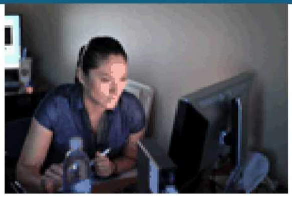
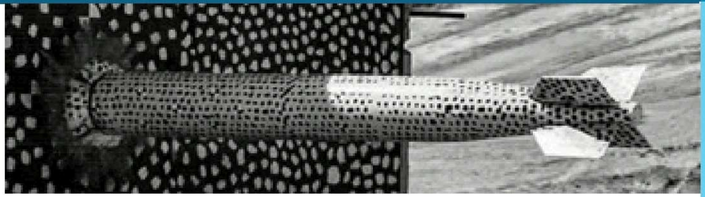
SAND2019-2463C

Identification of Candidate Species for Intercalation Doping of $ZrTe_5$





PRESENTED BY

Normand A. Modine, Department 1864

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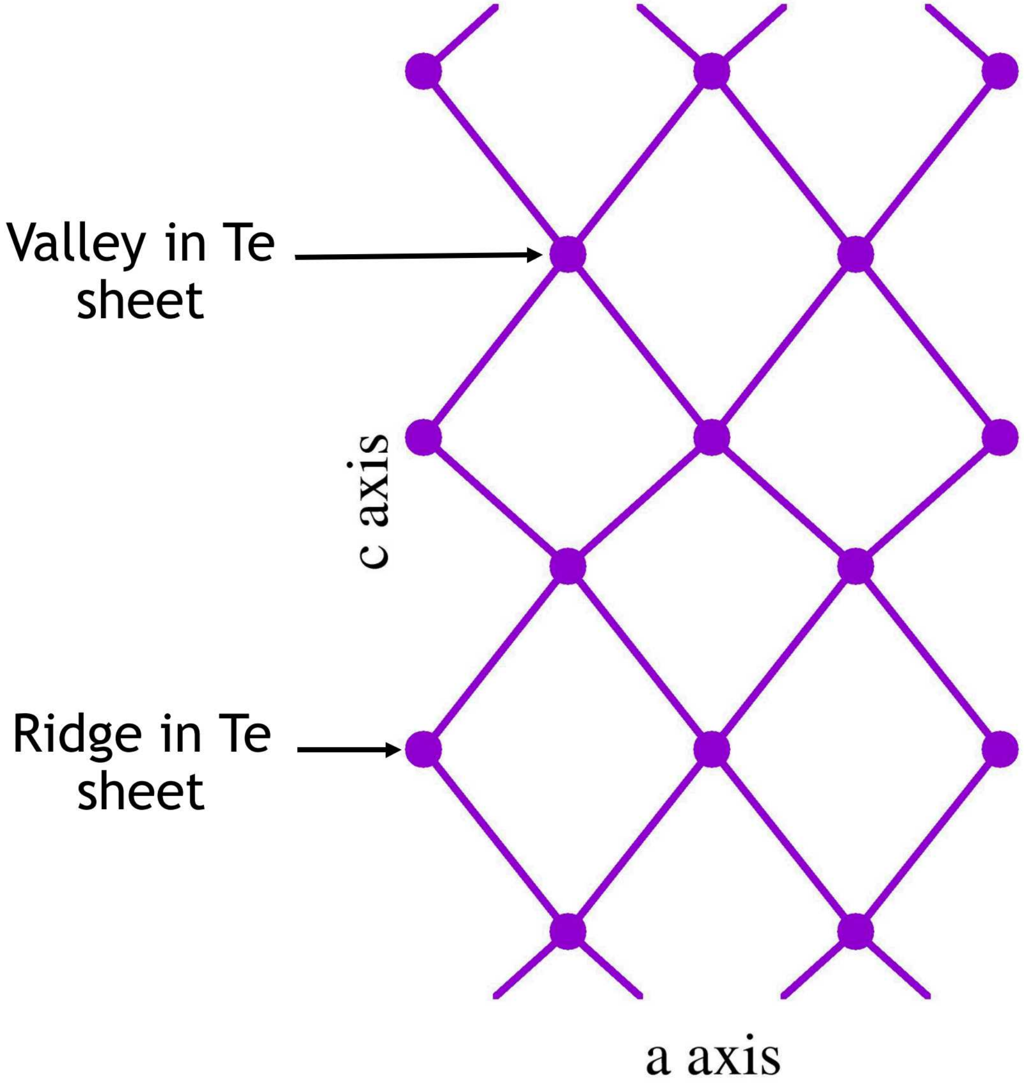
Research Motivation

