Project Title:

Structural, electronic and magnetic properties of newly discovered 2D MXenes

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There has been emerging interest in synthesis and exploring the potential properties and applications of two-dimensional (2D) systems other than graphene. In this regard, layered materials preserve a large source for the 2D systems. Recently, some of the layered systems, MAX phases -$M_{n+1}AX_n$ systems, where n= 1,2, or 3, "M" is an early transition metal, "A" is A group elements, mostly groups 13 and 14 elements, and "X" is carbon and/or nitrogen - have been exfoliated into 2D single- and multi $M_{n+1}X_n$ layers [Adv. Mater. 23, 4248 (2011); ACS Nano 6, 1322 (2012)]. The resulting 2D- $M_{n+1}X_n$ transitional metal carbides and nitrides were named as MXenes. Considering the large number of compositional possibilities of MAX phase compounds, the large number of MXenes with unprecedented properties could also be obtained in the future. The extraordinary properties of 2D MXenes have been intensely investigated since its successful synthesization experimentally. High conductivities and high elastic moduli are found in MXenes. Besides, the applications of MXenes in Li-ion battery anodes and hybrid electro-chemical capacitors are also demonstrated. Most of the synthesized $M_{n+1}X_n$ layers are metallic with no band gap, although it is shown that the electronic structure can be modulated by appropriate surface terminations such as oxygen and fluoride [Appl. Phys. Lett. 104, 133106 (2014)]. MXenes have experimentally found applications to store massive amount of energy as supercapacitors [Science 341, 1502 (2013); Nature 516, 78 (2014)]. Above applications motivate us to carry out more theoretical and computational studies. In this research subject, first-principles electronic structures calculations were employed to study the structural and magnetic properties of various functionalized MXene Cr_2X (X=C, N) by F, O and OH groups. Each Cr_2X system consists of tri-layer sheets with hexagonal unit cell (P63/mmc space group), where the X (C or N) layer is sandwiched between the two "Cr" transition metal as shown in FIG.1.

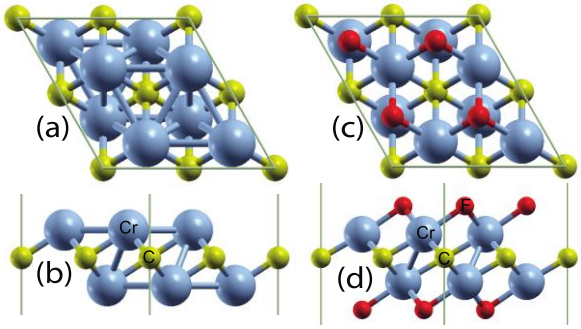

FIG. 1. The top views of 2x2 models of (a) the bare MXene Cr2C and (c) the MXene Cr2C functionalized by F group considered in this study. (b) and (d) show the side views of (a) and (c) structures, respectively.

It is found that in the MXene family, Cr_2C shows interesting magnetic properties upon different functionalization. Additionally, by using first-principles electronic structure calculations, we have studied the structural, magnetic, and elastic properties of Cr_2AX phases with different A (Al, Ge and Ga) and X (C, N and B) elements. In addition to the GGA approach, we have employed the GGA+U formalism to calculate the electronic structures and magnetic properties of Cr_2AX MAX phases to treat electron correlation effects in such multi-components carbides. We have studied the effect of applying Hubbard U interaction onto the Cr 3*d* electrons, on the lattice constants, magnetic ground states, magnetic moments, elastic parameters, and electronic density of states of the Cr_2AX MAX phases in detail. We have shown that it is very vital to consider different initial spin configurations — nonmagnetic (NM), ferromagnetic (FM), and antiferromagnetic (AFM) — to obtain the correct magnetic ground state in both GGA and GGA+U calculations. A schematic picture of all considered collinear AFM magnetic states as

well as NM and FM states in this study have been presented in FIG. 2.

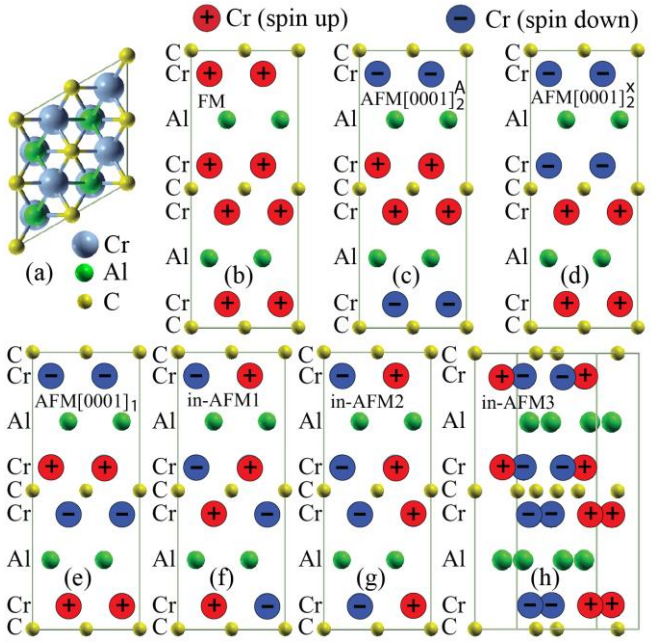

FIG. 2. Schematic representations of seven initial spin configurations considered for Cr_2AlC MAX phase.

It is found that the ground state of all Cr_2AX MAX phases in the level of GGA is in-plane AFM order, except for Cr_2AlB, Cr_2GeB, and Cr_2GaB that is NM. We have compared our computed lattice constants, bulk, shear, and Young modulus in the level of GGA and GGA+U with the corresponding available experimental data, which has been reported for Cr_2AlC, Cr_2GeC, Cr_2GaC, and Cr_2GaN so far. We conclude that the U- value should be less than 1 eV to find a good agreement with the experimental results.

Fiscal Year 2016 List of Publications Resulting from the Use of the supercomputer

[Publication]

Mohammad Khazaei, **Ahmad Ranjbar**, Mahdi Ghorbani-Asl, Masao Arai, Taizo Sasaki, Yunye Liang, and Seiji Yunoki, "Nearly Free Electron States in MXenes" *Phys. Rev. B.* 93, 205125 (2016).

Mohammad Khazaei, **Ahmad Ranjbar**, Masao Arai, and Seiji Yunoki, "Topological insulators in the ordered double transition metals M'$_2$M"C$_2$ MXenes (M'= Mo, W; M" = Ti, Zr, Hf)" *Phys. Rev. B.* 94, 125152 (2016).

Mohsen Babamoradi, Sussan Asgari, **Ahmad Ranjbar**, Rodion V. Belosludov, and Seiji Yunoki, "Many-electron states of the N2 and N3 color centers in diamond: A first- principles and many-body study" *Physica B.* 505, 17 (2016).

[Oral presentation at an international symposium]

Ahmad Ranjbar, Seiji Yunoki, "Structural, electronic and magnetic properties of Cr$_2$AX (A= Al, Ge, Ga; and X= C, N, B) MAX phases" *The 11th General Meeting of ACCMS-VO*, December 19-21, 2016, Sendai, Japan.