

Naturally occurring heterostructures in Van der Waals compounds

Mini project

By

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INTRODUCTION

Heterostructure materials represent a captivating avenue in materials science and semiconductor physics. Their ability to combine different materials with complementary properties opens up a wide array of possibilities for designing devices with tailored functionalities. The emergence of 2D materials, particularly graphene [1] and transition metal dichalcogenides (TMDs), has provided an exciting platform for constructing heterostructures due to their unique electronic and mechanical properties. [2]

The utilization of van der Waals (VdW) forces for bonding in heterostructures has enabled the assembly of atomically thin layers, allowing for precise control over the interface between materials. This method not only facilitates the creation of heterostructures with atomically sharp interfaces but also offers flexibility in stacking different materials with minimal lattice mismatch, reducing defects and strain within the structure.

The ability to control the twist angle between adjacent layers is a particularly intriguing aspect of heterostructure design. This twist engineering can dramatically alter the electronic properties of the system, leading to the emergence of unconventional phenomena such as moiré patterns, which can induce significant changes in the material's electronic structure and transport properties. Furthermore, the manipulation of twist angles has been demonstrated to drive transitions between different electronic phases, including superconductivity and correlated insulating states, providing a powerful tool for exploring novel quantum phenomena. [3]

Transition metal halides (TMHs) have also emerged as promising building blocks for heterostructures due to their tunability, electronic and magnetic properties. By combining TMHs with other materials, researchers can create heterostructures with unique functionalities not present in the individual components alone. Additionally, the ability to manipulate the composition and stacking sequence of TMH-based heterostructures offers further opportunities for tailoring their properties for specific applications, ranging from optoelectronics to quantum information processing.

Overall, the field of heterostructure materials continues to evolve rapidly, driven by advances in both experimental techniques and theoretical understanding. As researchers continue to explore the vast design space afforded by heterostructures, we can expect further breakthroughs in areas such as quantum computing, photonics, and energy harvesting, ultimately paving the way for next-generation semiconductor technologies.

What is misfit?

Misfit layer structures involve at least two somewhat independent layers, where one or two intralayer basic periodicities do not align. Over the last decade, there has been a rapid growth in the number of compounds exhibiting misfit layer structures. Initially discovered in natural minerals, significant examples were reviewed by Mako Vicky and Hyde. The term "misfit layer structure" was likely coined by Makovicky.[4]

A broader range of synthetic composite compounds with such structures has emerged, particularly in chalcogenide chemistry. These compounds consist of alternating layers, label as $A = MX$ and $B = TX_2$,

creating a composite structure defined by the formula $(MX)_{1+x}(TX_2)_m$, where $M = \text{Sn, Pb, Bi, Sb, rare earths}$; $T = \text{Ti, V, Cr, Nb, Ta}$; $X = \text{S, Se}$; $0.08 < x < 0.28$; and $m = 1-3$. Each set (A, B) has its own symmetry and unit cell parameters. Incommensurate behaviour arises from non-coinciding periodicities between the two sets, particularly along one direction. [5]

The compounds discussed in the review exhibit incommensurability along the crystallographic a direction, indicating that the ratio of a_A to a_B yields an irrational value. Periodicities along the b and c directions are either identical or multiples of each other, with c chosen as the stacking direction.

In these compounds, the MX part comprises a two-atom thick layer with a distorted NaCl structural type, while the TX_2 part forms a three-atom thick sandwich with T in either an octahedral (Ti, V, Cr) or trigonal prismatic (Nb, Ta) coordination with respect to the chalcogen atoms (X).

