Ab initio modelling and design of correlated materials

In this project we focus on the microscopic modelling of frustration, doping, pressure and temperature effects in a few classes of correlated electron systems investigated within the SFB/TR 49. We combine state-of-art *ab initio* density functional theory (DFT) calculations with molecular dynamics (MD) simulations and many-body techniques in order to describe the behaviour of the correlated materials, specially in the vicinity of a phase transition.
Building upon the experience and progress made in the first funding period both on methods and materials, we will extend our present studies to
• **(a)** the investigation of phonons, doping and chemical substitution effects on the electronic and magnetic properties of a few families of quantum spin systems: (a1) the triangular lattice family $Cs_2CuCl_{4-x}Br_x$ for $0 < x < 4$ and (a2) the spin dimer systems $BaCuSi_2O_6$, CuII- p-hydroquinonate coordination polymer TK91 and the organic tolane-bridged biradical MEAM 154 and variations of it.
• **(b)** the description of phase transitions in a large variety of charge-transfer salts with lattice ordering different than that of the family κ-(BEDT-TTF)$_2X$ (BEDT-TTF stands for bis(ethylenedithio)-tetrathiafulvalene, X for the anion) with complex pressure-temperature phase diagrams: (b1) the two-dimensional dual-layered material (BEDT-TTF)$_2$Ag$(CF_3)_4$(TCE) with alternating κ and α' phases in the structure, (b2) the frustrated square lattice β''-(BEDT-TTF)$_2SF_5RSO_3$ (R=CH_2CF_2, $CHFCF_2$, CHF) which shows a metal to insulator transition as a function of pressure, temperature or anion properties, (b3) $EtMe_3Sb[Pd(dmit)_2]_2$ (Et=C_2H_5, Me=CH_3) with a probable spin-liquid behaviour, (b4) the family (TMTCF)$_2X$ (C=S, Se, X=PF_6, AsF_6, SbF_6) with a rich phase diagram including charge ordered, Mott insulating, spin-Peierls, antiferromagnetic, metallic and superconducting phases (b5) (Tetrathiafulvalene)(tetracyanoquinodimethane) (TTF-TCNQ) under biaxial pressure and (b6) the spin-liquid κ-(BEDT-TTF)$_2Cu_2(CN)_3$. Understanding from first principles the microscopic origin of structural and magnetic phase transitions in correlated electron systems is at the centre of our investigations.
The starting point of our analysis are *ab initio* DFT calculations that we perform in a comparative scheme of different basis sets: Full potential linear augmented plane waves (FPLAPW), full potential localised orbitals (FPLO), projector augmented waves (PAW) and linear muffin-tin orbitals (LMTO) as well as exchange-correlation functionals: local density approximation (LDA), generalised gradient approximation (GGA), LDA+U, or the hybrid B3LYP (Becke, three-parameter, Lee-Yang-Parr). For our organic systems we also include a van der Waals correction to the total energy and perform some quantum chemistry calculations at the molecular level.
Pressure (hydrostatic and anisotropic) are simulated with Car-Parrinello molecular dynamics calculations as well as based on the Born-Oppenheimer potential energy surface. We also perform molecular dynamics calculations to study temperature effects.
One of our goals is the extraction of Hamiltonian model parameters out of electronic structure calculations to be used in the many-body-theory projects.