Quantum Materials Discovery by Combining Chemical and Physical Design Principles

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Abstract: Exploratory quantum materials discovery remains crucial to progress in material science. Due to the grand challenges that we are facing in predicting these materials and their properties from scratch, chemical design principles remain a key ingredient for the discovery of new materials. Chemical heuristics, structure, bonding, as well as global and local symmetries are at the very foundation of materials properties. In this regard, in this research, we aim to identify functional materials by composition-structure-property understanding. Materials discovery consists of a subset of methods and design principles that go hand in hand until a desired material or property is realized. However, materials synthesis is still far from a rational design approach. Rather, materials, and especially metastable materials, have to be accessed and synthesized in an exploratory, laboratory-intensive fashion. At the same time, quantum materials discovery is a vibrant highly active field of research that has seen various leaps of progress in recent years, and that holds the promise for many more in the coming years. Here, we lay out how we are discovering new materials and new materials physics in our and other chemical physics, or physical chemistry research groups, and how chemistry and chemical synthesis play a crucial role in this process.

Keywords: Materials science · Quantum materials · Solid-state chemistry

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Fabian O. von Rohr studied chemistry and physics at ETH Zurich. He carried out his Master's thesis at Princeton University, and received his PhD at the University of Zurich (UZH) for the work he accomplished with Prof. A. Schilling, in collaboration with Prof. R. Nesper (ETHZ). His PhD thesis was awarded the Mercator Award 2014 and was chosen as one of the best theses at UZH. During his postdoctoral research time, he joined the laboratories of Prof. Wolfgang Schnick at the LMU Munich and Prof. Robert J. Cava at Princeton University. In 2017, he became a group leader and in 2021 an assistant professor at UZH. In 2022, he joined the Department of Quantum Matter Physics at the University of Geneva as an associate professor.

1. Quantum Materials for Next-Generation Quantum Technologies

The discovery of materials with tailored properties has, time and time again, proven to be a crucial stimulus for technological advancement and, by implication, of societal progress – from superconductors used in magnetic resonance imaging (MRI) magnets to semiconductor materials that enabled modern computer technology. Currently, progress in the development of many technologically relevant materials is reaching a point of stagnation.[1–3] A prime example are silicon-based transistors, which are approaching a fundamental physical limit.[4] Progress based on silicon-based electronics is therefore bound to come to a halt. The consensus in the scientific and industrial communities is that we need to develop new device paradigms based on new materials to overcome this impasse. Quantum materials, in particular, are widely considered to have a key role in the development of such next-generation technologies that will meet the urgent technological demands for securing a sustainable and safe society.[3]

Quantum material is a collective term for a range of materials that display properties that cannot be described by semiclassical particles. These are materials that present strong electronic correlations or some type of electronic order, such as superconductivity, long-range magnetic order, or materials whose electronic properties are linked to non-generic quantum effects, *e.g.*, topological insulators. A common thread to all of these materials is the concept of emergence. With the term emergence, we mean systems in which the property of the whole is more than the sum of the properties of its constituents. There are many instances for this in nature, an obvious example is the behavior of a flock of birds. The interested reader is referred to the essay of the recently deceased Nobel laureate P. W. Anderson entitled: *More is different*.[5]

Most recently, there have been many exciting new quantum materials that have been discovered. These include – just to mention a few – the discovery of superconductivity in magic-angle graphene, demonstration of room-temperature two-dimensional (2D) ferromagnetism, high-temperature superconductivity in hydro-

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gen-rich materials under extreme pressures, topological materials with protected massless states, or the observation of the quantum anomalous Hall effect.[6–9]

The prediction of new quantum materials from scratch remains despite great progress in condensed matter theory and advanced computational methods challenging today. Quantum materials exhibit several competing and coexisting interactions that have to be considered. Therefore, exploratory quantum materials discovery remains crucial to progress in this field. In this essay, we lay out how new materials and new materials physics is discovered in our and other chemical physics, or physical chemistry research groups and how chemistry and chemical synthesis play a crucial role in this process.

2. How New Materials are Discovered and Improved

How do we discover new quantum materials? As often with discovery and exploratory chemistry this question boils down to the infamous needle in a haystack. The former Max-Planck director and pioneer in solid-state chemistry research Prof. Martin Jansen, illustrated this challenge following the discovery of the cuprate superconductor YBCO in a 2003 review article on solid-state synthesis.[10] He argued that in order to discover YBCO without prior knowledge, it would take 27,000 years even if one could produce and characterize the large number of 10^5 samples per day. Although one can argue about the precise number that has to go into such an estimation, this example in any case illustrates that we need different, more efficient tools to sample experimental space than a pure high-throughput undirected search.