- $ZrTe_5$ is a very interesting topological material that is reported to be a 3D Dirac semimetal near the boundary between strong and weak topological insulator phases
  - Weng, Dai, and Fang, PRX 4, 11002 (2014)
  - Zheng et al., PRB 93, 115414 (2016)
- It is desirable to be able to control the doping in topological materials in order to adjust the Fermi level relative to the bulk band gap / Dirac points
- $ZrTe_5$ is a layered material with weak interlayer bonding
- One possible approach to controlling the doping is to intercalate a dopant species between the layers
- We wish to identify species that are likely candidates for such “Intercalation Doping” of $ZrTe_5$

Research Techniques

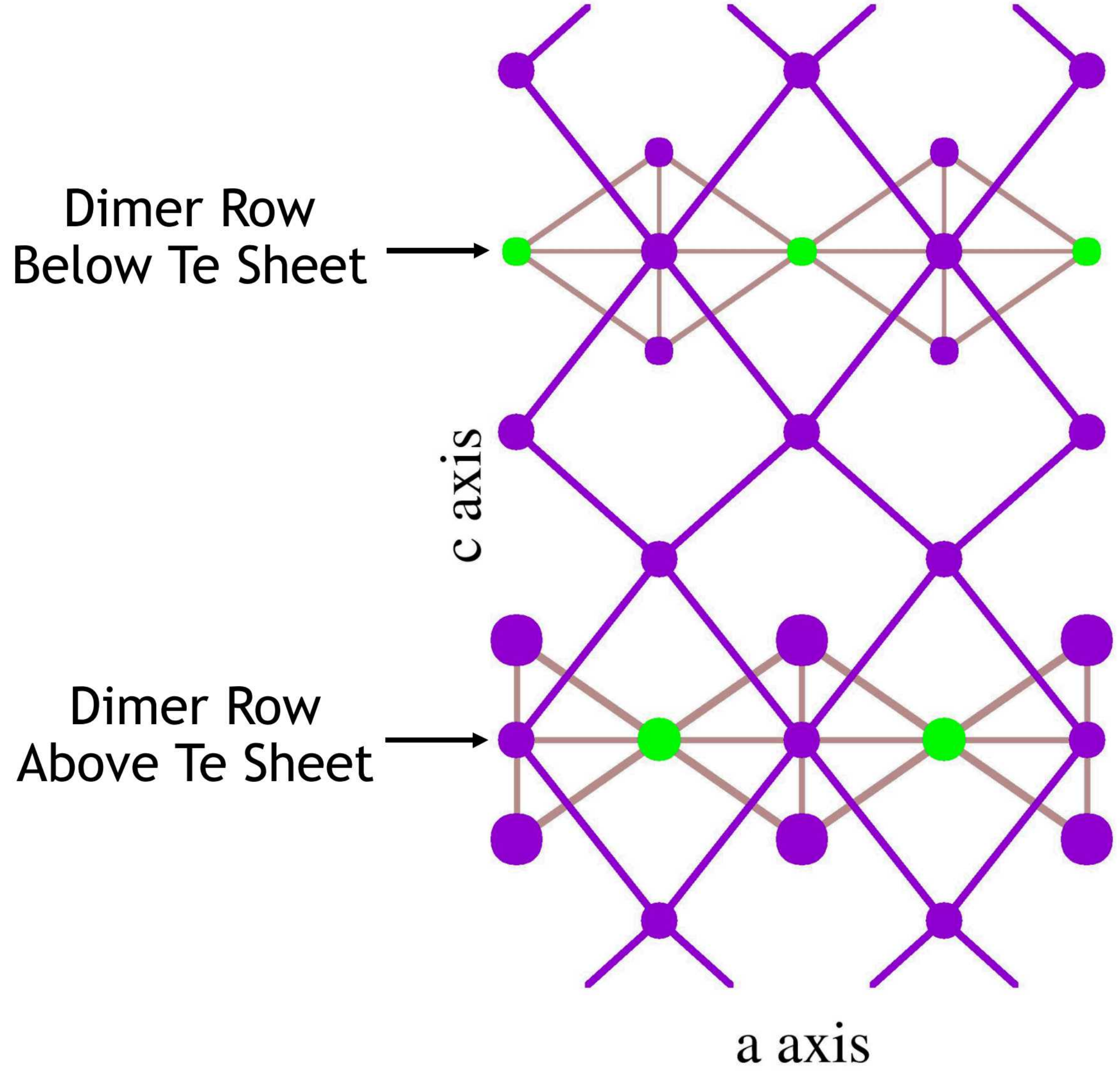
- Density Functional Theory (DFT) using the Vienna Ab initio Simulation Package (VASP)
- The optB86b—vdW exchange-correlation functional was used in order to capture dispersion interactions between the $ZrTe_5$ layers
  - Klimes, Bowler, and Michaelides, J. Phys.: Cond. Matt. 22, 022201 (2010); PRB 83, 195131 (2011)
  - Dion, Rydberg, Schroder, Langreth, and Lundqvist, PRL 92, 246401 (2004)
  - Roman-Perez and Soler, PRL 103, 096102 (2009)
- Projector Augemented Wavefunction (PAW) method
  - $4s^2 4p^6 5s^2 4d^2$ electrons in valence for Zr
  - $5s^2 5p^4$ electrons in valence for Te
  - $2s^1$ electrons in valence for Li
  - $2p^6 3s^1$ electrons in valence for Na
  - $3p^6 4s^1$ electrons in valence for K
  - $5s^1 4d^9$ electrons in valence for Pd

