$CrSiTe_3$ electronic-structure DFT calculation

Jan Kierzkowski

September 25, 2025

1 Introduction - $CrSiTe_3$ system characterization

$CrSiTe_3$ crystallizes in a rhombohedral structure with space group $R3^-$ (No. 148). In this lattice, Cr atoms are octahedrally coordinated by Te, and Si sits in between the layers. Because its layers are weakly bound by van der Waals forces, single or few-layer flakes can be mechanically exfoliated.

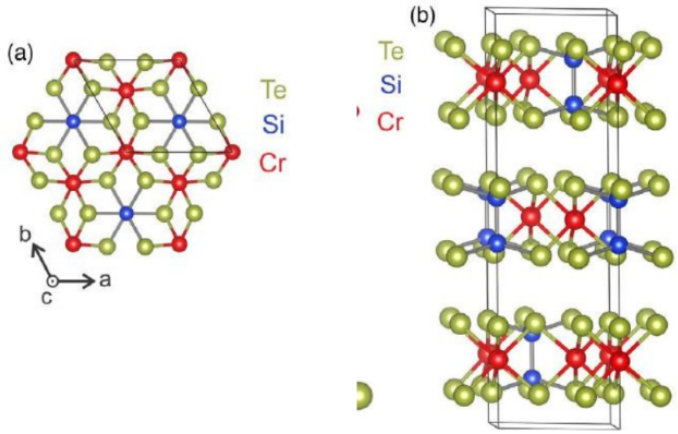

Figure 1: $CrSiTe_3$ A-B-C stacking system

It is ferromagnet with a transition temperature of 33 K, with magnetic moments primarily from the Cr^{3+} ions. Calculations and experiments indicate that the easy axis is along the crystallographic c-axis (out-of-plane). Therefore, we assume the spin direction along the c-axis (out-of-plane).Cr atoms carry the local magnetic moments (from the $3d^3$ configuration of Cr^{3+}).

Electronic-structure calculations indicate a narrow band gap - about 0.19 eV in spin-polarized calculations (SP+U=3eV) and ∼0.04 eV (SOC+U=3eV) - suggesting that it is nearly gapless (semimetallic behavior).

2 Calculations

The calculations were performed using **VASP (Vienna Ab-initio Simulation Package)** software. It is a DFT code based on the **PBE-GGA (Perdew–Burke–Ernzerhof-Generalized Gradient Approximation)** functional for exchange–correlation. The calculation parameters, including plane-wave cutoff, smearing method, Hubbard U corrections(U=3 eV for Cr atoms), and van der Waals interactions, were set in the **INCAR** file provided with this paper.
The simulation was divided into two parts:

1. Spin-polarized (SP)
2. Spin-orbit-coupled (SOC)

Each part has three stages:

- Cell geometry relaxation
- Static SCF (self-consistency cycle) calculation
- Density of State (DOS) and band structure calculation

2.1 Relaxation of the system

The first stage of each calculation (SOC and SP) was a cell geometry relaxation calculation (**ISIF=3** option in **INCAR** file). The starting lattice vectors and scaling factor were set to the same values (Table 1).

Table 1: Starting lattice vectors

Lattice vec.	Scaling factor = 1.0		
a	3.42	-5.92	0.0
b	3.42	5.92	0.0
c	0.0	0.0	21.77

After full relaxation can seen only minor adjustments has to be done relax the system (Tables 2 and 3). After relaxation in the SP calculation external pressure was 0.04 kB and pullay stress 0.00 kB. In the SOC calculation external pressure was 0.01 kB and pullay stress 0.00 kB. So that relax structure were achieved for the SP and the SOC calculations. The lattice vector lengths were compared with experimental values [1], showing a relative error of less than 2.5% (Table 4 in the Appendix). Then to obtain a converged WAVECAR file. In order to reach convergence more quickly, static SCF calculations were done in each part (SP and SOC).

Table 2: After cell relaxation in SP calculation

Lattice vec.	Scaling factor = 1.0		
a	3.39	-5.87	0.0
b	3.39	5.87	-0.0
c	0.0	0.00	20.21

Table 3: After cell relaxation with SOC calculation

Lattice vec.	Scaling factor = 1.0		
a	3.40	-5.88	-0.0
b	3.40	5.88	0.0
c	0.0	0.00	20.17

2.2 Bands Structure and Density of States

Firstly, it is necessary to determine the high-symmetry path along which VASP should compute the band structure. The high symmetry points and path was found by using SeeKpath online software[2]. The chosen high-symmetry path: $\Gamma - T - H_2 - H_0 - L - \Gamma - S_0 - S_2 - F - \Gamma$, is presented underneath on the Fig. 2.

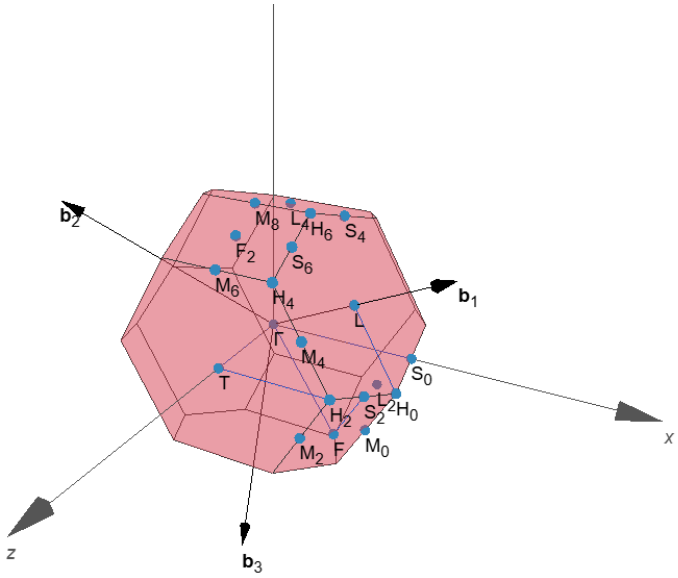

Figure 2: K-path on Brillouin zone[2]

Then band structure and DOS calculation were performed for each setup. The spin-up and spin-down bands are shifted related to each other (Fig.4). This is the signature of a ferromagnetic systems. The SP calculation shows narrow band gap - about 0.19 eV indicating semiconductor character. The SOC corrected calculation closed the band gap to -0.04 eV (Fig.1 in the Appendix) suggesting metallic character of the system. Near Fermi level bands

are primarily occupied by Cr (magenta color), Si (green) and Te (orange) are more present on higher bands, confirming that Cr carry the magnetic moment.

