# Crystal fields and superexchange interactions in localized f-electron compounds

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Strongly-correlated rare-earth (RE) and actinide compounds are studied by an ab initio framework that combines the density-functional theory (DFT) with a quasi-atomic (Hubbard-I) dynamical treatment for the on-site Coulomb repulsion between localized f states. We first discuss applications of this approach to the crystal-field (CF) splitting on RE ions in hard magnetic transition-metal-RE intermetallics. The CF splitting on the 4f shells in these compounds is known to determine the RE single-ion (SI) magnetic anisotropy, which is crucial for the hard-magnetic behavior of these systems. Within the present approach the 3d magnetism of the TM sublattice is treated by the standard local spin-density approximation (LSDA), while strong correlations on the RE 4f shell are included through Hubbard-I. We carefully remove an unphysical self-interaction contribution to CF due to the LSDA [1]; an optimal choice for the localized basis to represent 4f orbitals in solids is also discussed. The method is applied to RCo5 and RFe11MX families of ferromagnetic intermetallics (R=Nd,Sm, M=Fe,Ti, X=empty, N, Li), for which we analyze the impact of 4f-conduction states hybridization on the CF splitting and magnetic anisotropy.

In the second part of the talk we present a linear-response approach for extracting intersite exchange interactions between localized shells. Starting from the high-temperature local-moment paramagnetic state described within the DFT+Hubbard-I framework, we derive these exchange interactions by evaluating the response of DFT+ Hubbard-I functional to small fluctuations in atomic configurations on two neighboring sites [2]. Using this approach we evaluate the superexchange coupling between the dipole and quadrupole U moments in UO2 [3]. The calculated superexchange Hamiltonian has a non-collinear 3k antiferromagnetic (AFM) ground state, in agreement with the experimental magnetic structure of UO2. We find that the stabilization of 3k AFM is due to a subtle anisotropy of the quadrupolar superexchange lifting the degeneracy between various AFM structures on the frustrate fcc U sublattice. Applying the same approach to NpO2 we predict a purely multipolar low-temperature ordered state with no dipole magnetic moments and a primary triakontadipole rank-5 order parameter.

[1] P. Delange et al. PRB 96, 155132 (2017)
[2] L. V. Pourovskii PRB 94 115117 (2016)
[3] L. V. Pourovskii and S. Khmelevskyi, PRB 99, 094439 (2019).