Structure of the Te Sheet in a $ZrTe_5$ Layer



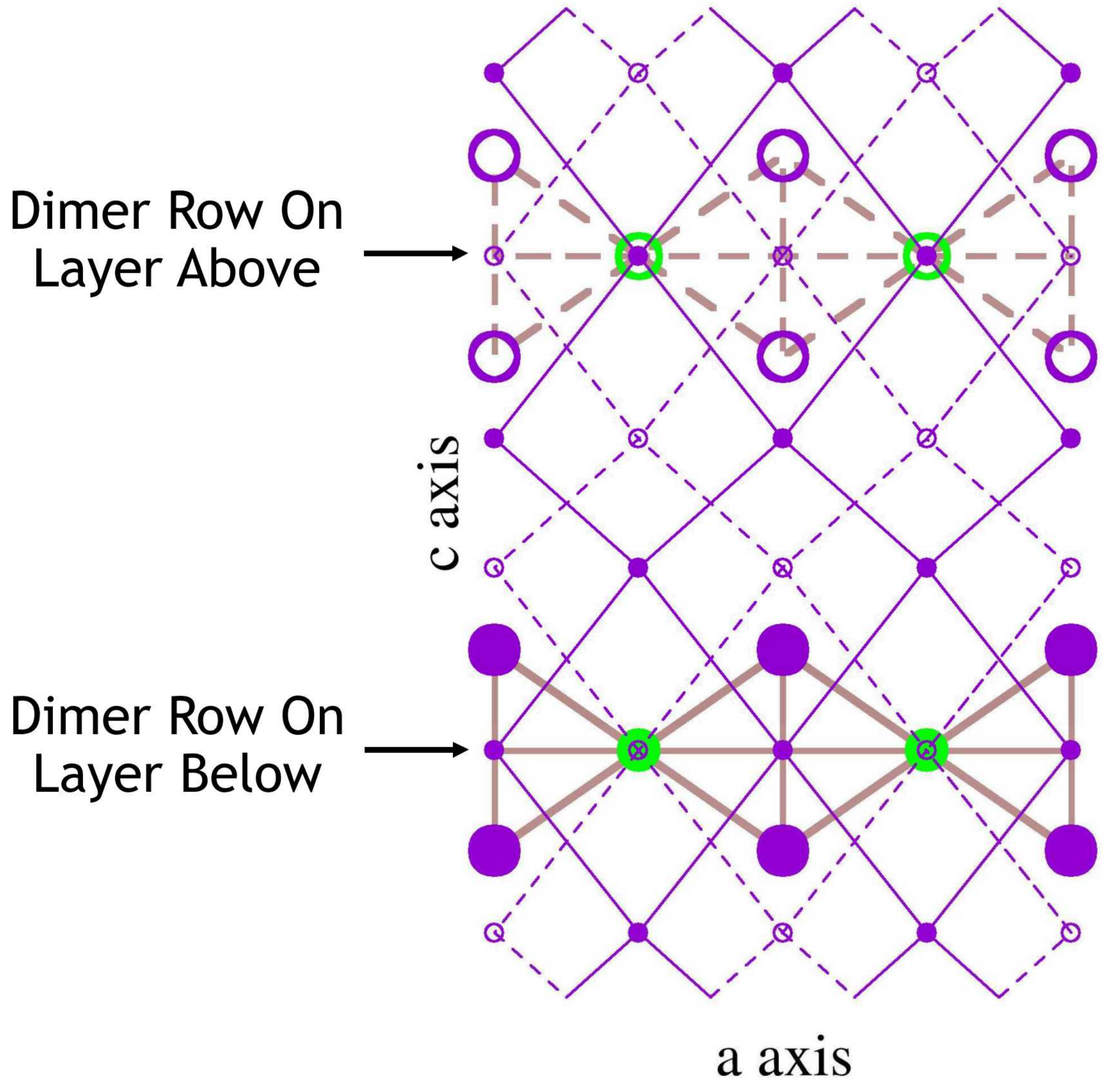
- Circles indicate Zr (green) and Te (purple) atoms.
- Purple lines indicate bonds of corrugated Te sheets.

Structure of a $ZrTe_5$ Layer