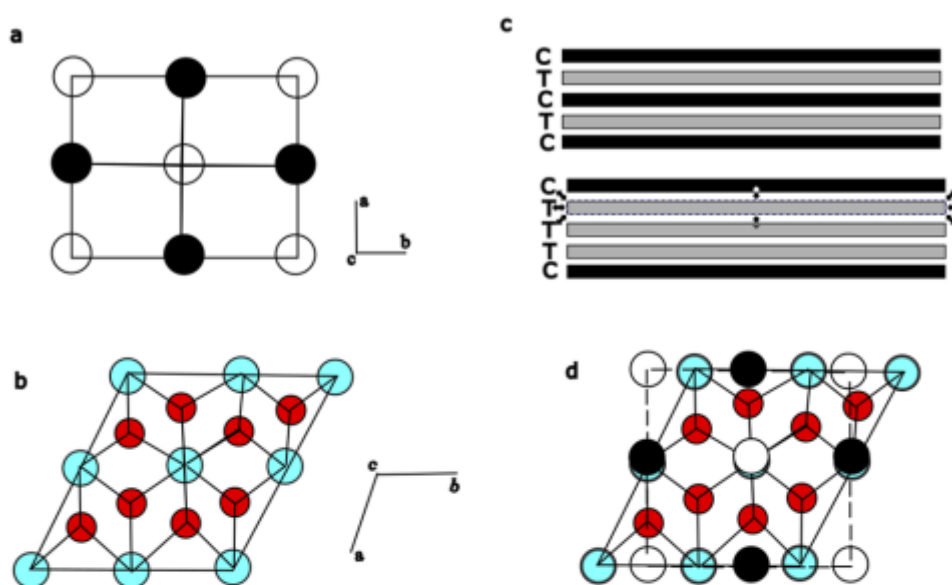


FIG. 1. (a) and (b) Illustration of the two generic components of a misfit layered compound. On top is the cubic rock salt sublattice and at the bottom is the trigonal TMDC sublattice. Colors are chosen arbitrarily to distinguish atoms for connectivity. The trigonal sublattice is oriented with the c axis directed out of the page. (c) Illustration of two different types of stacking order. C = cubic layer, T = trigonal layer. Other stacking orders exist, such as those having two or three cubic rock salt layers followed by a single trigonal layer. The layer stacking orders can be described similarly to the typical scheme used in polymers or "classical" layered materials such as graphite and MoS_2 . (d) Illustration of the stacked structure of a misfit layered compound. The cubic layer is overlaid on the trigonal layer, and the black and white spheres are atoms from this cubic layer.

The composition of the compound, determined by the ratio of MX to $[TX_2]$, is expressed by the variable x , derived from $x = (4/2)(a_2/a_1) - 1$; the factor $4/2$ accounts for the number of formula units per slab (4 for MX, 2 for TX_2). The m value ($m = 1, 2, 3, \dots$) represents the alternating type sequence, indicating the ratio between the number of TX_2 and MX entities along the stacking direction.

In misfit layer structures, one layer typically adopts a cubic rock salt-type structure, akin to materials like NaCl. The other layer typically exhibits a hexagonal or trigonal structure, similar to transition metal dichalcogenides such as NbSe₂. In epitaxial scenarios, the *c*-axis of the trigonal layer aligns with the (111) axis of the cubic structure due to their shared trigonal symmetry. However, in naturally occurring misfits, the *c*-axis of the trigonal unit and a principal (100) axis of the cubic structure align. Consequently, the coaxial fourfold and threefold rotational symmetries become incompatible, leading to an incommensurate matching of the two layers along at least one axis.

This mismatch means that the cubic (C) and trigonal (T) layers experience a misfit along one (or both) in-plane directions. Various stacking sequences along the layering direction are possible, such as the commonly observed patterns like ...-C-T-C-T-C-... and ...-C-T-T-T-C-T-T-T-C-.... These combinations result in significant structural complexity. Despite the dissimilarity between the two lattices, misfit layered compounds can still be thermodynamically stabilized and grown in single crystal form, which is quite unusual.

The recently discovered naturally occurred Heterostructure misfit compound Ba₆Nb₁₁Se₂₈ it discovered in recently in 2022.[6]