In our research, we aim to identify functional materials by composition-structure-property understanding. Materials discovery consists of a subset of methods and design principles that go hand in hand until a desired material or property is realized. An important part of this work is therefore exploratory in nature.

A usual work circle in our group looks as follows: First, we find inspiration for developing a new material by recognizing patterns in the chemical or electronic structures that we associate with a certain property. For this, we are using a combination of chemical concepts[11–15] in order to come up with what we call an 'educated guess' about the properties of a material. Second, the material and a set of variations of this material will be synthesized, and a full set of compounds, based on the original hypothesis, will be investigated. The new materials will be realized by state-of-the-art solid-state synthesis methods. Third, the crystal structure and materials properties will be characterized by X-ray scattering, magnetization, electrical transport, and other methods. These results will be compared with the original hypothesis, which then will be adapted correspondingly. Subsequently, this leads to a new well-informed hypothesis, for which again materials will be synthesized and characterized, leading to an iterative work cycle until qualitative (rather than incremental) progress is made. The combination of chemical synthesis and the measurement of physical properties is an integral part of this line of research because it allows for immediate feedback loops and hence fast improvements of the materials.

3. Tuning Structure and Composition towards New Functionalities

The search for new materials sometimes relies on compositional or structural modifications of well-known compounds. For a given chemical formula, the exploration of the chemical space by adjusting the synthesis conditions can lead to the stabilization of new polymorphs, that is, to compounds which present the same chemical composition but different crystal structures. As the crystal structure greatly defines the physical properties of materials, these can drastically change for different polymorphs. Control over the synthesis route is required in order to target and isolate the desired composition-structure match.

On one side, varying the synthesis conditions can allow access to metastable phases. For example, by applying high-pressure high-temperature conditions we found a new layered polymorph in the GeSe system, structurally and electronically related with the α-GeSe[16,17] and black phosphorus[18] semiconductors. The so-obtained β-GeSe phase[19] has also a pronounced 2D character, but displays a different corrugation of the GeSe layers, *i.e.* with a boat conformation as compared to the chair conformation of both α-GeSe and black phosphorus. The new β-GeSe polymorph is also a semiconductor, with a bandgap falling between that of α-GeSe and black phosphorus, making β-GeSe a promising candidate for 2D semiconductor applications.

Another example of the stabilization of a metastable polymorph through a careful adjust of the synthesis conditions can be found in the FeS_2 system. There are two naturally occurring iron disulfide polymorphs, pyrite and marcasite,[20] with related crystal structures based on octahedral $[FeS_6]$ units as shown in Fig. 1a. Both polymorphs are found to form in nature under hydrothermal conditions,[21] *i.e. via* precipitation from an aqueous solution at high temperatures and vapor pressures. The less common marcasite polymorph forms as a metastable phase under acidic conditions but transforms into the thermodynamic stable pyrite at high temperatures.[22] Therefore, laboratory-based synthesis of phase-pure marcasite proved challenging, preventing a proper characterization of its physical properties. Considering that pyrite stands as a promising candidate for electronic and photovoltaic applications,[23–26] the exploration of marcasite as a potential functional material retains a great interest.

We thus assessed the preparation of marcasite FeS_2 by systematically exploring the hydrothermal synthesis in acidic media.[27] We found that phase-pure marcasite can be obtained in a narrow pH window of 1.2–2.7 at moderate temperatures, between 190–225 ºC. Pyrite can be also isolated at lower pH values and higher temperatures, in an extremely narrow area of the synthesis map (in green in Fig. 1b). Outside these windows, a mixture of both polymorphs is obtained. The identification of these narrow experimental regions for phase-pure marcasite and pyrite allowed us to isolate both polymorphs, and to further characterize their fundamental physical properties.

Particularly, we measured the band gap of the two FeS_2 phases by X-ray absorption/emission spectroscopy as shown in Fig. 1c,d. We find that marcasite has a band gap of 0.73 eV, comparable to the 0.87 eV band gap of pyrite. This, together with its positive Hall coefficient and small transport gap E_g of 0.11 eV suggest that marcasite – that is now synthetically available in a straightforward fashion – is as equally promising as pyrite as candidate for various semiconductor applications based on earth-abundant elements.

