

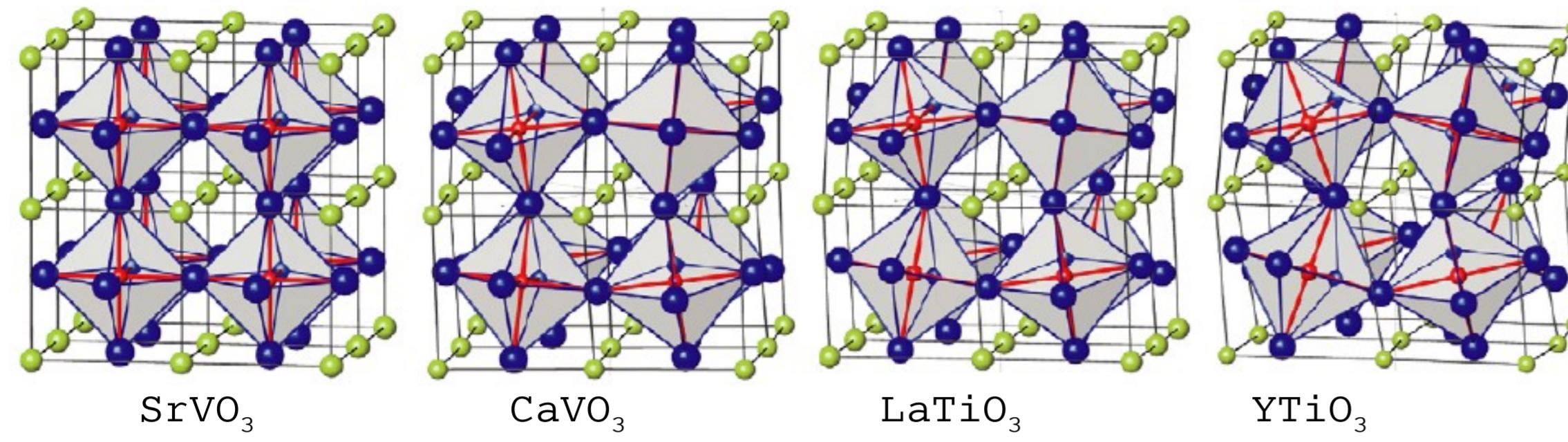
Correlated Band-Structure trend in 3d¹ perovskite series

