Ab initio Theory of Unconventional Superconductivity

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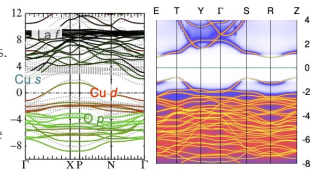
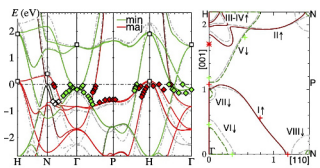
The Quasiparticle Self-Consistent *GW* Approximation (QSGW)

Green's-function methods supply *nonlocal, dynamical* self-energy *ab initio*

- **DFT**: insufficient fidelity for UCS, correlated systems. In La_2CuO_4, La(*f*) too high, Cu(*s*) too low, Cu(*d*) too wide, O(*p*) too high.
- **GW**: is simplest diagram.
- **Quasiparticle Self-consistency**: (QS*GW*) No reliance on Hartree Fock or DFT. Discrepancies with experiment become **uniform**, **systematic**, and traceable to specific diagrams
- **Systematically Improvable**: ladder diagrams greatly improve fidelity in both 1- and 2-particle properties
- G_0 **and** G: yields both optimal nonlocal noninteracting G_0 and interacting G
- **Response functions**: intrinsic to the theory, needed for UCS
- **Minimal Spin Fluctuations** QS*GW* handles most systems extremely well. Ladder diagrams needed to up short ranged part.
- **Fluctuating local moments** are missed by QS*GW*



La_2CuO_4 noninteracting G_0 and interacting G

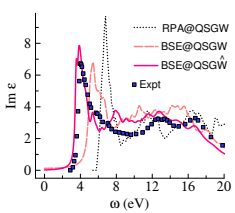


High fidelity QSGW description of Fe

Dynamical Mean Field Theory: for strong local correlations

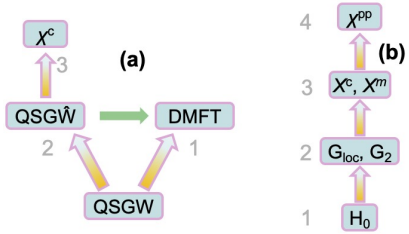
DMFT is needed because low-order QS*GW* has no diagram for spin fluctuations. Spin fluctuations responsible for incoherence, "bad metals."

- **Charge Fluctuations** long-ranged, high energy and very well described by QS$G\hat{W}$. Ladder diagrams patch up short ranged error in RPA very well.
- **Spin Fluctuations** usually short-ranged, low energy. No vertex like ladders in *GW*, and they are poorly described.



Dielectric function of NiO.
dotted black line: RPA
dashed orange line: BSE@QS$G\hat{W}$
solid red line: BSE@QS$G\tilde{W}$
dots: Experiment from Spicer

When the vertex in the spin channel is weak, we can use MBPT. Augment with DMFT when strong For superconductivity, need χ^m, χ^c. BSE gives Γ^{pp} and χ^{pp} needed for Eliashberg equation.



[1] M. Methfessel, M. van Schilfgaarde, and R. A. Casali. *A full-potential LMTO method based on smooth Hankel functions.* Lecture Notes in Physics, H. Dreysse. Springer-Verlag, Berlin, 2000.

[2] D. Pashov, S. Acharya, W. R. Lambrecht, J. Jackson, K. D. Belashchenko, A. Chantis, F. Jamet, and M. van Schilfgaarde. *Questaal: A package of electronic structure methods based on the LMTO technique.* Computer Physics Communications, 249:107065, 2020.

[3] Swagata Acharya et al., *Electronic Origin of Tc in Bulk and Monolayer FeSe* Symmetry 13, 169 (2021)

Origins of Superconductivity in FeSe

FeSe is a **Hund's metal** with d_{xz}, d_{yz}, 3_{xy} all present at E_F.

- Which orbitals drive superconductivity?
- How does nematicity (seen in ARPES, neutron measurements, etc) affect T_C?
- How applicable is BCS/BEC theory?
- Role d_{xy} hole pocket at Γ
- What causes T_c to increase five fold when intercalated with Li, Na, K, Cs, etc?
- When FeSe is deposited as a monolayer on $SrTiO_3$, why does T_c jump to ∼80K?
- Is superconductivity driven by instabilities at Fermi surface?

(See oral presentation for details)

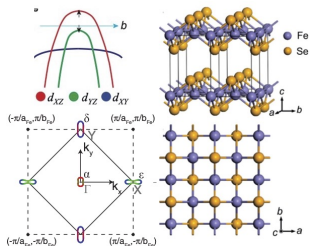


Figure 1. Top: d_{xz}, d_{yz}, 3_{xy} at Γ; Bottom: Fermi surface, showing pockets at Γ and M; Right: crystal structure.

Main findings for 1-particle properties:

- d_{xy} slightly above E_F, contrary to ARPES. True for both QSGW and QSGW+DMFT.
- QSGW+DMFT: d_{xy} becomes **very incoherent** — much more than d_{xz}, d_{yz}. Incoherence in d_{xy} is will be main driver for superconductivity.
- DFT *d* bandwidth much too wide

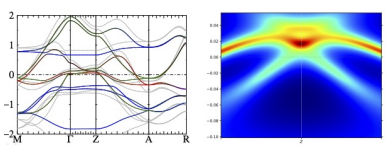


Figure 2. Left: QS*GW* band structure. Right QS*GW*+DMFT spectral function on R-Z-R line.

Crystal Structure and 2-particle properties:

- Position of d_{xy} relative to E_F very sensitive to Se height
- Incoherence in d_{xy} is very sensitive to proximity of d_{xy} to E_F.
- Therefore superconductivity is extremely sensitive to crystal structure.

