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Two-Dimensional Materials from Data Filtering and *Ab Initio* Calculations

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Progress in materials science depends on the ability to discover new materials and to obtain and understand their properties. This has recently become particularly apparent for compounds with reduced dimensionality, which often display unexpected physical and chemical properties, making them very attractive for applications in electronics, graphene being so far the most noteworthy example. Here, we report some previously unknown two-dimensional materials and their electronic structure by data mining among crystal structures listed in the International Crystallographic Structural Database, combined with density-functional-theory calculations. As a result, we propose to explore the synthesis of a large group of two-dimensional materials, with properties suggestive of applications in nanoscale devices, and anticipate further studies of electronic and magnetic phenomena in low-dimensional systems.

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I. INTRODUCTION

The recent explosion of scientific activity around graphene [1] has resulted in a large number of works [2–5] addressing its intriguing physical properties. More recently, efforts have been devoted to study similar, i.e., two-dimensional (2D), systems having a finite band gap, which is a desired feature for many technological applications. Among the directions that have been explored, chemically modified graphene (such as graphane [6,7], graphone [8], fluorographene [9–11], or graphXene [12]) and graphene on various substrates [13–16] and other 2D systems [17–22] (e.g., hexagonal boron nitride and transition-metal dichalcogenides) appear to be the most promising. Apart from technological possibilities with new two-dimensional structures, such materials open up new investigations of several unsolved questions, e.g., concerning the coupling between metallic conductivity and dimensionality. Abrahams *et al.* [23] pointed out that one should not find metallic conductivity as $T \rightarrow 0$ K in two-dimensional materials. Instead, the resistance is expected to grow logarithmically (in the so-called weak localization regime) or exponentially (for strong localization), becoming infinite for $T \rightarrow 0$ K. However, experiments primarily on semiconducting heterostructures challenge these theories, pointing to a much more complicated scenario. The possibility to widen the experimental platforms to new classes of two-dimensional materials is expected to shine light on this problem. Also, similar complex couplings between dimensionality and magnetism are well known [24].

However, from an experimental point of view, the study of even a single system requires the ability to synthesize and to characterize it, for example, by probing it with some relevant spectroscopy, such as x-ray surface diffraction, photoemission, low-energy electron diffraction, scanning-tunneling spectroscopy, etc. Such a task is unfortunately very time consuming, which makes it difficult to undertake a systematic study of a large number of systems. Computational studies, on the other hand, are somewhat simpler: Once the chemical composition and the crystal structure are known, a large number of properties can be obtained, provided that a reliable theoretical tool is used. This approach has been used, in combination with data-filtering techniques, to identify, e.g., new scintillator materials [25,26], novel superconductors [27], and topological insulators [28].

In the present work, we use the efficiency of density-functional theory to reveal the electronic and magnetic structures of candidates of new two-dimensional materials. We report here the parent crystal that is composed of two-dimensional building blocks that can be separated into a two-dimensional unit. We list the International Crystallographic Structural Database (ICSD) number of these crystals and we also provide the electronic structure of the two-dimensional building blocks that these crystals are composed of. Hence, the crystal and electronic structure of a large group of potential new two-dimensional materials is proposed here. These crystals have been identified by means of data-mining algorithms, of compounds listed in the ICSD, and we provide the details of the algorithms in the paragraph below.

