Electronic Supplementary Material (ESI) for Materials Advances.

Signature of magnetism in 2D-Chromia: 2D analog of natural α-Cr_2O_3 mineral and its heterostructure with graphene

Renu Singla,[1,2] Rahul Singla,[3] Sarvesh Kumar,[4] Timothy A. Hackett,[5] and Manish K. Kashyap[1,6,*]

[1]Department of Physics, Kurukshetra University, Kurukshetra 136119 (Haryana), India.
[2]Department of Physics, Daulat Ram College, University of Delhi, Delhi 110007, India.
[3]Department of Physics, Panjab University, Chandigarh 160014 India
[4]Inter-University Accelerator Centre (IUAC), Aruna Asaf Ali Marg, New Delhi 110067, India.
[5]Department of Biochemistry, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0664, United States.
[6]Renewable Energy Laboratory, School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India
*Corresponding author's e-mail: manishdft@gmail.com, mkkashyap@mail.jnu.ac.in

Supplementary Information (SI)

The thermal stability was ensured even after running 5000 steps, the variations in total energy was very small (Fig. S1) and no bond was broken. Also, the movements of Cr and O atoms were negligible with infinitesimally small change in bond length.

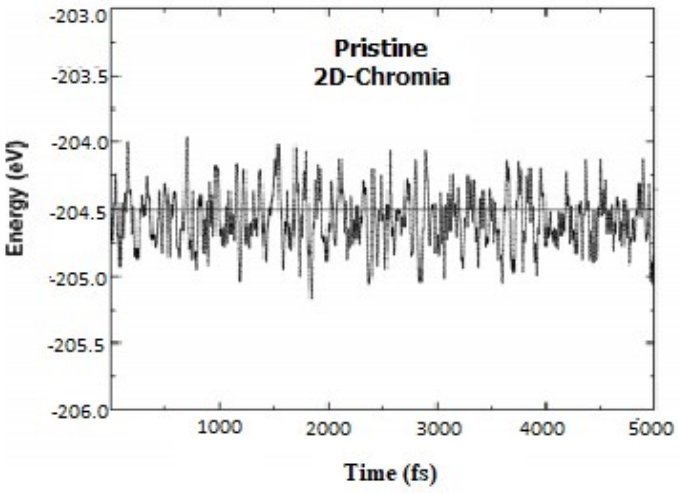

Fig.S1. Variations of total energy of 2D-Chromia in E_5 configuration during AIMD simulation with a time step of 1 fs at a temperature of 300 K.

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The pristine 2D-Chromia has only real phonon frequencies which demonstrates its dynamical stability as depicted in phonon dispersion curve (Fig. S2).

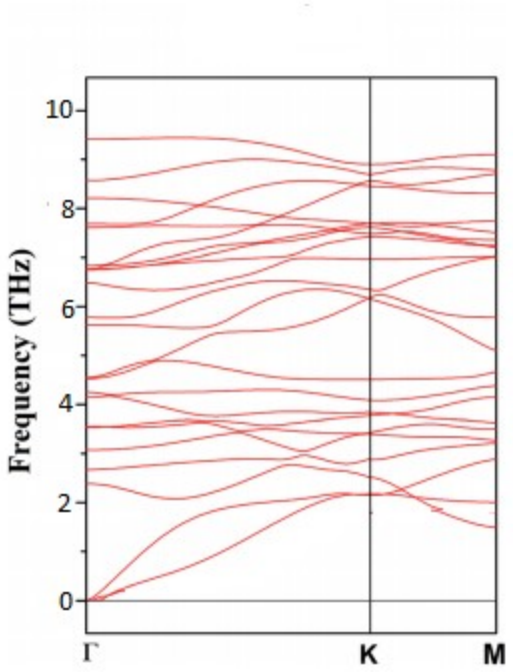

Fig.S2. Phonon dispersion curve for 2D-Chromia in E_5 configuration along the high symmetry k-points.

Unlike pristine 2D-Chromia monolayer which has large band gap and ferromagnetic in ground state, pristine graphene has zero band gap and zero magnetic moment (Fig. S3).

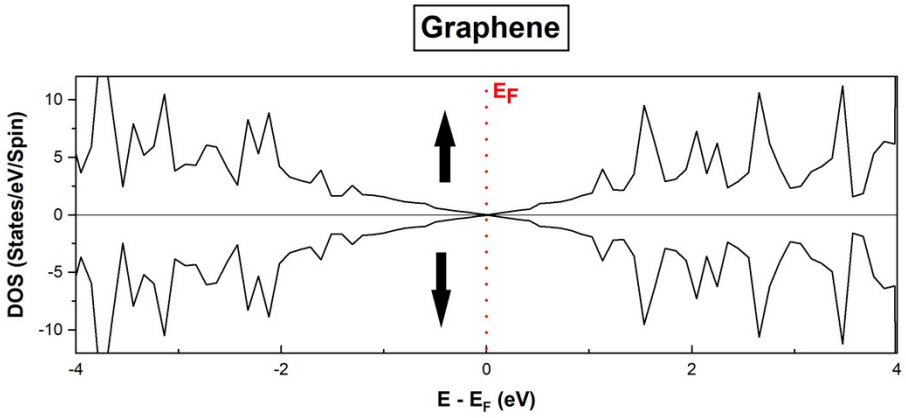

Fig.S3. Total Density of states plot (TDOS) for pristine graphene in majority (↑) and minority spin (↓) channels. E_F denotes the Fermi level and is shifted to zero.

Clearly, valance band and conduction band in both the spin channels touches at the Fermi level indicating zero band gap. Also, the TDOS is exactly symmetrical for both the spin channels which manifests zero

magnetic moment. Both these limitations abstain the application of graphene in transistors, optoelectronics and spintronics. Thus, in order to overcome these limitations, we made an attempt to make its heterostructure with 2D-Chromia.

In order to confirm the structural stability, the binding energy of Graphene/2D-Chromia heterostructure was calculated by using the formula as below:

$$E_{bind} = E_{graphene/2D\text{-}Chromia} - N_l E_{graphene} - N_l E_{2D\text{-}Chromia} \quad \text{(S-1)}$$

where $E_{gra/2D\text{-}Ch}$, $E_{graphene}$ and $E_{2D\text{-}Chromia}$ represent the ground state energy of Graphene/2D-Chromia heterostructure, pristine graphene and 2D-Chromia monolayers, respectively. The values of binding energies in all five configurations are depicted in Table S1.

Table S1. Calculated Binding Energy(eV), total magnetic moment (μ_S) and magnetic state of Graphene/2D-Chromia heterostructure for all possible combinations of states.

Configuration	**Cr_A**	**Cr_B**	**E_{bind}(eV)**	**$\mu_S(\mu_B)$**
E_1	↑↑	↓↓	Unstable and transform to E_5 after relaxation	
E_2	↑↓	↑↓	+0.249	0.00
E_3	↑↑	↑↓	+0.240	4.65
E_4	↑↓	↑↑	+0.123	4.70
E_5	↑↑	↑↑	-0.154	11.65

The binding Energy comes out to be negative only in case of E_5 configuration. This indicates that the resulting Graphene/2D-Chromia heterostructure is stable only in this state with almost same magnetic moment as that of pristine 2D-Chromia case.