D.Karmakar¹, E. Pavarini², A. Yamasaki¹, O. Jepsen¹, O. K. Andersen¹

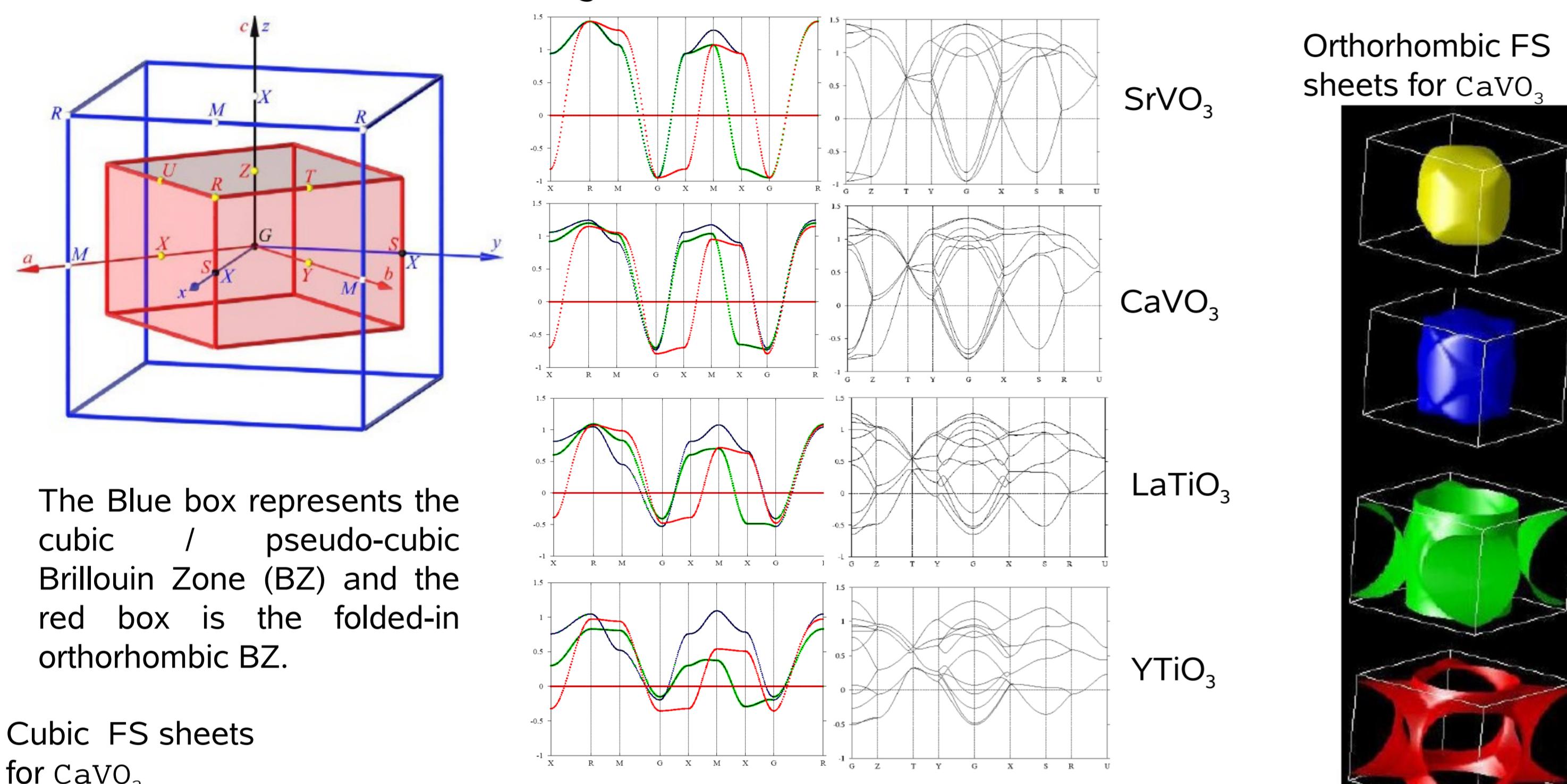
¹ Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

² Forschungszentrum Jülich GmbH, Institut für Festkörperforschung, D-52425 Jülich