Precise control over the polymorph formation therefore allows to enlarge the materials database, and to perform a rigorous characterization of the physical properties.

On the other side, small compositional changes while maintaining the same structural frame can also drive a drastic change in the physical properties.[28] One particularly attractive chemical approach relies on the incorporation of dopants into void positions or between layers in layered materials (see, *e.g.* refs [29–34]). Thereby, for example, superconductivity can be induced in otherwise non-superconducting materials, and the critical temperature may increase substantially in already superconducting systems. In this line, the nearly intermetallic η-carbide materials stand as an interesting system for further exploring the effect of filling void positions as a way of tuning the electronic properties. As shown in Fig. 2a, the crystal structure of η-carbides can be understood as a filled version of the Ti₂Ni-type structure, with light nonmetallic atoms (either carbon, oxygen or nitrogen) occupying interstitial positions.

η-Carbide-type compounds are a large family of materials, with more than 120 composition within the structural type.[35,36] Such a chemical versatility provides a wide range of physical
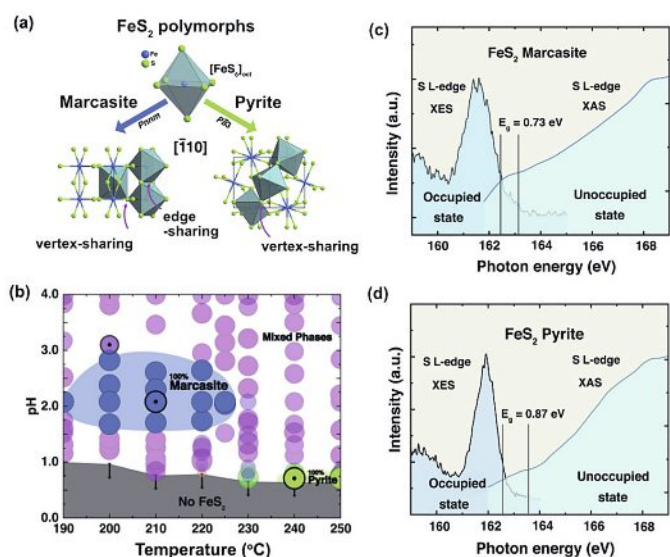
Fig. 1. (a) Crystal structures of the two FeS_2 polymorphs emphasizing the common $[FeS_6]$ octahedra building block. (b) Synthesis map of the hydrothermal synthesis of FeS_2 as a function of pH and temperature. Marcasite (in blue) and pyrite (in green) form in narrow separate windows of pH and temperature. The band gap values for the phase-pure marcasite (c) and pyrite (d) polymorphs as determined from combined X-ray absorption (XAS) and emission (XES) spectroscopy measurements. Figures adapted from ref. [27].
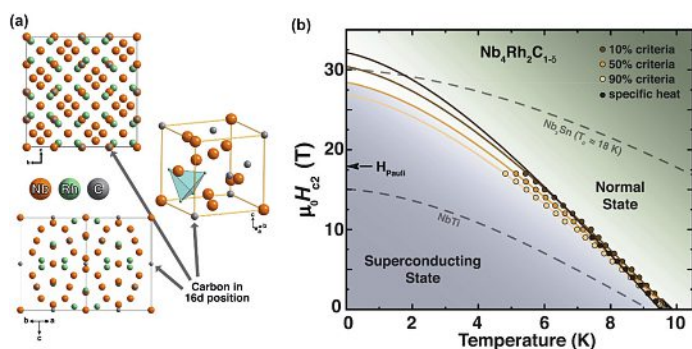

Fig. 2. (a) Crystal structure of the cubic $Nb_4Rh_2C_{1-\delta}$ η-carbide along three different crystal projections, emphasizing the crystallographic void positions occupied by C atoms. (b) Electronic phase diagram of $Nb_4Rh_2C_{1-\delta}$ showing the temperature dependence of the upper critical field μ_0H_{c2}. Fitting using the Werthamer-Helfand-Hohenberg formalism is shown as continuous lines. The critical field dependence of the NbTi and Nb_3Sn model compounds are shown as dashed lines for comparison. Figures adapted from ref. [39].

properties and, in particular, provides a promising frame for the investigation of emergent quantum properties.[35,37,38] Nonetheless, so far, only few superconducting η-carbides have been reported. Also, the physical properties of superconducting η-carbides have been somewhat overlooked. In particular, the role of the void-filling atoms remained uncertain.

