Designer quantum matter in van der Waals heterostructures

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Van der Waals materials can be easily combined in lateral and vertical heterostructures, providing an outstanding platform to engineer elusive quantum states of matter. However, a critical problem in material science is to establish tangible links between real materials properties and terms that can be cooked up on the model Hamiltonian level to realize different exotic phenomena. Our review aims to do precisely this: we first discuss, in a way accessible to the materials community, what ingredients need to be included in the hybrid quantum materials recipe, and second, we elaborate on the specific materials that would possess the necessary qualities. We will review the well-established procedures for realizing 2D topological superconductors, quantum spin-liquids and flat bands systems, emphasizing the connection between well-known model Hamiltonians and real compounds. We will use the most recent experimental results to illustrate the power of the designer approach.

I. INTRODUCTION

Two-dimensional materials are at a focus of intense research efforts, with the paradigmatic examples of graphene, hexagonal boron nitride, transition metal dichalcogenides, and transition metal trihalides. The genuine interest in these materials stems from the many high-quality synthesis possibilities, together with the richness of different behaviours. These compounds have been shown to realize properties starting from conventional insulating and metallic behaviour, all the way up to complex many-body ground states such as superconductors and topological insulators.

Besides their intrinsically interesting properties, layered 2D vdW materials can be easily combined in lateral and vertical heterostructures. As the layers only interact via the weak vdW forces, the individual layers can retain their intrinsic properties. This property alone allows creating combinations of electronic orders that no naturally occurring material possesses. This possibility has given birth to the field designer quantum materials, where heterostructures are exploited to realize elusive quantum phases of matter not present in conventional compounds. In this review, we present a quantum materials cookbook point of view on how to achieve this and use three elusive quantum states engineered in vdW heterostructures as examples: topological superconductors, quantum spin-liquids and flat band systems.

The creation of topological superconductivity represents the first paradigmatic example of the possibilities brought by this flexibility. It is well known that topological superconductivity can be artificially engineered by combining s-wave superconductivity, spin-orbit effects, and magnetism. Materials with these properties can be combined in heterostructures of 2D materials by using layered superconductors, monolayer magnetic materials, and strong spin-orbit effects as the necessary ingredients of realizing topological superconductivity.

A second example consists of engineered quantum spin-liquids, highly entangled quantum magnets. The emergence of quantum spin-liquids is known to require a fine-tuning between spin-interactions, which is one of the limitations to finding them in non-tunable compounds. VdW heterostructures provide a way around this, with their possibility of finely tuning magnetic interactions in a two-dimensional magnet by a proper choice of 2D substrate. We will also discuss the prospects of realizing QSL in artificial systems.

Finally, as the third example, we discuss how combining two-dimensional materials allow us to create dramatically new electronic dispersions beyond simple superposition of the electronic orders of parent compounds. The most dramatic case of this consists of the emergence of flat bands from a material with highly dispersive electrons. This is exemplified by structurally engineered on-surface graphene structures, and the whole family of twisted vdW heterostructures.

II. ARTIFICIAL VDW TOPOLOGICAL SYSTEMS

The engineering of novel topological states of matter[1, 2] represents one of the milestones of current materials engineering. While a variety of natural topological compounds have been identified in nature[3], artificial engineered systems open new prospects for potential technological applications with common compounds. Ultimately, this topological engineering can ultimately lead to the realization of states that no natural compound hosts. Topological states of matter encompass a wealth of states, including crystalline, higher-order and quasiperiodic topological states. Here, we will focus on two paradigmatic cases, namely quantum anomalous Hall insulators[4] and topological superconductors[5]. These two topological states represent critical milestones for the fields of electronics and topological quantum computing, respectively.
A. Artificial topological superconductors

The creation of topological superconductivity\cite{7} represents the first paradigmatic example of the milestones of artificial engineering\cite{5}. It is well known that topological superconductivity can be artificially engineered by combining s-wave superconductivity, spin-orbit effects, and magnetism\cite{5, 8–12}. Materials with these properties can be combined in heterostructures of 2D materials\cite{13} by using layered superconductors\cite{14}, monolayer magnetic materials\cite{15, 16}, and strong spin-orbit effects as the necessary ingredients to realize topological superconductivity.

Topological superconductivity represents one of the most pursued quantum states of matter in modern condensed matter physics. Besides the interest in this state sparked from the rise of topological insulators, topological superconductors represent one of the potential cornerstones for topological quantum computing\cite{17–19}. However, topological superconductors are extremely rare in nature, and thus a great amount of experimental efforts have been focused on engineering this state in a variety of platforms\cite{5, 12}. The fundamental requirements for creating topological superconductivity rely on creating an effective superconducting spin-triplet state\cite{7, 20} starting from a conventional spin-singlet s-wave superconductor. This can be achieved by creating fine-tuned spin textures in materials combining exchange fields and strong Rashba spin-orbit coupling. Based on this idea, a variety of proposals and realizations have been demonstrated in the last years in semiconducting nanowires\cite{9, 19, 21, 22}, atomically engineered chains\cite{23–30} and topological insulators\cite{10, 31}. In all these systems, the critical emphasis is put on combining different materials containing magnetism and superconductivity, a task in which interface physics is known to play a critical role\cite{32}. Two-dimensional materials provide a unique opportunity in this direction, due to the weak van der Waals forces that allow combining different layers\cite{13, 16}, namely superconducting and magnetic, on a single van der Waals heterostructure.