Introduction

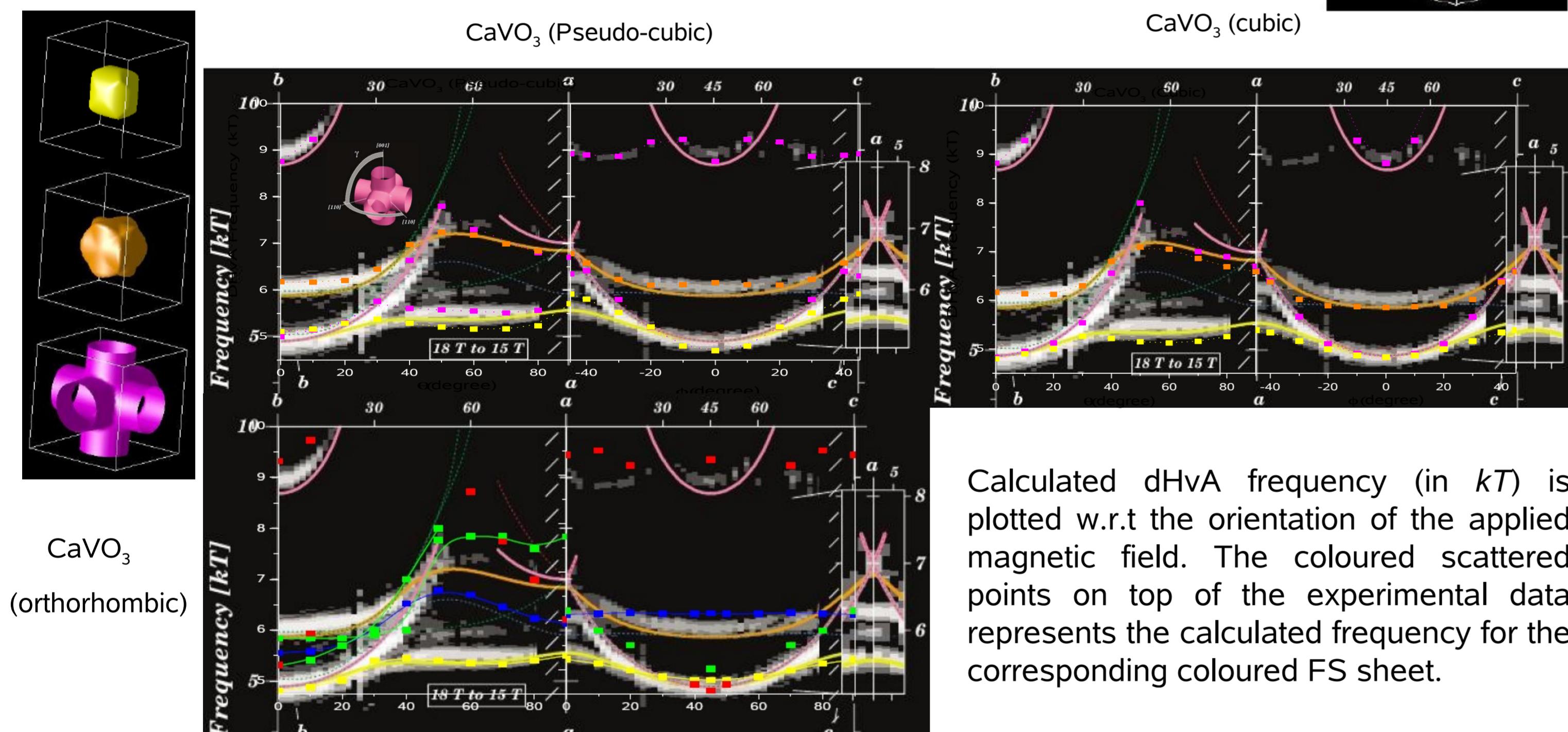


In the transition metal perovskite t_{2g}¹series, SrVO₃-CaVO₃-LaTiO₃-YTiO₃, the interplay among strong coulomb correlations, narrow 3d t_{2g} bands and GdFeO₃-type distortions lead to unusual electronic and magnetic properties [1]. Materials with similar electronic configuration exhibit different behaviour: SrVO₃ and CaVO₃ are strongly correlated metals with mass enhancements about 2, whereas LaTiO₃ and YTiO₃ are Mott insulators with gaps respectively of 0.2 and 1 eV. The Mott transition occurs between CaVO₃ and LaTiO₃. The GdFeO₃ distortion increases along the series and through cation-oxygen and cation-transition metal covalency, it lifts the t_{2g} degeneracy. In LaTiQ and YTiO₃, the crystal field splitting is about 200-300 meV and thus these Mott insulators are orbitally ordered. We use a combination of LDA and single-site dynamical mean-field (DMFT) approximation to investigate the evolution of the strongly correlated electronic structure along the series.



The Blue box represents the cubic / pseudo-cubic Brillouin Zone (BZ) and the red box is the folded-in orthorhombic BZ.

Cubic FS sheets for CaVO₃



Calculated dHvA frequency (in kT) is plotted w.r.t the orientation of the applied magnetic field. The coloured scattered points on top of the experimental data represents the calculated frequency for the corresponding coloured FS sheet.

$$\text{The LDA Bloch-transformed Hamiltonian in terms of } t_{2g} \text{ Wannier orbitals: } H_{R'm', Rm}^{LDA}(k) = \sum_T t_{m', m}^{R', R+T} e^{ik \cdot (T+R-R')}$$

$$\text{can be transformed to } k\text{-}Q \text{ representation: } \langle m', k-Q' | H | m, k-Q \rangle = \sum_R t^{(Q-Q'), R} e^{i(k-Q), R}$$

$$\text{with } t_{m', m}^{Q, R} = \frac{1}{4} \sum_R t^{R', R+R} e^{iQ, R'} \text{ and } Q = 000, \pi\pi0, 00\pi, \pi\pi\pi \text{ to obtain a single analytical band-structure}$$

model for all the four systems. The pseudo-cubic bandstructure in the folded-out blue Brillouin Zone(BZ) (shown in fig.) can be utilized to understand the evolution of three cubic, non-interacting, nearly two-dimensional xy, yz, zx bands for SrVO₃ into three distorted, monoclinic bands with the yz and xz level splitted by : $\pm[t_{yz,xz}^{000} + 2t_{yz,xz}^{001} \cos k_z + 2\langle t_{||z,\perp z} \rangle (\cos k_x + \cos k_y)]$

for the orthorhombic systems. These bands, when folded into the orthorhombic red BZ (in the fig.) results in 12 orthorhombic LDA bands. This pseudo-cubic band-structure is very useful for understanding of fermi-surface behaviour of metals. CaVO₃, although experimentally observed to be a strongly correlated metal of orthorhombic structure with V-O-V bond angle of $\sim 160^\circ$, the dHvA frequency measurement on the same system [2] has revealed that the LDA Fermi-surface (FS) is closer to the cubic structure than the orthorhombic ones. Our calculation of extremal areas of cubic and pseudo-cubic FS sheets shows that the non-cubicity introduced in the pseudo-cubic case in terms of non-equality of the onsite hoppings and optimized hybridization parameters are more effective for a better fitting with the experimental data.