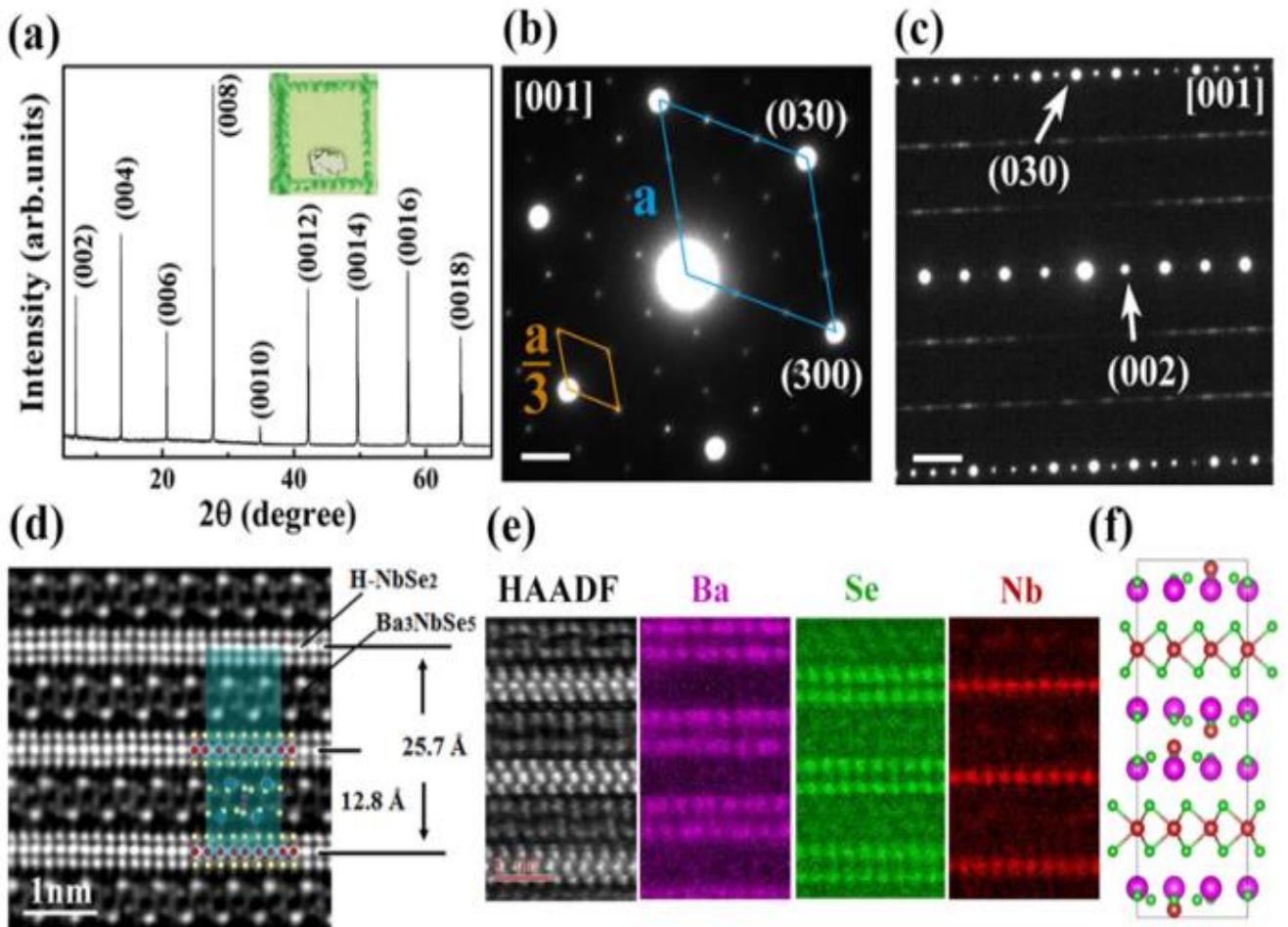


FIG. 2 (a) XRD pattern of Ba₆Nb₁₁Se₂₈ single crystal shows a series of (00*l*) diffraction peaks. The inset shows the single crystal; the green grid is 5 mm × 5 mm inside. Electron diffraction images taken looking along (b) the [001] axis and (c) [100] axis. The scale bar in both panels is 1 nm⁻¹. (d) HAADF-STEM image of Ba₆Nb₁₁Se₂₈ taken along the [110] axis (scale bar, 1 nm). A simulation of the model structure is overlaid with one unit cell shaded in blue. Ba, Nb, and Se atoms are depicted as blue, red, and yellow circles, respectively. (e) The EELS mapping of Ba₆Nb₁₁Se₂₈ (scale bar, 1 nm). (f) Crystal structure of Ba₆Nb₁₁Se₂₈ along [100].

Natural superlattice Ba₆Nb₁₁S₂₈

As outlined above, Ba₆Nb₁₁S₂₈ exhibits a superlattice structure with a 2D superconducting phase originating from the layers of TMD 2H-NbS₂. The anisotropic nature of the superconductivity arises due to the layered configuration of the crystals. The long mean free path relative to the coherence length of quasiparticles suggests the cleanliness of the 2D superconducting state. Modelling the Fermi surface as that of 2H-NbS₂ with zone-folding from the 3×3 superstructure of the Ba₃NbS₅ layer is supported by transmission electron microscopy (TEM) observations and quantum oscillations measured in transport experiments. The presence of quantum oscillations also indicates a high mobility exceeding that of bulk 2H-NbS₂. [7]

The superconductivity in Ba₆Nb₁₁S₂₈ surpasses the Pauli limit in the upper critical field, H_{c2} , suggesting evidence for a Fulda-Ferrell-Larkin-Ovchinnikov (FFLO) phase. Torque magnetometry reveals a sharp decrease in diamagnetism near H_{c2} , indicating the presence of this phase, which entails a spatially modulated superconducting state where Cooper pairs possess nonzero momentum.

