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, I have used first-principles electronic structures calculations for studying the structural and magnetic properties of various functionalized MXene Cr_2X (X=C, N) by F, O and OH groups. It is found that in the MXene family, Cr_2C shows interesting magnetic properties upon different functionalization. We have also investigated the electronic, magnetic and structural properties of Cr_2AlX (X=C, N or B) MAX phase, using first-principles density functional theory (DFT) in the framework of GGA and GGA+U approximations. We have shown that the proper treatment for the correlation among *d* electrons of Cr is needed to obtain a good agreement with available experimental data for the lattice parameters, magnetic moments, magnetic order, bulk modulus and electronic density of Cr_2AlX. We have considered nonmagnetic (NM), ferromagnetic (FM) and six anti-ferromagnetic (AFM) configurations. We have shown the importance of studying on a broad range of magnetic states in search for the ground states. We have performed several calculations, with either PBE or PBE+U method. The both PBE and PBE+ U results show that the ground state of Cr_2AlX is AFM. We have concluded that while GGA is not able to reproduce the equilibrium geometry and bulk modulus of Cr_2AlC, GGA+ U with U=1.95 eV and J = 0.95 performs much better.

Fiscal Year 2015 List of Publications Resulting from the Use of the supercomputer

[Publication]

Mohammad Khazaei, Masao Arai, Taizo Sasaki, Ahmad Ranjbar, Yunye Liang, and Seiji Yunoki, "OH-terminated two-dimensional transition metal carbides and nitrides as ultralow work function materials," *Phys. Rev. B.* 92, 075411 (2015).

Hongming Weng, Ahmad Ranjbar, Yunye Liang, Zhida Song, Mohammad Khazaei, Seiji Yunoki, Masao Arai, Yoshiyuki Kawazoe, Zhong Fang, and Xi Dai, "Large-gap two-dimensional topological insulator in oxygen functionalized MXene" *Phys. Rev. B.* 92, 075436 (2015).

[Oral presentation at an international symposium]

Ahmad Ranjbar, "Hydrogen Adsorption on Carbon-Based Materials: Application in Magnetism and Energy Storage" *The 9th General Meeting of ACCMS-VO*, 20th – 22th December 2014, OIST, Okinawa, Japan.

Ahmad Ranjbar, Seiji Yunoki, "GGA and GGA+U modeling for atomic, electronic and magnetic structures of Cr_2AlC" *The 10th General Meeting of ACCMS-VO*, 1st – 3rd November 2015, Sendai, Japan.