Effect of strong correlations on LDA band-structure: LDA +DMFT results

We use a combination of LDA and the single-site dynamical mean-field (DMFT) approximation to investigate the evolution of chemistry and strongly correlated behaviour along the series. By means of the N-th order Muffin-Tin Orbitals (NMTO) [3] method, we construct a set of symmetrically orthonormalized t_{2g} Wannier Orbitals and the corresponding material specific 3-band Hubbard model. We solve this model with DMFT and obtain the self-energy matrix. Next this self-energy matrix (transformed in the global cubic basis) is introduced in the $Q = 0$ and Q -coupled LDA hamiltonian to obtain the complete model-Hamiltonian for all four systems:

$$H_{LDA} + \Sigma(\omega, k)$$

Constructing the on-site Green's function

$$G_{Rm, R'm'}(\omega) = \sum_k [(w + \mu) - \Sigma(\omega) - H_{LDA}(k)]_{Rm, R'm'}^{-1}$$

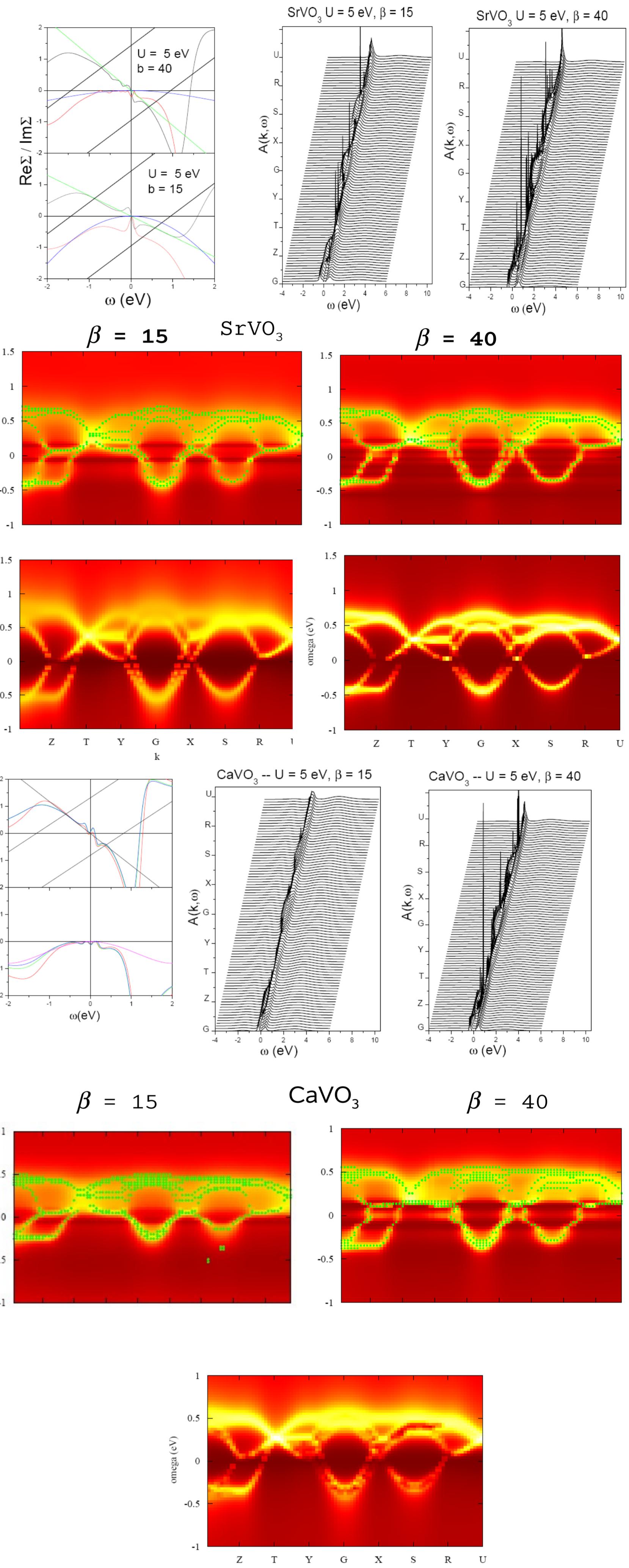
we obtain the k -resolved spectral densities