3 Summary

This work presents DFT calculations of the electronic structure of $CrSiTe_3$, a 2D van der Waals ferromagnet, using spin-polarized and spin-orbit-coupled methods. The results show a narrow band gap in spin-polarized calculations that closes under SOC, with Cr states dominating near the Fermi level and carrying the magnetic moment.

References

[1] Barat Achinuq et al. "Covalent Mixing in the 2D Ferromagnet CrSiTe3 Evidenced by Magnetic X-Ray Circular Dichroism". In: *physica status solidi (RRL) – Rapid Research Letters* 16.4 (2022), p. 2100566. DOI: https://doi.org/10.1002/pssr.202100566. eprint: https://onlinelibrary.wiley.com/doi/pdf/10.1002/pssr.202100566. URL: https://onlinelibrary.wiley.com/doi/abs/10.1002/pssr.202100566.

[2] Y. Hinuma et al. *Band structure diagram paths based on crystallography.* 2017. DOI: 10.1016/j.commatsci.2016.10.015. URL: https://arxiv.org/abs/1602.06402 (visited on 09/24/2025).

Appendix

Table 4: Comparison between calculated lengths of lattice vector to the experimental values[1]

	SOC (Å)	SP (Å)	experimental values[1]	SOC relative error (%)	SP relative error (%)
|a|	6.7922	6.7786	6.7699	0.33	0.13
|b|	6.7922	6.7786	6.7699	0.33	0.13
|c|	20.17	20.21	20.6825	2.48	2.28

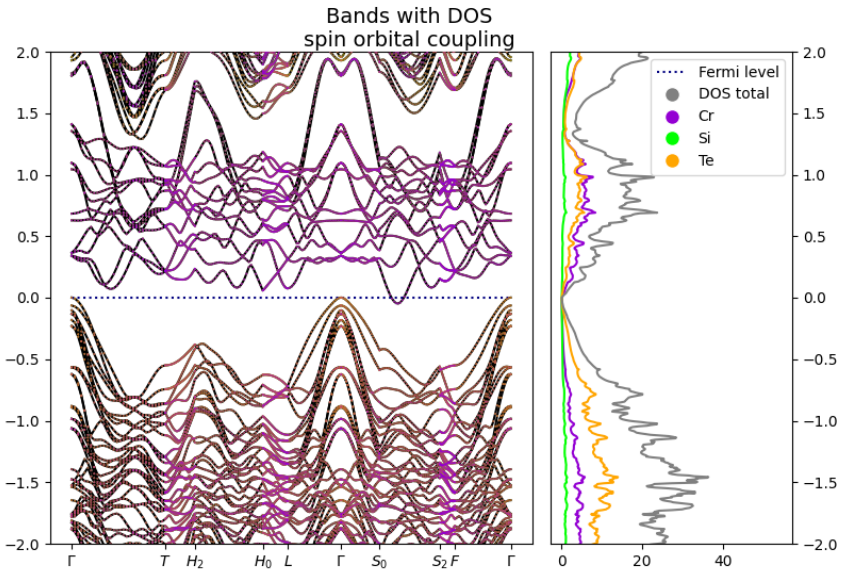

Figure 3: Band structure with density of state for spin-orbit coupling calculations.

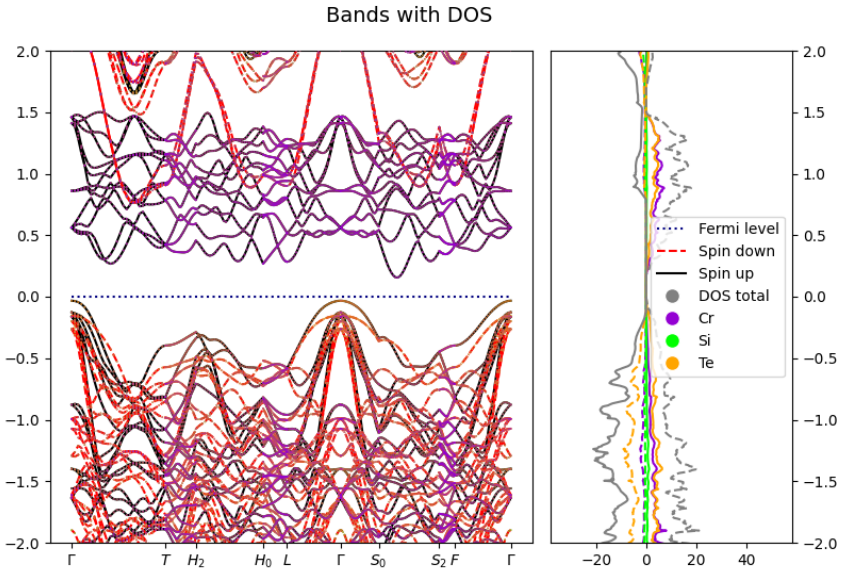

Figure 4: Band structure with density of states for spin-polarized calculation.