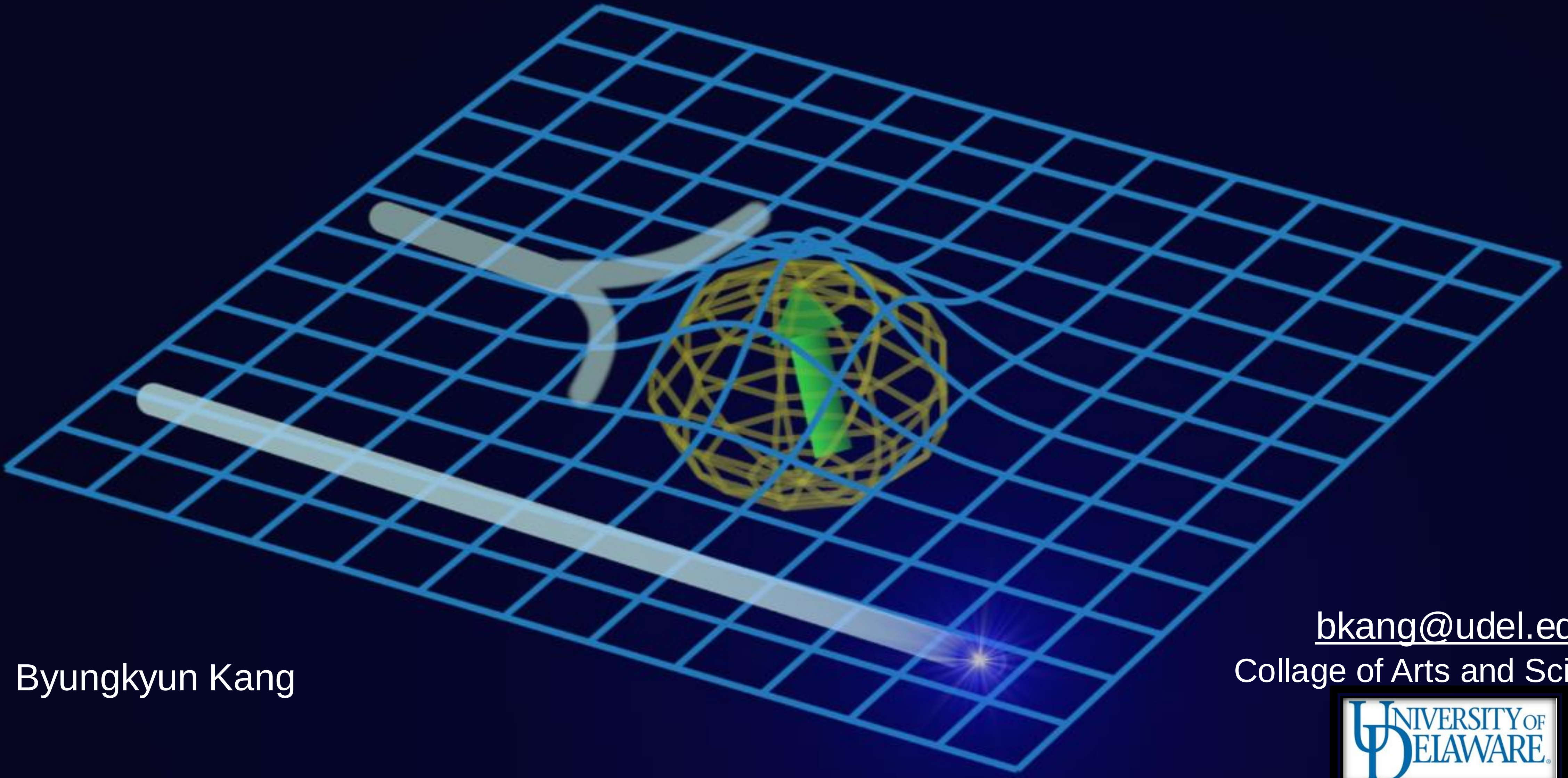
# Ab initio GW+DMFT applications in magnetism-related physics



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# Goal

- Applications of GW+DMFT

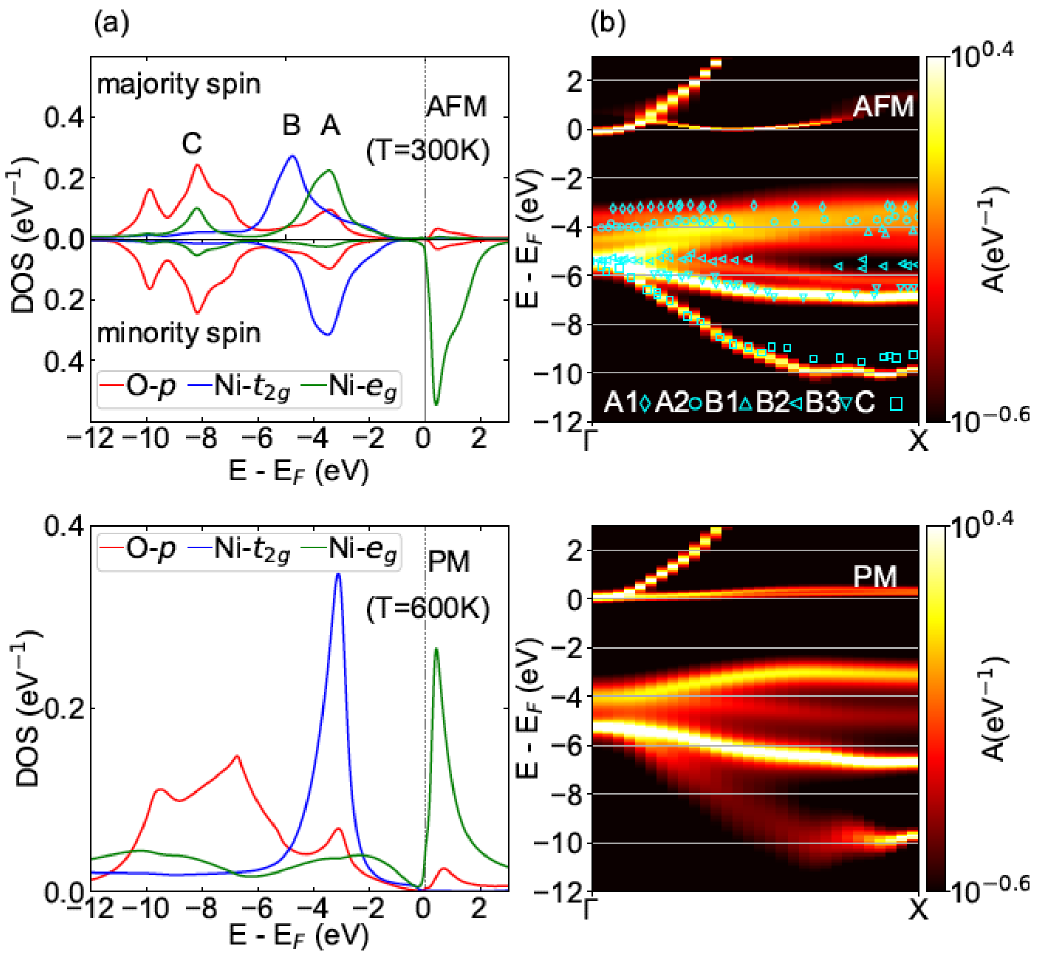
  Mott in NiO

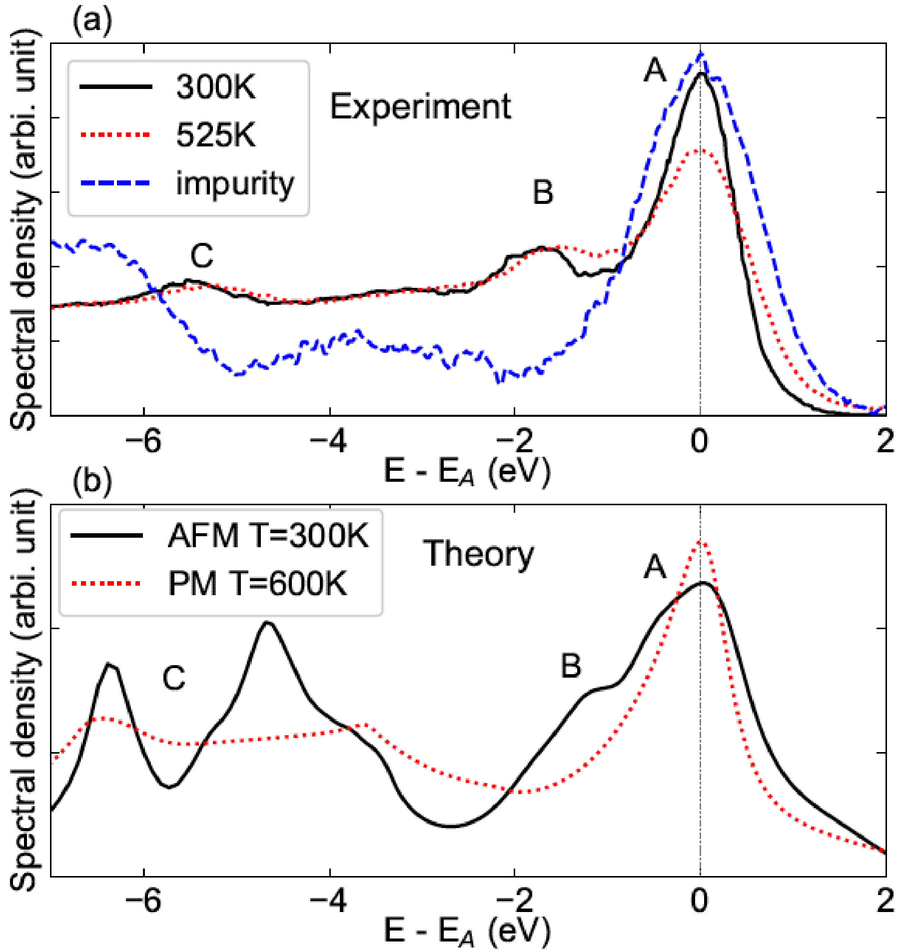
  Hund in $LaNiO_2$

  Crossover from Hund to Mott in FeSe

  Kondo effect in USbTe, $UTe_2$ and $NdNiO_2$

# Mott in NiO (LQSGW+DMFT)



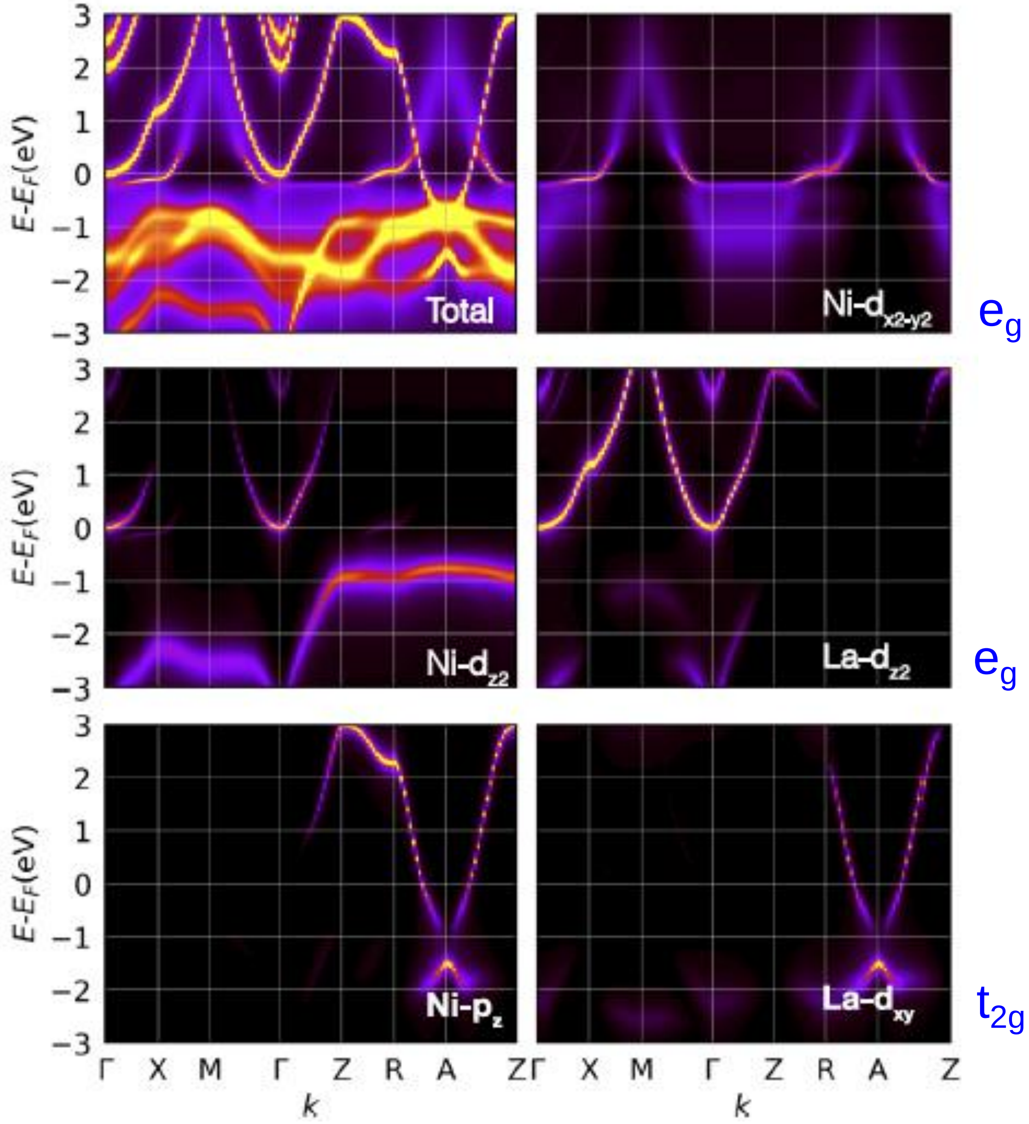




- Mott insulator appear in Paramagentic phase unlike non-magnetic LDA and LQSGW
- AFM agree with Angle Resolved Photoemission data.
- Hybridization between O-p and Ni-eg -> Majority-spin splitting explain two-peak structure.