The requirement of these different order parameters to engineer a topological superconductor can be easily rationalized. In short, engineering topological superconductivity requires creating an effective spinless superconductor, whose minimal model gives rise to a topological superconducting state. For this sake, let us start with the simplest model for topological superconductivity: the one-dimensional Kitaev model\cite{7}. This model considers spinless electrons on a one-dimensional chain in the presence of a finite first nearest-neighbor pairing, whose Hamiltonian takes the form

$$H = \sum_n t c_n^\dagger c_n + \sum_n \Delta c_n c_{n+1} + \text{h.c.} \quad (1)$$

It should be noted that for spinless fermions, on-site superconductivity is forbidden from the fermionic anticommutation relations. The previous Hamiltonian in Fourier space takes the form

$$H = \sum_n t \cos(ka) c_n^\dagger c_k + \sum_n \Delta \sin(ka) c_k c_{-k} + \text{h.c.} \quad (2)$$
giving rise to a fully gapped eigenspectra \( \epsilon_k = \sqrt{t^2 \cos (k a)^2 + \Delta^2 \sin (k a)^2} \). Despite its fully gapped structure, solving the Hamiltonian Eq. 1 with open boundary conditions gives rise to a zero mode, which in the case of the exactly solvable point \( \Delta = t \) has an associated eigenstate of the form \( \gamma = \frac{1}{2} (c_0 + c_0^\dagger) \). For finite chemical potential \( \mu \), an exponentially localized zero mode exists, yet with a more complex spatial profile. In contrast with conventional fermions, this creation operator is its own dagger \( \gamma = \gamma^\dagger \). This implies that these particles are their own antiparticles, which is expressed in this model through this mathematical property, as expected from a Majorana operator. Similar models can be written for a two-dimensional system, in which case the single-Majorana mode becomes a propagating Majorana edge state in an otherwise fully gapped spectrum.

The central question of artificial topological superconductivity is to find procedures of engineering an effective spinless superconductor, starting from spin-singlet superconductivity is to find procedures of engineering an effective edge state in an otherwise fully gapped spectrum. In this model through this mathematical property, as expected from a Majorana operator. Similar models can be written for a two-dimensional system, in which case the single-Majorana mode becomes a propagating Majorana edge state in an otherwise fully gapped spectrum.

The typical recipe for achieving topological superconductivity rely on designing a pseudo-helical electron gas[9, 10] (states crossing the Fermi level have a spin that is locked to their momentum, i.e. a certain momentum implies certain spin direction), yielding an effective single degree of freedom and with a finite projection on the spin-singlet state above, which interestingly could be directly engineered with two-dimensional van der Waals topological insulators[33]. The previous idea implies that the electronic modes must have a finite spin-momentum coupling so that the propagation direction depends on the spin channel. Such spin-momentum coupling can be realized by different forms of spin-orbit coupling[9, 10], or by exploiting non-collinear magnetic textures[23, 34]. It is interesting to note that these strategies work both in one and two-dimensions, and as a result, recipes for one-dimensional topological superconductivity can easily be generalized to two dimensions.

The typical recipe for achieving topological superconductivity is illustrated in Fig. 1a. Starting with a parabolic band, the addition of Rashba-type spin-orbit coupling and magnetization creates the type of band structure required for TSC as explained above. The addition of superconductivity completes the requirements and results in a system that realizes the phase diagram shown in Fig. 1b. When the chemical potential is tuned to the band crossing point at \( k = 0 \), even a very small magnetization is sufficient to drive the system into the topological phase. If the chemical potential is tuned away from this point, then stronger magnetization is needed. Although this procedure requires very precise fine-tuning between the system parameters, it has been successfully demonstrated for a variety of semiconductor devices[21, 22], and van der Waals materials[6]. Finally, it is worth emphasizing that besides the Majorana edge modes, topological superconductors are also expected to show Majorana excitations at domain walls[35, 36] and vortices[37–40].

### B. Materials for artificial topological superconductors

The section above lays out the rather stringent requirements for realizing topological superconductivity, and we need materials that will retain their magnetic and superconducting properties in a heterostructure. This strongly suggests using vdW materials: this allows for a rational design of the heterostructure as we expect to retain the intrinsic properties of the different constituents. Topological superconductivity has been realized in atomic-scale structures using conventional materials (e.g. iron atom chains on a lead or rhenium substrates[24–27], cobalt islands under a Pb monolayer, and iron islands on an oxygen-terminated rhenium substrate[32, 41]). However, considering the strong chemical bonding between the materials in this case, these systems are susceptible to disorder, and interface engineering might be required in some cases[32].