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II. METHODS

Most of the compounds known to exist in two dimensions (such as graphene) have a bulk form where the layers are held together by van der Waals forces. To identify new two-dimensional compounds, we have selected materials by screening compounds in the ICSD on the basis of purely geometrical considerations by gradually refining the selection with a filtering for certain characteristic properties of van der Waals bonded layered compounds. For computational reasons, only high-symmetry compounds are considered that yield square or hexagonal in-plane structures. A study of some known weakly bonded compounds showed that the structures of interest are found in a fairly narrow interval of the packing ratio, defined as the covalent volume (the sum of the volumes of the set of spheres of the covalent radii of each atom in the cell) divided by the total volume of the cell. Higher packing ratios tend to be close-packed structures, and lower packing ratios are typically molecular solids, and so the packing ratio is used as the most coarse selection tool. Packing ratios in the range 0.15–0.50 are selected, based on covalent radii compiled from various chemistry textbooks, and databases were obtained from Ref. [29]. In the second step of our procedure, we have selected from our preliminary list the compounds that have sufficiently large gaps (> 2.4 Å) between crystallographic planes along the c axis, the direction of stacking. In the last step, to further refine the selection thus obtained, we inspect the gaps found and identify covalent bonds, if any, that reach across the gaps of the crystal structure. This refinement is done by considering a distance between two atoms close to the sum of their covalent radii as being indicative of a covalent bond. If no bonds are found across the gap, we identify the material as likely to be a weakly bonded layered compound and we then extract the crystal structure of a single layer of this material. Notice that nonstoichiometric compounds, which are very common in mineralogy, are *de facto* excluded from our searching procedure. In the same way, layered compounds not having a sufficiently large gap along the c axis do not appear in our list. Examples of compounds that are layered, but not layered enough for the present search criteria, are, e.g., the high-temperature superconductors.

For each identified single layer, we have performed *ab initio* calculations within the framework of density-functional theory [30] with the Perdew-Burke-Ernzerhof (PBE) [31] version of the generalized-gradient approximation (GGA) [32] as the exchange-correlation potential, as implemented in the Vienna *Ab initio* Simulation Package (VASP) [33,34], and using the projector augmented-wave method [35] for the basis set. For each structure, we proceed in two steps: Starting from the 2D structure as obtained from the ICSD, we perform a geometry optimization until the force acting on each ion becomes smaller than 0.001 eV/Å. Then, this relaxed geometry is used as the fixed crystal structure for a second calculation that allows us to obtain the corresponding total density of states and band structure. In all our calculations, we have used a plane-wave cutoff of 500 eV, while the k-point mesh is generated automatically to have a distance Δk along each reciprocal lattice vector of $\Delta k = 0.2$ Å^{-1} during the geometry optimization process and then densified to $\Delta k = 0.15$ Å^{-1} to obtain precise total density of states and band structures. For materials containing either V, Cr, Fe, Co, or Ni, we checked for the possibility to obtain a spin-polarized ground state within the crystallographic cell. (To keep the calculations affordable, magnetic orders that can occur in a cell larger than the crystallographic cell are not considered.) Information from the ICSD was processed for geometrical analysis and setup of calculations using python classes from the program CIF2Cell [36].

III. RESULTS AND DISCUSSION

Applying our computational procedure, we have been able to identify 92 two-dimensional compounds, in total. Their chemical composition, the ICSD number of the corresponding bulk (3D) material, the value of the minimum band gap (if present), as well as the magnetic order of the ground state (when relevant), are presented in Tables I, II, and III. The band structure of these 92 materials is provided in the Supplemental Material [37]. As usual with a functional such as the GGA, the true value of the band gap is larger than the calculated one, which should therefore be seen as the lower bound, with a correction of between about 50% and 100% [38]. However, the GGA functional very often makes an accurate distinction between metallic and semiconducting materials and describes the structural parameters well.

During our search for new two-dimensional compounds, several already known materials emerge out of the data-filtering procedure. Hence, we successfully identify graphene (from graphite, ICSD number 617290) and 2D hexagonal boron nitride (from three-dimensional hexagonal boron nitride, ICSD number 035538) as relevant materials. At the same time, 2D Bi_2Se_3 (from bulk Bi_2Se_3,

TABLE I. List of known two-dimensional materials also found by our search algorithm. The ICSD number of the corresponding bulk material, the calculated band gap of the two-dimensional material, and its magnetic state (FM refers to a ferromagnetic ordering) are given as well. (The cells of the table are left blank if the material is not magnetically ordered.)

