Ab Initio Study of Mismatched Layered Superconductors

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Abstract

It has been found that the layered materials BiS + NbS$_2$ and BiSe + NbSe$_2$ superconduct at temperatures lower than 5 K.[3] These materials belong to a larger family of materials characterized by the formula BiX + NbY$_2$ where X= S, Se, Te and Y=S, Se. Here, we present how the superconducting transitions temperature of these six materials is enhanced from monolayer NbS$_2$ and NbSe$_2$ using Density Functional Theory. Using the simple Bardeen-Cooper-Schrieffer Theory as well as the density-of-states at the Fermi level, the superconducting enhancement was calculated. It was found that all six layered materials are projected to have higher superconducting transition temperatures than their monolayer NbS$_2$ or NbSe$_2$ counterpart.

Introduction

Superconductivity is a property of materials that has a wide range of applications, from Maglev trains to medical MRI machines.[1,2] The scientific community is constantly exploring the superconductive properties of different materials in the hopes of one day finding one that is effective at room temperature and room pressure. A new class of materials has been produced in lab and investigated to superconduct at temperatures lower than 5 K.[3] These materials consist of alternating layers of a rocksalt and a transition metal dichalcogenide (TMD.) However, the superconducting critical temperature for these materials is still much lower than room temperature.

Using Density Functional Theory (DFT), materials characterized by the formula BiX + NbY$_2$ where X= S, Se, Te and Y=S, Se, were studied to characterize their superconducting properties. Only materials where X=Y have been produced in a lab and thus the use of DFT allows for the study of layered materials where X≠Y. In this investigation, new values for the superconducting transition temperature, T_c, were estimated using DFT calculations of the density-of-states (DOS) at the Fermi level, $N(0)$. In addition, the material layers of these structures are incommensurate and the impact of this was explored.

Theory

The Bardeen-Cooper-Schrieffer (BCS) Theory is one of the main theories governing superconductivity:

$$k_B T_c = 1.134 \, E_D \, e^{-1/(N(0)V)}, \qquad (1)$$

where, k_B is the Boltzmann constant, T_c is the superconducting transition temperature, E_D is the Debye energy, V is the electron-phonon coupling, and $N(0)$ is the electron density-of-states at the Fermi level.

The goal of this study was to explore trends in the superconducting transition temperatures, T_c, of the six different materials with a focus on the density-of-states (DOS.) Since S, Se, and Te are found within the same group in the periodic table, the values of E_D are not expected to vary much. Due to the nature of the exponent, T_c is very sensitive to changes in $N(0)$ and V. The DOS was chosen as the initial focus of the study because the chemical bonding and thus electron-phonon coupling constants of the materials are expected to be very similar due to them being isovalent materials from the same group of the periodic table and because the density-of-states is initially easier to calculate than V.

The lattice constant of the TMD and rocksalt layers are generally not commensurate. Listed in Table 1 are the relative lattice sizes, known as the 1 + δ numbers (supercell lattice vector ratios) for the 6 materials.

1 + δ	BiS	BiSe	BiTe
NbS	1.109	1.034	0.930
NbSe	1.194	1.110	1.001

Table 1: *1+δ numbers.*

Next, to find the best approximate lattice matches, Farey numbers were used to find the best rational approximations for the 1+δ numbers for cells up to size 20. The ratios used for the various materials appear in Table 2.

1 + δ	BiS	BiSe	BiTe
NbS	10:9	1:1	13:14
NbSe	6:5	10:9	1:1

Table 2: *Farey rational approximations used to construct large unit cells.*

Finally, it is noted that for some cases, the best Farey approximation is actually 1:1. In these cases, the calculations were performed on supercells of size 6:6 to allow for any potential reconstruction.

Results

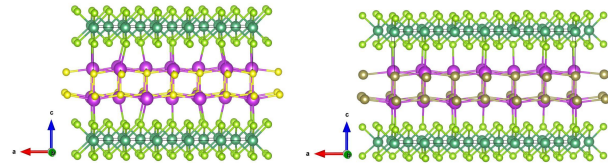Shown below are the first principles calculated structures of two of the materials.

Figure 1: *First principles calculated structures of BiS+NbSe$_2$ (left) and BiTe+NbSe$_2$ (right.)*

The BiS+NbSe$_2$ cell has six rocksalt layers to five TMD layers and the BiTe+NbSe$_2$ cell has 6 rocksalt layers to six TMD layers. The bonds drawn by the software are drawn based upon a preset distance between atoms that highlights the alignment of the layers in the BiS+NbSe$_2$ cell.