While many monolayer ferromagnet materials are available for exfoliation (e.g. CrI\(_3\)), they are very reactive, and accessing the topological edge modes in scanning probe microscopy and other experiments requires the system to have very clean edges. This points out towards the use of e.g. molecular-beam epitaxy (MBE) growth and luckily high-quality growth of several materials has been demonstrated (Fe\(_3\)GeTe\(_2\)[42], CrBr\(_3\)[43, 44]). For the superconductor material, typical suggestions would include the 2H phase of the NbS\(_2\), NbSe\(_2\), TaS\(_2\), and TaSe\(_2\)[45, 46]. The scheme for realizing TSC is also applicable to bulk superconducting substrates, there the magnetic layer will couple strongly to the top layer of the SC, and as long as the substrate has relatively weak interactions between the layers, it is expected to work similarly to the monolayer case[6, 47]. These real materials have hexagonal symmetry, which is reflected in the band structure. Instead of a single high-symmetry point in the Brillouin zone, there are several (\( \Gamma, M, \) and \( K \) points), and the topological superconducting phase can be realized at any of these points. This means that tuning the Fermi level across the relevant band (e.g. the Nb d-band in the case of NbSe\(_2\)), there are three different topological phases that have different Chern numbers as illustrated in Fig. 1c. In a real vdW heterostructure, the doping of the substrate will determine whether the system will enter a topological phase.

This route to TSC has been realized experimentally in CrBr\(_3\) / NbSe\(_2\) heterostructures[6, 44]. As can be seen from the calculated Nb d-band bandstructure shown in Fig. 1c, the \( M \) point is closest to the Fermi level, and it is likely that the topological phase arises from this point. Experimentally, the strongest signature is the
Majorana edge modes that appear at the interface between the trivial and topological phases. This is shown in Fig. 1d, which shows an STM topographic image of CrBr$_3$ island on a bulk NbSe$_2$ substrate and three d$I$/d$V$ spectra (the signal is proportional to the local density of states, LDOS, at the position of the STM tip): on the NbSe$_2$ substrate (blue), on the CrBr$_3$ island (red) and right at the edge of the island (green). The spectrum recorded on the island edge has a strong peak centered around the Fermi level (zero bias) consistent with the expected LDOS corresponding to the Majorana zero modes. Fig. 1e shows the measured (left) and theoretical LDOS (right) as a function of the energy. At the Fermi energy, both the bulk phases are gapped, and only the Majorana modes at the edges of the islands are visible. As the energy is increased, we eventually start to see excitations in the topological superconductor with the edge modes overlapping with bulk states. Finally, above the superconducting gap, all significant LDOS contrast is lost.

Comparison between theory and experiment allowed estimating the values of the model parameters, namely, the induced magnetization in the top NbSe$_2$ layer due to the proximity of the CrBr$_3$ layer and the magnitude of the Rashba spin-orbit coupling. These estimates suggest that the magnetization and the spin-orbit coupling are of a similar magnitude, a few tens of meV. This values were also consistent with density-functional theory (DFT) calculations and in-line with proximity induced exchange coupling in CrI$_3$/WSe$_2$ and CrBr$_3$/MoSe$_2$ heterostructures [48, 49]. Finally, the moiré pattern between CrBr$_3$ and NbSe$_2$ was suggested to further stabilize the topological superconducting state[50].

C. Artificial Chern insulators

Chern insulators [54] represent another paradigmatic state of matter in two-dimensional systems. Besides their conventional engineering by combining spin-orbit coupling and exchange fields [4], van der Waals materials offer a novel approach to engineered Chern insulators. This new approach to engineer Chern bands specifically exploits moiré patterns in twisted two-dimensional materials. The emergence of flat bands stems from a non-abelian elastic gauge field and will be further addressed in more detail in section IV E. As illustrated in Fig. 2a-c, the varying lattice registry in twisted bilayer graphene creates a long-wavelength moiré pattern. This moiré modulation creates moiré mini-Brillouin zones at the $K$ and $K'$ points of the two graphene layers. These valleys are well-separated, and each valley hosts two Dirac cones of the same chirality. The Dirac cones living at the $K^1$ and $K^2$ (and at $K'^1$ and $K'^2$) hybridize, and when the respective Dirac points are sufficiently close to each other in the $k$-space, this hybridization results in the formation of a flat band with narrow bandwidth that is well separated from other bands is formed as the twist angle $\theta$ is tuned across the magic angle (Fig. 2c). This emergence of flat bands in twisted graphene bilayers is the starting point for realizing the Chern insulator states[55, 56].

Flat bands in twisted bilayers can be interpreted as pseudo-Landau levels of an artificial gauge field, generated by the modulated stacking in the unit cell[57]. Close to charge neutrality, these flat bands yield an 8-fold manifold, two-fold degeneracy coming from electron-hole states, two-fold coming from valley symmetry and two-fold coming from spin[55, 56, 58, 59]. As Landau levels[57, 60], each flat band is expected to carry a non-trivial Chern number, analogous to conventional Landau levels of quantum Hall states[61]. However, the original system is time-reversal symmetric, implying that flat bands stemming from opposite valleys will carry opposite Chern numbers[57, 62]. This property suggests that if valley symmetry is spontaneously broken, for example, due to electronic interactions, twisted graphene bilayers become natural Chern insulators[63]. The breaking of valley symmetry takes place when electronic interactions create a spontaneous symmetry breaking, leading to a filling of just on the the valley flat bands. A specific feature that must be taken into account is that due to the existence of Dirac points in the electronic structure[55, 56, 58, 59], leading to the Chern insulator regime requires to first opening a gap at the Dirac points[63–65]. This is done by taking aligned hBN layers with the twisted bilayer that induce a small symmetry breaking in the twisted bilayer lifting the original Dirac points. Ultimately, in the presence of partial filling, this could lead to the emergence of fractional Chern states [66].

