Effective electronic model of a quasi-one dimensional compound: $Li_{0.9}Mo_6O_{17}$

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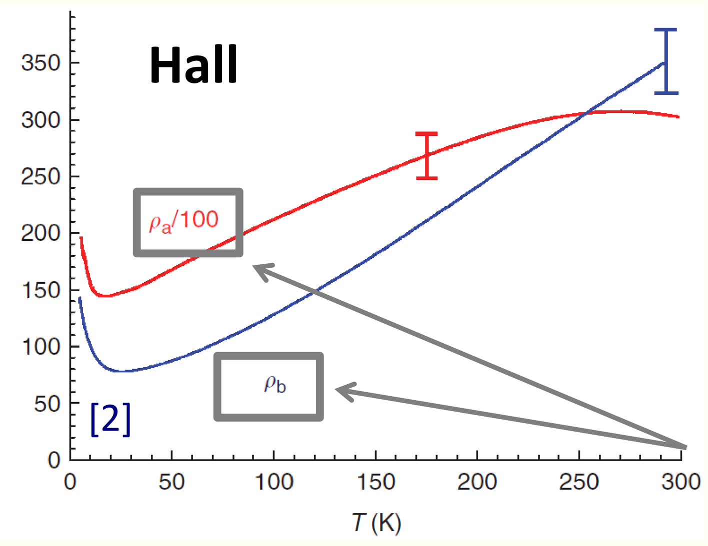
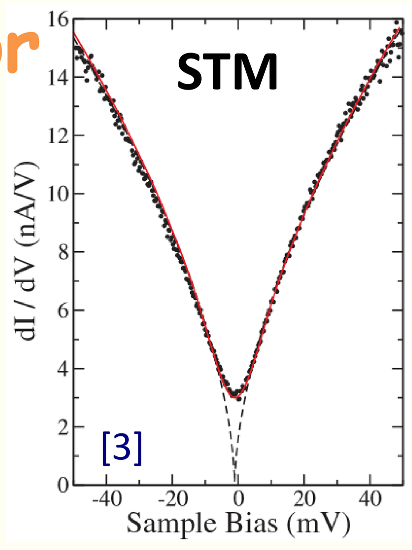
MOTIVATION



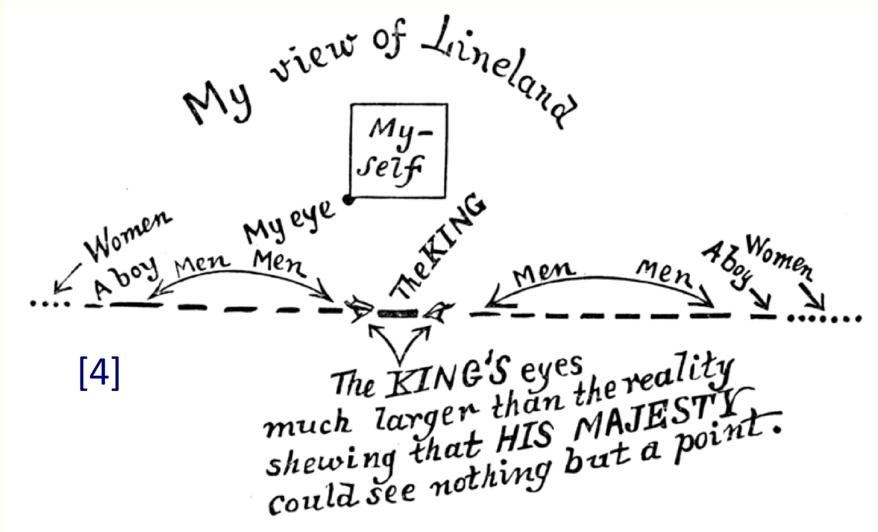
complex el. behavior

- metal-insulator-trans. @ 24 K
- superconductivity @ < 2 K
- possibly CDW
- highly anisotropic