2D chemical formula	3D ICSD number	Gap (eV)	Magnetism
C (graphene)	617290	0.0	
BN	035538	4.7	
Bi_2Se_3	165226	0.9	
Bi_2Te_3	158366	0.9	
FeSe	163559	Metal	
LiFeAs	610480	Metal	FM

ICSD number 165226) and 2D Bi_2Te_3 (from bulk Bi_2Te_3, ICSD number 158366), which are the building blocks of the corresponding bulk materials presenting electronic properties of topological insulators, as well as 2D FeSe and 2D LiFeAs, which belong to the group of iron-based superconductors, are found as well. In Table I, we briefly summarize some of their properties but we do not discuss here in detail their electronic structure, since it has already been reported [18,39,40].

An important family of two-dimensional compounds is the layered dichalcogenides, which appear as possible competitors to modified graphene, as it has been shown that they can be obtained in the form of two-dimensional sheets [17,19–21,41,42]. So far, only MoS_2, $NbSe_2$, WS_2, $MoSe_2$, $MoTe_2$, $TaSe_2$, $NiTe_2$, and VS_2 have been synthesized as nanosheets, but the family of dichalcogenides is known from bulk studies to be larger than that. The full list of dichalcogenides found from our data filtering of the ICSD database is presented in Table II. We have identified the following transition-metal dichalcogenides (with $X =$ S, Se, or Te): TiX_2, ZrX_2, HfX_2, VX_2, NbX_2, TaX_2, MoX_2, WX_2, PdX_2, and PtX_2 from their 3D counterparts in the ICSD database. CrX_2 compounds are also present in our list: While CrS_2 and $CrSe_2$ exist as bulk materials, we have obtained 2D $CrTe_2$ as a layered building block of bulk thallium chromium telluride ($TlCrTe_2$, ICSD number 152836; bulk $CrTe_2$ is not referenced in the ICSD database). We have also identified the sulfides YS_2, TcS_2, and ReS_2, the selenide $ReSe_2$, and the tellurides $CoTe_2$, $RhTe_2$, $IrTe_2$, and $NiTe_2$. Also, $SiTe_2$, SnS_2, and $SnSe_2$, which are layered dichalcogenides but not composed of a transition-metal ion, are reported. The electronic structure of MoX_2, NbX_2, WX_2, and TaX_2 has already been investigated in the past [43], and our computed band structures, which are essentially equivalent to earlier studies (see the Supplemental Material [37]), will not be commented upon any further here. From our calculations, the vanadium dichalcogenides VX_2 are all found to be ferromagnetic, a fact that was noticed before [44] for VS_2 and VSe_2. (VTe_2 was not studied in Ref. [44].) In the case of TaS_2 and $TaSe_2$, two polymorphs are found, depending on how the S and Se atoms are located on each side of Ta. In Table II, we use the symbol $-AA$ for the polymorph when the S (or Se) are on top of each other and $-AB$ when they are not. To our knowledge, the electronic structure of the other dichalcogenides in their two-dimensional form has not been presented so far. While a majority of them are metals (see Table II), several present a sizable band gap, and in particular $ZrSe_2$ and $HfSe_2$ have a GGA band gap very close to the one obtained for bulk silicon ($ZrSe_2$ and $HfSe_2$ have calculated band gaps of 0.4 and 0.6 eV, respectively, while bulk silicon has a GGA band gap of 0.46 eV).

TABLE II. List of two-dimensional dichalcogenides found by our search algorithm with the ICSD number of the corresponding bulk material and with our calculated band gap (in eV). The corresponding magnetic state (AFM refers to an antiferromagnetic ordering) is given in the last column to the right. (The cells of the table are left blank if the material is not magnetically ordered.)

