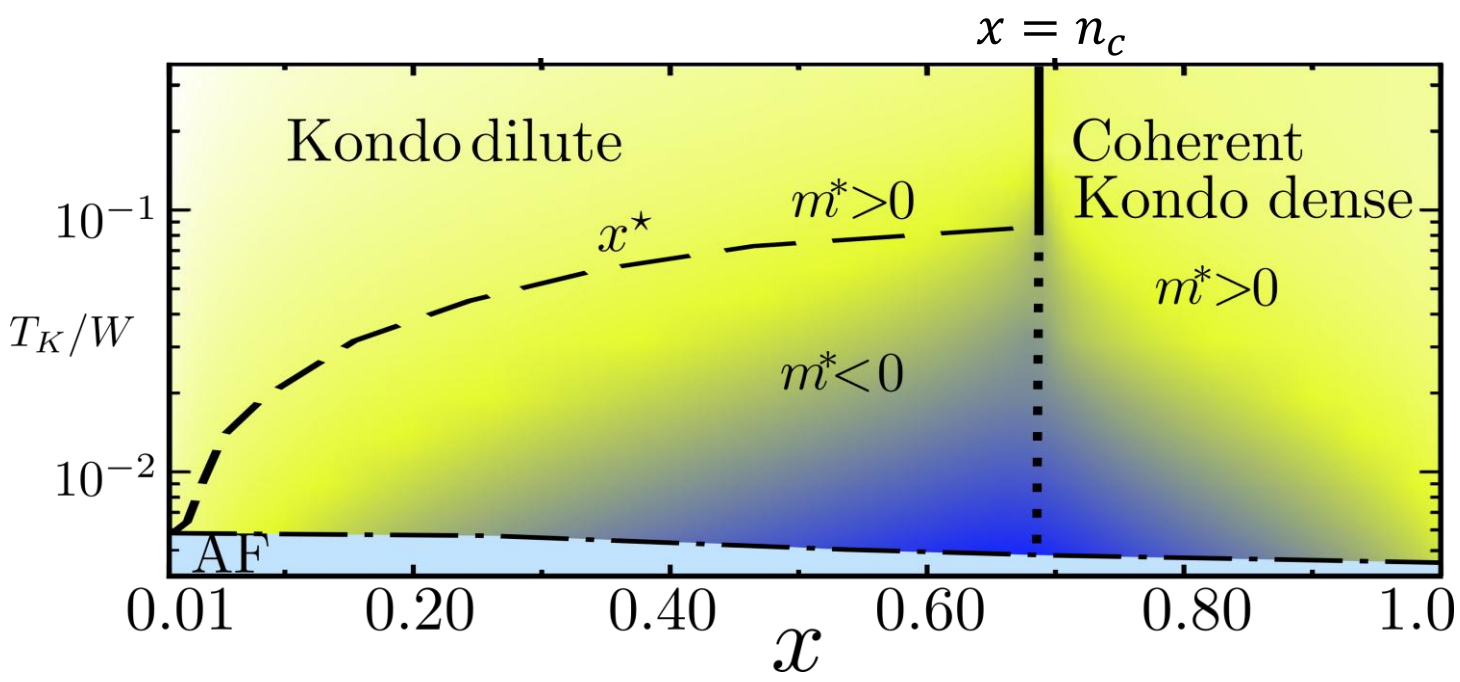


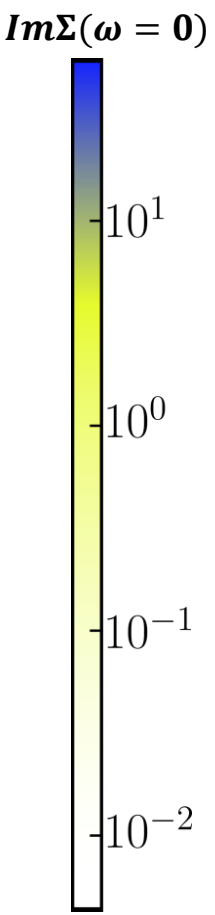
two-branch dispersion

Phase diagrams of Kondo alloys

Poudel *et al.* arXiv'21

Effective mass $\mathbf{m}^\star = \partial Re\Sigma(\omega)/\partial\omega|_{\omega=0}$

