

# AB INITIO STUDY OF EXCHANGE INTERACTIONS IN THE MAGNETIC SYSTEMS BASED ON $\text{Cr}_2\text{Ge}_2\text{X}_6$ (X=S, Se, Te)

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## I. INTRODUCTION

Limiting the dimension of the spin sublattice (atoms with a magnetic moment) leads in most cases to the destruction of the long-range magnetic order. However, layered van der Waals structures (vdW materials) appear low-dimensional magnetism. Recently, the first 2D magnet based on an intrinsic vdW Ising compound ( $\text{FePS}_3$ ) was exfoliated [1] from crystals with 3D magnetic order. 2D vdW monolayers are a promising class of materials as a functional layer in spintronic devices.

Magnetic exchange energy plays a key role in the formation of magnetic order. This type of energy is determined by the integral of the exchange interaction, which characterizes the degree of the electrons wave functions overlap (or atomic orbitals).

The exchange interaction integral is calculated using *ab initio* simulation and the Heisenberg model. The values of the exchange interaction integral can help to establish the ground magnetic order. The article presents the results of the exchange interaction integrals calculations for  $\text{Cr}_2\text{Si}_2\text{X}_6$  (where X = S, Se, Te) compounds.

## II. RESULTS

We calculated the exchange interaction integral by using first principles calculations and the classical Heisenberg model together. This technique contains two main stages: quantum mechanical (QM) calculations and calculation of the exchange interaction integral using the Heisenberg magnetic model.

At the QM stage, we calculated the energies of atomic systems for various magnetic configurations at absolute zero temperature. The ground and excited magnetic states of the system were determined. These calculations were performed in a software package VASP, which implements the density-functional theory (DFT) method [2]. The main source of DFT simulation errors is inaccurate accounting of correlation energies. This is one of the terms that make up the total energy of the system. This should be especially taken into account in materials with localized (narrow) zones (d-electrons) due to an increase in the proportion of the correlation component in the energy characteristic of the system. The most common way to help neutralize this simulation flaw is the local spin-density approximation (LSDA+U), which was used in this work (U = 3 eV, J = 0.9 eV for Cr d-orbitals).

Exchange interaction integrals are calculated by comparing the value of the total energy obtained in the quantum mechanical simulation with the corresponding expression in the Heisenberg model.

Atomic systems were simulated with preliminary structural relaxation taking into account spin polarization. Next, static self-consistent calculations were performed to determine the electronic properties.

The magnetic exchange interaction weakens sharply with an increase in the distance between interacting electrons. Often, this type of interaction is significant only between the two nearest neighboring atoms. While

for all other combinations, it does not need to be taken into account due to a too small value. However, in this work, to analyze the spin-dependent properties in 2D vdW structures ( $\text{CrSiX}_3$ , where  $X = \text{S, Se, Te}$ ), the exchange interaction integrals were calculated for atoms located at a distance of up to 10 Å. This was done in order to confirm the absence of competition between magnetic orders.

To calculate the exchange interaction integrals ( $J_n$ , where  $n=1, 2, 3$  the order of the atom in accordance with the distance),  $2 \times 2 \times 1$  supercells were created. The orientations of the magnetic moment (spin) were set for each Cr atom in such a way as to form 4 magnetic configurations. One of which had a ferromagnetic order (FM), the other three were antiferromagnetic (AFM). Structural optimizations were performed for all configurations.

Table 1. Simulation results of  $\text{Cr}_2\text{Si}_2\text{X}_6$  structure, magnetic and electronic properties

Compound	$J_1$ , meV	$J_2$ , meV	$J_3$ , meV	$\frac{J_{1, \text{meV}}}{\text{Hubbard correction}}$	$\frac{J_{2, \text{meV}}}{\text{Hubbard correction}}$	$\frac{J_{3, \text{meV}}}{\text{Hubbard correction}}$	Magnetic order
$\text{Cr}_2\text{Si}_2\text{S}_6$	2.47	-0.07	0.04	0.84	-0.22	0.01	AFM
$\text{Cr}_2\text{Si}_2\text{Se}_6$	-1.59	0.18	-0.04	-1.88	0.13	-0.06	FM
$\text{Cr}_2\text{Si}_2\text{Te}_6$	-1.26	0.09	-0.17	-1.61	0.05	0.60	FM

Here, if the exchange integral is less than zero, then the atomic system is a ferromagnet. According to results,  $\text{Cr}_2\text{Si}_2\text{Se}_6$  and  $\text{Cr}_2\text{Si}_2\text{Te}_6$  structures ferromagnets. However,  $\text{Cr}_2\text{Si}_2\text{S}_6$  exhibits strong antiferromagnetic order. Almost all the contribution to the exchange interaction is made by  $J_1$ . Therefore, there is no competition in the magnetic moment.

### III. CONCLUSIONS

Structural and geometric optimizations were carried out using quantum mechanical simulation in a specialized software package VASP, and the main magnetic states were established. Two structures under study demonstrate ferromagnetic order ( $\text{Cr}_2\text{Si}_2\text{Se}_6$  and  $\text{Cr}_2\text{Si}_2\text{Te}_6$ ) and one structure is antiferromagnet.

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### REFERENCES

- [1] J. U. Lee, S. Lee, J. H. Ryoo, S. Kang, T. Y. Kim, P. Kim, C. H. Park, J. G. Park, H. Cheong, Nano Lett. 16 (2016) 7433.
- [2] G. Kresse and D. Joubert, Phys. Rev. B 59 (1999) 1758.