D. Materials for artificial Chern insulators

These predictions were realized in twisted graphene bilayers with the twist angle ($\theta = 1.15^\circ$) tuned to yield flat bands in the electronic spectrum [53, 55, 56]. The sample fabrication followed the usual “tear and stack” process [52, 67, 68], but TBG was aligned with the underlying h-BN layer. The alignment with BN turns out to be critical in lifting the low energy Dirac points, allowing for the emergence of a valley polarized state. These state-of-the-art devices typically use TBG encapsulated by h-BN layers, and atomically smooth graphite flake is used as the gate electrode (see Fig. 2a). Finally, the stack is electrically contacted using so-called edge contacts, which have high transparency and avoid unwanted doping of the TBG [51]. Fig. 2d shows the longitudinal ($R_{xx}$) and the Hall ($R_{xy}$) resistances measured as a function of the carrier density on a magic angle TBG device at $T = 1.6$ K and under an external magnetic field of $B = +150$ mT [53]. As expected for a quantum Hall state, $R_{xx}$ reaches $\hbar/e^2$ and $R_{xy}$ approaches zero when the electron density is tuned to filling factor $\nu = 3$ ($\nu$, where $\nu$ is the number of the electrons in the flat band per moiré unit w.r.t. no external doping, i.e. $\nu$ can have values between -4 and 4). The previous phenomena
are the hallmarks of the QAHE state, and most importantly it is retained in the absence of the field as shown in Fig. 2 - The Hall resistivity is hysteretic (Fig. 2e), with a coercive field of several tens of millitesla. The Hall resistivity is quantized ($R_{xy} = h/e^2$) and the longitudinal resistivity remains small through zero external magnetic field, which demonstrates that the quantum anomalous Hall state is stabilized by spontaneously broken time-reversal symmetry. In particular, this time-reversal symmetry breaking is purely associated to the valley sector, where spontaneous symmetry-breaking leads just one of the valley filled as sketched in Fig. 2f. Finally, it worth to note that it is quite typical in the TBG experiments that the observed phenomena are device-specific, with minor differences in the device parameters being decisive which states are formed. For example, robust, thermally activated, trivial insulator behavior and the QAHE state can occur in very similar devices.

Typically, the Chern number can be estimated from the value of the quantized Hall conductance, but this measurement requires working on a transport setup, and it would be extremely interesting to be able to somehow measure the Chern number directly and independently. Precisely this was done by a scanning tunneling microscopy-based technique to directly measure the Chern numbers of the different Chern insulating states [69]. The topological gaps can be identified by measuring the LDOS as a function of the electron density in the system (controlled through external doping) at different external magnetic fields. If the electron density at which gap opening and closing takes place depends on the external magnetic field, the transition can be identified as a topological transition. Tracking the electron densities at which these transitions happen as a function of the magnetic field $B$, gives direct access to the associated Chern number $C$ via $\frac{\partial n}{\partial B} = C/\Phi_0$, where $\Phi_0$ is the magnetic flux quantum. In addition to a host of levels arising from the zeroth Landau level at $\nu = 0$ with Chern numbers $C = 0, \pm 1, \pm 2, \pm 3, \pm 4, \pm 8, \pm 12$, the authors observe a hierarchy of correlated Chern insulating phases with Chern numbers $C = \pm 1, \pm 2, \pm 3$ emerging as a function of magnetic field from the different filling factors $\nu = \pm 3, \pm 2, \pm 1$, respectively. All these phases are stabilized by a magnetic field.

In addition to the example above, the Chern insulating state and the quantum anomalous Hall effect has also been realized in rhombohedral (ABC-stacked) graphene trilayers and twisted monolayer - bilayer graphene samples [70–72] and recent experiments on magic-angle bilayers also suggest the possibility of realizing fractional Chern insulator states [73].

III. VDW QUANTUM SPIN-LIQUIDS

Quantum spin-liquids[78, 79] are highly entangled quantum magnets, characterized by the emergence of novel fractionalized particles. These many-body states are classified to their pattern of long-range entanglement. In terms of their excitation spectrum, quantum spin-liquids can be classified in gapped or gapless, and in a minimal picture, this is ascribed to a gapless or
A. QSL from frustrated anisotropic interactions

A first strategy to design quantum spin-liquids is to focus on models showing highly directional interactions that are inherently frustrated [85]. This can be done, for example, by taking square [86] or honeycomb lattices [85], and imposing anisotropic spin-spin interactions that are dependent on the bond considered. As magnetic interactions in materials are rotational symmetric in the absence of spin-orbit coupling, these mechanisms are expected to be realized in materials containing heavy atoms in which spin-orbit effect compete and even overcome other interactions present [87–90].