**B. Kang** and S. Choi, H. arxiv.1908.05643

# Hund: Electronic structure of LaNiO2 (LQSGW+DMFT)

$La_{0.8}Ba_{0.2}NiO_2$




Strong two-dimensional character

Self-doping band,
Strong hybridization between other Ni orbitals and La orbitals
Three-dimensional character

- Ni two bands cross the Fermi level

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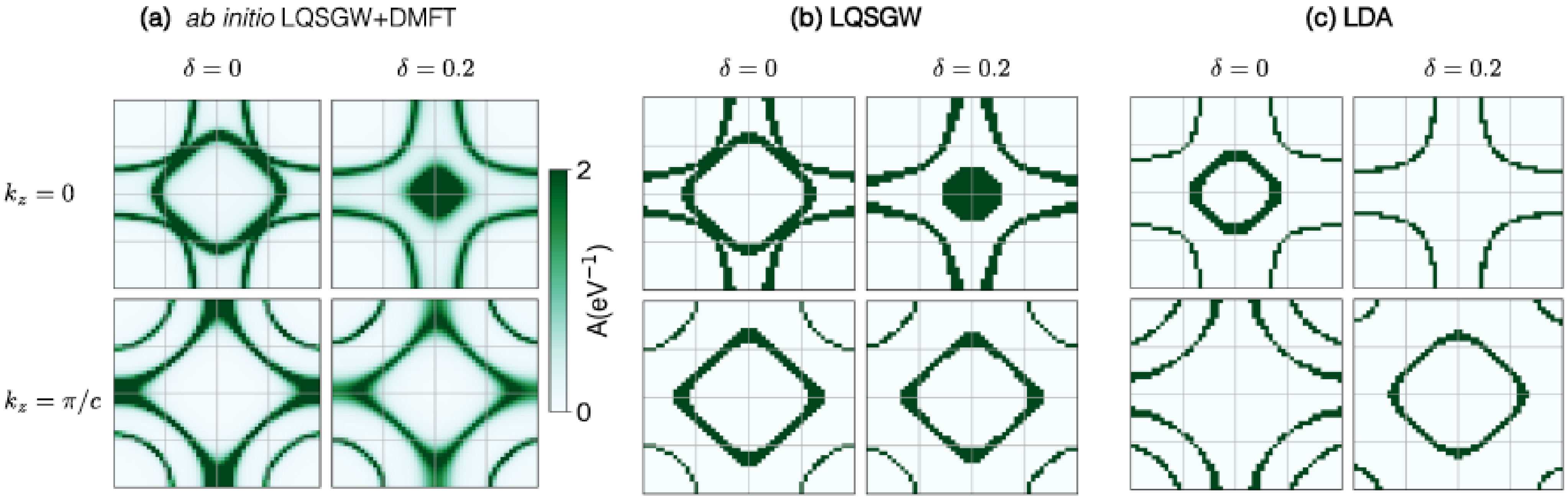
# Methods dependent

Fermi surface $La_{1-\delta}Ba_{\delta}NiO_2$

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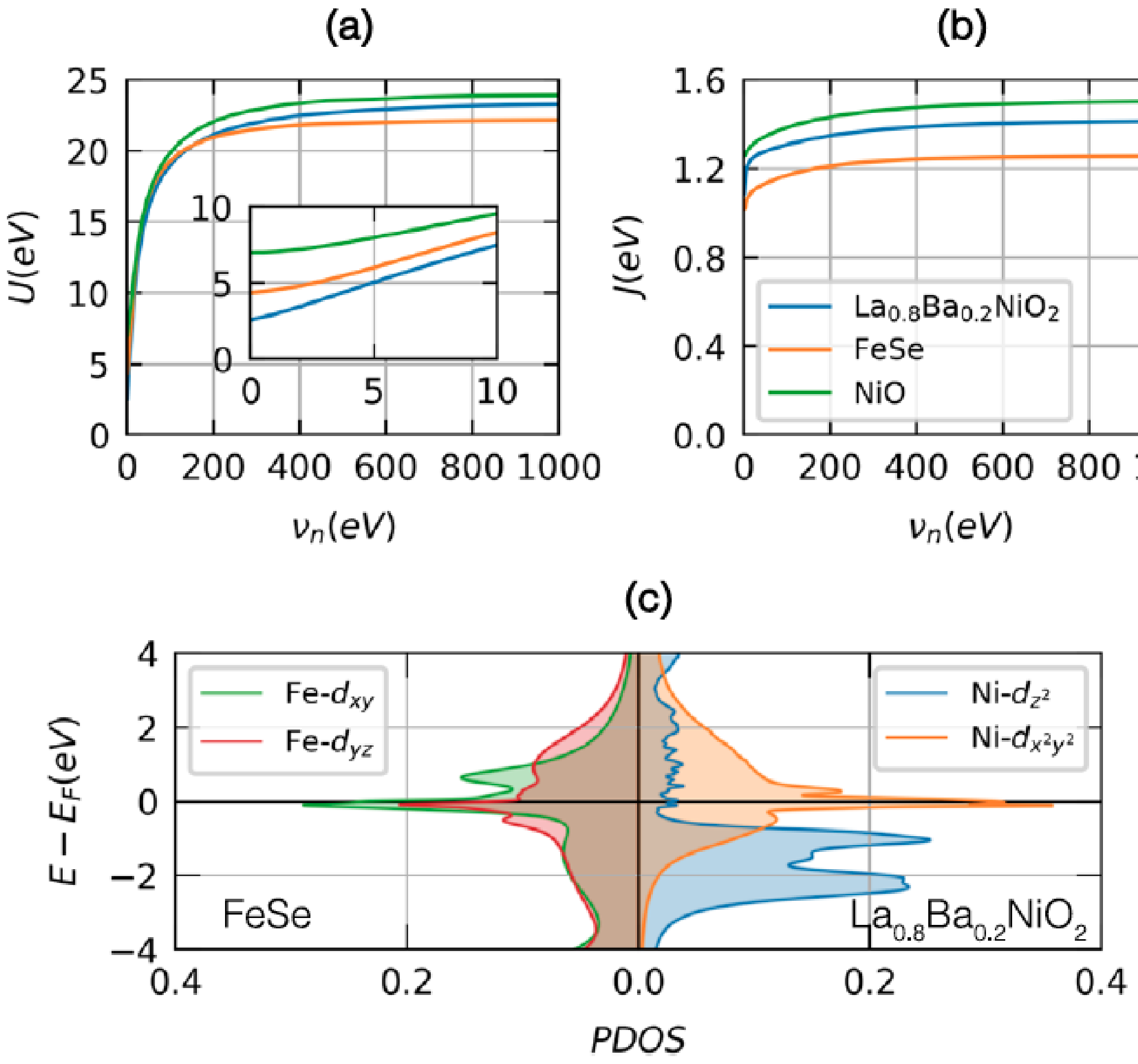
# Hundness evidence i)




Same band width albeit large static U of Fe-d => “Hundness over Mottness”

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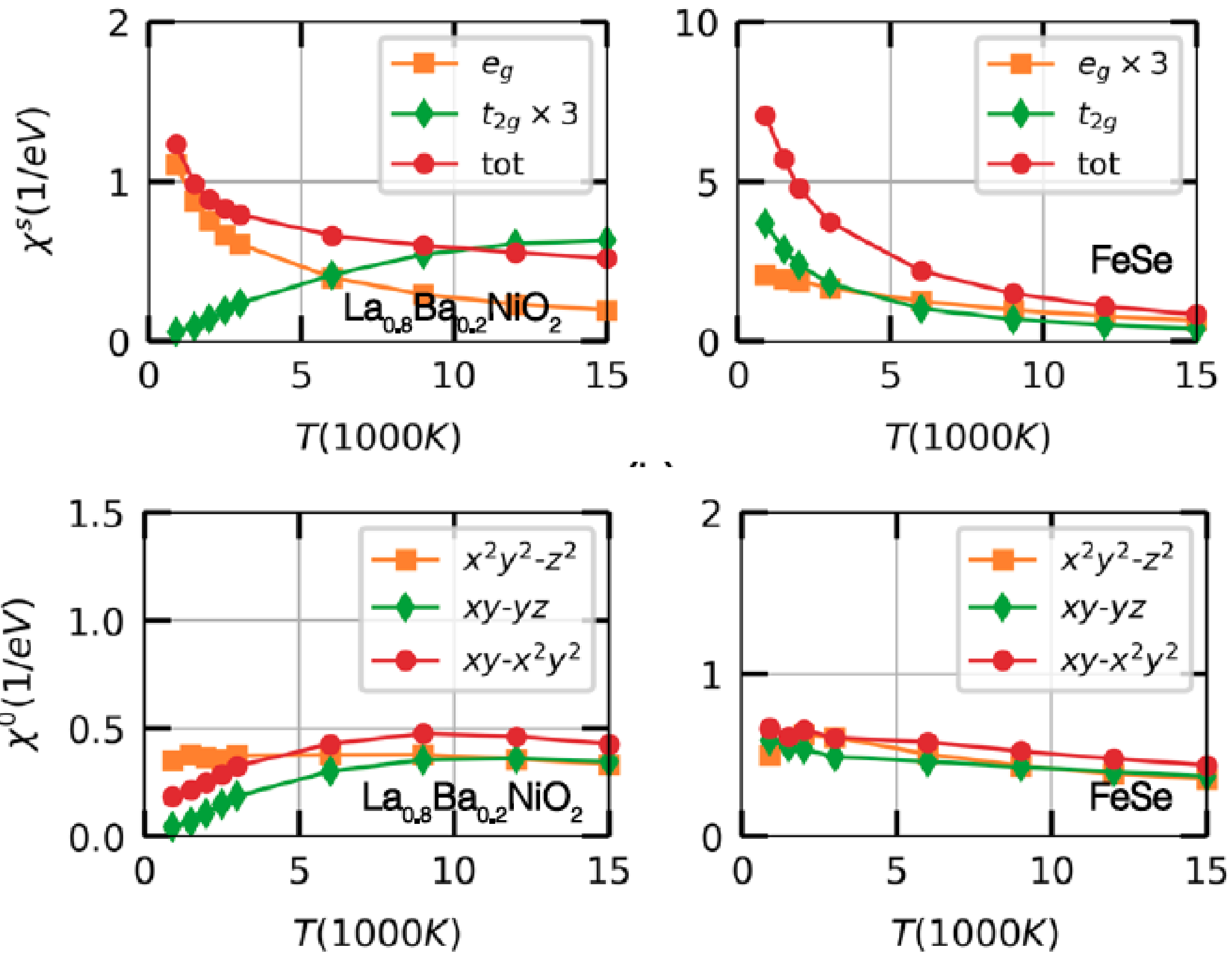
# Hundness evidence ii)




- T onset: deviates from Curie-like behaviors
  T orbital >> T spin
- Hundness: spin-orbital separation: a two-step screening process in which local spin moment is screened at much lower temperature than local orbital polarization.