Our detailed analysis of the physical properties of the $Nb_4Rh_2C_{1-\delta}$ η-carbide[39] evidences bulk superconductivity below $T_c = 9.75$ K, as demonstrated by specific heat measurements. The normalized specific heat jump of $\Delta C/\gamma T_c = 1.64$ lies indeed close to the weak-coupling BCS value of 1.43. We also found $Nb_4Rh_2C_{1-\delta}$ to be an extreme type-II superconductor which displays a surprisingly high upper critical field of $\mu_0H_{c2}(0) = 28.5$ T. The critical field of $Nb_4Rh_2C_{1-\delta}$ is indeed comparable to the most currently used superconducting magnet for higher-field applications, Nb_3Sn ($T_c = 18$ K),[40] with $\mu_0H_{c2}(0) \sim 30$ T (see Fig. 2b). And, further-

more, it represents a clear violation of the Pauli paramagnetic limit H_{Pauli}, which is the theoretical maximum critical field of a BCS-superconductor that can be estimated according to $\mu_0H_{Pauli} \sim 1.86$ [T/K]·T_c. Within the weak-coupling BCS theory, a paramagnetic pair-breaking effect is expected for applied magnetic fields above this limit, standing as the main practical limitation for the achievable magnetic field in superconducting magnets.

The possibility to surpass the Pauli limit in an otherwise conventional BCS superconductor opens exciting possibilities for improving superconducting magnets for MRI or nuclear magnetic resonance (NMR) applications, among others.

In this line, we find the related Ti_4Co_2O ($T_c = 2.7$ K; $\mu_0H_{c2}(0) = 7.08$ T) and Ti_4Ir_2O ($T_c = 5.3$ K; $\mu_0H_{c2}(0) = 16.06$ T) η-carbides also show upper critical fields well beyond the Pauli limit, suggesting a more general trend in this family of compounds.

In view of these exceptional superconducting properties, we further investigated the effect of the filling-void atoms on the superconducting properties by synthesizing Ti$_2$Ni-type Ti_2Co (*i.e.* with empty void positions) and η-carbide-type Ti_4Co_2O single crystals.[41] The transport measurements of such crystals show a clear impact of the oxygen incorporation into the void positions. In the normal state, the metallic character of Ti_4Co_2O contrasts with the semimetallic behavior of Ti_2Co. Moreover, while Ti_4Co_2O shows a sharp superconducting transition below $T_c = 2.7$ K, the critical temperature is depressed below $T_c < 0.5$ K in Ti_2Co. Therefore, despite the effect on the crystal structure being small, the incorporation of oxygen in the voids plays a crucial role in the physical properties and particularly, in the superconducting properties. The incorporation of electron-acceptor atoms in Ti$_2$Ni-type structures, and eventually in other materials with crystallographic void positions, thus seems a promising strategy in the design of new superconductors with exceptional magnetic performance.

4. Transition Metal Dichalcogenides: Challenges and Prospects

The realization of graphene-based devices has boosted the exploration of materials with two-dimensional (2D) character.[42] In particular, van der Waals materials turn out to be the most promising source of 2D-materials, as they can be easily exfoliated to obtain atomically thin layers.[43] The possibility to isolate and combine atomically thin layers into complex heterostructures leads to exceptional functionalities covering electronic, photonic and optoelectronic applications[44,45] for the next generation of electronic nanodevices.

Among van der Waals materials, layered transition metal dichalcogenides (TMDs) with general formula MX_2 where M is a transition metal and X is a chalcogen atom (*i.e.* S, Se, or Te) stand out. They are known to display rich electronic properties, from superconductivity to exotic topological phases.[29,46–48] TMDs exist for the majority of the group IV and group V transition metals, and different coordination geometries within the MX_2 layers (*i.e.* tetrahedral, octahedral and distortions thereof) are preferred depending on the M and X pair. Moreover, further complexity arises from the different possible stackings of the individual MX_2 layers which are bonded *via* van der Waals interactions along the stacking direction.