Due to their interacting nature, the solution of quantum spin-liquid models represents one of the open problems in many-body physics. A great amount of insight can, however, be obtained from finely tuned models that allow for an exact solution. Among these specially tuned models, we encounter the Toric code and the anisotropic Kitaev honeycomb model [85]. In particular, the Kitaev model realizes a highly anisotropic spin model in a honeycomb lattice that takes the form

\[ H = \sum_{\langle ij \rangle} S_i^\gamma S_j^\gamma \]  

where \( \langle ij \rangle \) denote first neighbors and \( \gamma \) labels the spin-component that interacts for each bond as depicted in Fig. 3a). The genuine feature of the Kitaev honeycomb model stems from the possibility of obtaining an exact solution in terms of single particle excitations. Remarkably, the single-particle excitation are of Majorana type, and depending on the parameter regime, realize gapless or gapped Majorana states [85].
B. Experiments QSL with anisotropic interactions

Interestingly, the Kitaev honeycomb model [85] (illustrated in Fig. 3a) can be potentially adiabatically connected to quantum spin-liquid states realized in $\alpha$-RuCl$_3$ (RuCl$_3$) [74, 89, 91–93], and thus in the following we will focus on this compound. RuCl$_3$ is a layered Mott insulator with significant spin-orbit interactions that is in the close proximity to the quantum spin-liquid ground state [66, 74, 75]. However, these materials often host complex Hamiltonians having several contributions beyond the Kitaev exchange, including first, second and third neighbor exchange, and symmetric off-diagonal exchange [74, 94–96]. The model typically employed for this compound gives rise to the phase diagram sketched in Fig. 3b as a function of the first neighbor couplings, keeping the second and third neighbor exchange finite [74]. In Fig. 3b, the x-axis represents the ratio of the Heisenberg ($J$) to Kitaev -type ($K$) spin coupling and $\Gamma$ is symmetric off-diagonal exchange coupling. The whole diagram has been evaluated with the ratio of Hund’s coupling ($J_H$) to the Coulomb on-site interaction ($U$) of $J_H/U = 0.2$, which can be estimated from ab initio calculations. It can be seen that the phase diagram hosts ordered magnetic phases ranging from ferromagnetic (FM) and antiferromagnetic (AFM) to more complicated zigzag (ZZ), 120 and incommensurate order (I) phases, even without considering variations in the further neighbor exchange. The best estimate for the parameters corresponding to bulk RuCl$_3$ is shown as a red diamond. This implies that the ground state of RuCl$_3$ is actually an ordered magnetic phase, which has been experimentally confirmed using, e.g., thermal Hall conductance measurements [77].

Remarkably, it was experimentally demonstrated that applying a sufficient in-plane magnetic field can destroy the long-range order of the magnetic ground state and give rise to quantum spin-liquid behavior [77]. This is illustrated in the phase diagram shown in Fig. 3c, where the boundaries of the different phases have been followed using thermal Hall conductance measurements. Interestingly, the Majorana edge modes that arise in the quantum spin-liquid ground state can be directly verified as half-integer quantized thermal conductance, which is observed in the region shaded with red in Fig. 3c. Further increasing the lateral magnetic field gives rise to a phase transition to some other non-topological phase.

In addition to the application of a lateral magnetic field, many other routes (e.g. external pressure and chemical doping) are being tested to suppress magnetism, enhance the pure Kitaev interactions and drive the system towards the quantum liquid state. In the spirit of the designer material principles, we highlight a couple of theoretical ideas where heterostructures could be used to promote the quantum spin-liquid state. It has been proposed that monolayer RuCl$_3$ on graphene (illustrated in Fig. 3d) would result in a system with enhanced Kitaev type interactions [75, 76]. By using ab initio calculations, it was shown that the RuCl$_3$ becomes strained and doped in this heterostructure. This might even drive an insulator-to-metal transition and help to realize predicted, exotic superconducting states in quantum spin-liquids [75, 101]. In any case, the strain and doping are predicted to enhance the Kitaev interactions (increasing $K$ and decreasing $J$ and $\Gamma$) and move the system closer to the Kitaev. RuCl$_3$/graphene heterostructures have been also realized experimentally [102, 103], but not yet down to the monolayer limit. However, experiment on thicker RuCl$_3$ layers already give indications of the charge transfer and hybridization between the RuCl$_3$ and graphene bands [102, 103].