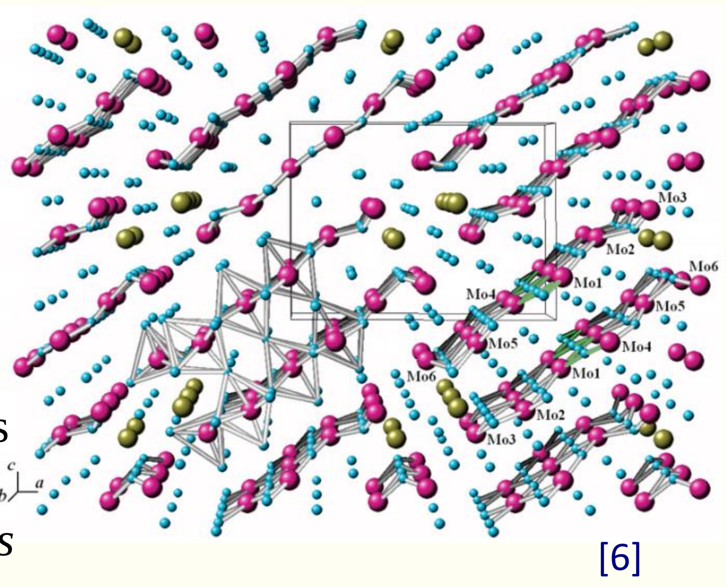
quasi - 1D

- real life realization of a Tomonaga-Luttinger-liquid?
- DOS(ω =0) ~ T^α, ($\alpha \approx 0.6$, STM [3]) ($\alpha \approx 0.9$, ARPES [7,8])
- 1D robust @ T = 300 K



1) Crystal structure [6,7]

- 48-atom unit cell
- monoclinic $P2_1/m$
  - $a \approx 12.8$ Å
  - $b \approx 5.5$Å
  - $c \approx 9.5$Å
  - β = 90.6°, Z = 2
- conduction e^- on 4 octahedral *Mo* sites
- double zig-zag chains along *b axis*



Literature

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5) Transport

$$\mathcal{A}^n_{\nu\mu} = N_{\text{spin}} \pi\hbar \int_{-\infty}^{\infty} d\omega\, (\beta\omega)^n\, p_{\text{FD}}(\omega,\mu,\beta) p_{\text{FD}}(-\omega,-\mu,\beta) \Gamma_{\nu\mu}(\omega,\omega)$$

