Hessisches Kompetenzzentrum
für Hochleistungsrechnen

Interplay Between Spin Orbital and Lattice Degrees of Freedom in Multiferroic and Orbital Ordered Oxide Systems

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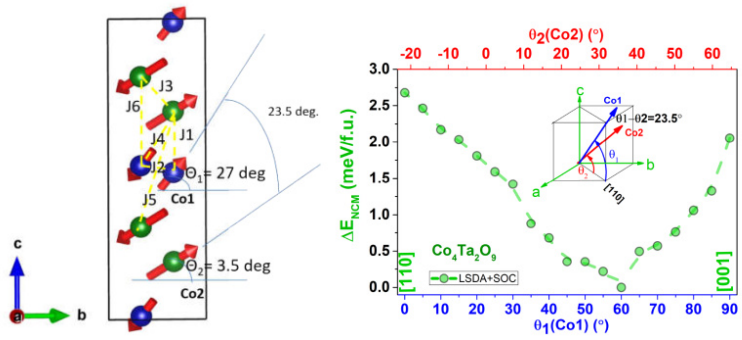
Project Term
2020 - 2021

Clusters
Lichtenberg Cluster Darmstadt

Software
WIEN2K

Additional Software
WIENNCM



Introduction

The electronic structure plays a key role in determining the physical properties of highly correlated electron systems. This may initiate various exotic phenomena such as multiferroicity, superconductivity or quantum criticality. We investigate two such phenomena, namely multiferroicity and spin reorientation transition in oxides. Multiferroicity has novel applications such as in MERAM utilizing the coupling between ferroic orders. Neutron scattering experiments revealed a variety of ground states (GS) in multiferroic $(Mn,Co)_4Ta_2O_9$. In our previous projects, we concluded that the different GSs are determined by a competition between different magnetic anisotropies such as spin-orbit coupling (SOC) and magnetic dipole-dipole interaction. Very recently a new high temperature magnetic phase was reported in $Co_4Ta_2O_9$ and we were interested in this.
The orthoferrites $RFeO_3$ (R: rare earth) are known for spin reorientation transitions at lower temperatures. Although the magnetic structures and spin wave excitations of the Fe lattice are known, the behavior of R and the interaction between them are less explored. Very recently, lower energy modes from Yb, due to Fe-Yb interaction was reported in $YbFeO_3$. We investigate the single particle crystal field (CF) excitation of a R-member and the results will be used to investigate the Fe-R interaction and the transformation of the CF excitation to a possible many-particle dispersive excitation.

Methods

We performed collinear and non-collinear spin polarized electronic structure calculations based on density functional theory (DFT) to determine the non-collinear anisotropy contributions and exchange constants to describe the GSs and the phase transitions.

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Calculation of the CF levels of f electron systems using a Hamiltonian mapping technique: The localized f Hamiltonian is obtained after a regular DFT run, transformed to real space and finally expanded in a series of spherical tensor operators, whose coefficients are the CF parameters.

Results

We were able to determine both magnetic phases by our neutron diffraction experiments (magnetic symmetry *C2/c'* and *C2'/c*). Our results indicate a competition between out of plane anisotropy and in plane anisotropy in the *ab* plane. Our non-collinear DFT calculations unambiguously showed the existence of an out of plane anisotropy due to SOC and this is in good agreement with the GS of $Co_4Ta_2O_9$ described by *C2/c'* (Figure 1). In order to understand the magnetic phase transition between *C2/c'* and *C2'/c,* exchange constants between Co-Co were calculated. The transformation between the magnetic structures are realized through spin reversal on half of the J2 (Co1-Co1) and J3 (Co2-Co2) exchange bonds. Surprisingly J2 (~-0.08 meV) and the in plane anisotropy within *ab* are very small. We also extracted the magnetic diffuse scattering from neutron diffraction pattern and determined the spin-spin correlation functions, which should reflect the exchange interactions. They are in good agreement with the calculated trend.
Point charge model is the method of choice to calculate the CF levels of R ions. However, the level splitting strongly depends on the effective charges. Here, we use a more accurate Hamiltonian mapping method to calculate the CF level splitting. We were able to reproduce the GS and the first excitation level very accurately by only varying the hybridization parameter Δ (Figure 2). Furthermore we were able to get estimations of the exchange constants of Fe-Fe, R-R and Fe-R using DFT calculations.