2D chemical formula	3D ICSD number	Gap (eV)	Magnetism	2D chemical formula	3D ICSD number	Gap (eV)	Magnetism
YS_2	651404	Metal		$CrTe_2$	152836	Metal	FM
TiS_2	651178	0.02		MoS_2	644245	1.6	
$TiSe_2$	173923	Metal		$MoSe_2$	644334	1.4	
$TiTe_2$	653071	Metal		$MoTe_2$	015431	1.15	
ZrS_2	651465	1.1		WS_2	202366	1.8	
$ZrSe_2$	652236	0.4		WSe_2	040752	1.5	
$ZrTe_2$	653213	Metal		WTe_2	073323	Metal	
HfS_2	638847	1.3		TcS_2	081816	1.2	
$HfSe_2$	638899	0.6		ReS_2	075459	1.4	
$HfTe_2$	638959	Metal		$ReSe_2$	081813	1.3	
VS_2	651361	Metal	FM	$CoTe_2$	625401	Metal	
VSe_2	652158	Metal	FM	$RhTe_2$	650448	Metal	
VTe_2	603582	Metal	FM	$IrTe_2$	033934	Metal	
NbS_2	645307	Metal		$NiTe_2$	159382	Metal	
$NbSe_2$	645369	Metal		PdS_2	166276	1.1	
$NbTe_2$	645529	Metal		$PdSe_2$	170327	1.3	
TaS_2-AB	651089	Metal		$PdTe_2$	649016	0.2	
TaS_2-AA	651092	Metal		PtS_2	649534	1.8	
$TaSe_2$-AB	651948	Metal		$PtSe_2$	649589	1.4	
$TaSe_2$-AA	651950	Metal		$PtTe_2$	649747	0.8	
$TaTe_2$	014390	Metal		$SiTe_2$	652385	Metal	
CrS_2	075420	Metal	AFM	SnS_2	650992	1.6	
$CrSe_2$	626718	Metal	FM	$SnSe_2$	651910	0.8	

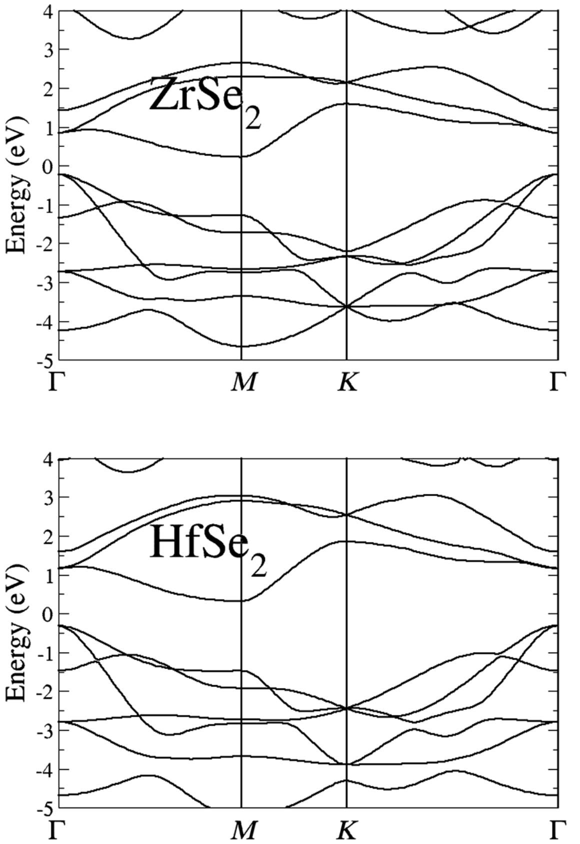

FIG. 1. Computed band structure of $ZrSe_2$ (left) and $HfSe_2$ (right) along the high-symmetry directions Γ-M-K-Γ. The Fermi level is at zero energy.