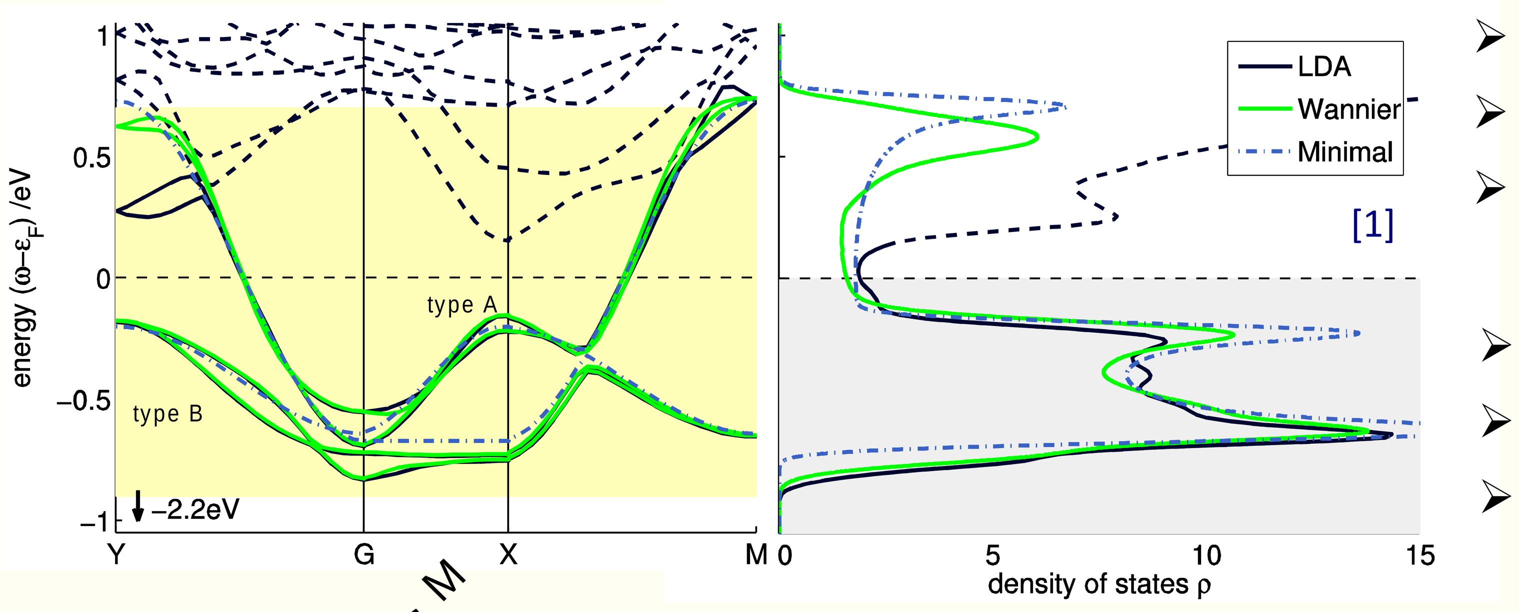
$$\Gamma_{\nu\mu}(\omega_1,\omega_2) = \frac{1}{V}\frac{1}{N_{1.\text{BZ}}} \sum_{\mathbf{k}\in 1.\text{BZ}} \text{Tr}\left[v_\nu(\mathbf{k}) A(\omega_1,\mathbf{k}) v_\mu(\mathbf{k}) A(\omega_2,\mathbf{k})\right]$$

p_{FD}: Fermi-Dirac; v_v velocity dir. v; A(ω,k): spectral function

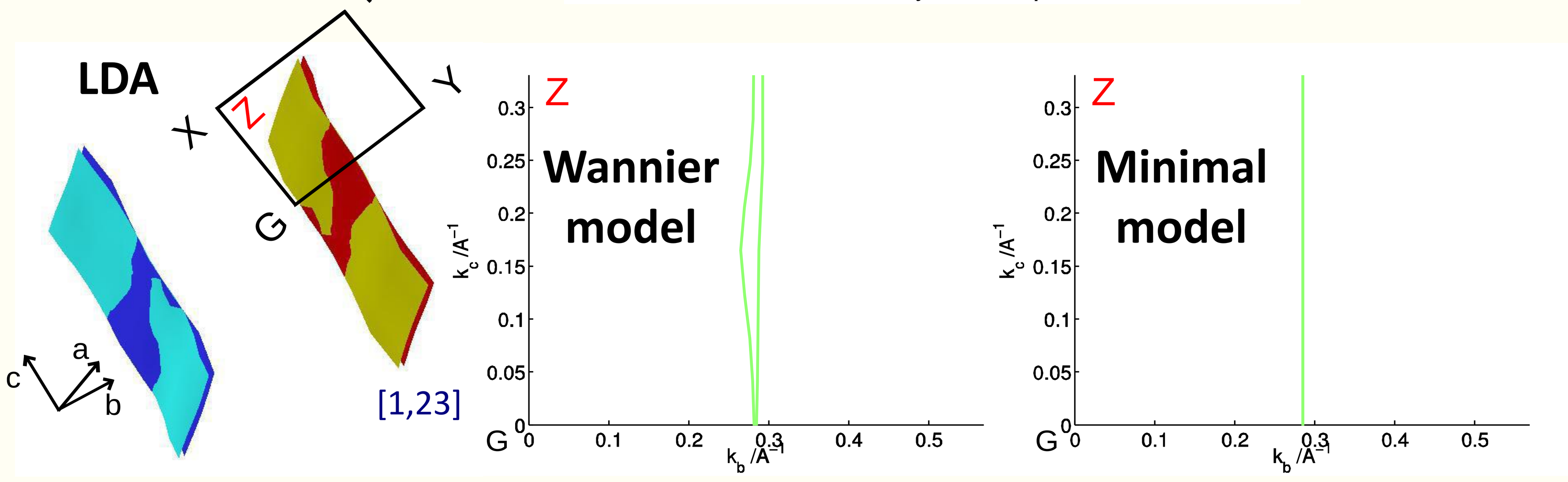
- low temperature, small scattering $\gamma \approx 0.05$eV
- minimal model $\sigma_{bb} = \frac{4e^2}{h\gamma}\frac{b}{ac}\sqrt{(2t_{AA})^2-\mu^2} \overset{\mu\approx 0}{\approx} N_{\text{spin}} N_{\text{band}} \frac{D}{R_K\gamma}\frac{b}{ac}$
- anisotropy via numeric solution of full Wannier model