The strong effect of the polymorphism in the electronic properties is clear for the molybdenum ditellurides, with the 1T'-$MoTe_2$ polymorph (with octahedral coordination) being a Weyl semimetal displaying low-temperature superconductivity,[49] while the 2H-$MoTe_2$ polymorph (with trigonal prismatic coordination) is a semiconductor with a rather large indirect band gap of ~1 eV.[50] Polytypism – meaning different combinations of the same layered structural units – is expected to have a less pronounced effect on the physical properties. For example, only a slight depression of the T_c is found from the 2H-$NbSe_2$ ($T_c = 7.2$ K) to the 4H-$NbSe_2$ ($T_c = 6.5$ K) polytypes.[51]

In some cases, the energy barrier for the formation of the different polymorphs and polytypes is very small, making a targeted synthesis challenging. This complicates the analysis of the physical properties and can lead to misleading interpretations. In this regard, NbS_2 stands as a representative example of the synthetic challenges for a proper understanding of the physical properties of TMDs. There are three different known polymorphs for NbS_2, the two polytypes $3R-NbS_2$ and $2H-NbS_2$, and the polymorph $1T-NbS_2$, which so far has only been synthesized in thin film form. In both $3R-NbS_2$ and $2H-NbS_2$, the Nb atoms adopt a trigonal prismatic coordination within the NbS_2 layers, but differ in the layer stacking sequence (see Fig. 3a). $2H-NbS_2$ is known to be a $T_c \sim 6$ K superconductor, while some reports claimed the $3R-NbS_2$ to be also superconducting with a similar critical temperature. However, the challenging preparation of phase-pure samples complicates the analysis of the physical properties of the individual polytypes.
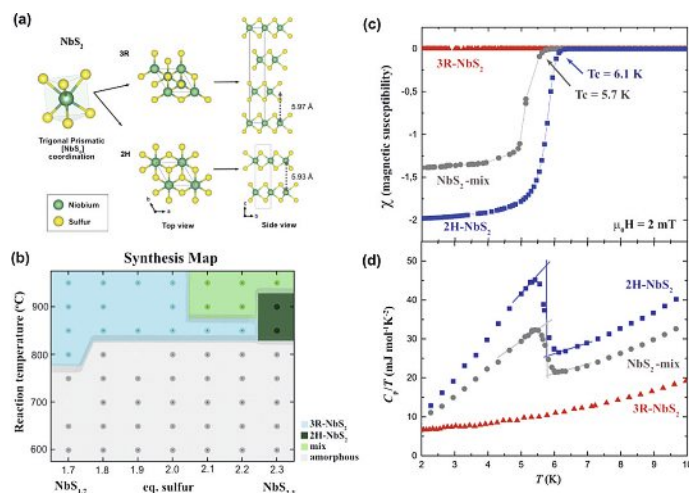
Fig. 3. (a) Crystal structure of the $3R-NbS_2$ and $2H-NbS_2$ polymorphs, showing the common trigonal prismatic coordination for the Nb atoms. (b) Synthesis map of the NbS_2 system as a function of the temperature and sulphur molar equivalents. Phase-pure samples of $3R-NbS_2$ and $2H-NbS_2$ polymorphs are shown in blue and dark green, the mixed-phase region in light green, and the amorphous region in grey. (c) zero-field-cooling temperature-dependent magnetization and (d) specific heat $C(T)/T$ of a phase pure $2H-NbS_2$ (blue), a phase pure $3R-NbS_2$ (red), and a sample consisting of both polytypes (grey). Figure adapted from ref. [52].

We addressed this by systematically exploring the solid-state synthesis of both polytypes, by varying the stoichiometry (*i.e.* the sulfur molar equivalents) and the temperature for the reactions.[52] The resulting synthesis map is shown in Fig. 3b. We find that the $2H-NbS_2$ polytype can be obtained as a single phase only in a very narrow region of temperature and under a slight excess of sulfur of ~15%. On the other hand, stoichiometric or slightly deficient reactant mixtures result in phase-pure $3R-NbS_2$ samples for a wider range of temperatures. It is worth stressing that the phase-pure regions for the two different polytypes are separated by a wide region of mixed-phase formation, emphasizing that a delicate adjustment of the synthesis conditions is crucial for isolating the two polytypes and avoiding the intergrowth of the two.

We further characterized the physical properties of the two phase-pure polytypes, together with a mixed-phase sample. The magnetic susceptibility measurements (Fig. 3c) evidence that $2H-NbS_2$ is superconducting with $T_c = 6.1$ K, in agreement with previous reports. On the other hand, the phase-pure $3R-NbS_2$ sample did not show any sign of superconductivity above $T > 1.75$ K.