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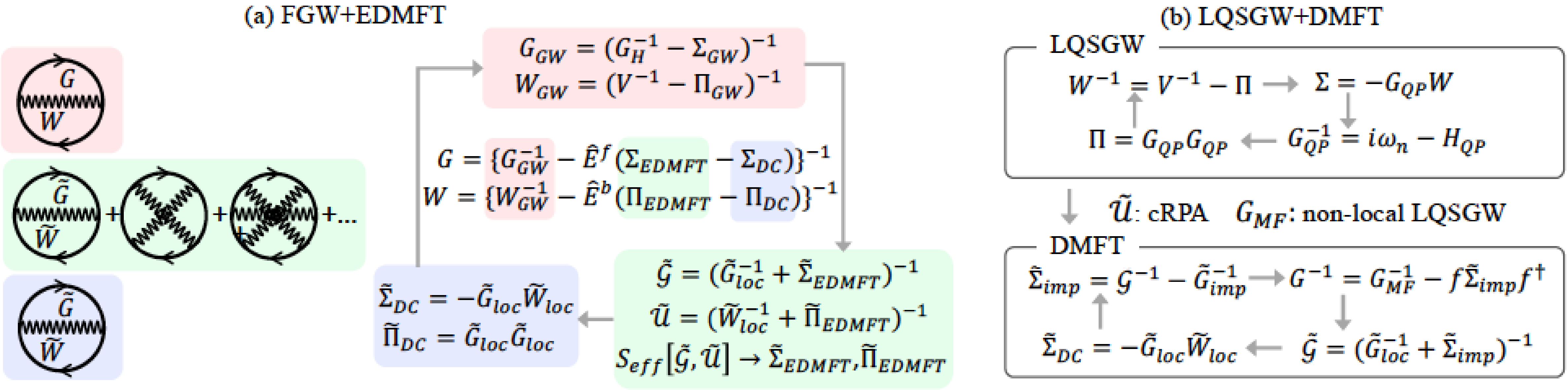
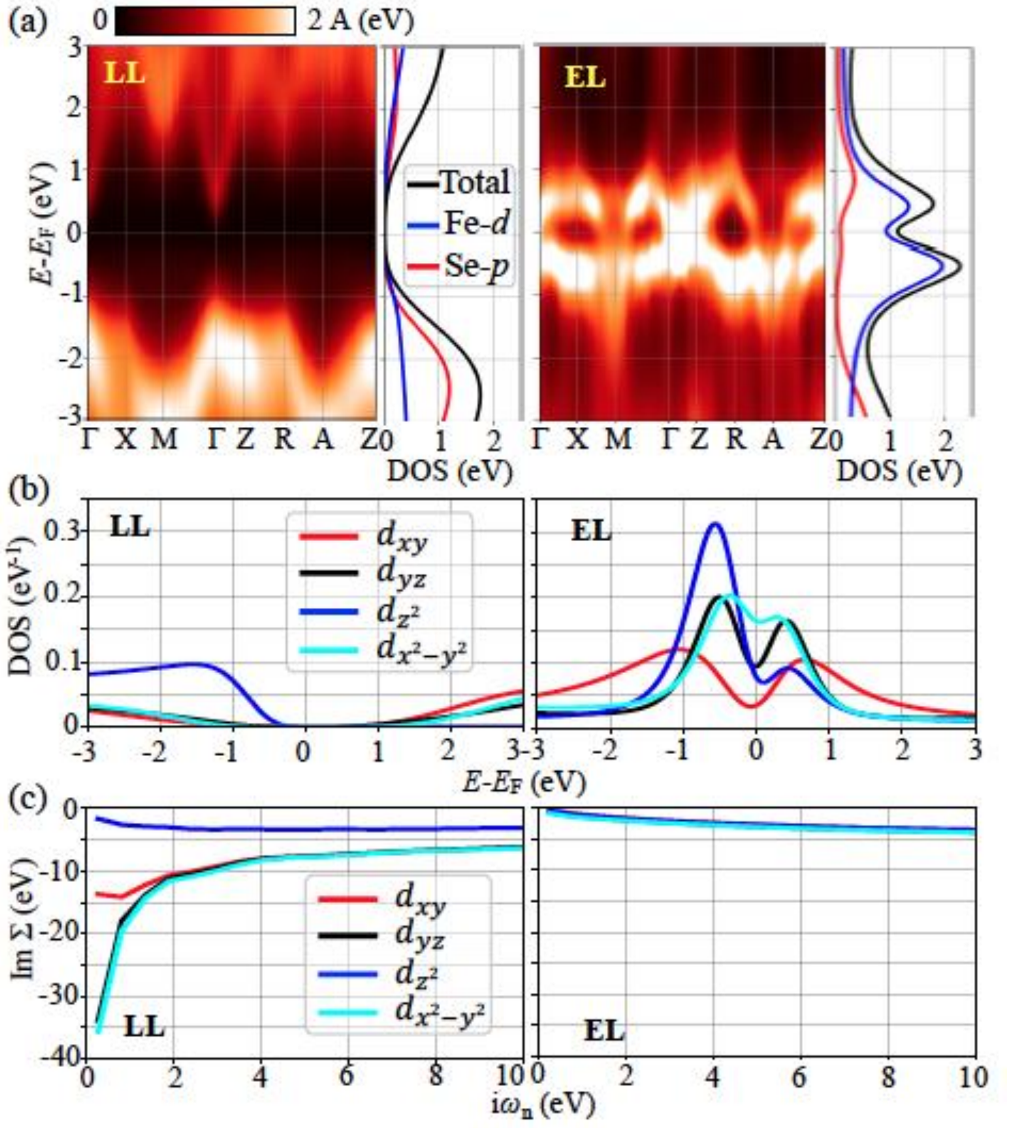
# First developing fully self-consistent GW+EDMFT




**B. Kang*** et al. Phys. Rev. Lett. 132, 266506 (2024)

# FGW+EDMFT capture Mott-insulator state of 2D FeSe

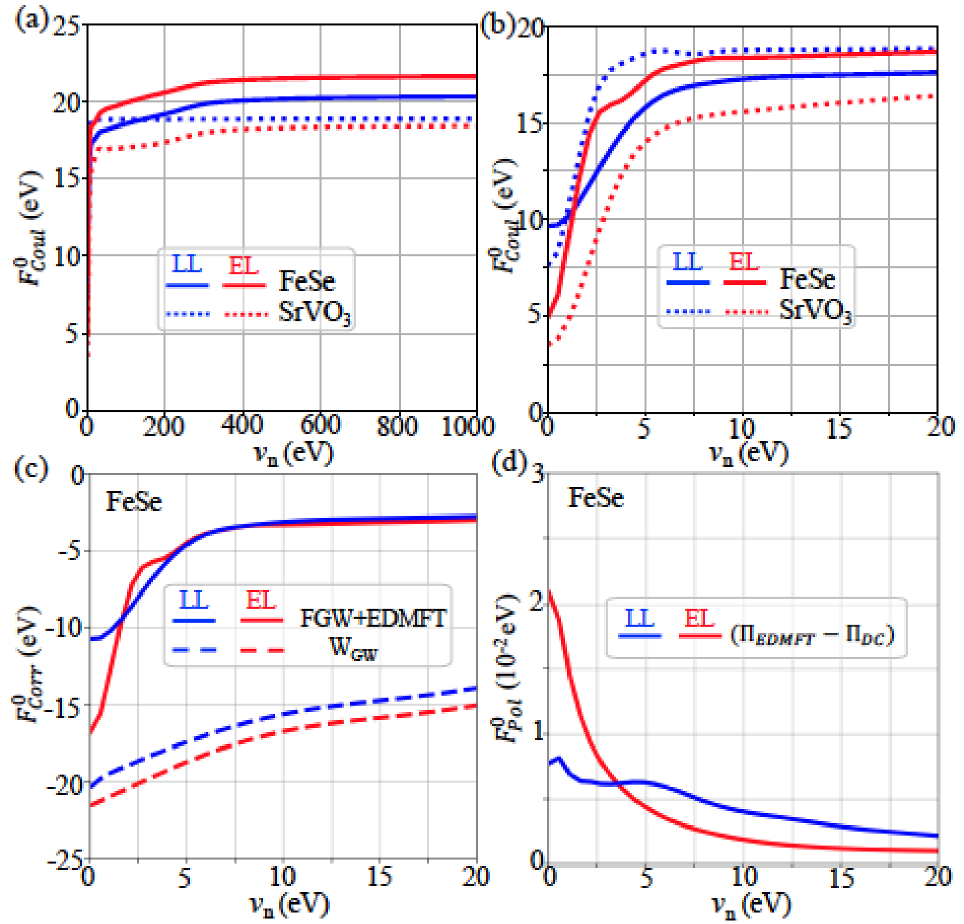


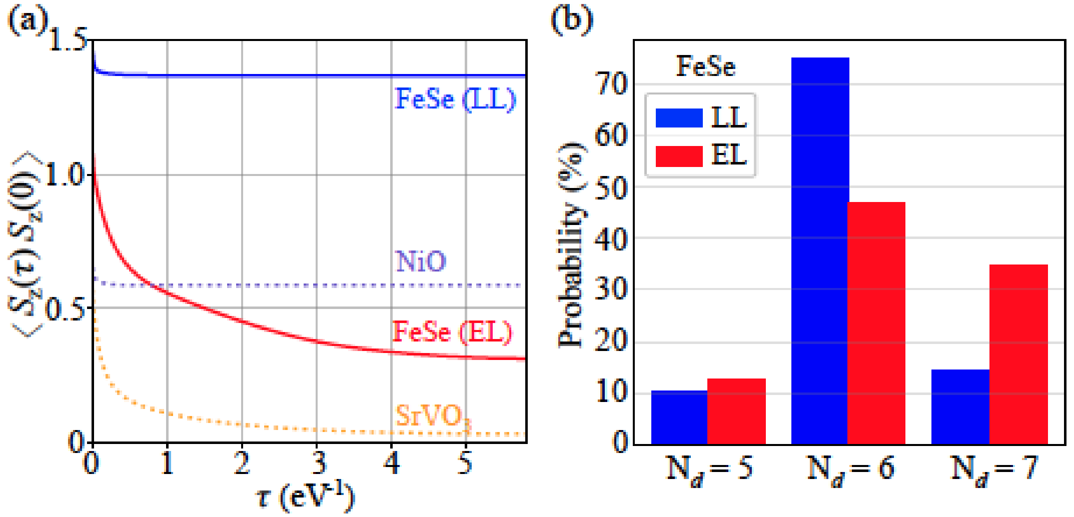

| | FGW+EDMFT | | LQSGW+DMFT | |
|---|---|---|---|---|
| | Large LC (LL) | Exp. LC (EL) | Large LC (LL) | Exp. LC (EL) |
| $d_{xy}$ | 1.02 | 1.22 (0.52) | 1.40 (0.40) | 1.22 (0.58) |
| $d_{yz}$ | 1.02 | 1.12 (0.49) | 1.12 (0.32) | 1.18 (0.52) |
| $d_{z^2}$ | 1.94 | 1.58 (0.49) | 1.26 (0.33) | 1.40 (0.49) |
| $d_{xz}$ | 1.02 | 1.12 (0.49) | 1.12 (0.32) | 1.18 (0.52) |
| $d_{x^2-y^2}$ | 1.02 | 1.24 (0.49) | 1.08 (0.25) | 1.28 (0.52) |

- For large lattice, strong self-energy appear leading to Mott gap.
- Half filling occupancy support the Mott phase.
- Mott state appeared only within FGW+EDMFT

**B. Kang*** et al. Phys. Rev. Lett. 132, 266506 (2024)

# FGW+EDMFT capture Mott-insulator state of 2D FeSe



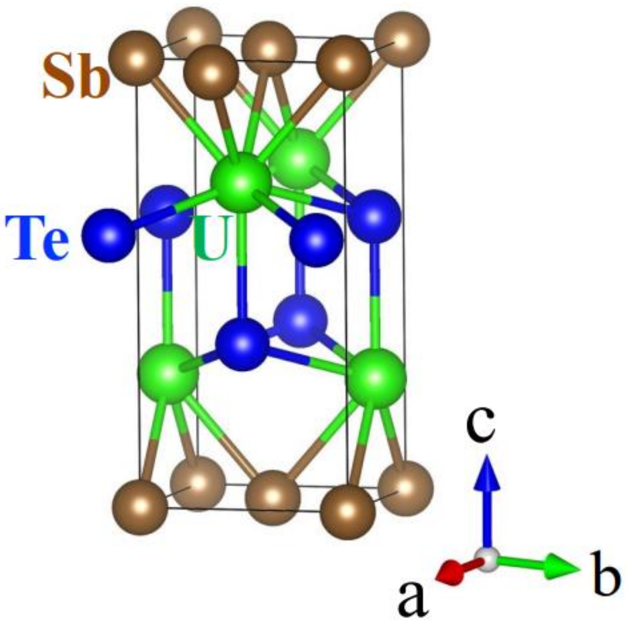



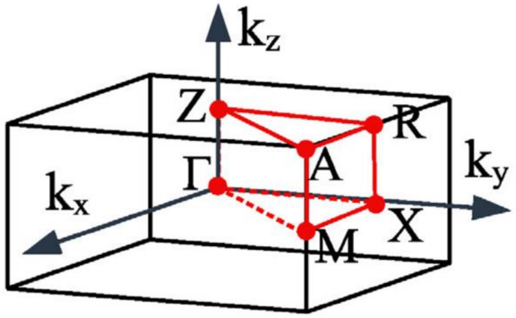

- Large lattice exhibit strong U (not in LQSGW+DMFT)
- Mott state appeared accompanied with weaking of both local and non-local screening.
- Strong local magnetic moments in LL FeSe

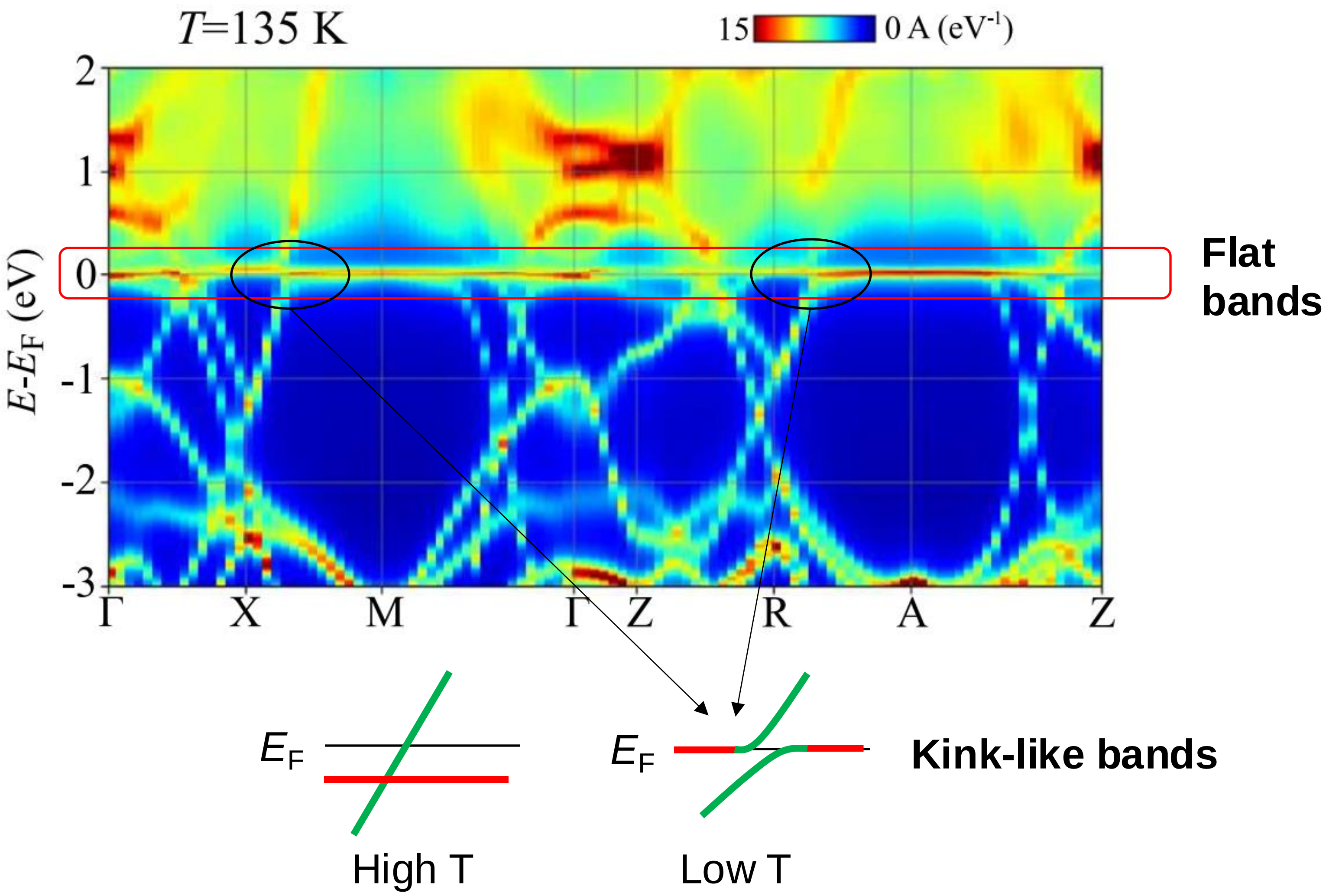
**B. Kang*** et al. Phys. Rev. Lett. 132, 266506 (2024)