Because of the similarity in calculated band gap to that of Si, we suggest that $ZrSe_2$ and $HfSe_2$ are potentially interesting for applications. In Fig. 1, we present their band structures along the high-symmetry directions Γ-M-K-Γ in the 2D Brillouin zone. The two band structures are very similar, with a maximum of the valence band at the Γ point and a minimum of the conduction bands at the M point. Furthermore, the GGA band gap of the tellurides $PdTe_2$ and $PtTe_2$ is less than 1 eV, suggesting that the true band gap is probably between 0.5 and 1.5 eV. It can be modulated to a suitable value by alloying the telluride with the corresponding sulfide or selenide, which are insulating. Also, the series of chromium dichalcogenides appears to be particular, since it presents magnetic ordering: antiferromagnetic for CrS_2 and ferromagnetic for $CrSe_2$ and $CrTe_2$.

In Table III, we present a list of materials that have not been previously reported as possible two-dimensional compounds and that are outside the group of dichalcogenides. This list includes materials with a relatively simple chemical composition and structure, such as GaS, FeS, FeTe, and PbO, and more complex compounds, such as P_2AgSe_6Bi, P_2CuSe_6Bi, ScP_2AgSe_6, and $K_2C_9FeO_3N_6$. Therefore, it is not surprising that the different electronic structures span everything from metals to wide-band-gap insulators, also including materials showing a magnetic order, such as FeTe, $FeBr_3$, and $CrSiTe_3$. These last two compounds present a moderate band gap (0.5 eV for $FeBr_3$ and 0.6 eV for $CrSiTe_3$) that could be of interest for electronics applications. The details of their electronic

TABLE III. List of compounds found by our algorithm that do not belong to the family of dichalcogenides. The chemical formula, the ICSD number of the corresponding bulk material, the value of the minimum band gap, and an eventual magnetic ordering are given in the different columns from left to right. (The cells of the table are left blank if the material is not magnetically ordered.)

2D chemical formula	3D ICSD number	Gap (eV)	Magnetism	2D chemical formula	3D ICSD number	Gap (eV)	Magnetism
PbIF	150193	2.3		$PbSb_2Te_4$	250250	0.8	
HgI_2	150345	1.8		$KC_6FeO_3N_3$	280850	4.5	
ZrClN	151468	1.9		MgI_2	281551	3.6	
BaIF	155006	4.3		BiIO	391354	1.5	
SrIF	155009	4.5		$FeBr_3$	410924	0.5	AFM
$AlCl_2$	155670	Metal		$MgPSe_3$	413165	2.1	
Ag_2ReCl_6	156662	Metal		IYGa	417149	Metal	
Ni_2Te_2Sb	158485	Metal		PTe_2Ti_2	418978	Metal	
$Bi_{14}Te_{13}S_8$	159356	0.9		ScP_2AgSe_6	420302	1.8	
$MgBr_2$	165972	4.8		$CrSiTe_3$	626809	0.6	FM
Cu_2S	166578	Metal		$FePSe_3$	633094	0.05	
P_2AgSe_6Bi	170640	1.4		FeS	633302	Metal	
P_2CuSe_6Bi	170642	0.8		FeTe	633877	Metal	FM
YI_3	170773	3.1		$Sb_2Ge_2Te_5$	637823	0.2	
GaS	173940	2.4		SbSiNi	646436	Metal	
VCl_2	246905	Metal		PbO	647260	2.5	
VBr_2	246906	Metal		CdI_2	655780	2.5	
VI_2	246907	Metal		GaSe	660262	1.8	
PFeLi	247089	Metal		$ZnIn_2S_4$	660273	Metal	
$PbBi_2Te_4$	250249	1.0		$Zn_2In_2S_5$	660333	Metal	