Ref.	ρ_a	ρ_b	ρ_c	ratio
	mΩcm	mΩcm	mΩcm	
Greenblatt etal. SSC 51, 681 (1984)	2470	9.5	-	260:1:-
Chen etal. EPL 89, 67010 (2010)	64.5	16	854	4.5:1:50
Choi etal. PRB 69, 085120 (2004)	-	1.7	-	-:1:-
Luz etal. PRB 76, 233105 (2007)	110(40)	19(1)	47(5)	6(2):1:2.5(4)
Mercure etal. PRL 108, 187003 (2007)	30	0.4	600	80:1:1600
Xu etal. PRL 102, 206602 (2009)	-	0.4	-	100:1:>100
Wakeham etal. NatComm. 1, 2 (2011)	-	-	-	100:1:-
Wannier model	$\approx 430\gamma$	$\approx 1.8\gamma$	$\approx 600\gamma$	240:1:330
minimal model	- [1]	$\approx 2\gamma$	-	-:1:-

2) *Ab-initio* electronic structure

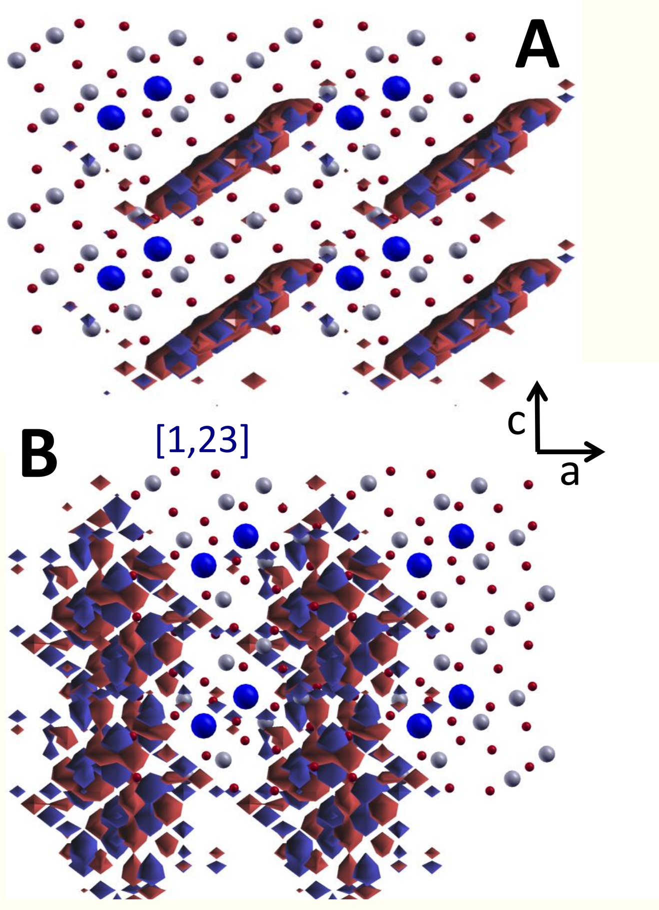


- Density Functional Theory (DFT)
- Local Density Approximation (LDA)
- all electron, Wien2k, FP-LAPW
- 4 bands close to Fermi energy
- *Mo 4d* character
- originate from 4 *Mo* atoms in unit cell