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

The poles of onsite Green's function using only the real part of self energy yields the correlated bands for each system.

d¹ metals- SrVO₃ and CaVO₃,

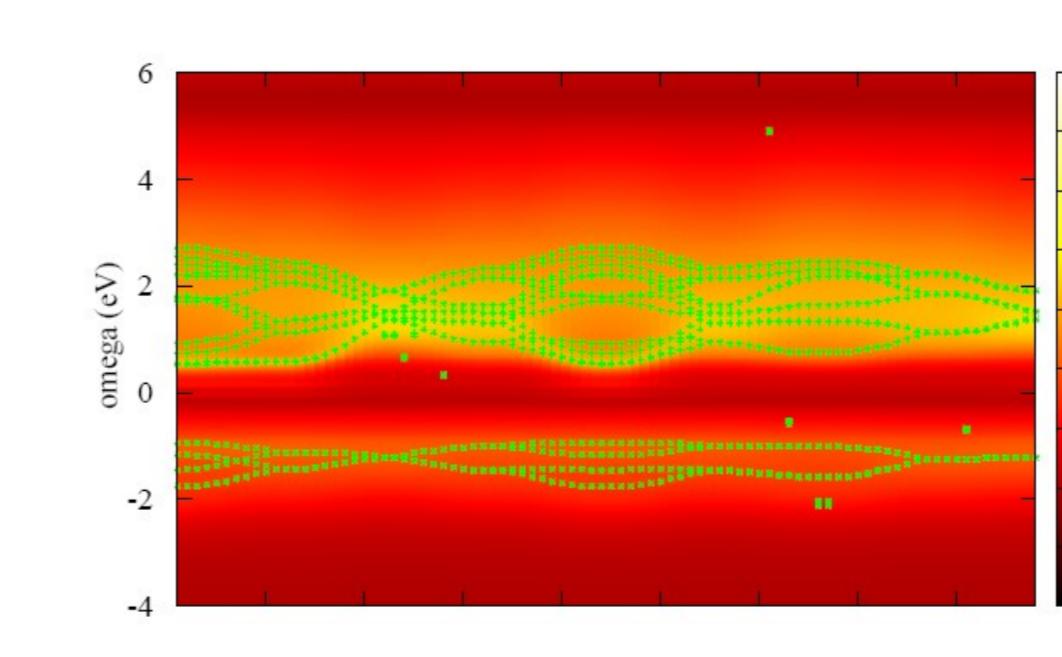
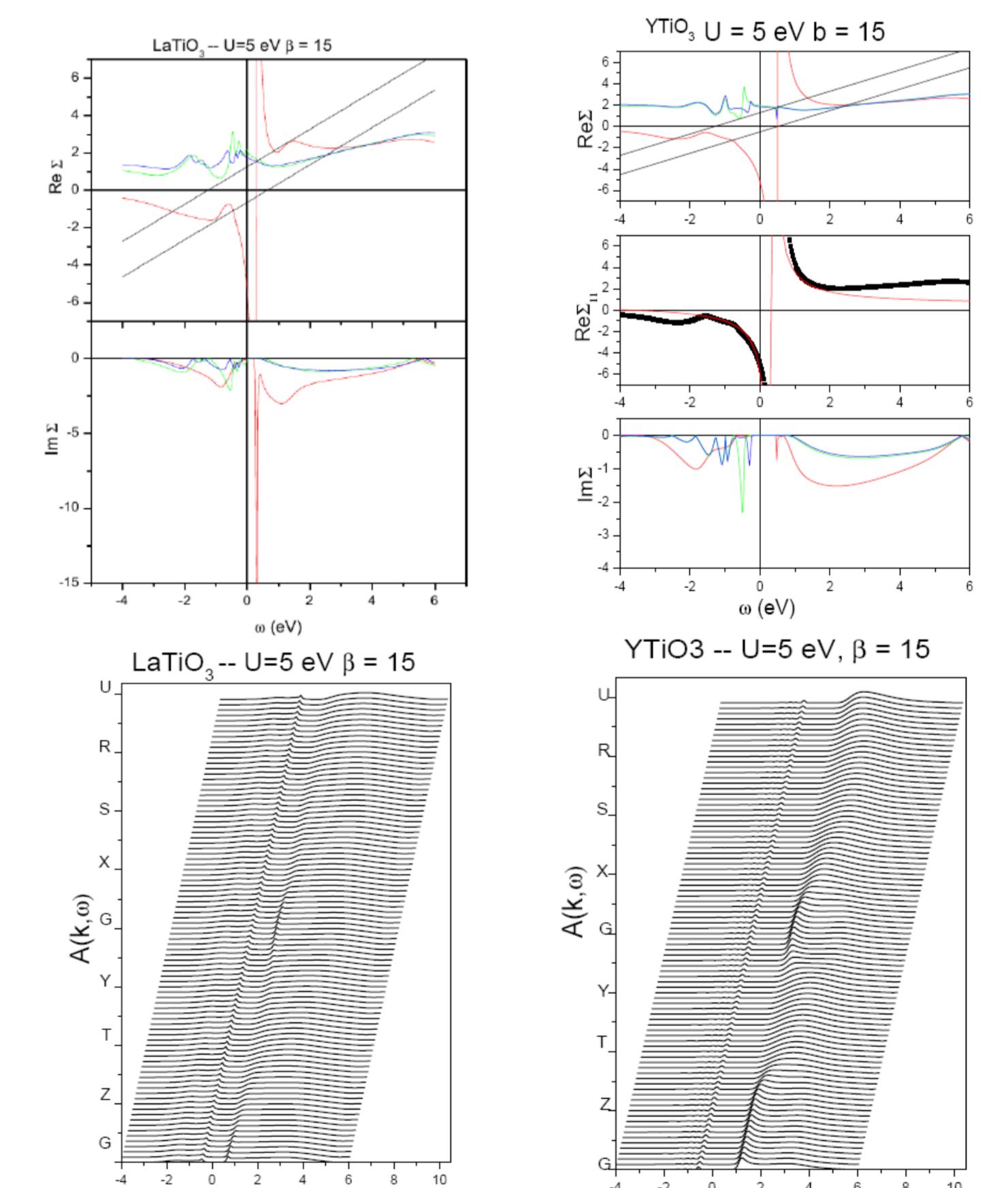
We investigate the metals – SrVO₃ and CaVO₃ in the Q -coupled representation in two temperatures. $\beta = 15$ and 40 corresponds to 800 K and 300 K respectively. The U and J values used in this calculations are 5 eV and 0.62 eV. The smoothed (green line) self energy at $\beta = 0$ reveals the effective mass m^* value to be 1.65 and 2.12 for $\beta = 15$ and 40 respectively, which are close to the experimental values. The LDA +DMFT bands and intensity plot of the k -resolved spectral functions for both temperatures shows the band-width reduction after introduction of the strong correlation. The total DOS plots shows a clear formation of lower and upper hubbard bands with the quasiparticle-peak near zero energy.