# Kondo: USbTe – calculated electronic structure (LQSGW+DMFT)

P4/nmm



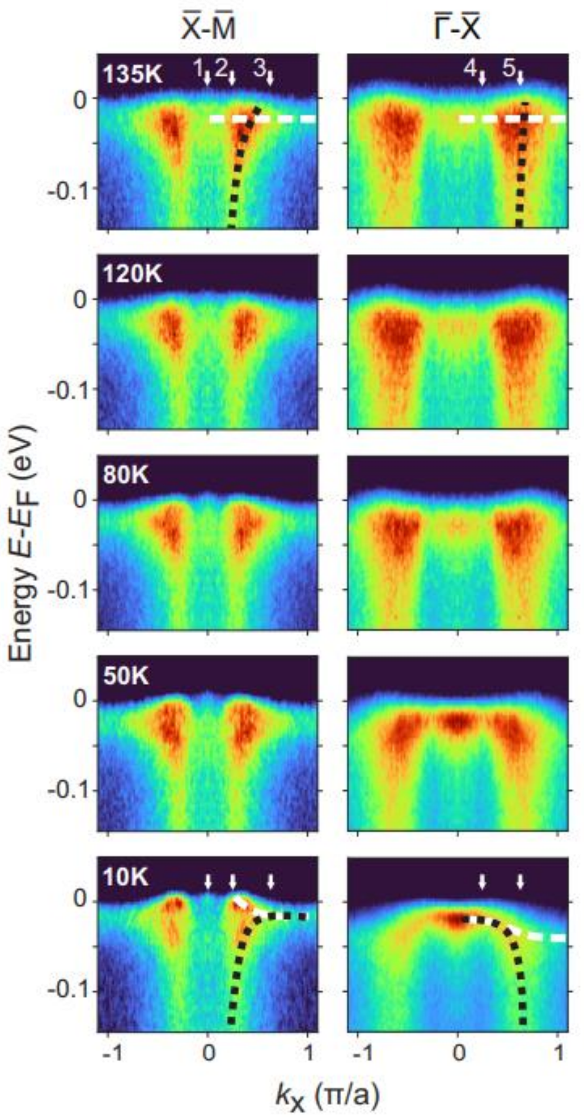




- Distorted conduction bands and flat *f* bands at the Fermi level are a feature of Kondo effect

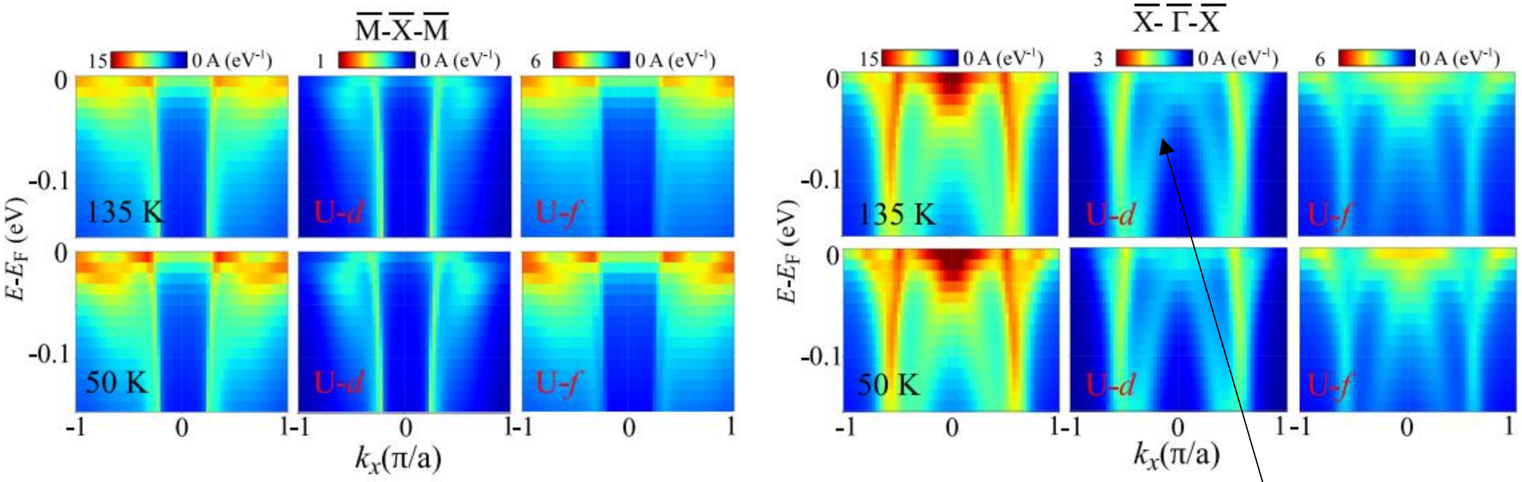
H. Siddiquee, C. Broyles, E. Kotta, S. Liu, S. Peng, T. Kong, **B. Kang**, Q. Zhu, Y. Lee, L. Ke, H. Weng, J. D. Denlinger, L. A. Wray, S. Ran Nature Communications 14, 527 (2023)

# Kondo effect in USbTe

ARPES (Experiment)




LQSGW+DMFT (Theory)



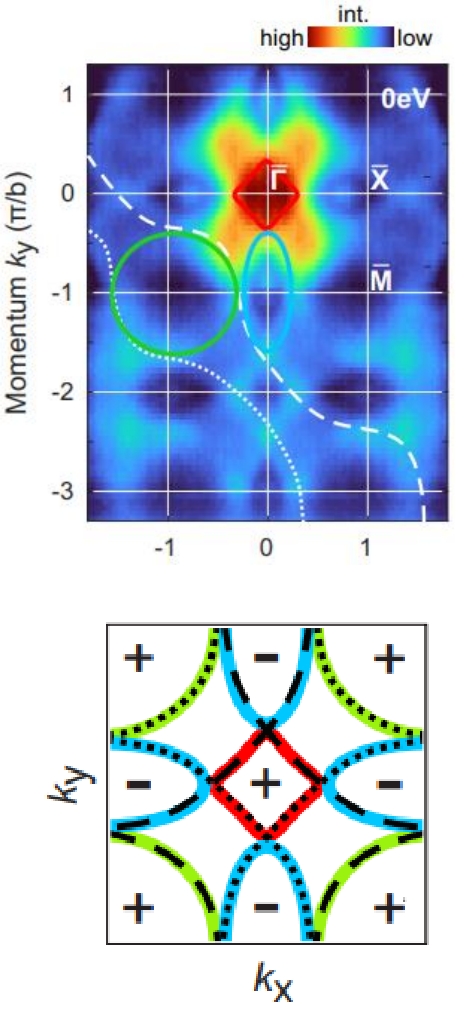

Not observed in ARPES due to incident energy dependence

- U-*d* consist of one parabolic band
- More dispersion of U-*d* at 50K
- Agree with ARPES
- Kondo hybridization stronger at 50K
- Significant contribution of flat *f* bands to the Fermi leve

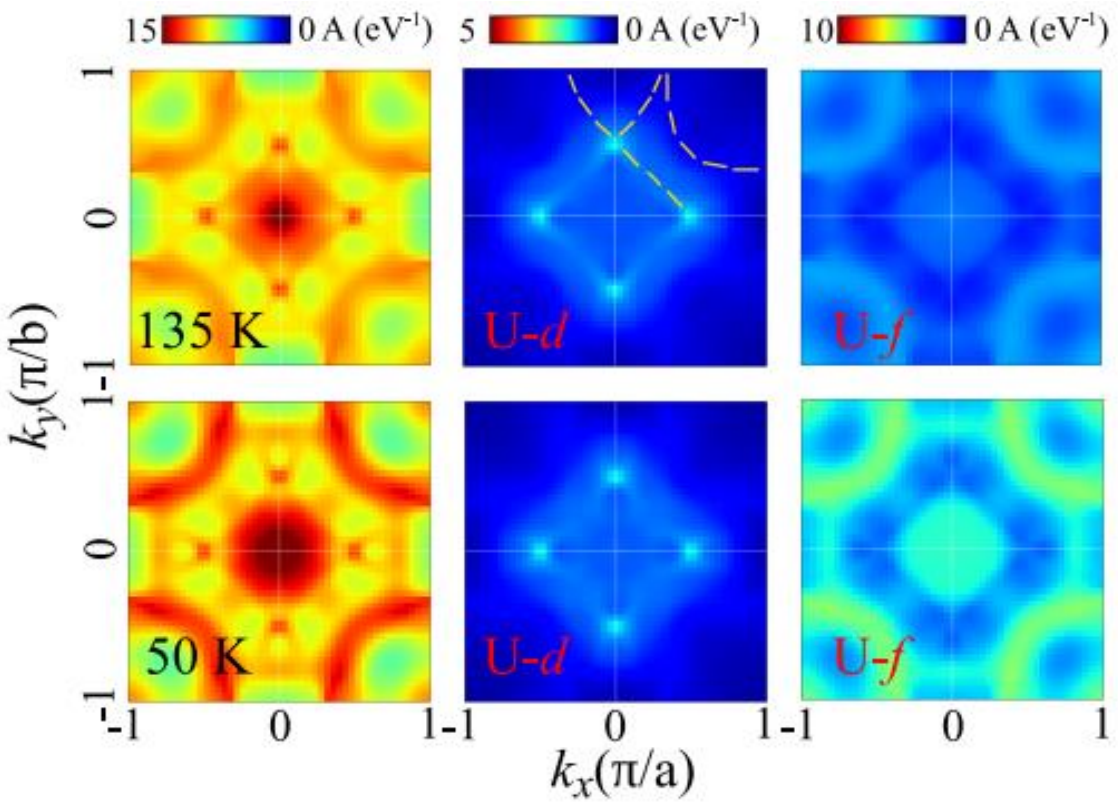
H. Siddiquee, C. Broyles, E. Kotta, S. Liu, S. Peng, T. Kong, **B. Kang**, Q. Zhu, Y. Lee, L. Ke, H. Weng, J. D. Denlinger, L. A. Wray, S. Ran Nature Communications 14, 527 (2023)

# Fermi surface of USbTe

## ARPES (Experiment)




## LQSGW+DMFT (Theory)




- U-*d* FS similar with ARPES
- Strong U-*f* on FS at 50K captured in theory
- Progress of coherent f bands

H. Siddiquee, C. Broyles, E. Kotta, S. Liu, S. Peng, T. Kong, **B. Kang**, Q. Zhu, Y. Lee, L. Ke, H. Weng, J. D. Denlinger, L. A. Wray, S. Ran Nature Communications 14, 527 (2023)

# Orbital selective Kondo: $UTe_2$-Calculated electronic structure

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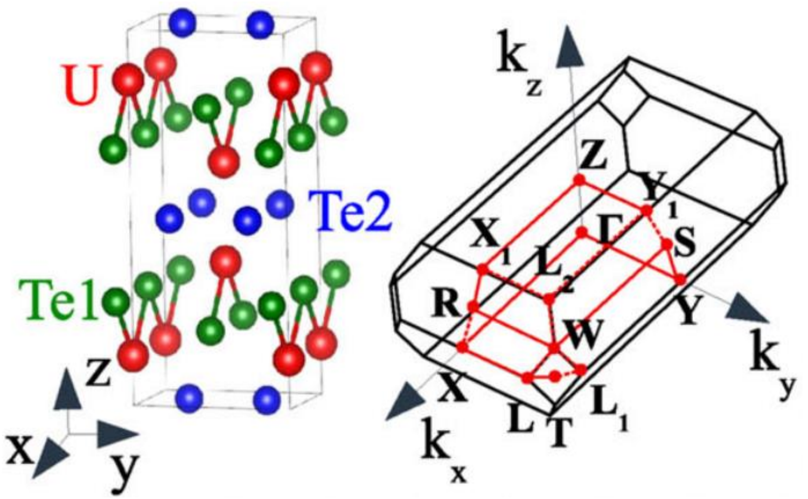