- 1D Fermi surface
- $v_F \approx 1.10^5$ m/s = bad metal
- W ≈ 1.5 eV

3) Effective Wannier model

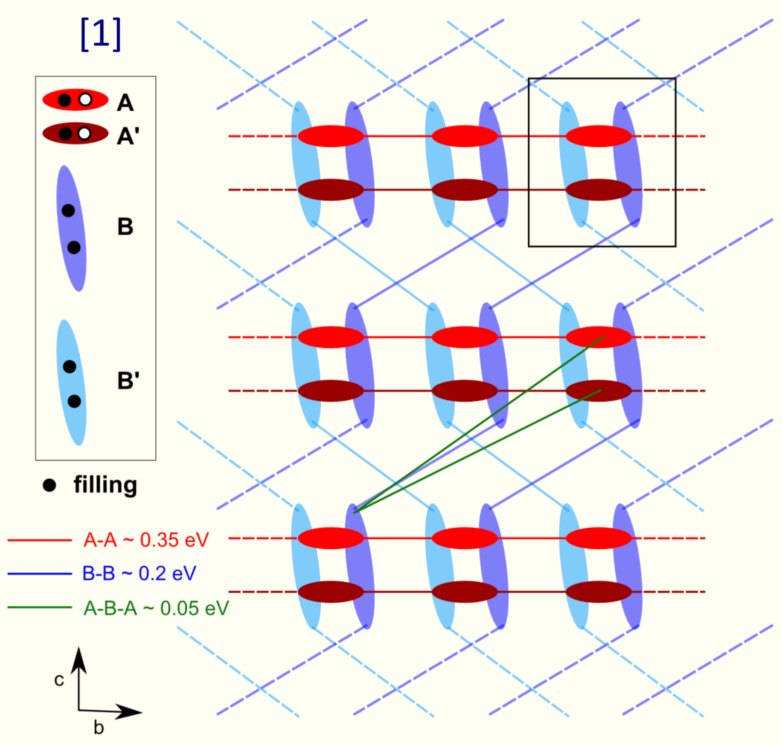


- maximally localized Wannier functions
- Wien2Wannier + Wannier90

$$H_{\text{Wannier},\alpha\beta}(\mathbf{k}) = \langle\omega_\alpha|\hat{\mathcal{H}}_{\text{Wannier}}(\mathbf{k})|\omega_\beta\rangle = \sum_{\delta\mathbf{R}} e^{-i\mathbf{k}\cdot\delta\mathbf{R}} M_{\mathbf{R}\alpha\mathbf{R}'\beta}$$

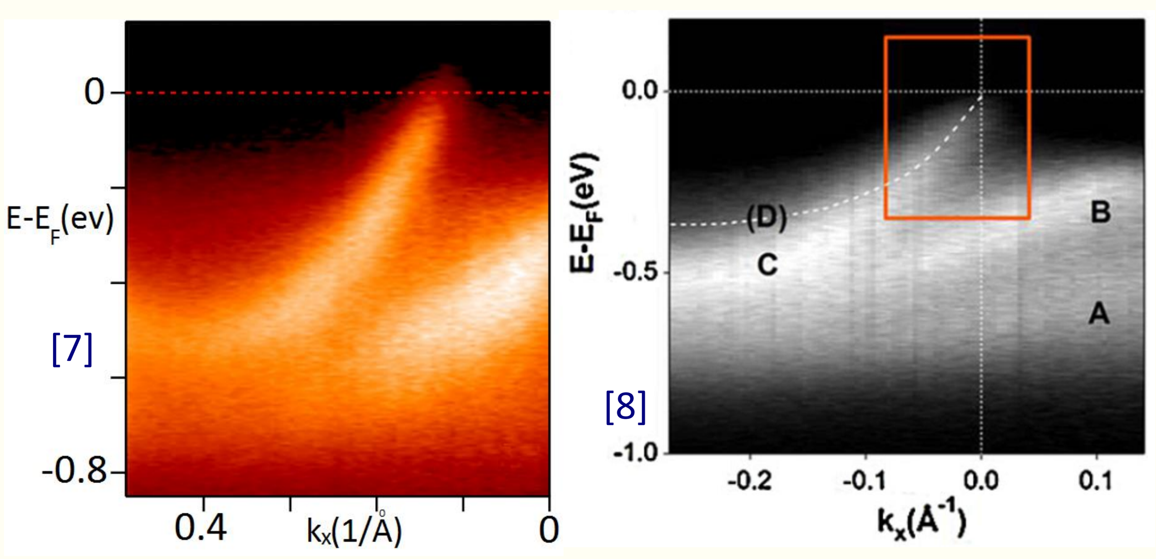
2 *half-filled* orbitals A, A‘: chains in *b*
2 *filled* orbitals B, B‘: „perpendicular"
2160 matrix elements