C. QSL from geometric frustration

Geometric frustration can lead to spin-liquid behavior and considering a simple picture of three spins can give a flavor of the general idea: Consider three spins at the corners of a triangle with antiferromagnetic interactions. This system does not have a configuration where all antiferromagnetic interactions can be simultaneously satisfied, i.e. the system is frustrated. This geometric frustration leads to unusually large ground state degeneracies, already at the classical level. The situation described above corresponds to a classically frustrated system, in which quantum entanglement between sites is not considered and corresponds to the so-called spin ice models. In the quantum realm, an effective strategy to realize quantum spin-liquid physics is to focus on models realizing non-bipartite lattices, such as triangular and kagome lattices. Kagome lattice models [104–106] have been known to be a paradigmatic platform for quantum spin-liquid physics. Since triangular lattices are often more common in the van der Waals world, we will in the following focus on that case. Focusing on the triangular lattice model, in the minimal case in which only first neighbor interactions are considered, the ground state is actually an ordered state with 120 degrees spin spiral [107–109]. However, this model can be pushed to a more frustrated regime by including additional interactions [110, 111], and in particular, a second neighbor exchange coupling [112], driving the system to a quantum spin-liquid ground state. Although an exact solution cannot be obtained in this limit, tensor network calculations have shown strong signatures of a gapless QSL liquid state in this regime, featuring gapless Dirac spinons [112].

The low energy excitations of these models in terms of chargeless emergent fractionalized excitations with $S = 1/2$ known as spinons. We start with a Heisenberg model of the form $H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$ where $\vec{S}_i$ are the local spin operators. Assuming a quantum spin-liquid ground state, we can express the localized spins as emergent chargeless $S = 1/2$ fermions of the form $S^\alpha_i = \sum_{s,s'} \sigma^\alpha_{s,s'} f_{i,s}^\dagger f_{i,s'}$, where $f_{i,s}^\dagger$ denotes the creation operator of a fermionic spinon in site $i$, and $\sigma^\alpha_{s,s'}$ are the spin Pauli matrices. The localized moment is implemented by enforcing having a
FIG. 4. (a) Schematic of a quantum-spin-liquid with frustration stemming from geometric frustration. (b) STM image recorded on a bulk 1T-TaS₂ surface showing the characteristic “Star-of-David” charge-density wave pattern [97]. (b) Calculated band structure of 1T-TaS₂ with the $\sqrt{13} \times \sqrt{13}$ “Star-of-David” reconstruction and including the Ta spin-orbit coupling reveals a single distinct split-off band at the Fermi level (green line) [98]. Including the Hubbard U-term splits this band into a lower and upper Hubbard band (d) STM $dI/dV$ measurements on a related compound 1T-TaSe₂ monolayer showing LHB and UHB [99]. The panels on the right show constant height $dI/dV$ maps that allow direct visualization of the LHB and UHB wavefunction symmetries. (e) Illustration of the low-temperature state of 1T-TaS₂ with spatially random pairs of “Star-of-Davids” forming singlets. It is possible to form gapless, low-energy fractional excitations (blue arrows) [100].

The necessary ingredients for a QSL - triangular lattice with frustrated magnetism (Fig. 4a) - can be realized in van der Waals materials. This has been demonstrated in the 1T phase of TaS₂ (1T-TaSe₂ is expected to be similar), where the presence of various charge-density wave (CDW) states (depending on the temperature) has been known for some time [114–116]. The low-temperature CDW state results in a $\sqrt{13} \times \sqrt{13}$ reconstruction of the 1T-TaS₂ lattice that has a 13 Ta atom “star of David” unit cell [97, 98, 114–116] as illustrated in Fig. 4b. This causes folding of the band structure and, together with modified hoppings caused by the reconstruction and the presence of spin-orbit coupling, results in a single band with a relatively flat dispersion at the Fermi level [98]. In the presence of strong electron-electron interactions ($U$ larger than the bandwidth of band at Fermi level), the system will undergo a Mott metal-insulator transition and instead of the single band at the Fermi level, there will be a fully occupied lower Hubbard band (LHB) below the Fermi level and a fully unoccupied upper Hubbard band (UHB) above it.

D. Experiments on geometrically frustrated QSLs

The single fermion in each site $f^\dagger f = 1$. At the mean field level, the Heisenberg Hamiltonian a tight binding model of free propagating $S = 1/2$ spinons of the form

$$H = \sum_{ij} \chi_{ij} f_{i,s}^\dagger f_{j,s'}$$

where $\chi_{ij}$ are the mean-field parameters of the mean-field Hamiltonian. The spinon excitations of the quantum spin-liquid state can thus be understood from the spinon dispersion. For example, gapless Dirac quantum spin-liquid have an associated spinon model featuring Dirac points,[112] whereas models with a finite spinon Fermi surface are stem from model with a finite Fermi surface. This classification is often used when characterizing quantum spin-liquid ground states, and has direct impact on the temperature-dependence of the thermal conductivity[113].
In the case of 1T-TaS₂, this Hubbard band correspond to a single unpaired electron per CDW “star of David” unit cell, which are the building block of the quantum spin-liquid state in this material. The Hubbard bands have been demonstrated in bulk 1T-TaS₂ [97, 118] and also in monolayer 1T-TaSe₂ [99]. As illustrated for 1T-TaSe₂ in Fig. 4d, tunneling spectroscopy allows direct verification that the system is gapped and the energies of the LHB and UHB can be easily probed. In addition, by mapping the spatial variation of the tunneling conductance $dI/dV \propto \text{LDOS}$, the spatial symmetries of the states can be probed. In the case of 1T-TaSe₂ monolayer, it can be seen that the orbital texture of LHB and UHB are different (right side of Fig. 4d).