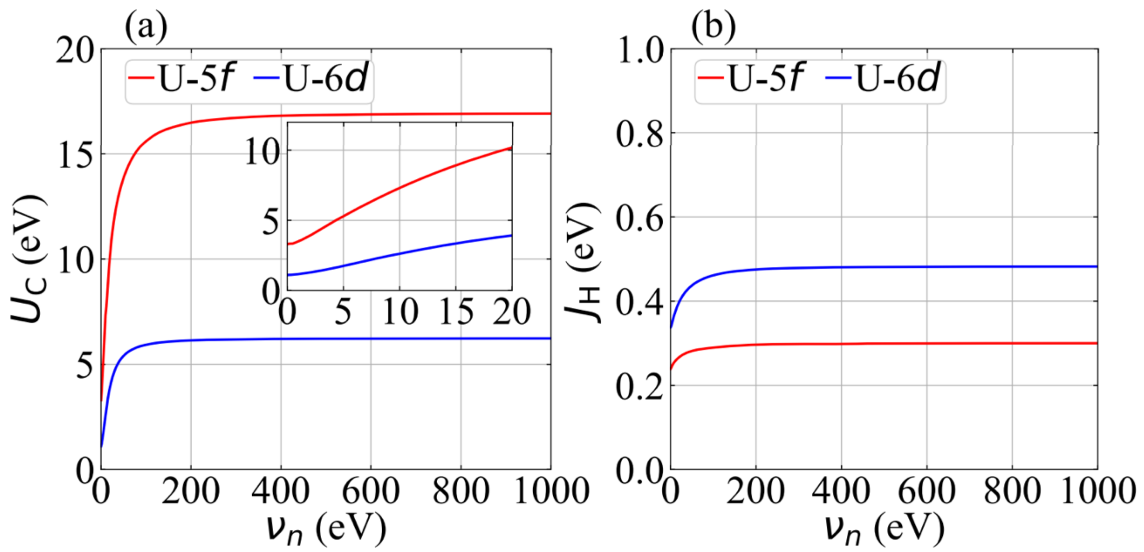



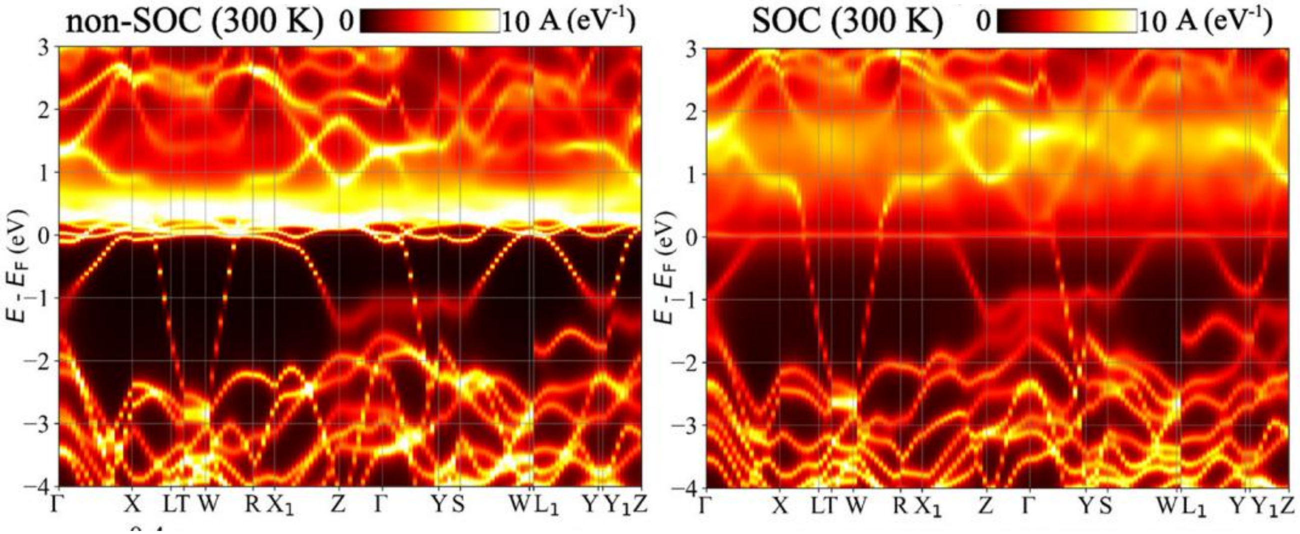

**Table 1.** Calculated electron occupation of U-5*f* orbitals in $UTe_2$ at $T = 300$ K. U-5*f* orbitals are labelled for convenience in this work.

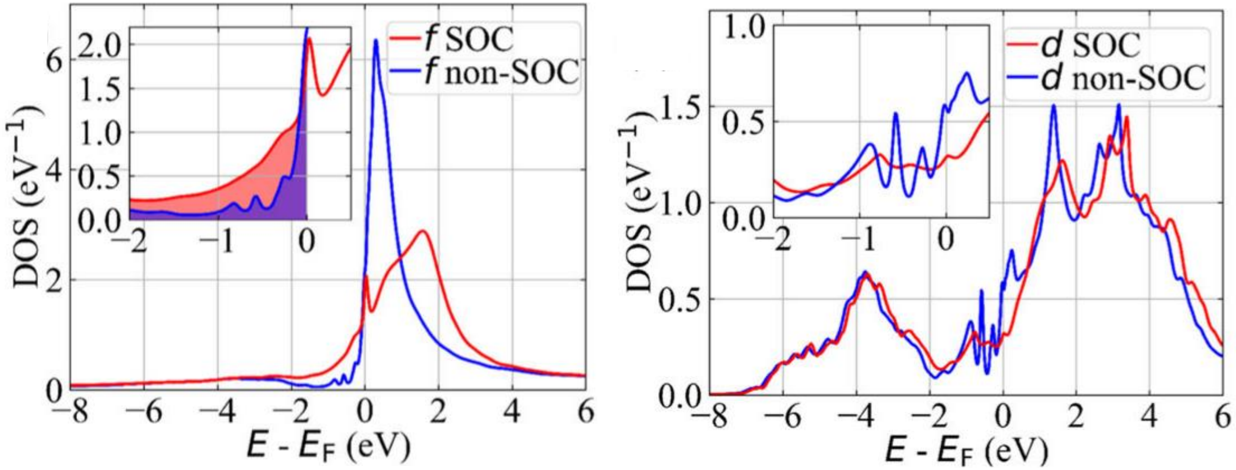
| *j* | 5/2 | | | | | | 7/2 | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $j_z$ | −2.5 | −1.5 | −0.5 | 0.5 | 1.5 | 2.5 | −3.5 | −2.5 | −1.5 | −0.5 | 0.5 | 1.5 | 2.5 | 3.5 |
| Occupation | 0.39 | 0.22 | 0.39 | 0.40 | 0.22 | 0.42 | 0.02 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.02 |
| Label | $f_1$ | $f_2$ | $f_3$ | $f_4$ | $f_5$ | $f_6$ | $f_7$ | $f_8$ | $f_9$ | $f_{10}$ | $f_{11}$ | $f_{12}$ | $f_{13}$ | $f_{14}$ |

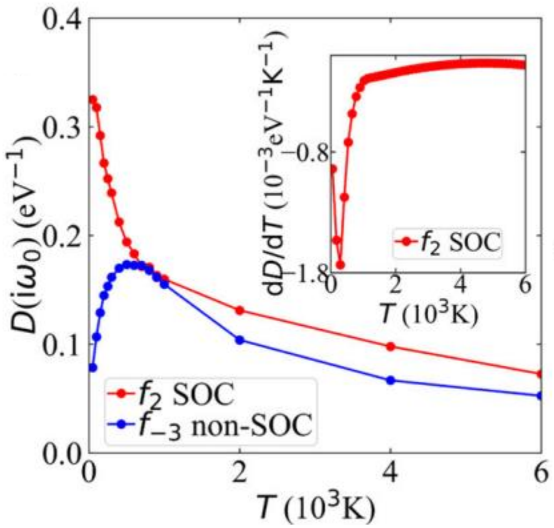
- No purposive judgements, such as intentional selection of on-site Coulomb interaction and disregarding spin-orbit
- Total occupation in U-*f* is 2.27, indicates significant local magnetic moment.

**B. Kang*,** S. Choi, H. Kim npj Quantum Materials 7, 64 (2022)

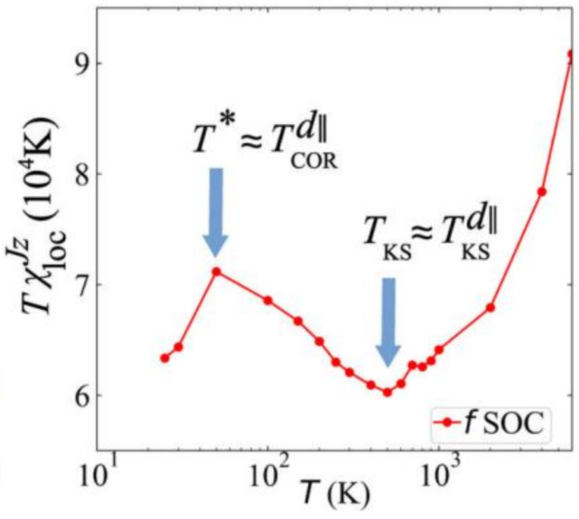
# $UTe_2$-SOC boosted Kondo effects




$$D(i\omega_0) = -\frac{1}{\pi}\mathrm{Im}G(i\omega_0)$$




$$\chi^{J_z}_{loc} = \int_0^\beta d\tau \langle J_z(\tau) J_z(0)\rangle$$

- SOC (non-SOC) : coherent (incoherent) electronic structure
- Flat f bands and peak at $E_F$ within SOC
- Deviates from Curie-Weiss behaviors: onset Kondo scattering

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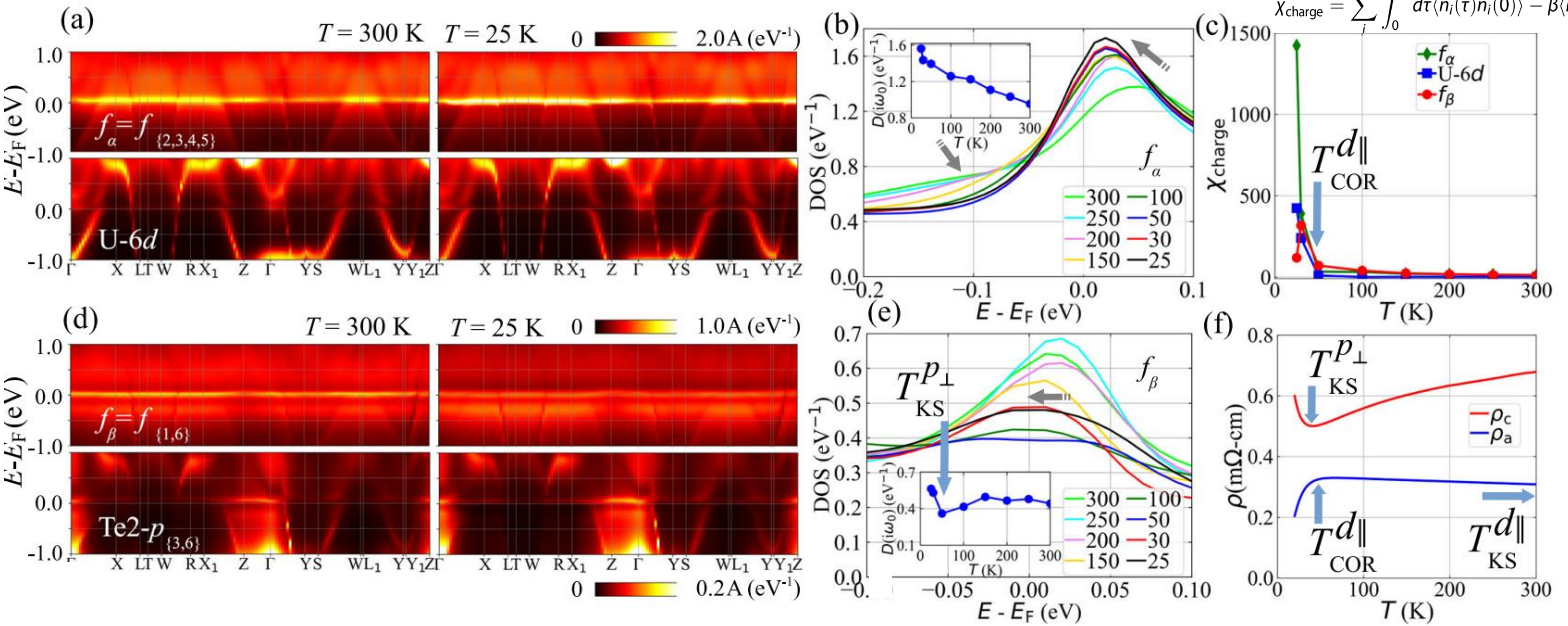
# UTe$_2$- Orbital selective Kondo effect

$$\chi_{\text{charge}} = \sum_i \int_0^\beta d\tau \langle n_i(\tau) n_i(0) \rangle - \beta \langle n_i \rangle^2$$




- $f_\alpha$ exhibits coherent Kondo lattice below 50 K; rapid drop in measured resistivity along the a axis
- $f_\beta$ exhibits onset of Kondo scattering around 50 K; resistivity upturn in measured resistivity along the c axis

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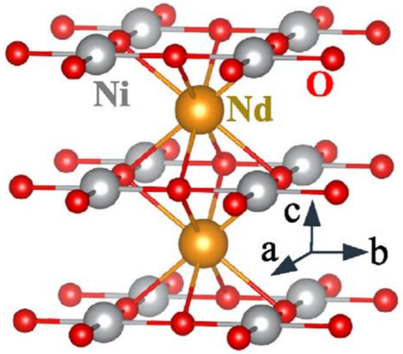
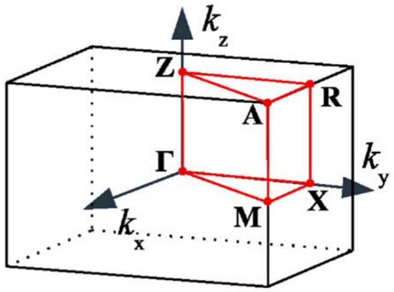
# Kondo: $NdNiO_2$- Calculated electronic structure