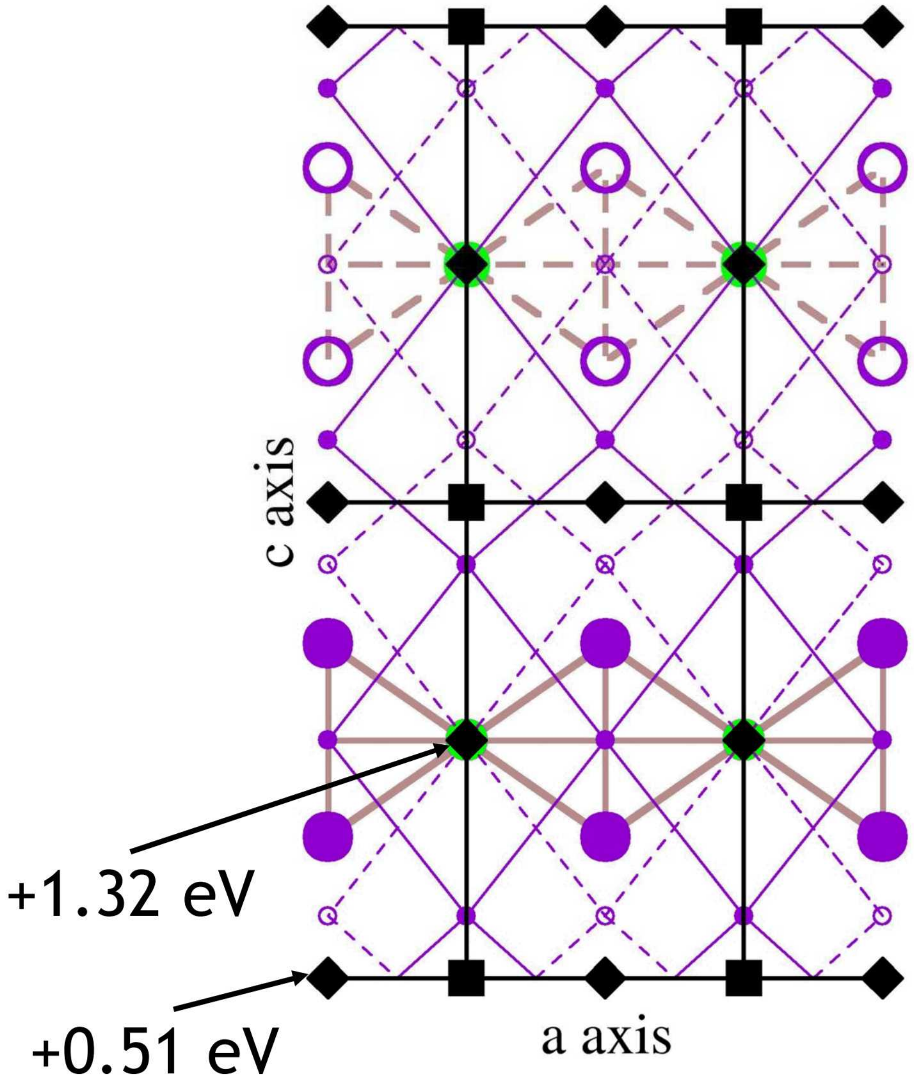
- Circles indicate Zr (green) and Te (purple) atoms.
- Purple lines indicate bonds of corrugated Te sheets.
- Brown lines indicate bonds of $ZrTe_3$–like dimer rows.