In the case of the mixed-phase sample, a superconducting transition with a slightly depressed $T_c \sim 5.7$ K is observed. Considering the structural similarity of both polytypes, which also leads to similar X-ray powder diffraction patterns, it is not surprising that the superconducting behavior of such a mixed-phase sample could be easily mistaken as a bulk superconducting $3R-NbS_2$ phase. Our results, however, clearly evidence that the superconducting signal cannot be assigned to the $3R-NbS_2$ polytype, but more likely reflect a defective $2H-NbS_2$ phase. The absence of superconductivity in $3R-NbS_2$ is further confirmed by the specific heat measurements shown in Fig. 3d. Also, they confirm the bulk nature of superconductivity in $2H-NbS_2$, with a normalized specific heat jump at the T_c of $\Delta C/\gamma T_c = 1.30$, in good agreement with the BCS theory. On the other hand, the mixed-phase sample displays a reduced value of $\Delta C/\gamma T_c = 0.98$, which can be rationalized to reflect a ~75% volume fraction of $2H-NbS_2$.

These results highlight the importance of a careful synthesis and chemical characterization of TMDs prior to the analysis of the physical properties.

Exotic superconducting states are also found among various TMDs.[53,54] For example, the Weyl semimetal $1T'-MoTe_2$ is a superconductor below $T_c \sim 0.1$ K at ambient pressure, and below $T_c \sim 4$ K under ~ 2 GPa.[49,55,56] It thus stands as a rare example of a superconducting material with Weyl fermions at the Fermi-level. Even indications of a topologically non-trivial superconducting state has been observed by means of muon spin relaxation spectroscopy (μSR).[55] Surprisingly, the superfluid density is found to scale linearly with the critical temperature, a trend in close resemblance to the Uemura relation which holds for cuprate superconductors, and that is considered a hallmark feature of unconventional superconductivity. To further elucidate the electronic mechanism for superconductivity in these TMDs, we extended our study to the $NbSe_2$ system (see Fig. 4), where a coexistence of superconductivity and a two-dimensional charge density wave (CDW) is found in both $2H-NbSe_2$ and $4H-NbSe_2$ polytypes.[51]

In our μSR analysis we evidenced how the superfluid density, as determined from the magnetic penetration depth $\lambda(T)$, also scales linearly with the critical temperature in both $2H-NbSe_2$ and $4H-NbSe_2$ (Fig. 4c).
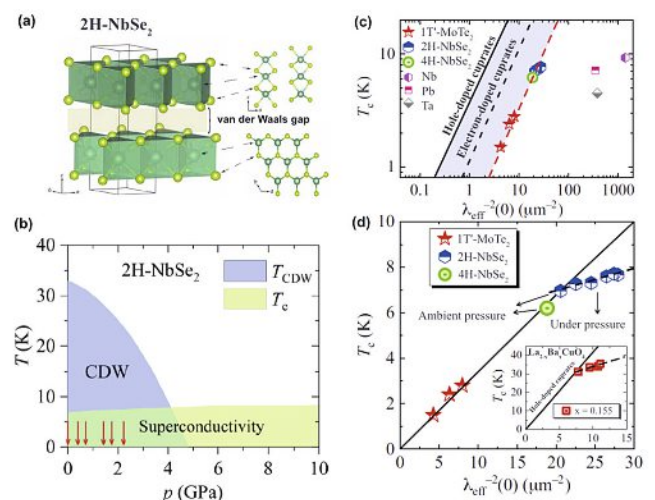
Fig. 4. (a) Crystal structure of $2H-NbSe_2$ and (b) electronic phase diagram. (c) Logarithmic plot of the T_c against the magnetic penetration depth in 2H- and $4H-NbSe_2$, and $1T'-MoTe_2$. The Uemura relation for hole and electron-doped cuprates is also shown for comparison. (d) Linear plot of T_c against the magnetic penetration depth showing the effect of the hydrostatic pressure in $2H-NbSe_2$. The inset shows the analogue deviation from the Uemura relation in optimally doped $La_{2-x}Ba_xCuO_4$ (x = 0.155) under pressure.[57] Adapted from ref. [51].

