2022 Workshop on Recent Developments in Electronic Structure (ES22), Columbia University

Title: Antiferromagnetic Spin Ordering in Two-dimensional Honeycomb lattice of SiP_3

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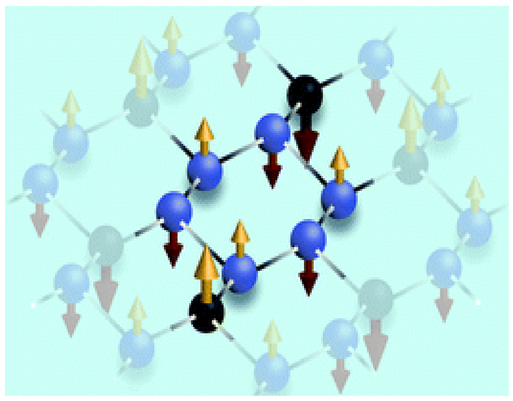
Abstract

Si-doped blue-phosphorene: Layered magnetic material

- According to Mermin-Wagner theorem, magnetic ordering is unlikely in two-dimensional system.
- Using density functional theory, we propose a buckled honecomb lattice of SiP_3 exhibits an antiferromagnetic ground state with itinerant electrons.
- Elemental Si substitution in pristine blue phosphorene to downshift the Fermi energy and induce the Fermi instability that results in a magnetic state.
- Inclusion of electronic correlation and charge transfer from P to Si induces magnetic ordering.



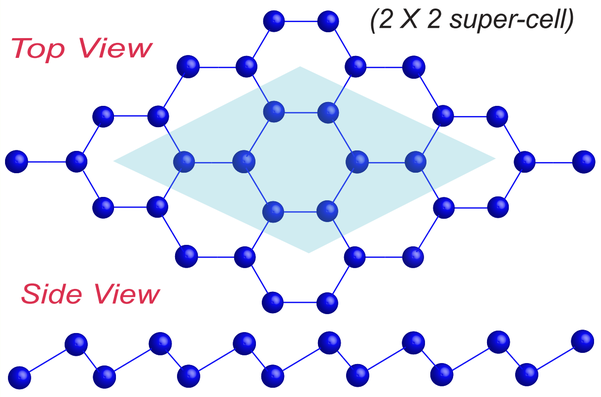
Finding: *Defect or transition metal free magnetism*

Introduction

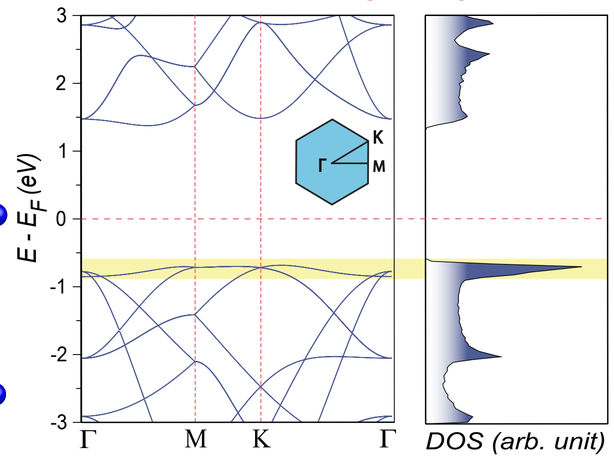
Pristine Blue Phosphorene

Top View *(2 X 2 super-cell)*

Side View



Band structure: *Non-magnetic ground state*



- Widely indirect band gap
- High charge carrier mobility
- Synthesized by molecular beam epitaxy method

Diversing-density of states below the Fermi-level

Hole doping or external electric field

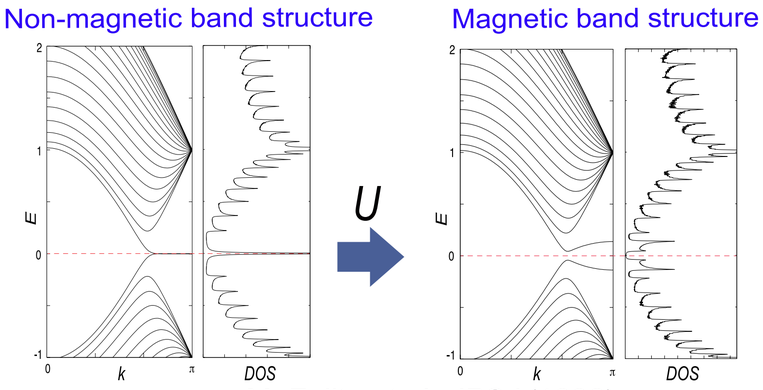
Brings the diverging DOS at Fermi energy and hence induces Fermi instability, that may lead to magnetism