Structure of a Interlayer Space in $ZrTe_5$



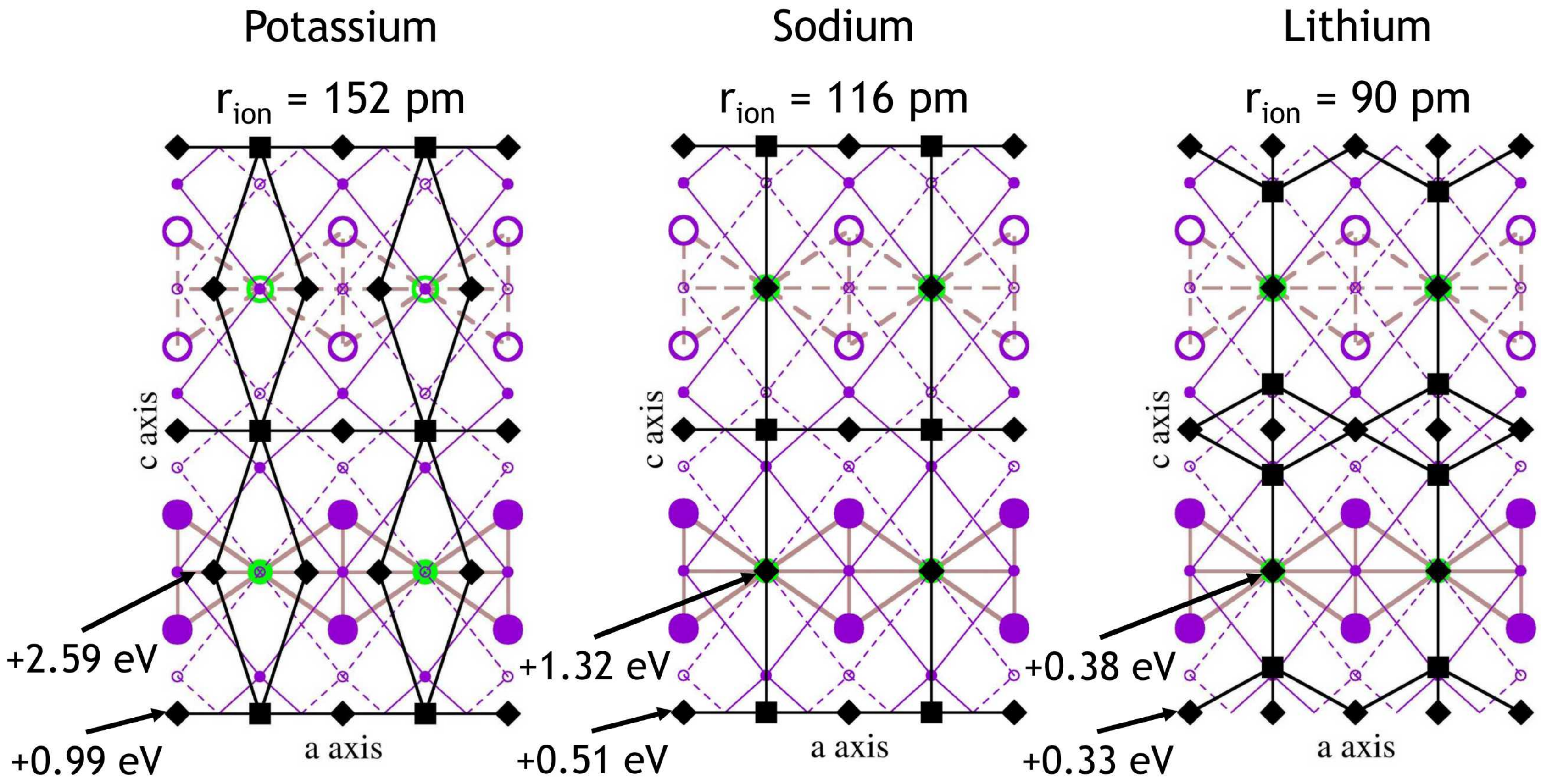
- Circles indicate Zr (green) and Te (purple) atoms.
- Purple lines indicate bonds of corrugated Te sheets.
- Brown lines indicate bonds of $ZrTe_3$–like dimer rows.
- Filled circles and solid lines indicate layer below
- Empty circles and dashed lines indicate the layer above.