While STM and tunneling spectroscopy can be used to probe the Hubbard bands, it is difficult to directly use these techniques to probe the spin-liquid state. This is usually done with neutron scattering, where the “smoking gun” for the QSL state is the lack of magnetic order down to the lowest temperatures. The other option is muon-spin-relaxation, which has been applied to bulk 1T-TaS₂ to show that the spin excitations are gapless, and there is no long-range order in temperatures of at least down to 70 mK [100]. Those experiments show that below 55 K, there is a broad distribution of the relaxation times indicating a highly inhomogeneous magnetic phase at all Ta sites. This is strong evidence that there is growing randomness in the spin system as temperature decreases below 55 K. The observed slowing down of spin fluctuations is consistent with the freezing of singlets as illustrated in Fig. 4e. Interestingly, for 1T-TaS₂ the resonant valence bonds are formed between magnetic moments with an extension of the enlarged unit cell generated by the CDW, in comparison with the atomic-like moments of bulk QSL candidates.

The problem with the bulk probes such as neutron scattering or muon spin-relaxation is that they are typically not sufficiently sensitive to probe monolayer samples. There are theoretical suggestions that tunneling spectroscopy could be used for this even though the magnitude of the predicted signal would depend on the measurement geometry (e.g. 2D junction vs. STM) and the type of the spin-liquid [120]. In addition, and despite their chargeless nature, signatures of spinon interference can be potentially probed by inelastic transport spectroscopy [121]. Finally, muon spin-relaxation has been used to probe the Kondo effect with spinons [122], by probing the existence of a spinon-Kondo cloud around magnetic impurities, and this technique could perhaps be extended to monolayer samples.

IV. NEW VDW FLAT BANDS

The engineering of flat bands has been at the forefront of condensed matter physics for a long time. Flat band systems are characterized by having almost dispersionless states, which in the presence of any residual interactions are expected to be prone to a variety of electronic instabilities [123–125]. In the following, we will discuss several directions that van der Waals materials provide towards the realization of flat band systems.

A. Generating flat bands from geometric frustration

The simplest instance in which flat bands appear in electronic systems are tailored lattices leading to destructive interference [123, 126]. Paradigmatic examples of these flat band models are Lieb and kagome lattices [126]. In this system, electron propagation is quenched due to the existence of complementary paths that interfere destructively. This destructive interference can be often weakened by adding additional perturbations. For example, next nearest neighbour (NNN) hoppings interactions will often cause the flat bands to acquire dispersion, as flat bands are localized eigenstates on “disconnected” lattice sites and NNN hoppings connect these sites and make the flat band dispersive. In the case of the Lieb lattice, the existence of a flat band can also be understood from Lieb’s theorem [124]. In its general form, this theorem states that for a fully bipartite lattice, the number of flat bands will be $|N_A - N_B|$, [124] where $N_A$ is the number of removed sites from sublattice $A$, and $N_B$ the number of sites removed of sublattice $B$. In particular, this implies that generic bipartite lattices in which one site is removed will show a flat band. The Lieb lattice can be built by removing one site from the square lattice, leading to the existence of a flat band. In this very same fashion, other flat band models can be systematically constructed by removing a certain number of sites. These types of flat bands have been realized in artificial systems based on atomic lattices [127–131]. They can also be formed in suitable engineered, chemically synthesized lattices, where covalent organic frameworks and metal-organic frameworks are especially attractive systems for realizing these artificial models [132–140].

B. Experiments flat bands from frustration

There have been many theoretical proposals on metal-organic frameworks (MOFs) with kagome structure that should result in flat bands in their band structure [132, 133, 135]. However, the experimental demonstration has proven difficult. If the assembly is carried out directly on a metal substrate (typically Au(111), Ag(111) or Cu(111)), it is relatively straightforward to realize MOFs with a large degree of structural perfection [134]. Unfortunately, the relatively strong interaction with the underlying metal substrate typically masks the intrinsic electronic structure of the MOF. On the other hand, the formation of the high-quality MOFs on weakly interacting substrates is much more challenging [141, 142].
and unambiguous proof of the flat bands has not been demonstrated.

The other chemical strategy for synthesizing two-dimensional networks on surfaces relies on the formation of covalent carbon-carbon bonds and structures called covalent organic frameworks (COFs). While there are extensive results on the formation of the one-dimensional nanocarbons (graphene nanoribbons) [131, 143–147], challenges remain to create two-dimensional assemblies with very high quality [148–150]. However, there are recent experimental results that are pushing this field towards higher quality samples towards the formation of flat bands in the MOF or COF band structure [119, 140, 142, 151]. The realized strategies rely on making a molecular network with a kagome lattice with one of the examples highlighted in Fig. 5g-i. In particular, on-surface polymerization was used to realize a high-quality two-dimensional polymer poly-trioxaazatriangulene network [119]. This sample was of sufficiently high quality to allow angle-resolved photoemission spectroscopy (ARPES) experiments that can be used to directly probe the structure of the occupied bands as shown in Fig. 5. This shows the folded bands of the valence band of the polymer that match the expected results well (calculated bands shown by dotted lines). The kagome flat band is expected to be at the bottom of the conduction band and cannot be directly probed by ARPES experiments. These results are along the path towards tuneable 2D organic or metal-organic structures with engineered flat bands. The incorporation of metal atoms with magnetism or a large spin-orbit interaction opens additional possibilities in realizing topological materials [132, 133, 152, 153].