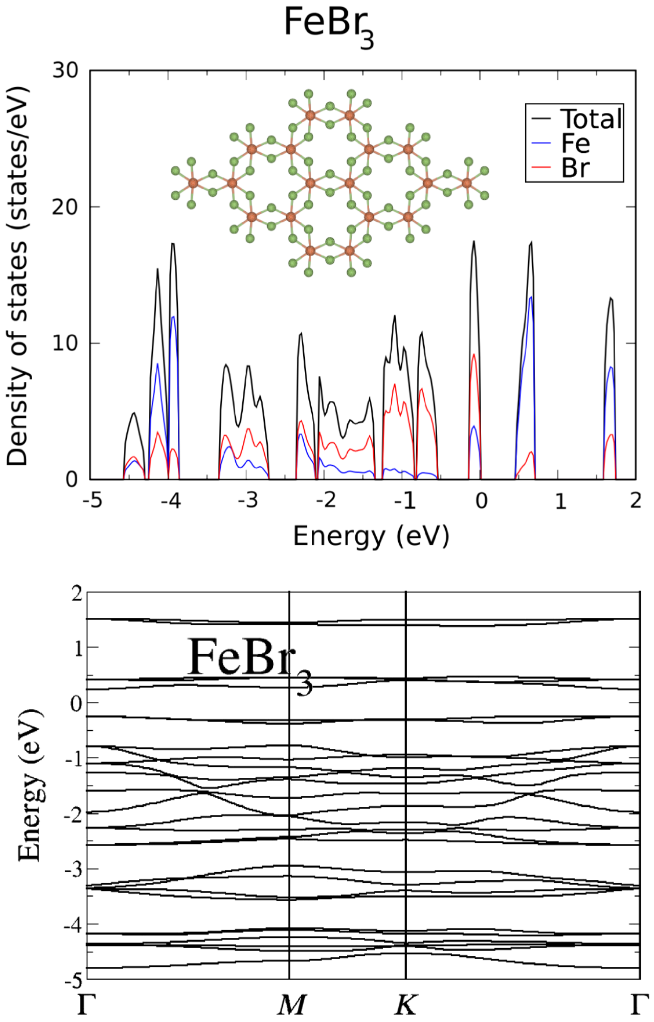

FIG. 2. Density of states (upper panel) and band structure along the high-symmetry directions Γ-M-K-Γ (lower panel) of antiferromagnetic $FeBr_3$. Note that the density of states is also shown as an atomic projected property. Also, the Fermi level is at zero. In the inset of the upper panel, the two-dimensional crystal structure is also shown.