The FFLO state comprises Cooper pairs with momentum $\Delta \propto \exp(i\mathbf{Q} \cdot \mathbf{R})$, while the LO state consists of Cooper pairs with $\Delta \propto \cos(\mathbf{Q} \cdot \mathbf{R})$. The crystals of Ba₆Nb₁₁S₂₈ can be exfoliated, presenting an opportunity to investigate these phenomena as a function of carrier doping.

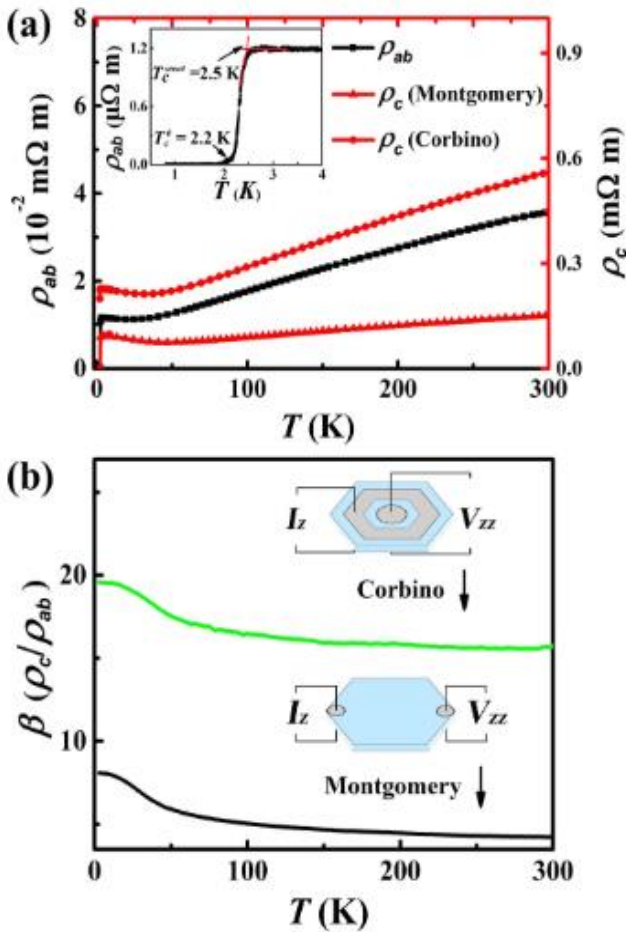


Fig. 3. (a) Temperature-dependent in-plane (black) and out-of-plane resistivity of Ba₆Nb₁₁Se₂₈ which were taken using the Montgomery (red) and Corbino (green) method. (b) The resistivity anisotropy $\beta = \rho_c/\rho_{ab}$.