For CaVO₃, from the room-temperature self energy plots, the effective mass is calculated to be 2.183. Similar bandwidth reductions are observed in CaVO₃. It may be seen that with the introduction of strong correlations the LDA+DMFT bands for SrVO₃ and CaVO₃ are very similar.

d¹ insulators – LaTiO₃ and YTiO₃

The self-energy in the crystal-field basis depicts a diverging behaviour for the occupied orbital and the two unoccupied orbitals are almost degenerate energetically. This is a non-spin polarized calculation for both of the materials and therefore a comparison with LSDA+U calculation reveals that both of the systems have four bands below Fermi level, which is the correct ground state for YTiO₃ and LaTiO₃ non-spin-polarized case. Since the self-energy of unoccupied orbitals are nearly degenerate, calculated poles for the unoccupied bands are energetically at the same position. Hence, we get less number of bands in comparison to LSDA+U calculations.



Band gaps of 0.2 eV for LaTiO₃ and 1 eV for YTiO₃ are reproduced from the LDA + DMFT results.

References:

- [1] E. Pavarini, A. Yamasaki, J. Nuss and O. K. Andersen, New J. Phys. 7, 188 (2005), E. Pavarini et. al. Phys. Rev. Lett. 92, 176403 (2004).
- [2] I. H. Inoue et. al. Phys. Rev. Lett. 88, 236403 (2002).
- [3] O. K. Andersen and T. Saha-Dasgupta, Phys. Rev. B 62, 16219(2000).