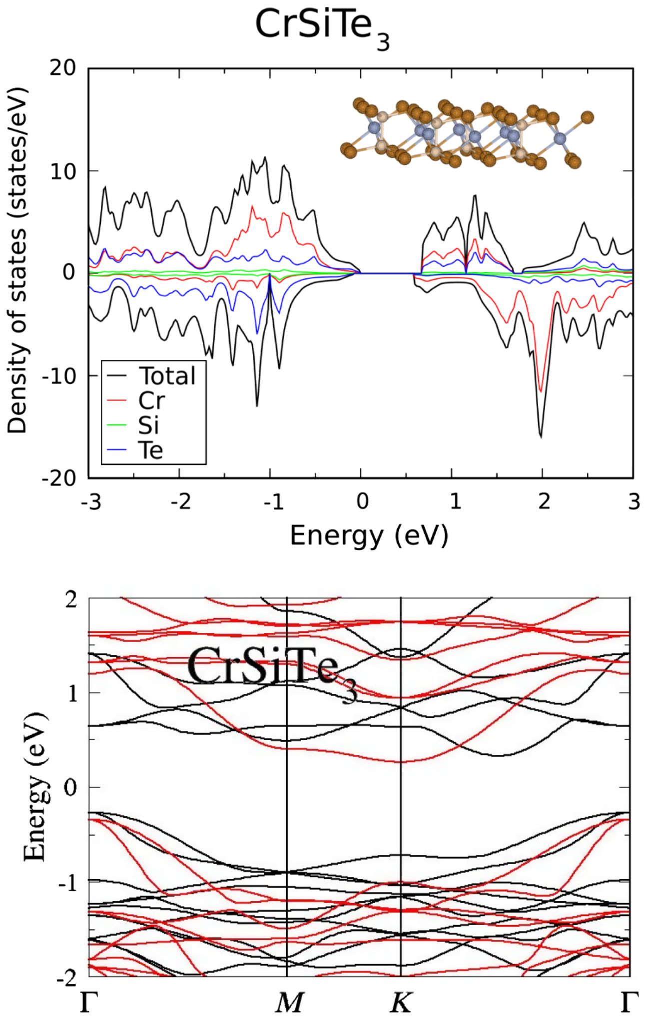

FIG. 3. Density of states (upper panel) and band structure along the high-symmetry directions Γ-M-K-Γ (lower panel) of ferromagnetic $CrSiTe_3$. Note that the density of states is also shown as an atomic projected property. The Fermi level is at zero energy. In the inset of the upper panel, the two-dimensional crystal structure is also shown.

structures are presented in Figs. 2 and 3. As for $FeBr_3$, which we find to be an antiferromagnet, the top of the valence band is composed of Fe and Br derived states, while the bottom of the conduction band arises mainly from Fe states. The band gap is found to be indirect, with a maximum of the valence band along the K-Γ direction, while the minimum of the conduction band is at Γ. $CrSiTe_3$ is found to be ferromagnetic, with the top of the valence bands and the bottom of the conduction bands made from Cr and Te states. Because of the magnetic moment of the Cr atoms, the Te and Si atoms become slightly spin polarized as well, as shown by the exchange splitting between spin-up and spin-down states. For this compound, the top of the valence band is at the Γ point with a majority spin polarization, while the bottom of the conduction band is at the K high-symmetry point with minority spin polarization. Also, $Bi_{14}Te_{13}S_8$, P_2CuSe_6Bi, $PbSb_2Te_4$, and $Sb_2Ge_2Te_5$ have a GGA band gap lower than 1.0 eV and are of interest as well. Our calculated band structures for the compounds presented in Table III are given in the Supplemental Material [37].

IV. CONCLUSION

Beside our proposed list of compounds, further materials are anticipated: The transition-metal dichalcogenides NiS_2, $NiSe_2$, $ReTe_2$, $TcSe_2$, and $TcTe_2$ have not been reported experimentally (and are therefore not present in the ICSD as bulk materials), but they are likely to exist. Another direction to explore is the synthesis of hybrid structures: This synthesis was achieved recently [45] with h-BN or graphene. The concept can be extended to other two-dimensional structures provided that a suitable match between the different subsystems is found. Also, these new materials can be put on substrates, as is currently done with graphene. Note that the two-dimensional materials that we find to be metallic should not be disregarded, since their electronic structure may be modified by functionalizing the material in the same way that graphene is nowadays modified in graphane or flurographene. Transparent metals also are sought after, e.g., as replacements to indium tin oxides [46]. In addition, as discussed above, metallic conductivity in two dimensions is far from understood, and the identification of new two-dimensional materials enables further studies of this complex phenomenon. Similar conclusions can be made about the complex coupling between magnetic properties and dimensionality. Notice that our proposed list certainly does not cover all possible 2D materials, since, as explained in Sec. II, we have only searched through a subset of the ICSD.

From our results, it appears that the number of 2D materials and the variety of their electronic structures is quite large: The list of possible 2D dichalcogenides has only begun to be explored by experimentalists [17,19,41], while the compounds presented in Table III have not been thought to exist as 2D crystals. Since the compounds that we propose have already been synthesized in bulk form, their 2D counterparts can probably be obtained using the same techniques [1,17,19] that are now routinely used to obtain graphene, 2D *h*-BN, and 2D transition-metal dichalcogenides. Therefore, the path now seems to be open to explore and engineer these new materials.

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