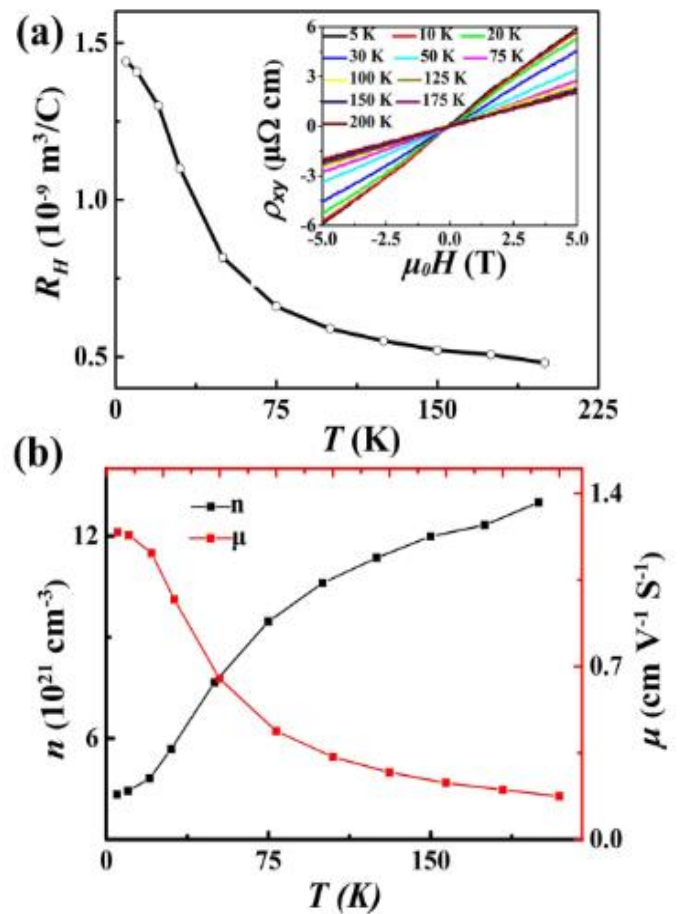


Fig. 4. (a) Temperature dependence of Hall coefficient R_H determined from linear fitting $\rho_{xy}(H)$ data. The inset is Hall resistivity vs magnetic field at various temperatures, and the data above 5 K are obtained in PPMS system up to 5 T. (b) Temperature dependence of carrier concentration (black) and mobility (red).

Moreover, the discovery of a natural, commensurate crystal with encapsulated TMD layers suggests the possibility of other variants. By altering the chemical composition of the constituent TMD, it may be feasible to explore the monolayer electronic properties of other TMDs within a natural bulk crystal. Additionally, by controlling the composition of the spacer, one can tune the properties of the TMD through interfacial interactions. Subsequent chapters will delve into the superlattices synthesized from these natural TMDs and their relationship to the materials discussed here.

Figure 3(a) depicts the temperature variation of the in-plane resistivity, denoted as $\rho_{ab}(T)$, which was measured under zero magnetic field conditions.

$(\text{LaSe})_{1.27}(\text{SnSe}_2)_2$ is a misfit compound composed of alternating layers of lanthanum selenide (LaSe) and tin selenide (SnSe_2). Misfit compounds are characterized by the arrangement of two distinct crystalline structures, leading to a unique blend of properties. [8]