- $|t_{max}|$=0.35eV: A-A hopping in *b* dir.
- long range perpendicular hopping
- direct A-A‘ processes negligible
- A-B-A type processes important but suppressed due to filling of B
- robust 1D physics



6) El. dynamics & correlations

- ARPES



- Theory – adding interactions to Wannier Hamiltonian

ad 2) DFT part: Solving Kohn-Sham equations

$$\left[-\frac{\hbar^2}{2m_0}\nabla^2 + V_{KS}(\mathbf{r})\right]\psi_{\mathbf{k}}(\mathbf{r}) = \varepsilon_{n\mathbf{k}}\psi_{\mathbf{k}}(\mathbf{r})$$

Calculation performed within the Local Density Approximation (LDA), using Wien2k

ad 3) Interfacing: Constructing projection operators on the correlated states

$$\hat{P}^\alpha(\mathbf{k}) = \sum_{m\in\mathcal{C}} |w^\alpha_{\mathbf{k}m}\rangle\langle w^\alpha_{\mathbf{k}m}|.$$

Wannier-like orbitals obtained with a projection scheme

ad 6) OR

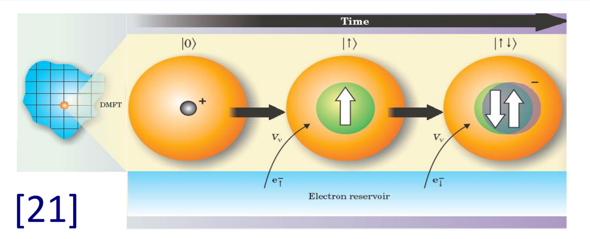
$\Sigma = \Sigma'(\omega, k)$

[18, 19] (e)VCA part: (extended) Variational Cluster Approach

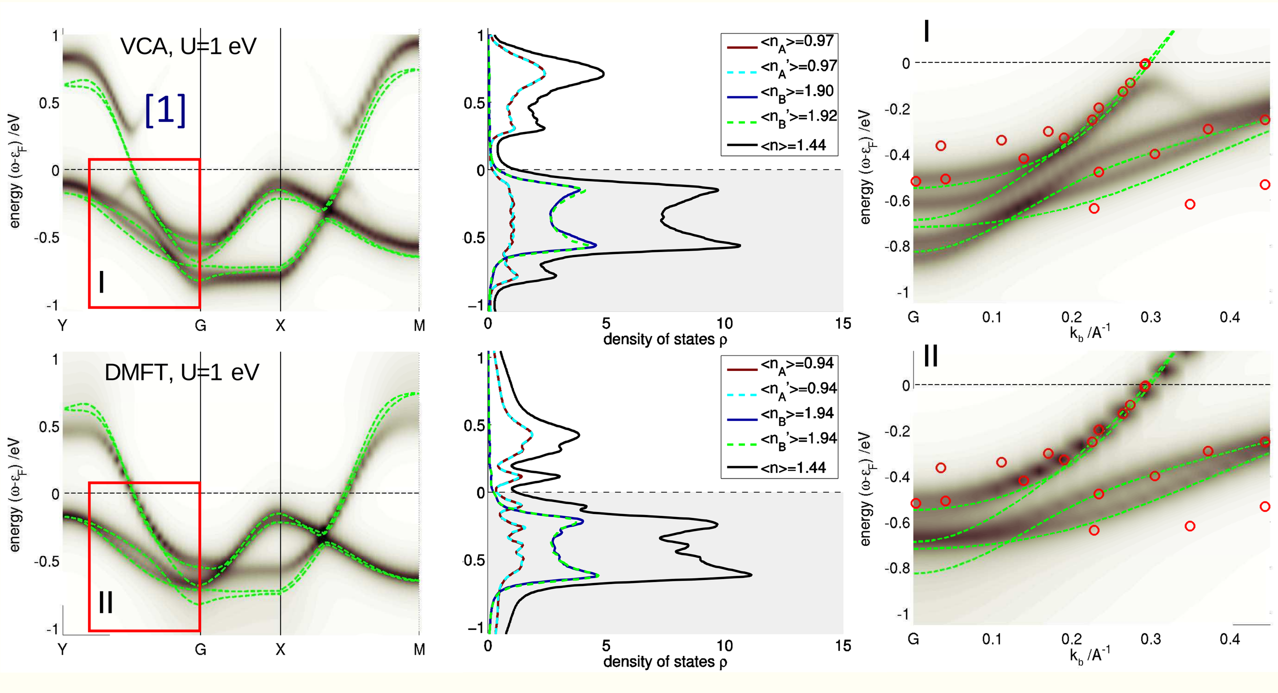
- Cluster Perturbation Theory [15,16]
- ED solver = real freq. correlators
- 8 orbitals – 2 unit cells in *b* dir.
- + variational principle: [17] Self Energy Functional Appraoch
- short range correlations = a non-local self-energy ∑

$\Sigma = \Sigma(\omega)$

[20] DMFT part: Performing the DMFT loop



[21]
DMFT calculation performed with an impurity solver (CTQMC algorithm)
[22]