P4/mmm



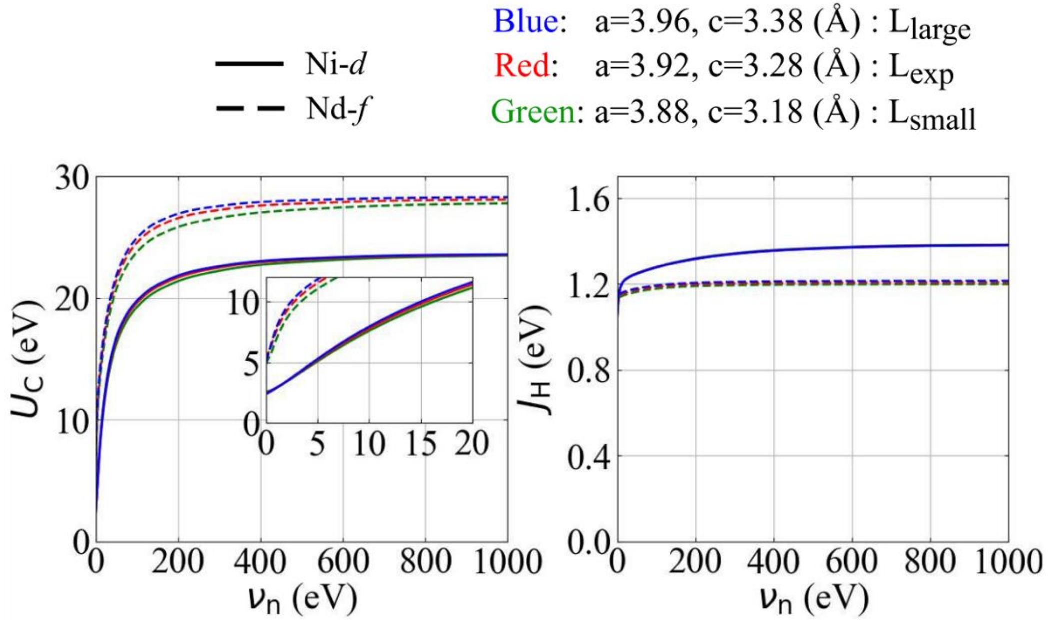




**Table 1. Calculated electron occupation of Nd-4*f* and Ni-3*d* orbitals in $NdNiO_2$ at $T$ = 300 K**

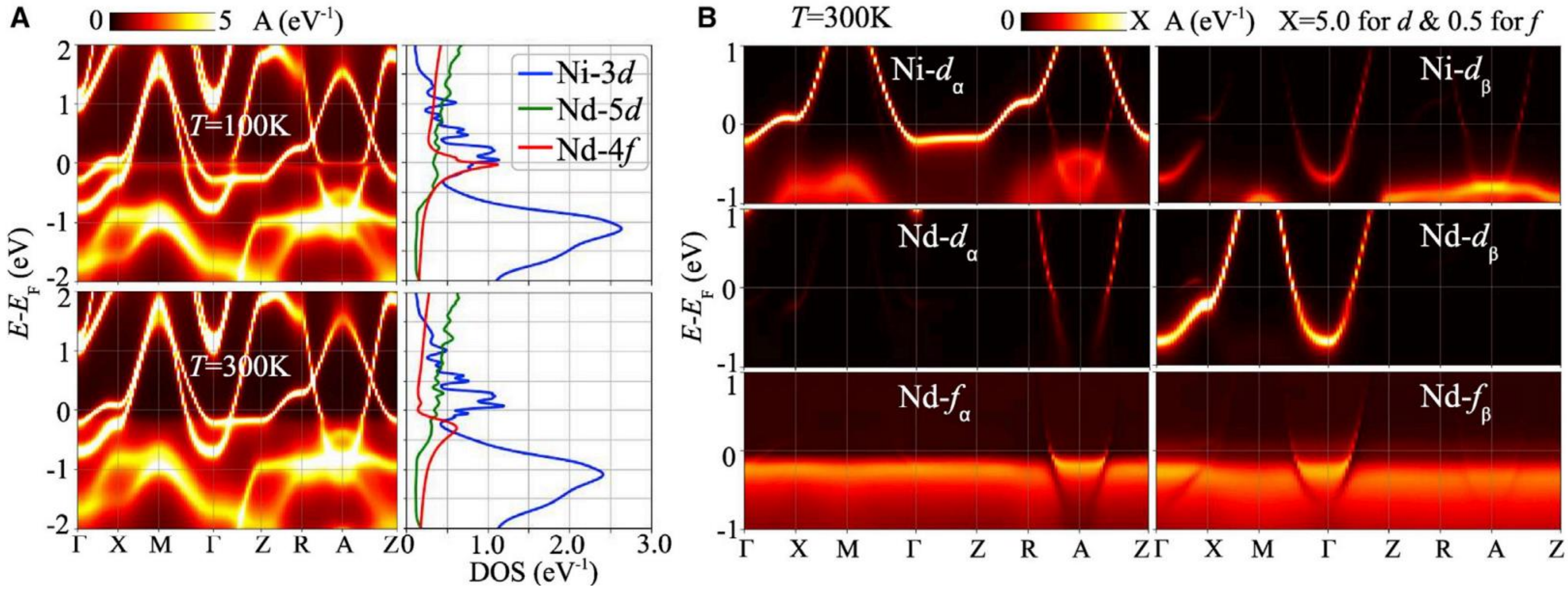
| | Nd-4*f* | | | | | | | | | | | | | | Ni-3*d* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *j* | 5/2 | | | | | | 7/2 | | | | | | | | 3/2 | | | | 5/2 | | | | | |
| $j_z$ | −2.5 | −1.5 | −0.5 | 0.5 | 1.5 | 2.5 | −3.5 | −2.5 | −1.5 | −0.5 | 0.5 | 1.5 | 2.5 | 3.5 | −1.5 | −0.5 | 0.5 | 1.5 | −2.5 | −1.5 | −0.5 | 0.5 | 1.5 | 2.5 |
| Label | $f_1$ | $f_2$ | $f_3$ | $f_4$ | $f_5$ | $f_6$ | $f_7$ | $f_8$ | $f_9$ | $f_{10}$ | $f_{11}$ | $f_{12}$ | $f_{13}$ | $f_{14}$ | $d_1$ | $d_2$ | $d_3$ | $d_4$ | $d_5$ | $d_6$ | $d_7$ | $d_8$ | $d_9$ | $d_{10}$ |
| Occupation | 0.36 | 0.13 | 0.42 | 0.43 | 0.13 | 0.37 | 0.01 | 0.01 | 0.01 | 0.02 | 0.02 | 0.01 | 0.01 | 01 | 0.91 | 0.89 | 0.89 | 0.91 | 0.73 | 0.93 | 0.84 | 0.84 | 0.93 | 0.73 |

Nd-4*f* and Ni-3*d* orbitals are labeled for convenience in this work.

- static U of ~5 eV & occupancy of 1.9 for Nd-*f*
- Possibility of localized magnetic moment ?
- Nd-4*f* affects the Fermi surface ?

**B. Kang*,** H. Kim, Q. Zhu, C.H. Park Cell Reports Physical Science 4, 101325 (2023)

# $NdNiO_2$- Nd-4*f* at the Fermi level




**Significant Nd-4*f* bands shift towards the Fermi level upon cooling!**

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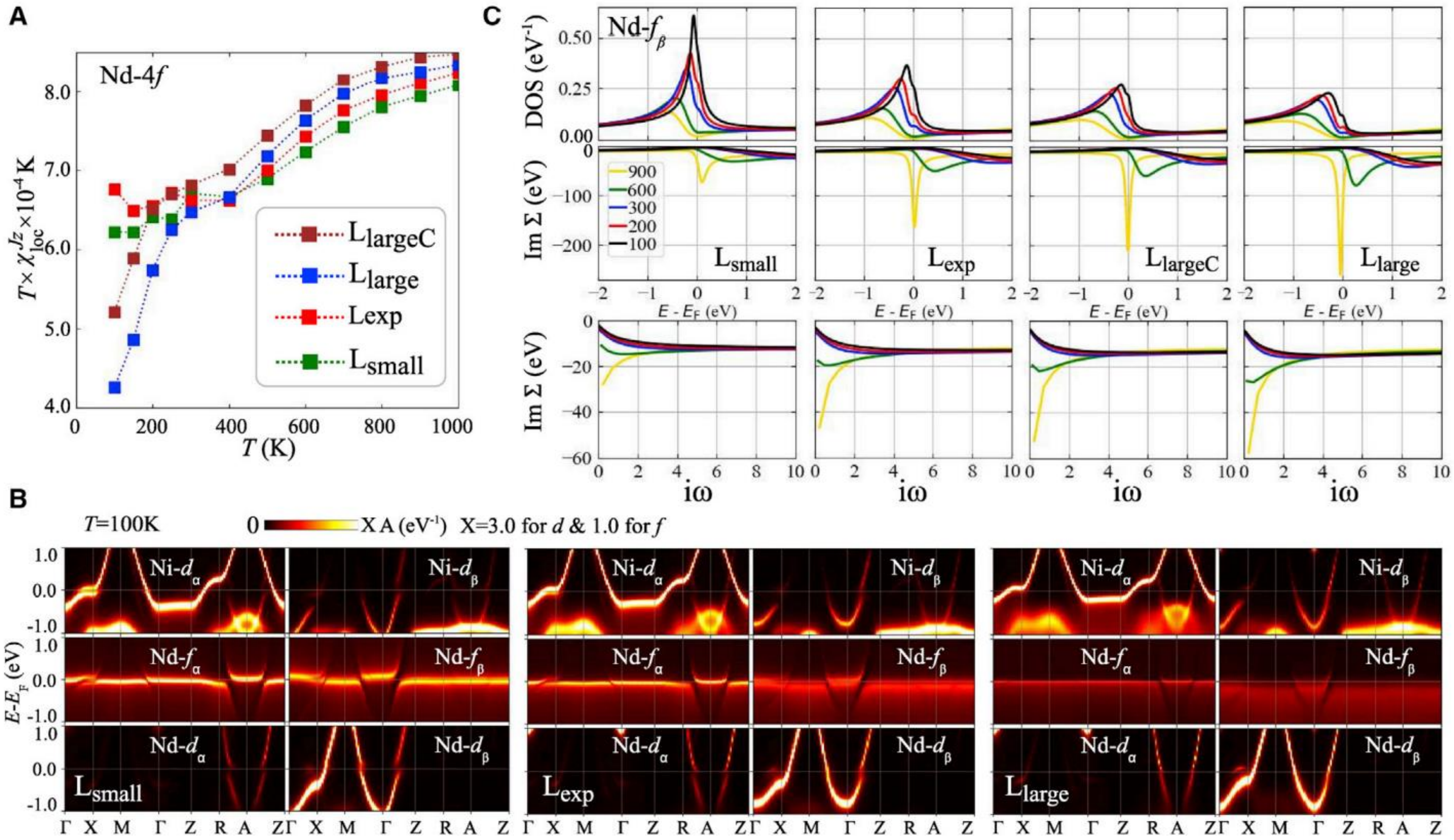
# $NdNiO_2$- Kondo effect under lattice modulation




- Kondo effect driven by the strong correlation of Nd-4*f* and Ni-3*d* electrons
- Kondo effect is easily destroyed by lattice modulation

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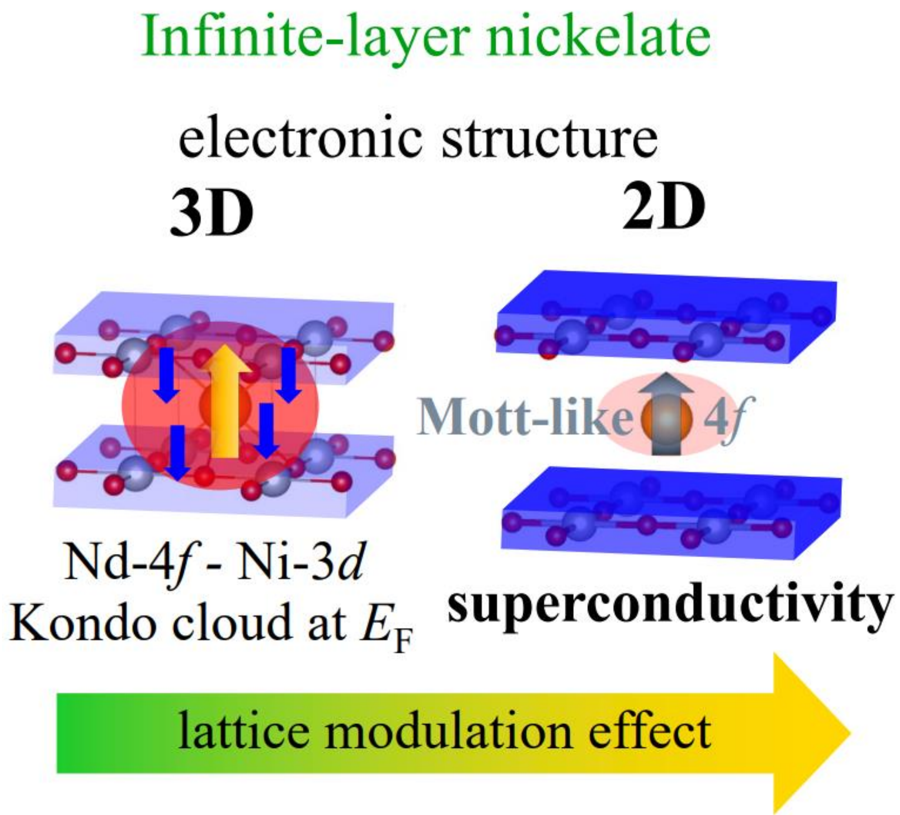
# $NdNiO_2$ - proposed impact of kondo cloud on superconductivity