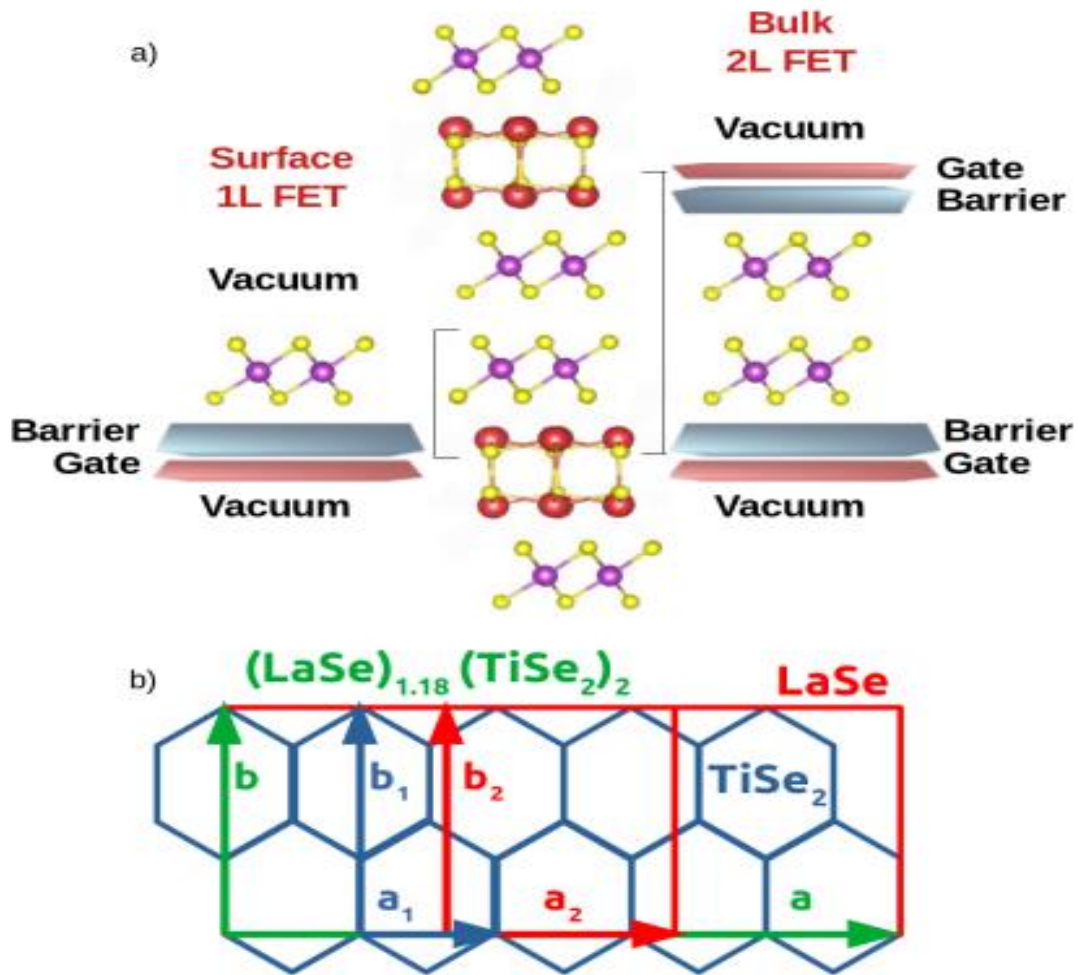


Figure. 5. a) Bulk structure of $(\text{LaSe})_{1.18}(\text{TiSe}_2)_2$. The field-effect modeling scheme is depicted for the case of the most common TiSe_2 terminated surface (left) and for bulk TiSe_2 (right). b) Sketch of the unit cell of $(\text{LaSe})_{1.18}(\text{TiSe}_2)_2$ misfit layer compound (\mathbf{a} , \mathbf{b}) compared with the ones of a TiSe_2 monolayer (\mathbf{a}_1 , \mathbf{b}_1) and of a LaSe unit (\mathbf{a}_2 , \mathbf{b}_2).

In this compound, the ratio $(\text{LaSe})_{1.27}(\text{SnSe}_2)_2$ indicates that there are approximately 1.27 layers of lanthanum selenide for every two layers of tin selenide. This non-stoichiometric composition implies a deviation from a simple integer ratio between the two constituent materials.

The properties of misfit compounds like $(\text{LaSe})_{1.27}(\text{SnSe}_2)_2$ can vary considerably depending on factors such as the exact composition, layer thickness, and crystal structure. These materials often showcase intriguing electronic, magnetic, and optical characteristics, rendering them appealing for diverse applications, including electronics, thermoelectric devices, and catalysis.

COMPOUNDS AND THEIR PROPERTIES

$(\text{Bi}_2\text{Ba}_{1.8}\text{Co}_{0.2}\text{O}_4)(\text{CoO}_2)_2$

Properties- The presence of cobalt suggests interesting magnetic properties.

$\text{Ca}_{25}\text{Co}_{22}\text{O}_{56}(\text{OH})_{28}$

The combination of calcium, cobalt, and oxygen suggests interesting thermoelectric properties.

$(\text{Ca}_2\text{CoO}_3)_{0.62}(\text{CoO}_2)$

Known as a member of the thermoelectric oxide family with high thermoelectric performance.

$(\text{Bi}_{2.08}\text{Sr}_{1.67}\text{O}_x)_{0.54}(\text{CoO}_2)$

Incorporates bismuth and strontium, suggesting potential for superconductivity or other interesting electronic properties.

$(\text{Bi}_{1.94}\text{Ba}_{1.83}\text{O}_y)_{0.56}(\text{RhO}_2)$

Includes bismuth, barium, and rhodium, indicating potential for unique electronic and magnetic properties.

Tellurium-based

$((\text{PbTe})_{1.17})_m(\text{TiTe}_2)_n, m:n = 1:1, 1:3, 1:5, 5:1, 3:1, 1:2$

Titanium-containing compounds often exhibit interesting magnetic behaviours.

Selenium-based

$(\text{LaSe})_{1.14}(\text{NbSe}_2)$

The combination of lanthanum selenide and niobium diselenide may lead to unique electronic and magnetic properties.

$(\text{LaSe})_{1.14}(\text{NbSe}_2)_2$

The 1:2 ratio suggests a different stoichiometry compared to the 1:1 ratio, which may result in altered structural and electronic properties.

$(\text{Pb}_{1-x}\text{Sn}_x\text{Se}_2)_{1.16}(\text{TiSe}_2)_2,$

Involves lead-tin selenide and titanium selenide, suggesting potential for interesting electronic and thermal properties.

(SnSe)_{1.16}(NbSe₂)

The ferecrystalline compound is 1.6 times more conductive than [the misfit layer compound](#).

(SnSe)_{1.18}(TaSe₂)

The combination of tin selenide and tantalum di-selenide may result in interesting electronic, thermal, or magnetic properties.