- moderate on-site interaction U ~ 1 eV appropriate
- off-diagonal Coulomb terms less important
- hybridization mechanism A-B bands

4) Minimal model

- neglect hopping < 0.1$|t_{max}|$
- + restrict to nearest neighbor in **b-c plane**

$$H_{\text{minimal}} = \begin{pmatrix} t_{AA} 2\cos(k_b b) & 0 \\ 0 & \epsilon_B + t_{BB} 2\cos(k_b b \pm k_c c) \end{pmatrix}$$

two degenerate chains at Fermi energy, t_{AA}=-0.35 eV
+ two degenerate bands below, t_{BB}=-0.11 eV, e_B=-0.45 eV

accurate band structure and DOS, but fully separates

- estimation of perpendicular processes:

$$\tilde{\hat{\mathcal{H}}}_{AA} = \hat{\mathcal{H}}_{AA} + \hat{\mathcal{H}}_{AB}\left(\omega - \hat{\mathcal{H}}_{BB}\right)^{-1}\hat{\mathcal{H}}_{BA} \Longrightarrow \begin{matrix} t_{AA} \approx -0.02 \text{ eV} \\ t_{AA'} \approx -0.005 \text{ eV}\end{matrix}$$

7) Conclusions

- obtained *ab-initio*, general purpose, unbiased **model** for electronic properties of $Li_{0.9}Mo_6O_{17}$
- 2 large **half-filled** orbitals (A) = **chains** in b dir.
- 2 large **filled** orbitals (B) perpendicular
- **1D** physics stable: indirect hopping A-A' via filled B
- highly anisotropic **transport** char. reprod. exp. trend
- moderate on-site **U** appropriate

We acknowledge support of the Austrian Science Fund (FWF) P24081 and SFB ViCoM projects F04103 and F04104. For a parts of our calculations we used the Vienna Scientific Cluster VSC-II.