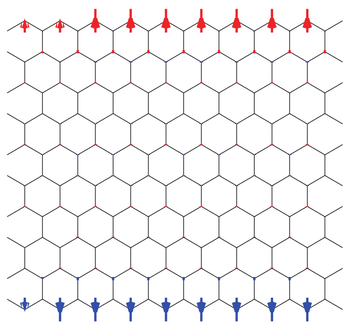
Nano Lett., **16** 4903, 2016

Objective

Zigzag Graphene Nanoribbon (ZGNR, N_z = 20)

Non-magnetic band structure → *U* → Magnetic band structure





Antiferromagnetic coupling between two edges

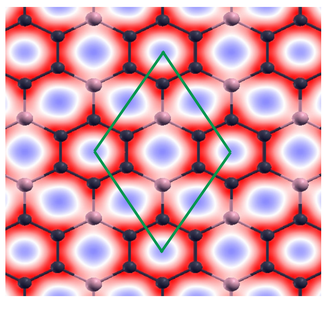
M. Fujita et al. *JPSJ* (1996)
K. Wakabayashi et al. *JPSJ (1998)*

- Diversing-density of states at the Fermi-level (Fermi instability).
- Inclusion of correlation in terms of Hubbard U, open gap and stabilize in a magnetic ground state.
- The magnetic ground state of ZGNR has been experimentally synthesized in room temperature (*Nature*, **514** 608-611, 2014)

Fermi instability induced magnetism in ZGNR

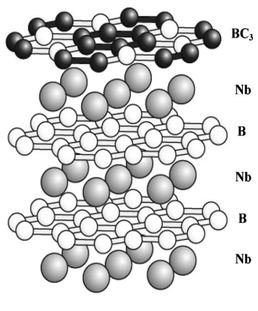
Motivation

2D lattice of BC_3



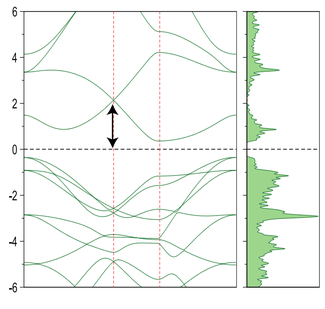
J. Mater Chem., **22** 20881, 2012

Experimental Realization



Surf. Sci., **600** 4072, 2006

Band structure of BC_3



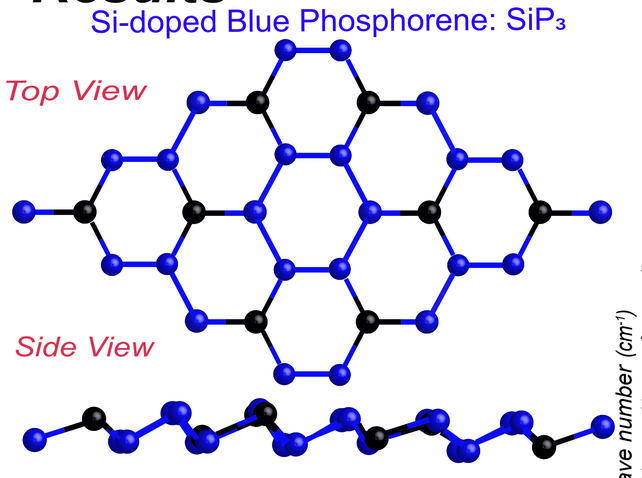
- BC_3, a 2D semiconductor with planer honeycomb structure.
- Carbon substitution in boron honeycomb layer over NbB_2 (0001) surface.
- Boron brings hole in graphene, results in ~ 2 eV Fermi level shift (black arrow) in BC_3.

Results

Si-doped Blue Phosphorene: SiP_3

Top View

Side View



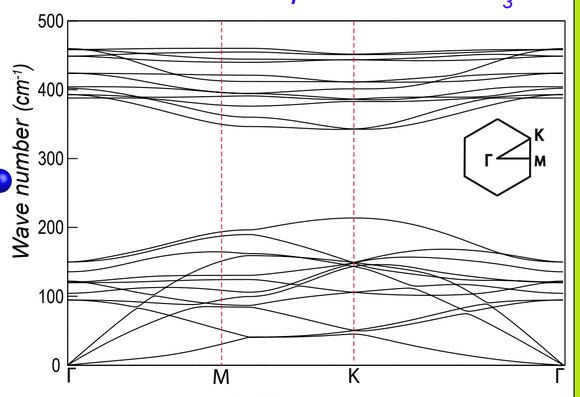
- No imaginary phonon frequency in phonon dispersion indicates stability of SiP_3
- Acoustic and optical modes are well separated