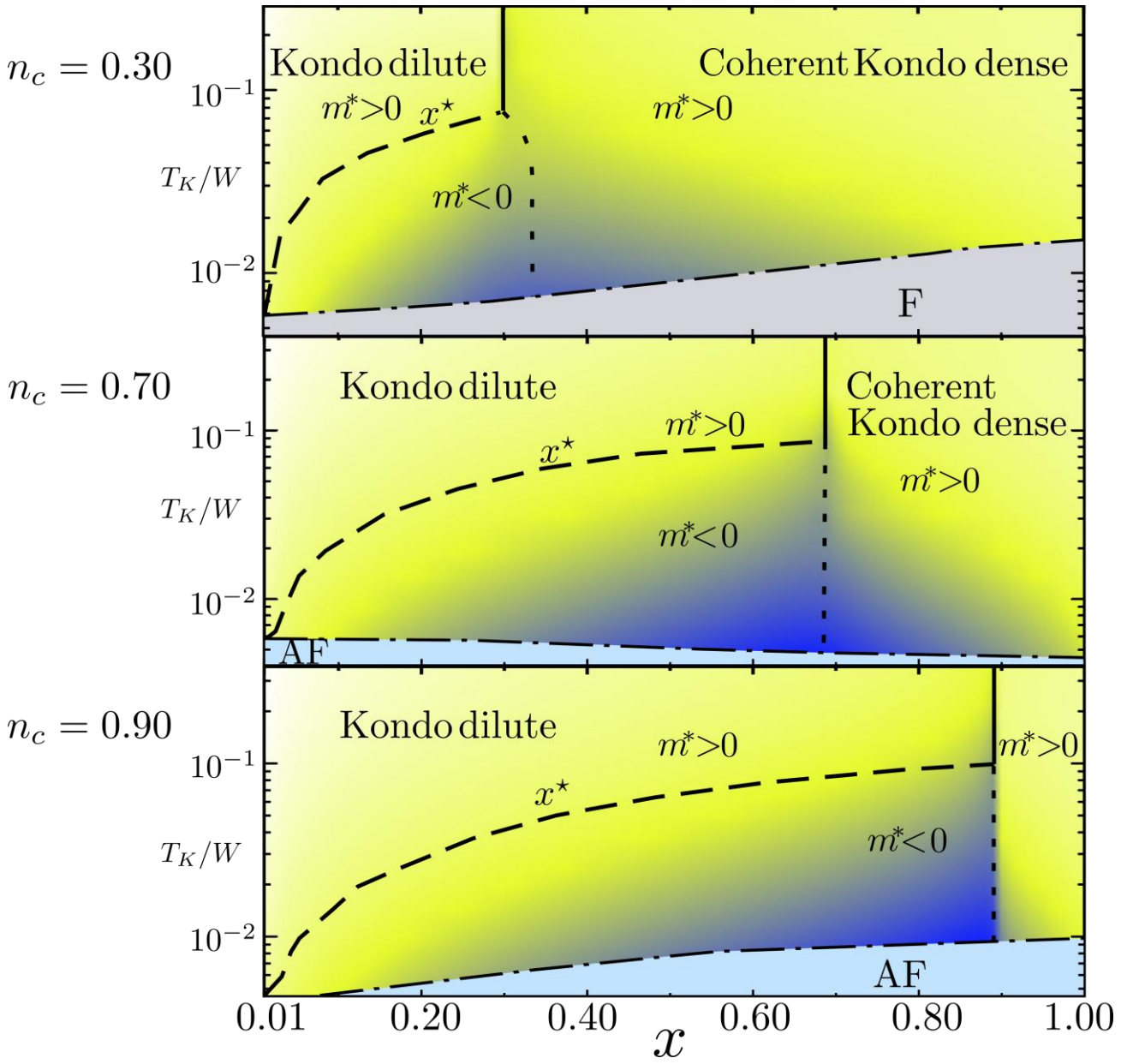


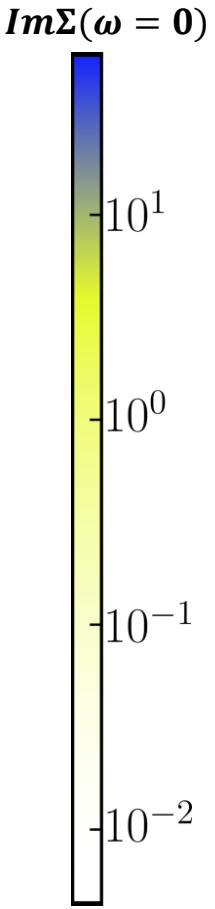


- $x = n_c$: **Lifshitz-like transition for strong T_K**
  Crossover for intermediate T_K with peudogap
- $x = x^\star$: **Merging of two branches in ARPES dispersion**
  Vanishing of effective mass $m^\star$
- **Shortened finite life-time of quasi particule around $x = n_c$**
- **Magnetic ordered phase dominates the phase diagrams at weak T_K**

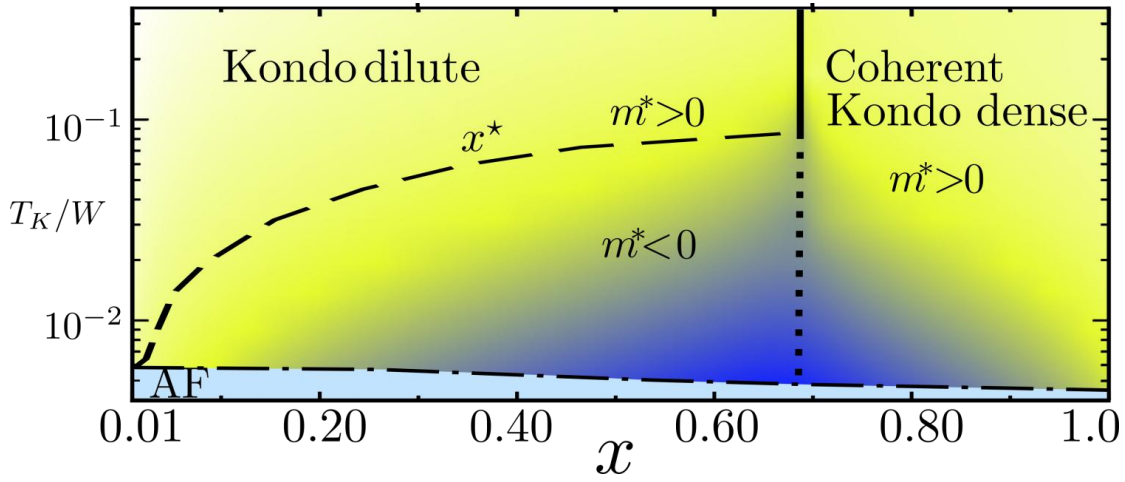
Phase diagrams of Kondo alloys

Poudel *et al.* arXiv'21





Conclusions, perspectives and acknowledgement



- **A Lifshitz-like transition is observed in Kondo alloys at strong coupling for $x = n_c$**
- **Another transition is observed at $x = x^\star$ in Kondo alloys at intermediate coupling with $m^\star = 0$**
- **This transition is « unusual » but experimental signatures are expected:**

  Pseudo gap for $x > n_c$ versus normal metal for $x < n_c$

  Particule versus hole excitations

  ARPES signatures are expected

- **Acknowledgement:**
  I would like to thank to Sébastien Burdin for his daily guidance, and Gertrud Zwicknagl, Christoph Geibel and Claudine Lacroix for their precious remarks.

université de BORDEAUX

UMR5798 Laboratoire Ondes et Matière d'Aquitaine

ANR

Fermi-NESt

DFG Deutsche Forschungsgemeinschaft

cnrs

Thank you for your attention

Breakdown of coherence in Kondo alloys

Burdin et al., PRB'2007 **Burdin** et al., PRL'2013 **José-Luiz Ferreira thesis** '2016 **Burdin** et al. 2019

Systems: rare earth alloys of the kind $Ce_xLa_{1\_x}$[...]