The calculation of the chemical shift for different local structure models (for NMR) in low level Li doped $BaTiO_3$ and Al and Ta co-doped TiO_2 were completed during this period, which were started during the previous period.

Discussion

The magnetic phase transition between *C2/c'* and *C2'/c* takes place possibly due to a partial rotation of the spins assisted by a very weak J2 value. This transition is not limited to $Co_4Ta_2O_9$ but extends to up to $Co_3MnTa_2O_9$. The calculated exchange constants will be used as starting values to model the spin waves in $Co_4Ta_2O_9$, obtained from inelastic neutron scattering. The CF coefficients are important to fix the magnetic anisotropy to model the spin wave like excitations. The calculated exchange constants were used as starting values for the modeling of spin wave like excitations in $RFeO_3$.
The DFT and related calculations are extremely time-consuming in serial form and resource-wise expensive. Therefore we utilized the k-point as well as the hybrid MPI/OpenMP parallelization options available in our DFT codes. The Lichtenberg HPC is an

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ideal system to perform such computationally intensive calculations.

Figures

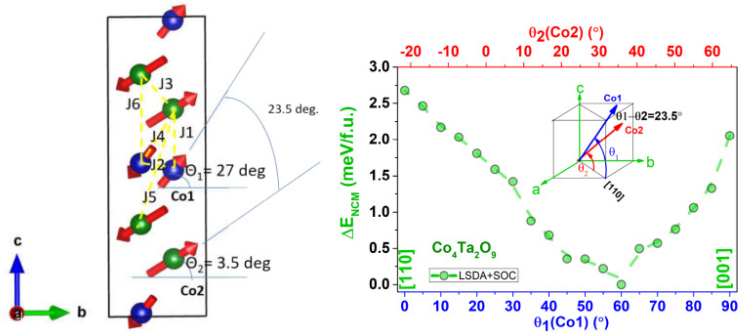

Figure 1: Non-collinear ground state magnetic structure of Co4Ta2O9 described by C2/c' used for non-collinear DFT calculations with important exchange paths J_i (left) and magnetic anisotropy within the non-collinear setup (right). Our calculations show an out of plane anisotropy.

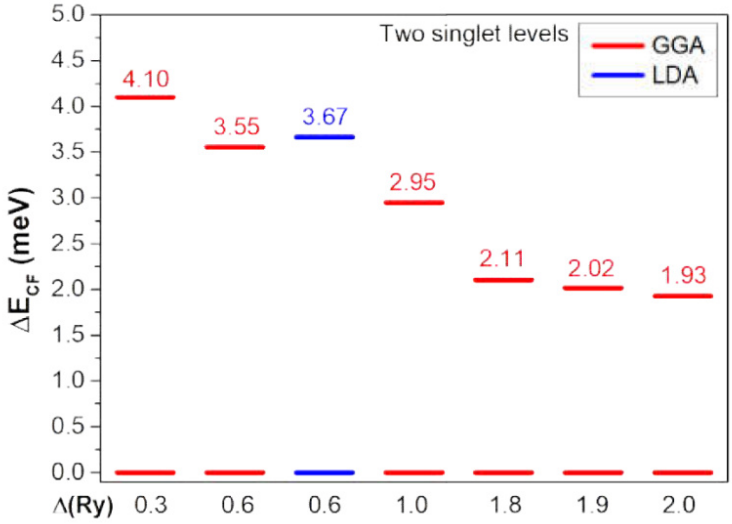

Figure 2: CF levels of J=4 ground state for LDA and GGA and for different values of Δ: Only the lowest two singlets adjusted to the experimental values are shown.

Publications

Narayanan, N., Lou, Q., Rawal, A., Lu, T., Liu, Z., Chen, J., Langley, J., Chen, H., Hester, J., Cox, N., Fuess, H., Mcintyre G.J., Li, G., Yu, D., Liu, Y.: "Defect structure and property consequence when small Li+ ions meet BaTiO3, Physical Review Materials 4, 084412, 2020
https://doi.org/10.1103/PhysRevMaterials.4.084412

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