Theoretical Observations

The ground state of SiP_3 stabilizes in anti-ferromagnetic state

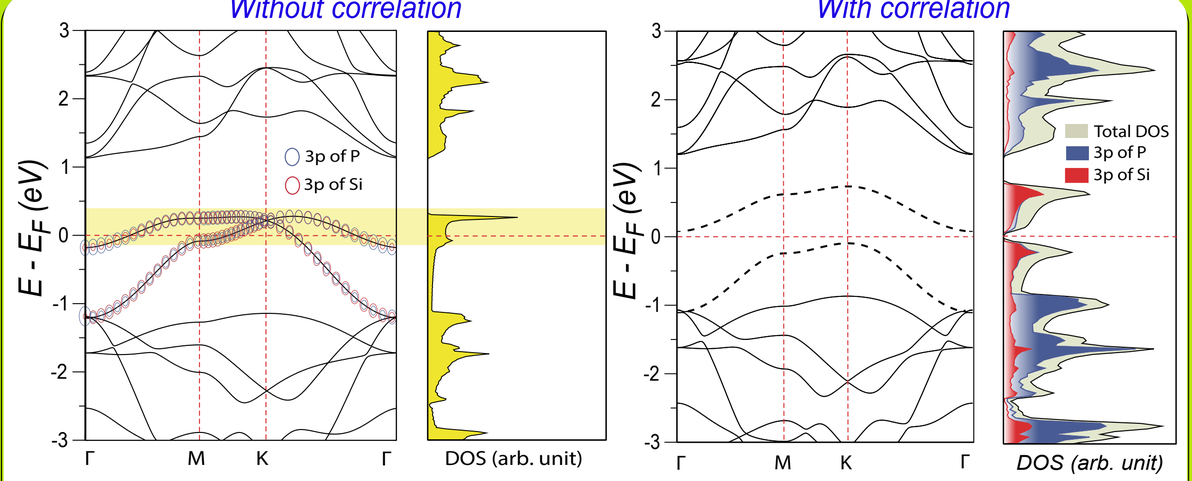
SiP_3 is more stable as compared to blue phosphorene

Phonon dispersion of SiP_3



2D monolayer of SiP_3 that can be obtained experimentally

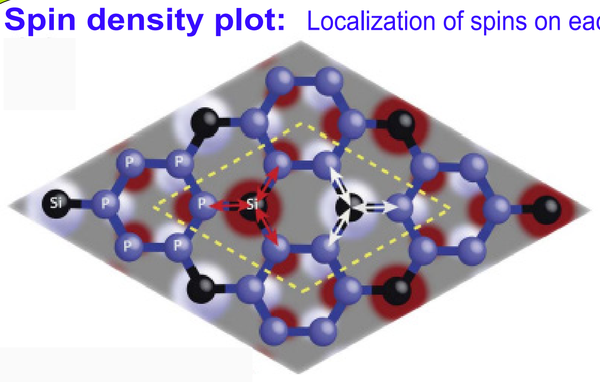
Without correlation *With correlation*



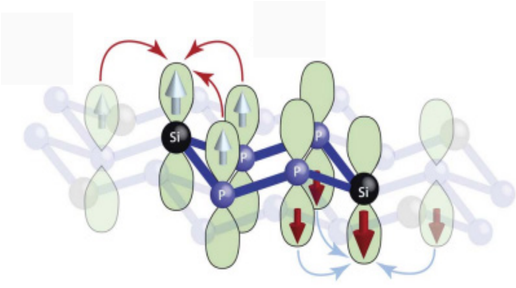
- *Alignment of high DOS near the Fermi-level i.e., Fermi instability.*
- *Two dispersive bands near Fermi energy arise from hybridized 3p orbitals of both P and Si atoms*
- *The bands near Fermi energy get separated, creating a semiconducting gap of 0.172 eV.*

Antiferromagnetic ground stand

Spin density plot: Localization of spins on each atom



- Phosphorous hexagons preserve the bipartite nature of spin alignment
- The dopants (Si) are showing ferromagnetic coupling with the neighboring P atoms
- Overall anti-ferromagnetic ordering



System (element)		$s + p_x + p_y$	p_z spin-up	p_z spin-down	s-character in (%)
Graphene (C)		3.000	0.467	0.467	33.33
Blue-phosphorene (P)		3.466	0.614	0.614	46.10
SiP_3	*(P)*	3.478	0.614	0.573	44.51
	(Si)	2.887	0.242	0.631	46.31

- Charge transfer from P to Si is about 0.04

Charge transfer from P to Si induces magnetic ground state

Further, we have observed, in case of CP_3, GeP_3, SnP_3 (other group IV elments) does not show magnetic grpund state, no charge transfer from P to C, Ge, and Sn

Conclusion

2D antiferromagnetism in monolayer SiP_3 with small semiconducting band gap can be potential applications in spintronics, memory storage and quantum informations

Computational Details

Density Functional Theory as implemented in SIESTA with GGA-PBE exchange and correlation functional, magnetic ground state also verified in VASP

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