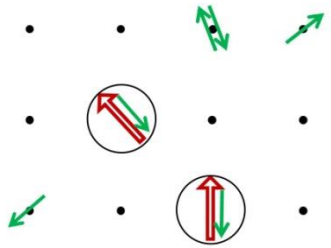
x electrons $4f^1$ from Cerium (Kondo magnetic impurities)
n_c conduction electrons from other orbitals

Normal metal when $x = 0$

A transition is predicted for $x = n_c$

Kondo lattice when $x = 1$

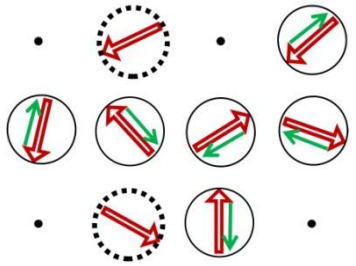
- *Diluted impurities* $x < n_c$



Cerium= hole doping

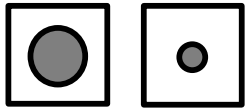
$n_c - x$ quasi-particules

- *Dense impurities* $x > n_c$

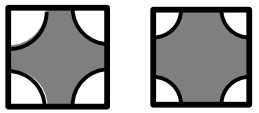


Cerium= electron doping

$n_c + x$ quasi-particules



Consequence: unconventional physical properties may emerge



First question: Is this transition still present at small Kondo coupling?

Kondo alloys

Prototype example of system: **metal + quantum magnetic impurities**

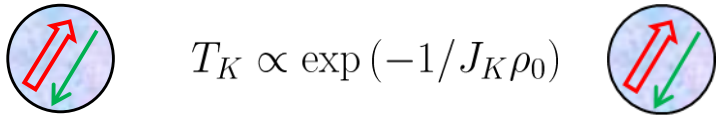
Kondo Hamiltonian: $H = H_{\mathrm{e-cond}} + J_K \sum_i \vec{S}_i \cdot \vec{\sigma}_i$

Conduction electrons
"free" density of states ρ_0

Quantum spins 1/2
Kondo Interaction (antiferromagnetic) between impurities and electrons

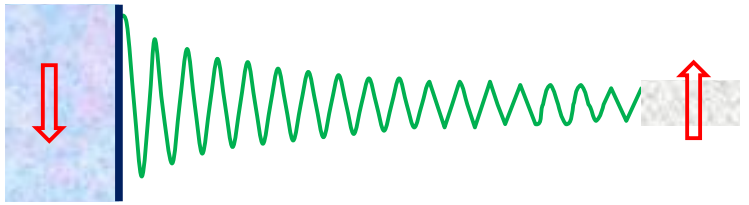
Several energy scales emerge at low temperature, including:

- *The Kondo temperature* T_K *(crossover)*
  characterizing the formation of local singlets



$$T_K \propto \exp\left(-1/J_K\rho_0\right)$$

- *The RKKY magnetic interaction energy*



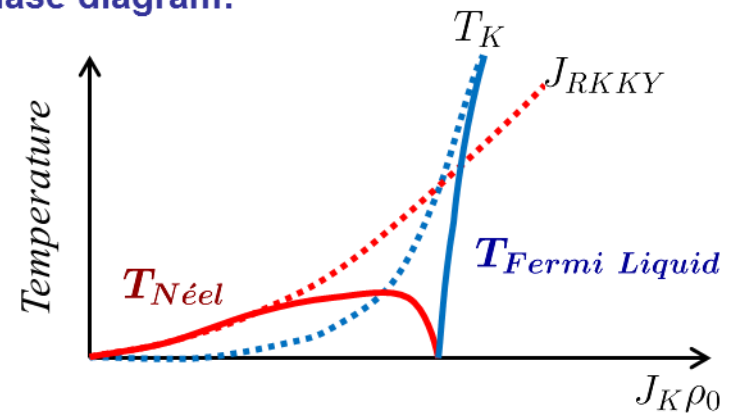
$$J_{RKKY} \propto J_K^2$$

Kondo alloys: effect of RKKY interaction

Poudel *et al.* '20 (to appear in JMMM)

Doniach'77

Phase diagram:



- $T_K \propto \exp\left(-1/J_K\rho_0\right)$
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