After the cells relaxed using DFT and the atomic structures were visualized, band structure calculations were done and band structure diagrams were created for all materials. Shown in Figure 2 below are the band structure diagrams for $BiS+NbSe_2$ and $BiTe+NbSe_2$.
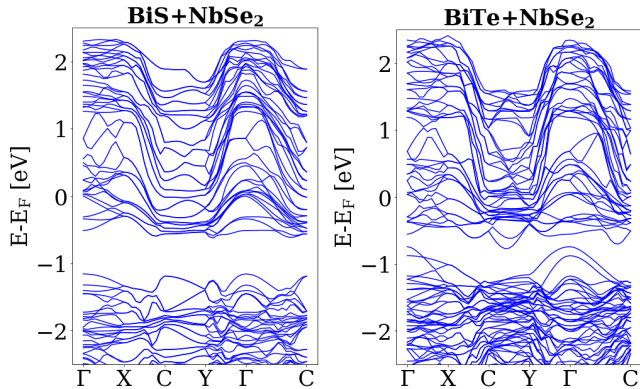

Figure 2: *Band structure diagrams for $BiS+NbSe_2$ (left) and $BiTe+NbSe_2$ (right.)*

We see that, indeed, replacement with atoms from the same column of the periodic table yields quite similar band structures, but with some salient differences. In particular the material with the heavier element (right panel) tends to be more compressed together in energy, particularly near the Fermi level.

Next, density-of-states (DOS) plots were created for all six materials. Displayed in Figure 3 are the DOS plots for $BiS+NbSe_2$ and $BiTe+NbSe_2$. As expected from the relative compression of the electronic bands, the Fermi-level DOS is higher for the compound with the heavier atoms.
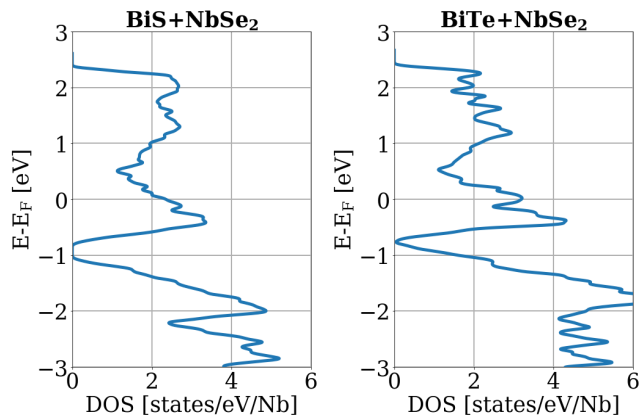

Figure 3: *DOS plots for $BiS+NbSe_2$ (left) and $BiTe+NbSe_2$ (right.)*

The calculations for the DOS at the Fermi level for all six materials are listed in Table 3.

[states/eV/Nb]	BiS	BiSe	BiTe
NbS	2.06	2.53	2.02*
NbSe	2.82	3.04	3.20

Table 3: *DOS at Fermi level (*results for $BiTe+NbS_2$ are preliminary)*

There is a clear trend in the table that the DOS at the Fermi level increases as larger atoms are used in the layered material.

Finally, from these DOS calculations as well as the experimentally calculated T_c values for monolayer NbS_2 and $NbSe_2$, the T_c enhancements can be estimated. The superconducting transition temperatures for monolayer NbS_2 and $NbSe_2$ are 1.75 K and 3.1 K respectively.[4,5] Listed in Table 4 are the factors by which the superconducting temperature of each material would be expected to improve compared to its respective monolayer TMD and the BCS equation (1).

	BiS	BiSe	BiTe
NbS	4.99	9.30	4.62*
NbSe	6.84	8.05	8.88

Table 3: *T_c enhancements as factors of respective monolayer TMDs (*results for $BiTe+NbS_2$ are preliminary)*

Conclusion

Throughout this project, the relaxed structures of six different misfit layered compounds were created using Density Functional Theory. From these relaxed ionic positions, the band structures and ultimately the density-of-states plots were created. It was discovered that the DOS at the Fermi level of these materials generally increases with the size and weight of the elements used to create the material. Finally, it was discovered that, based on the DOS calculations and BCS Theory, $BiTe+NbSe_2$ has the highest projected T_c value for all of the materials in the studied family.

Future work on this project includes fitting wannier bands to the already created band structure diagrams to gain additional accuracy to the DOS calculations. These large cells can also be compared to already calculated smaller 2:2 (rocksalt:TMD) cells to identify the improved accuracy of the Farey number approximation method for the larger cells. Finally, the electron-phonon coupling and Debye energy can be calculated for these materials in order to produce more accurately estimated T_c values.

Acknowledgements

References

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