We furthermore find an increase in the superfluid density in $2H$-$NbSe_2$ under the application of hydrostatic pressure, with a concomitant deviation from the linear relation, as shown in Fig. 4d. This behavior is analogue to that observed in optimally doped cuprate superconductors, as shown in the inset of Fig. 4d. The strong similarity with other unconventional superconductors in the relation between the superfluid density and the critical temperature strongly suggests that these TMDs may also exhibit unconventional superconducting properties. Our findings – essentially based on our ability to control polymorph and polytype formation in a chemically precise manner in these systems – therefore encourage further elucidation of the underlying quantum physics in layered TMDs.

5. Magnetic van der Waals Materials

A key step forward for the development of new quantum technologies can be clearly found in the field of van der Waals and/or 2D materials that display intrinsic long-range magnetic order.[58–60] These materials hold the promise to open up new possibilities for the electrical manipulation of spins for use in memory devices, quantum computing, or spin filtering devices.[61] Recently, the first experimental realization of intrinsic magnetic monolayers was indeed attained, *via* mechanical exfoliation of insulating $Cr_2Ge_2Te_6$ and CrI_3, and metallic Fe_3GeTe_2 van der Waals magnets.[6,62,63] Since then several new materials have been identified and characterized as 2D magnetic materials.

The development or discovery of new 2D magnetic materials with large bandwidth and high ordering temperatures is one of the great challenges in this area of research. The most recently identified, mixed-anion van der Waals material $CrSBr$[64] stands out as a promising candidate. The combination of halogen and chalcogen anions in this material results in a large band dispersion while retaining a suitable direct bandgap of ~1.8 eV.[65] Its high magnetic ordering temperature of T_N = 130 K and its substantial air-stability reinforce the potential of $CrSBr$ as component for electronic devices. A pronounced magnetoresistance has been indeed demonstrated below the ordering temperature.[66]

The mixed-anion character of the material has also a profound impact on the magnetic, structural and transport properties of $CrSBr$. Interestingly, an anisotropic band structure is observed, with flat bands along the real space *a* direction but dispersive bands along the *b* direction.[67] Concomitantly, strongly anisotropic transport properties are observed within the in-plane directions, with high conductivity values along the *b* direction but an insulating character along the *a* axis leading to a 1D electronic transport.[68]

Regarding the magnetic properties, $CrSBr$ also shows a substantial in-plane uniaxial magnetic anisotropy, with the easy magnetic axis lying in-plane along the *b* direction.[66,69] Altogether, the highly anisotropic electronic, magnetic and transport properties make this system being better described as a quasi-1D system.[68-70] Moreover, an anomalous change on the sign of the magnetoresistance is observed below T* = 40 K,[71] followed by a subtle increase in the magnetization, suggesting additional magnetic complexity at low temperature. We studied the origin of this hidden order by combining neutron powder diffraction (NPD), synchrotron X-ray diffraction, magnetization measurements, and μSR. Our results indicate that $CrSBr$ displays a complex dynamic magnetism,[69] enlarging its potential application as a component for spin-based electronic devices.

$CrSBr$ thus stands as a promising van der Waals magnet with a strong uniaxial character in the magnetic, structural, as well as in the transport properties. Furthermore, the robustness of the high temperature magnetic correlations within the monolayers,[69] and the additional magnetic complexity at low temperature, may open the door for exploring new applications, such as ultra-compact spintronics. On a broader scope, we believe that the inclusion of mixed-anion chemistry stands as a promising route for the design of new van der Waals materials with low dimensional magnetic character.

6. Conclusions and Outlook

In this overview we have highlighted some of our recent work on quantum materials discovery. Thereby, we have put special emphasis on the challenges that this field of research is currently facing regarding the conceptualization, as well as the realization and synthesis of metastable phases. Specifically, we have discussed (i) the case of the hydrothermal synthesis of the metastable marcasite FeS_2 polymorph that allowed us to precisely measure its semiconducting properties, (ii) the discovery of new superconductors and superconducting properties by a void-filling approach in η-carbide-type phases, (iii) the challenges regarding the preparation of phase-pure samples of different polytypes in the NbS_2 and $NbSe_2$ systems, and (iv) the realization of next generation van-der-Waals and 2D magnets by a mixed-anion chemistry approach. These studies exemplify the importance of chemical design and chemical synthesis for the realization of these highly complex materials that are needed for the next generation of technologies, *i.e.* quantum sensing and quantum technologies in general. At the same time these materials display a large variety of intriguing, intertwined properties that are of fundamental interest.

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