(SnSe)_{1.17}(TaSe₂)

Physical vapor deposition of tin selenide and tantalum di-selenide in a 1:1 ratio may yield thin films with tailored properties, potentially useful for electronic or optoelectronic applications.

(PbSe)_{1.14}(NbSe₂)

Lead selenide (PbSe) is known for its high thermoelectric performance. Combining it with niobium di-selenide (NbSe₂) may result in enhanced or unique thermoelectric properties.

Physical vapor deposition (PVD) often allows for the growth of thin films. This compound synthesized using PVD may exhibit unique structural and electronic characteristics due to the thin film nature.

(BiSe)_{1.15}(TiSe₂)

Bismuth selenide (BiSe) is another material known for its thermoelectric properties. The combination with titanium selenide (TiSe₂) may result in enhanced thermoelectric performance.

Cu_x(BiSe)_{1.15}(TiSe₂)₂

The addition of copper may introduce interesting electronic, magnetic, or structural properties. Copper-containing compounds often exhibit diverse behaviours.

(BiSe)_{1+x}(TiSe₂)_n, n = 2–4

The ability to vary the ratio (1:n) and the use of physical vapor deposition provide tunability, allowing researchers to explore a range of compositions and properties.

“MTX₃” (M = RE + Bi, T = Ti, V, Nb, Ta, X = Se)

The combination of rare earth elements, bismuth, transition metals, and selenium suggests potential for diverse electronic, magnetic, or structural properties. The compound may exhibit multifunctionality.

“NbBiSe₃” = (BiSe)_{1+δ}(NbSe₂)

Potentially leading to interesting electronic and structural characteristics due to variations in charge density.

$((\text{PbSe})_{1.14})_m(\text{NbSe}_2)$, $m = 1-6$

Varying the ratio $m:1$ allows for the **tuning** of electronic and structural properties.

Niobium di-selenide (NbSe_2) is a known superconductor, and the compound may exhibit interesting superconducting behaviours.

Compounds and their properties

compound	magnetic	Growth	electronic	superconductivity	thermoelectric	link
Oxygen based						
$(\text{Bi}_2\text{Ba}_{1.8}\text{Co}_{0.2}\text{O}_4)(\text{CoO}_2)_2$	yes(antiferromagnetic)	solid state reaction	yes(metallic)	N/A	yes	yes
$\text{Ca}_{25}\text{Co}_{22}\text{O}_{56}(\text{OH})_{28}$	yes(The compound shows Curie-Weiss paramagnetism with an antiferromagnetic)	High pressure solid state reaction	N/A	N/A	YES	yes
$(\text{Ca}_2\text{CoO}_3)_{0.62}(\text{CoO}_2)$	N/A	Hydrothermal , FLUX	N/A	N/A	yes	yes
$(\text{Bi}_{2.08}\text{Sr}_{1.67}\text{O}_x)_{0.54}(\text{CoO}_2)$	The broadened profiles can be decomposed into the ferromagnetic and superparamagnetic components	SSR	yes	yes	N/A	yes
$(\text{Bi}_{1.94}\text{Ba}_{1.83}\text{O}_y)_{0.56}(\text{RhO}_2)$	yes	SSR	yes(semiconductor)	N/A	N/A	Yes
Tellurium-based						
$((\text{PbTe})_{1.17})_m(\text{TiTe}_2)_n$, $m:n = 1:1, 1:3, 1:5, 5:1, 3:1, 1:2$	yes	physical vapour transport	yes	N/A	yes	Yes
Selenium-based						
$(\text{LaSe})_{1.14}(\text{NbSe}_2)$	N/A	solid state reaction	yes	yes	N/A	Yes
$(\text{LaSe})_{1.14}(\text{NbSe}_2)_2$	N/A	solid state reaction	yes	N/A	N/A	