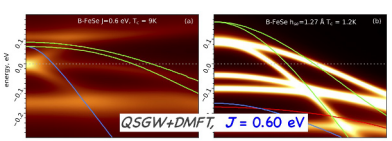


Figure 3. Left: QS*GW* band structure. Right QSGW+DMFT spectral function on R-Z-R line.

Role of Hunds J:

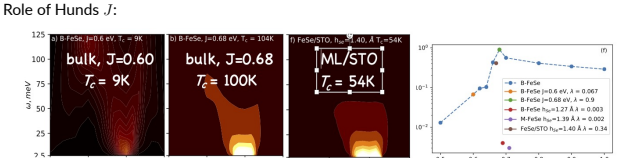


Figure 4. Left: Dynamical spin susceptibility χ^m for : (left) pristine FeSe @ cRPA J=0.60 eV (middle) pristine FeSe @J=0.68 eV (right) ML FeSe/$SrTiO_3$ @cRPA J=0.66 eV (far right) leading eigenvalue under different geometries as a function of J.

- Screening J critically affects incoherence. J=0.60→J=0.68 causes 10-fold increase in T_c!
- ML FeSe/$SrTiO_3$ has high T_c mostly because reduced screening increases J
- Intercalated FeSe has high T_c because (1) increase in Se height (changes position of d_{xy}, and also (2) expanded lattice parameter reduces screening of J.
- Nematicity changes *shape* of e.g. χ^m, but only slightly affects T_c
- **Conclusions**: (a) Two key factors are (1) proximity of d_{xy} to E_F and (2) value of J.
  (b) BCS theory does not apply — property of **vertex**, not density-of-states at F_F
  (c) d_{xy} need not be at E_F, because of ω-dependence of χ^m.

Properties of YFe_2Ge_2 (collaboration with Dessau group CU Boulder)

Overarching quemastion: is it a spin triplet superconductor?

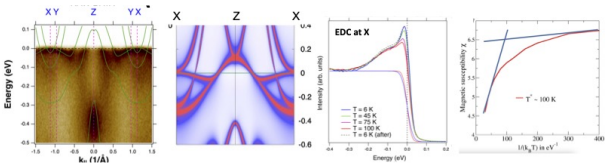


Figure 5. Left: QS*GW* band structure. Right QS*GW*+DMFT spectral function on R-Z-R line.

- ARPES shows very flat band at X ... but QS*GW* already provides a good description of it
- Slight band renormalizations needed (supplied by DMFT)
- Flat band not a property of Hundness, but originates from cancellation in hopping matrix elements, much like twisted graphene. Not strongly correlated.
- Flat band still gives rise to Kondo physics
- One-particle theory shows strong tendency to FM, which would yield spin triplet superconductivity. DFT predicts YFe_2Ge_2 to be FM!
- But, Kondo physics intervenes below Kondo temperature, prevents FM (In accord with experiment).
- Preliminary calculations show spin triplet does not survive because of Kondo physics.

Is $TiSe_2$ an excitonic insulator?

Overarching quemastion: Why is $TiSe_2$ an insulator with a gap of 0.1 eV? $TiSe_2$ can be be driving superconducting with pressure or Cu doping. But one-particle properties still not understood.

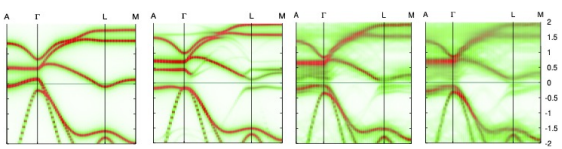


Figure 6. QS*GW* spectral function of $TiSe_2$ in a supercell folded to the Brillouin zone of the high-symmetry $P\bar{3}m1$ structure for (a) the ideal $P\bar{3}m1$ structure, (b) the $P\bar{3}c1$ charge-density wave, (c) statistically averaged snapshots from the 120 K *ab initio* molecular dynamics simulations of 96-atom cells and (d) MD simulations at 300 K.

- Long-standing conjectdure that $TiSe_2$ is an **excitonic insulator**. (They are **rare**.)
- Important, if true, because implies storng electron-hole coupling ⇒ strong many-body effects
- Evidence comes from presence of band at L (seen in ARPES), missing from band theory.
- $TiSe_2$ undergoes transition from high-symmetry $P\bar{3}m1$ structure to CDW $P\bar{3}c1$ around 200 K
- QS*GW* theory says: *metal* in high-symmetry $P\bar{3}m1$ (Fig. 6a) but becomes narrow gap insulator as CDW (Fig 6b). In unfolded Brillouin Zone of $P\bar{3}m1$, Umklapp processes make it appear as though a band appears beloe E_F
- We do AIMD simulations at finite temperature, and QS*GW* calculations at snapshots. A "memory" of the CDW remains, at 120 K and also at 300 K.
- Conclusion: $TiSe_2$ is a **band insulator**, because of (dynamic) symmetry breaking. It is the only instance to our knowledge where a metal-insulator transition forms from dynamical nuclear fluctuations.

[4] S. Acharya, D. Pashov, m van Schilfgaarde, *Role of nematicity in controlling spin fluctuations and superconducting T_c in bulk FeSe*, Phys. Rev. B 105, 144507 (2022).

[5] D. Pashov, R.E. Larsen, S. Acharya, M van Schilfgaarde, *Lattice fluctuations, not excitonic correlations, mediated electronic localization in $TiSe_2$* Preprint https://arxiv.org/abs/2311.08015

[6] S. Acharya, D. Pashov, E. Chachkarova, M van Schilfgaarde, and C. Weber, *Electronic structure correspondence of singlet-triplet scale separation in strained Sr_2RuO4_4*, Appl. Sci. 11, 508 (2021).

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