DFT Modeling of Sodium Diffusion in $ZrTe_5$



- Circles indicate Zr (green) and Te (purple) atoms.
- Purple lines indicate bonds of corrugated Te sheets.
- Brown lines indicate bonds of $ZrTe_3$–like dimer rows.
- Filled circles and solid lines indicate layer below
- Empty circles and dashed lines indicate the layer above.
- Dopant sites are indicated by squares (stable states) and diamonds (dominant transition states).
- Black lines indicate dopant diffusion network.

DFT Modeling of Alkali (K, Na, Li) Diffusion in $ZrTe_5$



- Unlike Na, the larger K atom squeezes around the Zr atoms during diffusion along the c-axis
- Unlike Na, the smaller Li atom can lower its ground state energy by displacing toward the dimer row on one side or the other

Results for Alkali (K, Na, Li) Absorption and Diffusion in $ZrTe_5$

	Absorption Energy into $ZrTe_5$	Cohesive Energy of Elemental Metal
Potassium	2.67 eV/atom	0.93 eV/atom
Sodium	2.34 eV/atom	1.11 eV/atom
Lithium	2.79 eV/atom	1.63 eV/atom

- Intercalation of K, Na, and Li in $ZrTe_5$ should be energetically favorable
- Room temperature diffusion rates for Li should be reasonable; Intercalation of K and Na should be much slower
- We also obtain a 2.54 eV adsorption energy and 0.28 eV (x-axis) and 0.52 eV (z-axis) diffusion barriers for Li at the $ZrTe_5$ surface

	A-Axis Diffusion Barrier	Estimated A-Axis Room-T Diffusivity	C-Axis Diffusion Barrier	Estimated C-Axis Room-T Diffusivity
Potassium	0.99 eV	1.9×10^{-6} nm^2/s	2.59 eV	1.7×10^{-33} nm^2/s
Sodium	0.51 eV	2.4×10^{2} nm^2/s	1.32 eV	4.9×10^{-12} nm^2/s
Lithium	0.33 eV	2.7×10^{5} nm^2/s	0.38 eV	3.8×10^{4} nm^2/s