Ab-intio studies of Chromia : Calculation of Hubbard U parameter

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Abstract

Cr_2O_3 (Chromia) is one of the fascinating materials because of its magnetoelectric properties and potential applications in spintronic devices like SpinFET. It has been studied experimentally as well as theoretically using density functional theory. But it is known that the conventional density functional theory (DFT) fails for the materials that have strongly correlated electrons [1]. This failure is caused by significant inaccuracies in the intra-atomic Coulomb and exchange interactions. Through the use of empirical parameters, the DFT+U approach provides better results but still it is a parameter dependent approach. Therefore, to make the DFT+U method a truly first-principles technique, a systematic ab initio strategy for assessing the intra-atomic Coulomb interaction(U) is being used, where the Hubbard U value is calculated through Density Functional Perturbation Theory (DFPT) using the Linear response theory for the compound of interest [2], that is, chromia.

Introduction

- Antiferromagnetic Cr_2O_3 (Chromia) has been a material of significance to be exploited in spintronics logic devices and memory [3]. It can be used the material for building gate of the spin-FET. Previous works have shown that the magneto-electric properties of Chromia can be exploited in the ME-RAM (Magneto-Electric RAM) [4].

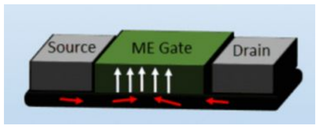

FIG. 1 Spin-FET where the spin of carriers is manipulated by interaction with a magnetoelectric (ME) gate (green).

- Results obtained from conventional DFT are inaccurate and different from the experimentally properties (such as the band gap of 1.4 v/s 3.2eV), resulting in a poor understanding of such complex oxides [1].
- Understanding the intra-atomic interactions is helpful in making the results accurate. An approach to compute the Hubbard U value through Linear response theory has been successfully proposed and done for other oxides such as MnO_2 [5].
- In this work, the Hubbard U value is computed from the fully first principles calculations.

Structure

- The antiferromagnetic structure of Cr_2O_3 has lattice parameters a = c = 5.070 Å and b = 5.473 Å and crystallizes in the rhombohedral corundum-like structure with *R3c* symmetry. The linear magneto-electric effect is possible due to the alternate '+' and '-' spins of the Cr atoms along the Rhombohedral axis.

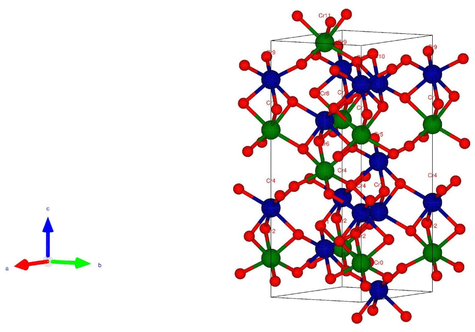

FIG. 2. Unit cell of Antiferromagnetic Cr_2O_3. Green (for '+') and Blue (for '-') atoms represent Cr atoms.

- The 3d shell is partially filled and spin polarised and these electronic correlations are poorly understood.

Theoretical Method

- Bulk Cr_2O_3 with antiferromagnetic ordering is investigated using first principle calculations, employing the Generalized Gradient Approximation (GGA) for exchange and correlations [7]. The calculations are extended using the linear response theory to find the Hubbard U value.
- The calculations are done using the Plane Wave representations using the ultrasoft pseudopotentials implemented in the Quantum Espresso package.
- The crystal is relaxed (volumetric and ionic relaxations) for every spin configuration to get the optimized lattice parameters.
- The energy, force and pressure thresholds for convergence are kept at 10^{-8} Ry, 10^{-5} Ry/Bohr and 1Kbar respectively.
- The Brillion-zone integration is performed using a 8x8x5 Monkhorst-Pack *k*-point grid for bulk Chromia.
- To represent the electronic wave functions, we have used the energy cutoff of 50 Rydberg and the charge density cutoff is kept at 400 Ry.
- The calculations for Hubbard U parameter are done using Marzari-Vanderbilt smearing along with a k-point mesh of 12x12x7 and the energy cutoff of 75 Ry.

Results

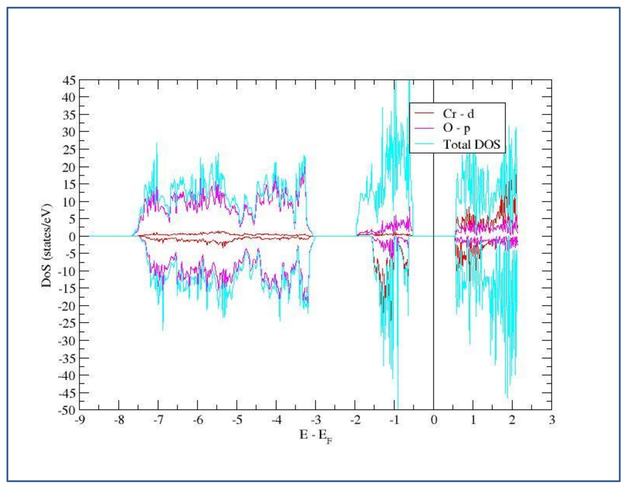

FIG. 3. Total density of states(TDOS) & PDOS for bulk Cr_2O_3. Fermi level is $E = 0$.

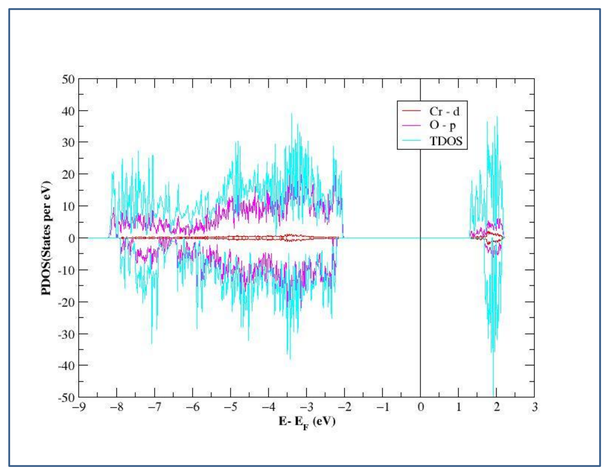

FIG. 4. Total DOS & PDOS after DFT + U (with U = 6.62eV)

Table 1 : The calculated parameters for Cr_2O_3

	Hubbard U (eV)	Magnetic Moment per cr atoms (μ_B)	Band Gap (eV)	Band Gap (DFT + U) (eV)
Our Work	6.62	3.08	1.07	3.34
Experimental work [8]	NA	3	3.3	NA
Other's work on DFT[1]	4	2.9	~ 0.8	3.07

- The ab-initio computed value of onsite coulomb interaction parameter U is 6.62 eV .
- The underestimated band gap obtained from DFT calculations is 1.07 eV.
- The computed band gap after DFT + U is 3.34 eV which is very close to the experimental value of ~ 3.3 eV [8].
- The magnetic moment of 3.2 μ_B for Cr atoms is very close to the experimental value of $3\mu_B$ [9].

Conclusion

- In conclusion, the first-principle calculations have been used to investigate the electro-magnetic properties of bulk Cr_2O_3 with antiferromagnetic ordering.
- The Hubbard U is computed ab-initio and is found to be 6.62 eV, which is in agreement with the literatures that have used the U parameter empirically.
- The linear response theory is found to be a good formalism for computing the onsite coulomb interactions ab-initio which otherwise is not possible.
- The antiferromagnetic ordering of Cr_2O_3 carries interesting properties and should be studied further.

References

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