(Pb_{1-x}Sn_xSe₂)_{1.16}(TiSe₂)₂,	N/A	solid state reaction/Chemical vapour transport	yes(insulator)	yes	N/A	Yes
(SnSe)_{1.16}(NbSe₂)	N/A	solid state reaction	yes(metallic)	yes	N/A	Yes
((SnSe)_{1+x})_m(NbSe₂)_n m = n = 1–20	N/A	physical vapour decomposition	N/A	N/A	N/A	
(SnSe)_{1.18}(TaSe₂)	yes	solid state reaction	yes	yes	yes	Yes
(SnSe)_{1.17}(TaSe₂)	N/A	solid state reaction	N/A	N/A	N/A	Yes
(PbSe)_{1.14}(NbSe₂)	N/A	solid state reaction/Physical vapour decomposition	yes	N/A	Yes	Yes link
(BiSe)_{1.15}(TiSe₂)	diamagnetic	chemical vapour transport	yes(metallic)	N/A	yes	Yes
Cu_x(BiSe)_{1.15}(TiSe₂)₂	yes	chemical vapour transport	yes	N/A	N/A	
(BiSe)_{1+x}(TiSe₂)_n, n = 2–4	N/A	physical vapour decomposition	yes(metallic)	N/A	N/A	Yes
“MTX₃” (M = RE + Bi, T = Ti, V, Nb, Ta, X = Se)	yes	chemical vapour transport/CsCl/KCl flux	yes	N/A	N/A	
“NbBiSe₃” = (BiSe)_{1+δ}(NbSe₂)	N/A	physical vapour decomposition	yes	yes	N/A	Yes
((PbSe)_{1.14})_m(NbSe₂)_m, m = 1–6	N/A	physical vapour decomposition	yes	yes	N/A	Yes
sulfur-based	layer ratio					
“LaNbS₃” = (LaS)_{1.14}(NbS₂)	01:01	CVT	yes	yes	N/A	
“LaCrS₃” = (LaS)_{1+x}(CrS₂)	01:01	CVT	yes	N/A	N/A	
(LaS)_{1.18}(VS₂)	01:01	CVT	N/A	N/A	N/A	

(YbS)1.25(CrS2)	01:01	CVT	N/A	yes	N/A	
(PbS)1.18(TiS2)	01:01	CVT	N/A	N/A	N/A	
(PbS)1.12(VS2)	01:01	SSR CVT	N/A	N/A	yes	
(SmS)1.19(TaS2)	01:01	CVT	N/A	N/A	N/A	
(SnS)1.15(TaS2)	01:01	CVT	N/A	yes	N/A	
((Nb _{1-y-z} Pb _y BizS)1.5) _{1+x} (NbS ₂)	01:01	CVT	N/A	N/A	N/A	
(PbS)1.13(TaS2)	01:01	SSR CVT	yes	N/A	N/A	
(PbS) _{1+x} (NbS ₂) _n	1:n	SSR CVT	N/A	N/A	N/A	
(EuS)1.15(NbS2)	01:01	SSR	N/A	yes	N/A	
“SnTiS3” = (SnS)1.2(TiS2)	01:01	SSR CVT	yes	N/A	N/A	
(CeS)1.15(TaS2)	01:01	SSR/CVT	N/A	N/A	yes	
“BiTi2S5” = (BiSe)(TiSe2) ₂	01:01	SSR	yes	N/A	N/A	
“LnMS3” = (LnS) _{1+δ} (MS ₂)	01:01	SSR/CVT	N/A	yes	N/A	

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More about some definition

Superconductivity: Superconductivity is the ability of certain materials to carry direct current (DC) electricity without any loss of energy when they are cooled below a critical temperature (referred to as T_c). These materials also exhibit the phenomenon of expelling magnetic fields as they undergo the transition into the superconducting state.

Magnetic properties: The magnetic characteristic of a material arises from the atomic or subatomic reaction to an applied magnetic field. This response is a consequence of electron spin and charge interactions, leading to the formation of a dipole moment ^{*}.

Air stability: Chemical stability encompasses the likelihood of alterations occurring in the composition of a substance due to various chemical reactions. These reactions, which include oxidation, polymerization, and hydrolysis, have the potential to induce significant changes in the chemical structure and properties of the substance. Factors such as environmental conditions, presence of catalysts, and the nature of the substance itself can influence its susceptibility to these reactions and thus impact its overall stability ^{*}.

Thermoelectric: Thermoelectric (TE) materials possess the ability to transform heat into electricity, offering opportunities to enhance fuel efficiency and establish a reliable alternative energy source across diverse applications. By harnessing wasted heat, these materials play a pivotal role in advancing energy solutions. Constructing high-performance TE devices necessitates targeting superior TE materials through a range of strategies. [link](#) [9]

CONCLUSION

Misfits represent an intriguing category of naturally formed heterostructures that have remained relatively unexplored compared to conventional crystalline material families. Challenges in characterizing their structures and linking them to emerging physical properties have hindered their study thus far. However, recent theoretical and experimental breakthroughs have enabled the investigation of the structure, electronic, and magnetic properties of heterostructures with mismatched layer alignments. This opens new possibilities for significantly broadening the range of 2D and heterostructure phenomena.

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