Materials Science & Engineering Doctoral Defense

Exploring Pentagonal Geometries for Discovering Novel Two-Dimensional Materials

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Abstract

Single-layer pentagonal materials have received limited attention compared with their counterparts with hexagonal structures. They are two-dimensional (2D) materials with pentagonal structures, that exhibit novel electronic/optical/magnetic properties. There are 15 types of pentagonal tessellations which allow plenty of options for the construction of 2D pentagonal lattices. Few of them have been explored theoretically or experimentally. The study on this new type of materials with density functional theory (DFT) would inspire the discovery of new 2D materials and open up the application of these materials in electronics and magnetic devices. In this dissertation, DFT is applied to discover novel 2D materials with pentagonal structure. Firstly, I examine the possibility of forming a 2D nanosheet with the vertices of type 15 pentagons occupied by boron, silicon, phosphorous, sulfur, gallium, germanium or tin atoms, obtaining different rearranged structures such as gallium with triangular patterns. Then the exploration expands to other 14 types of pentagons, leading to the discovery of carbon nanosheets with Cairo tessellation (type 2 pentagons) and other patterns. They exhibit diverse electrical properties. Then I reveal the hidden Cairo tessellations in the pyrite structures and discover a family of planar 2D materials (such as PtP$_2$), with a chemical formula of AB$_2$ and space group $pa\bar{3}$. The combination of DFT and geometries opens up a novel route for the discovery of new 2D materials. Following this path, a series of 2D pentagonal materials are revealed with electronic/magnetic applications, such as 2D CoS$_2$. The DFT calculations show that CoS$_2$ is an antiferromagnetic semiconductor with a band gap of 2.24 eV, and a Néel temperature of $\sim$20 K. In order to enhance the (super-)exchange interactions between the ions in this binary compound, I explore the ternary 2D pentagonal materials CoAsS, which lacks the inversion symmetry. As expected, CoAsS exhibits higher Curie temperature of 95 K and a sizable piezoelectricity (d$_{11}$=-3.52 pm/V). In addition to CoAsS, 34 ternary 2D pentagonal materials are discovered, among which I focus on FeAsS. FeAsS is a semiconductor showing strong magnetocrystalline anisotropy and sizable Berry curvature. Its magnetocrystalline anisotropy energy (MAE) is 440 $\mathrm{\mu}$eV/Fe ion, higher than other 2D magnets that have been found. Overall, this work provides insights into the structure-property relationship of 2D pentagonal materials and opens up a new route of studying 2D materials by combining computational materials science and geometry. It holds promise for the future applications of 2D pentagonal materials in electronic and magnetic devices.

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