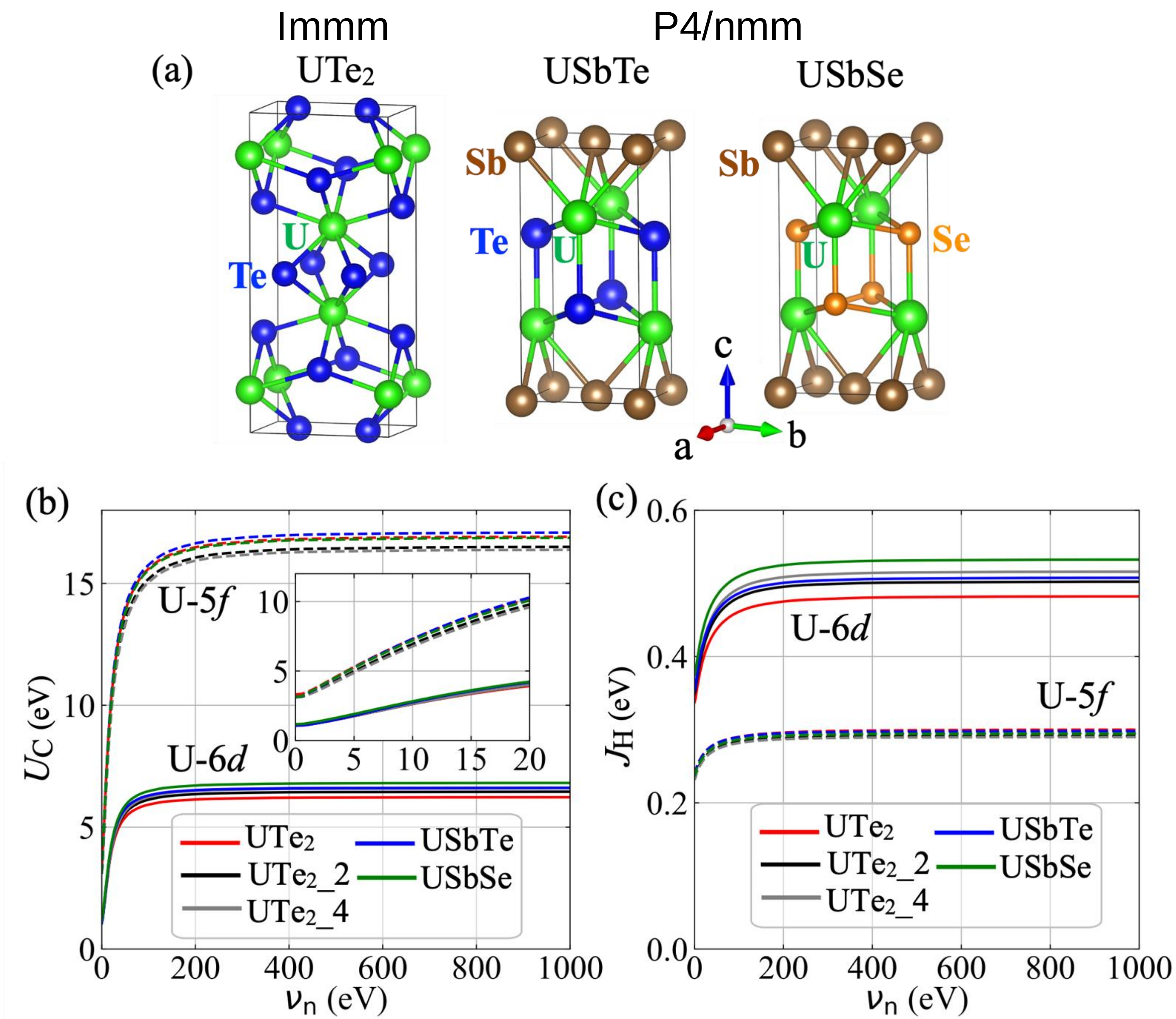



- 3D nature of Kondo effect driven by the strong correlation of Nd-4*f* and Ni-3*d* electrons
- Quasi-2D Fermi Surface is a strong restriction on the pairing symmetries
- 3D Kondo effect disappears with lattice modulation, which may affect superconducting instability.
- Calling for ARPES

[1] D. Li et al. Physical Review Letter 125, 027001 (2020) [2] Q. Li et al. Communications Materials 1, 16 (2020) [3] S. Zeng et al. Nature Communications 13, 743 (2022) [4] **B. Kang*,** H. Kim, Q. Zhu, C.H. Park Cell Reports Physical Science 4, 101325 (2023)

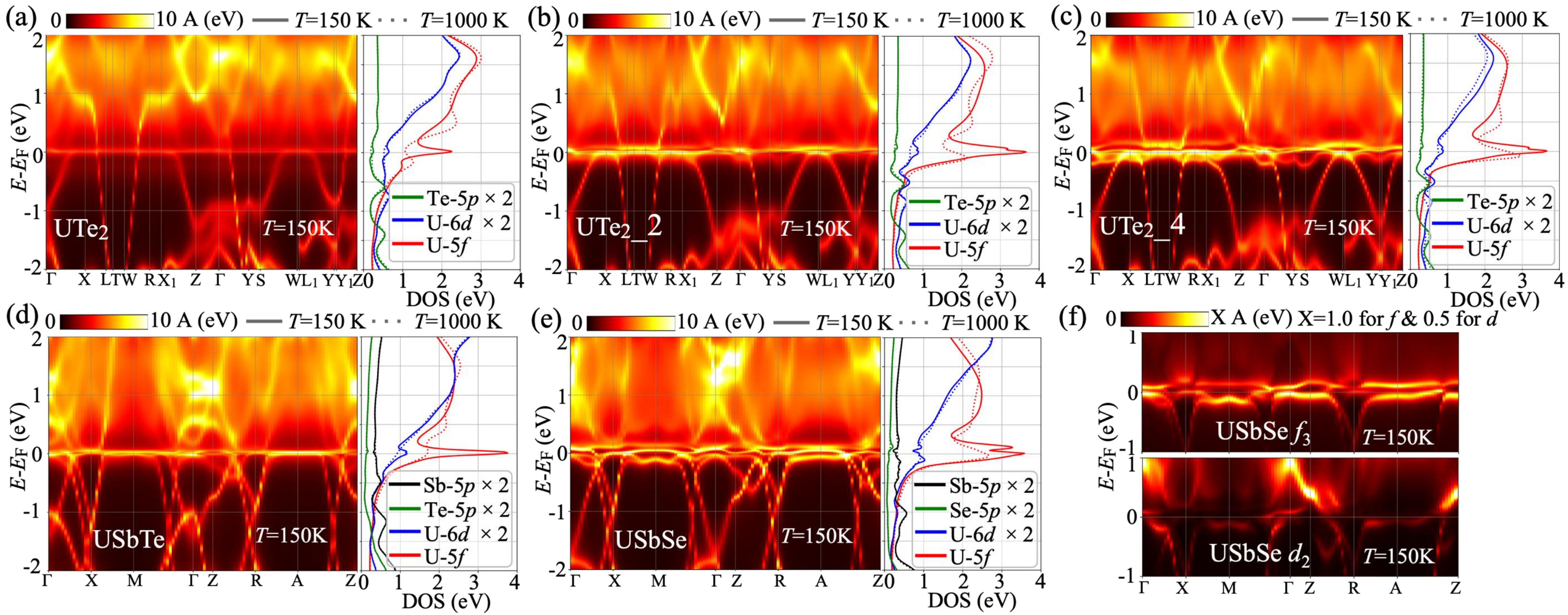
# Dual nature of magnetism – $UTe_2$, USbTe, USbSe

Immmm P4/nmm



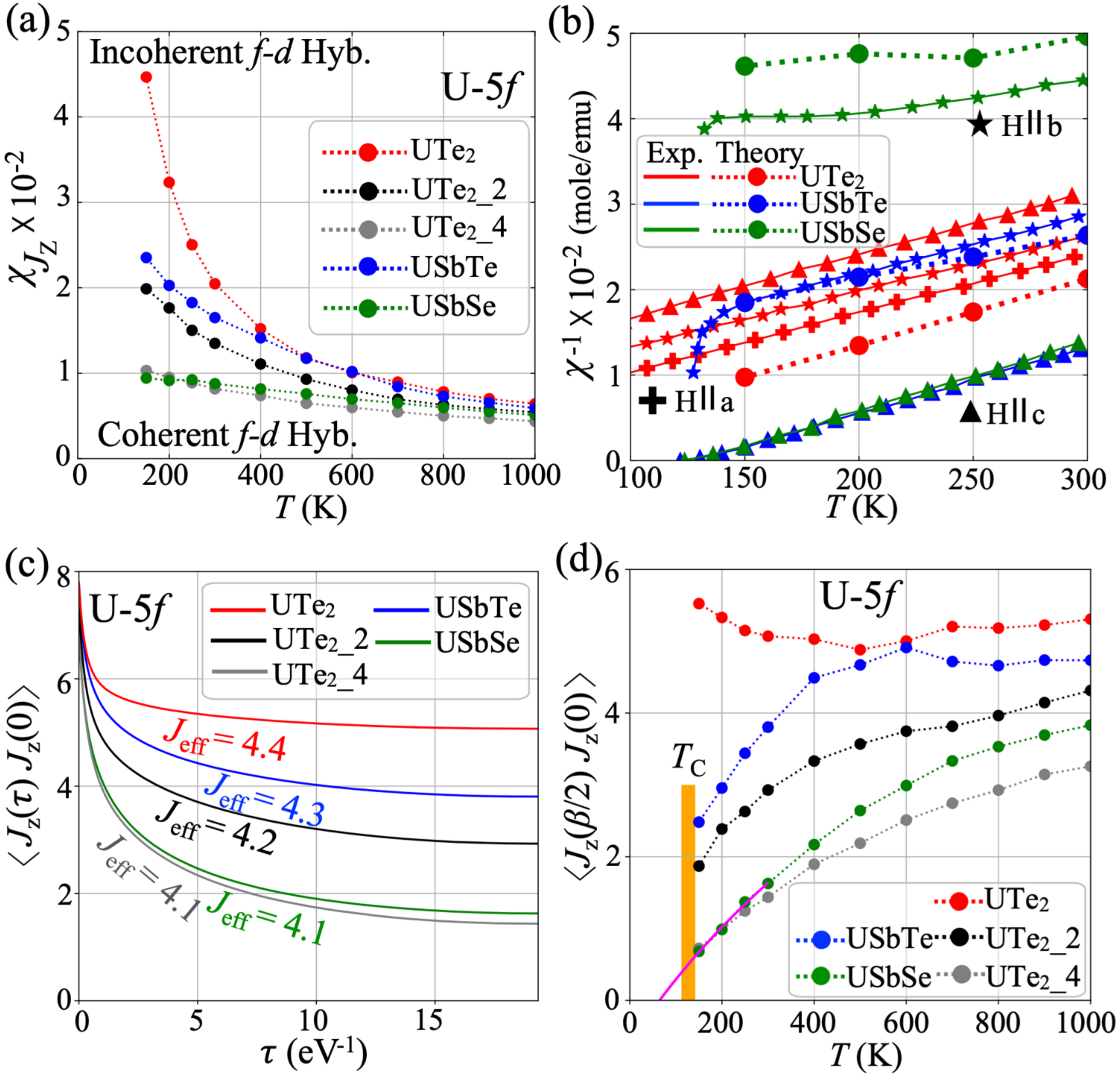

- The static $U_C$ of U-6d and U-5f orbitals were found to be comparable across the compounds studied

# Dual nature of magnetism – electronic structure




- Strong U-5f peak in the vicinity of the Fermi level at low temperature.
- Hybridization between U-5f and U-6d
- Flat f bands and kink-like band structure=> f-d Kondo effect.

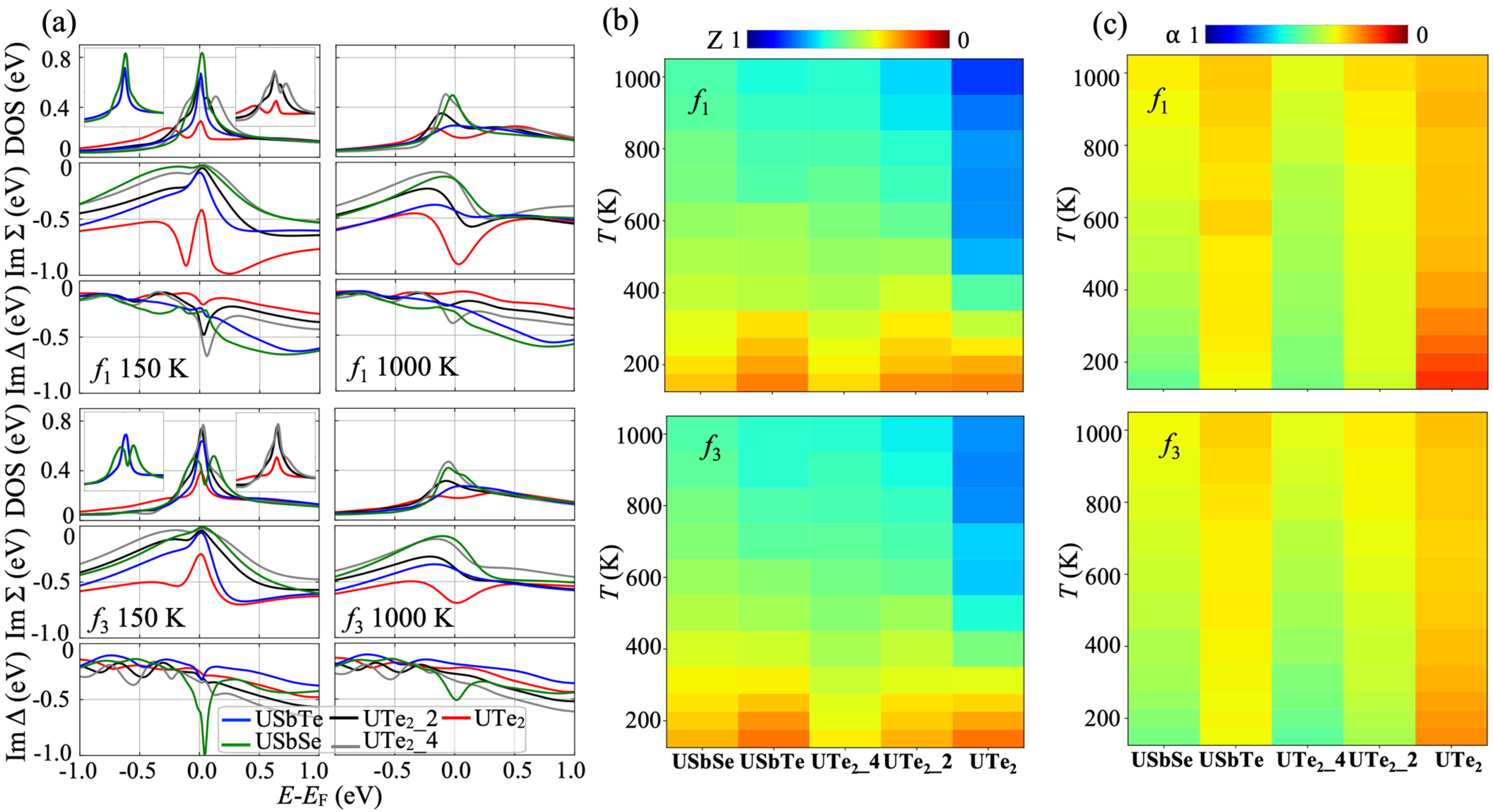
# Dual nature of magnetism – crossover incoherent to coherent




- Crossover from incoherent to coherent f-d Kondo hybridization
- Consistent with experiment
- Clear dual nature of Pauli-like susceptibility and local magnetic moments in USbSe

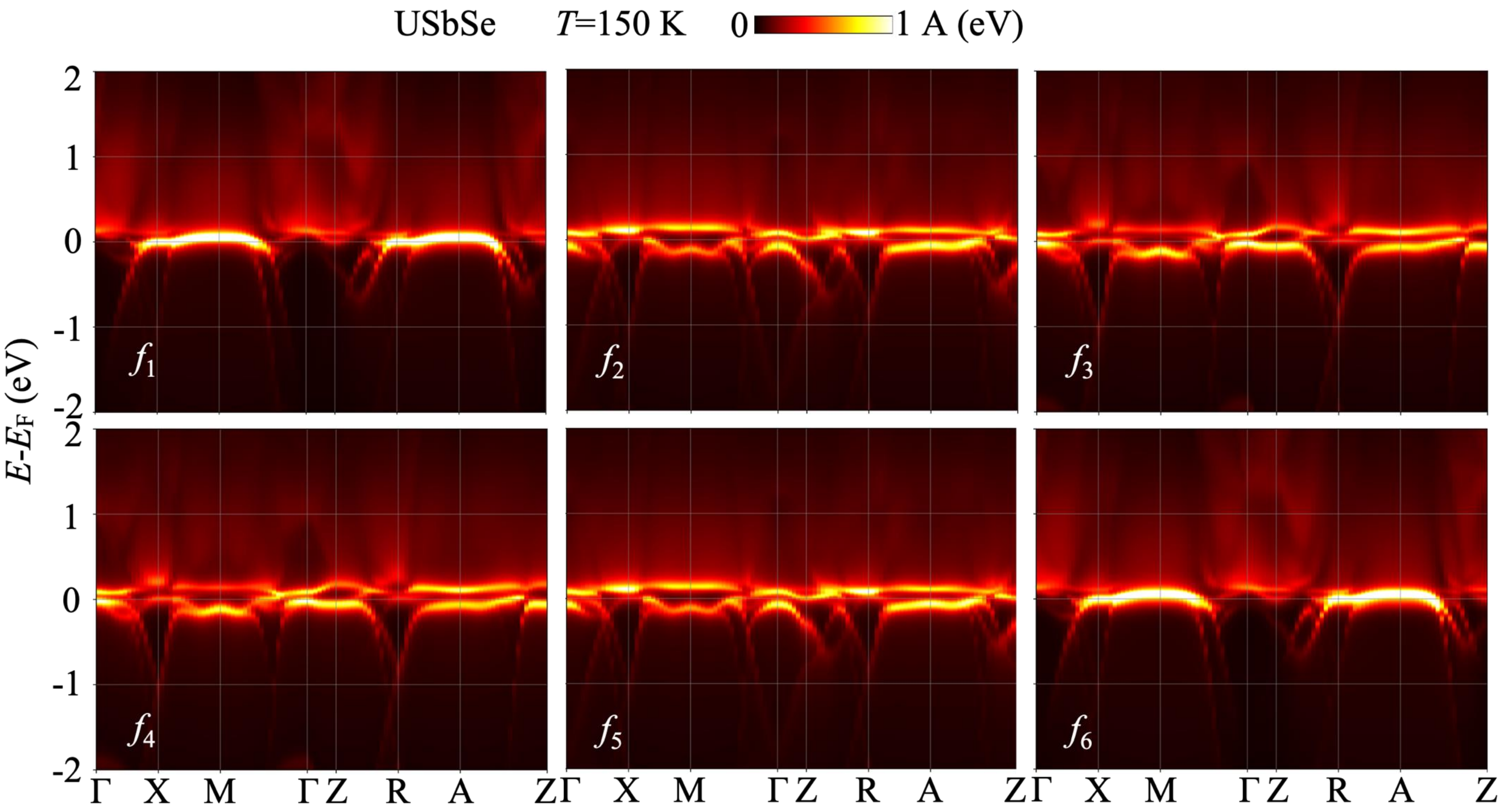
**B. Kang*,** Y. Lee, L. Ke, H. Kim, M-H. Kim arXiv preprint arXiv:2305.08003 (2023)

# Dual nature of magnetism – quasi-particle



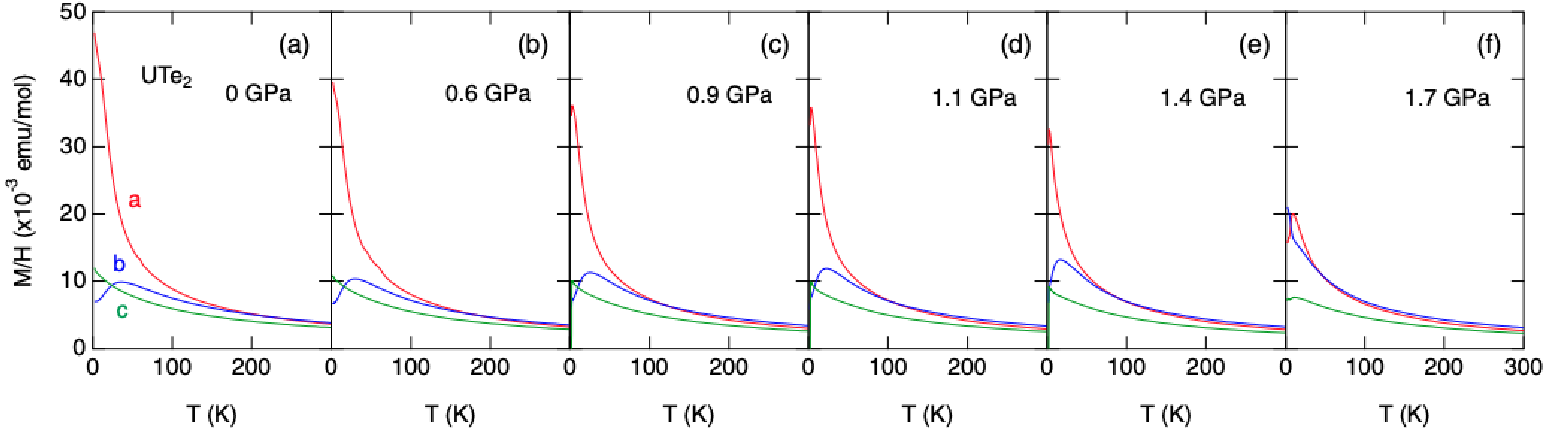

- No orbital selective Mott transition
- All U-f orbital contribute to local magnetic moments

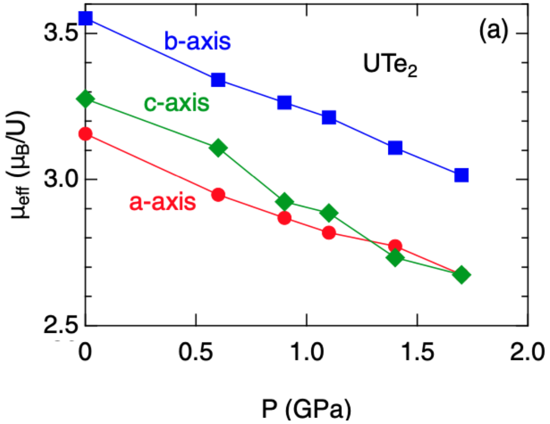
# Dual nature of magnetism – momentum dependent f-d Kondo hybridization

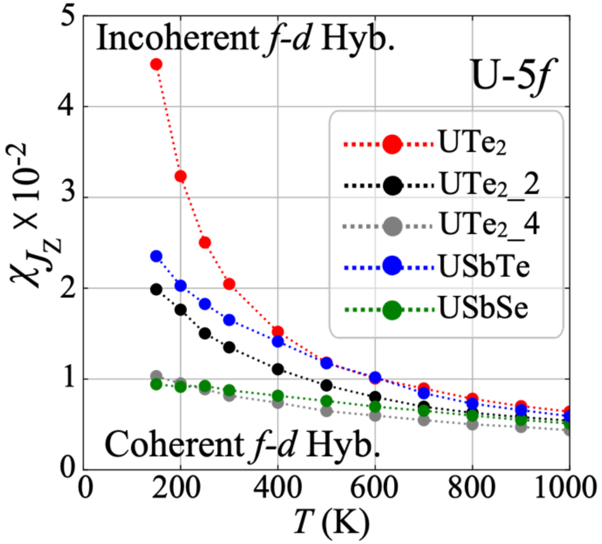


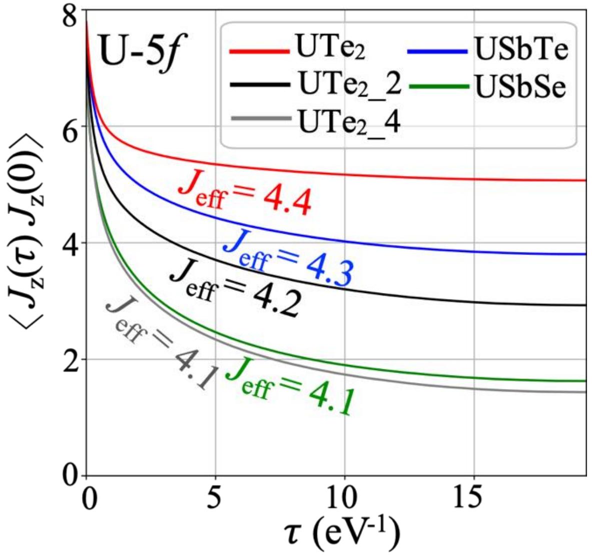

- The flat f bands are subject to coherent f-d hybridization
- That is momentum dependent and forms partially flat bands along X-M-Γ and R-A-Z symmetry lines, resulting in the emergence of van Hove singularity
- Pauli-like magnetic susceptibility and local magnetic moments of U-5f electrons in the coherent regime.

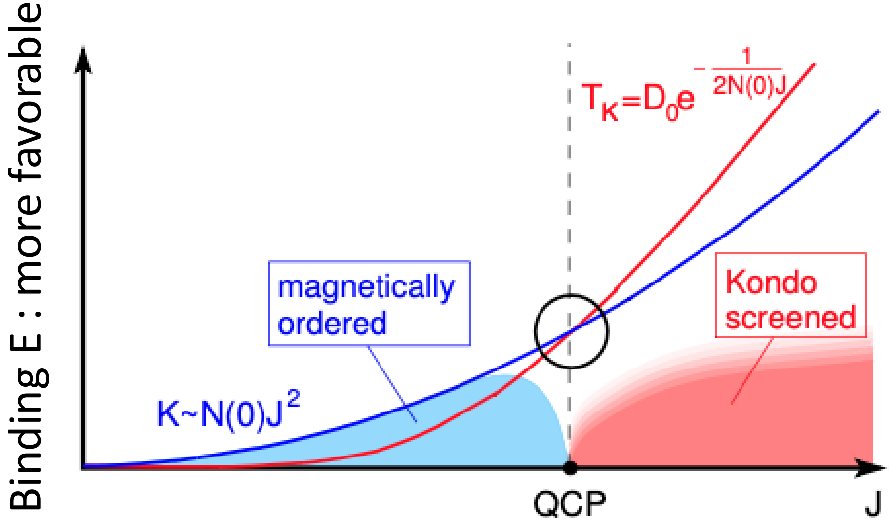
# Dual nature of magnetism – $UTe_2$ under pressure




- Pressure up => susceptibility and local magnetic moments down (both theory and experiment)
- Pressure up => J increase => Kondo effect more prominent than RKKY
- VHS in coherent UTe2 is responsible for the long-range magnetic ordering under pressure


[1] D. Li et al. J. Phys. Soc. Jpn 90, 073703 (2021)

Thank you