C. Generating flat bands from long wavelength modulations

A simple way of generating nearly flat bands consists of weakly coupling quantum dot states. In this picture, the bandwidth is determined by the coupling between the quantum dots - the weaker it is, the flatter the resulting bands will be. A convenient way of achieving this in a real material in a large scale is by exploiting moiré patterns [154–160]. The fundamental idea relies on the locally modulated stacking over the moiré pattern that causes a spatial modulation of the conduction and valence band edges and leads the formation of a large scale array of quantum dots in twisted van der Waals superlattices. The mechanism for flat band generation can be rationalized from the decoupled limit, in which the system consists of decoupled quantum dots. The twist angle between the layers changes the size and separation between the quantum dots, promoting a finite hybridization between them that leads to nearly flat bands[159, 161, 162]. It is worth noting that this mechanism holds when there is a bandgap in the original materials (e.g. twisted h-BN and twisted dichalcogenide systems). This mechanism also requires the existence of a confinement gap. As a result, semimetals like graphene, in which electrons cannot be electrostatically confined, require a different mechanism for flat band generation. We will illustrate the use of gauge fields for this in section IV E.

D. Experiments flat bands from quantum dots

As we discussed above, flat bands can be realized in gapped, twisted moiré systems and this has been demonstrated in several experiments. An early experiment by
Zhang et al. relied on direct growth of rotationally aligned MoS₂/WSe₂ heterostructure, where the lattice mismatch then creates a moiré pattern [163]. While not directly resolving the flat bands spectroscopically, they demonstrated that the system had the necessary ingredients for their existence: the modulated interlayer coupling giving rise to a modulation of the conduction and valence band edge energies. They showed that the valence and conduction band edges are located at different layers and that the local bandgap was periodically modulated with an amplitude of $\sim 0.15$ eV, leading to the formation of a two-dimensional electronic superlattice.

The flat bands were directly identified in a later study [164], which concentrated on a twisted bilayer WSe₂ samples with twist angles of $3^\circ$ and $57.5^\circ$. By using scanning tunnelling spectroscopy, it was possible to directly map the spatial extent of the wavefunctions at the flat-band energy and to show that the localization of the flat bands depends on the twist angle. The observed flat bands originated from the highest valence band at the $\Gamma$ point (the conduction band onset varies very little over the moiré pattern and hence does not result in the type of quantum dot states required for the formation of the flat bands). The flat band in $3^\circ$ twisted bilayer is localized on the hexagonal network separating the AA sites where as in the $57.5^\circ$ systems, it is localized on the AB sites. These observations match well with the results of earlier atomistic calculations [154].

While the basic physics of these systems can be understood with only considering the spatially varying stacking, in real materials, additional effects are expected to take place. For example, it is likely that there are some atomic-scale structural relaxations over the moiré pattern. This is precisely the effect that was assessed in the paper by Li et al. [165], who focussed on the twisted WSe₂/WSe₂ system and used a combination of scanning tunneling spectroscopy (STS) experiments and ab initio simulations of TMD moiré superlattices. They find a strong 3D buckling reconstruction together with large in-plane strain redistribution in their heterostructures. Using STS imaging, they identify different types of flat bands originating either from the $K$-point at the valence band edge or from the $\Gamma$-point that gives rise to more deep-lying moiré flat bands. By analyzing the origin of these flat bands in detail, it is revealed that the $K$-point flat bands are mainly a result of the deformation of the monolayer. Similar behavior can be reproduced by considering only a puckered monolayer WSe₂. On the other hand, the $\Gamma$-point flat bands are more in-line with the idea of the moiré induced, weakly coupled array of quantum dots. We will discuss the effects of periodic strain in more detail in section IV E.

The flat bands in the twisted TMD bilayers where the electron kinetic energy is suppressed are of course, fertile ground for realizing systems where interactions play a dominant role. There have been several publications on e.g. realizing different kinds of correlated states, correlated insulators and Wigner crystals in WSe₂/WSe₂ and WSe₂/W₂ moiré superlattices [166–168]. However, the moiré flat band systems can also have exciting optical effects and this has given birth to a field studying moiré excitons [169–172].

When the moiré period is larger than the exciton Bohr radius (around $\sim 1 - 2$ nm in e.g. MoSe₂ and WSe₂), the excitons will experience a spatially modulated periodic potential from the moiré. The other design parameter in heterobilayers is the relative alignment of the conduction and valence band edges, which allows the formation of inlateral excitons (e.g. WSe₂/WSe₂ system where the electron and the hole reside in the same layer [171]), interlayer excitons (e.g. MoSe₂/WSe₂ system where the electron and the hole exist in different layers [169, 170]) and hybridized excitons (e.g. MoSe₂/W₂ where the electron (for this system) is delocalized in the two layers [172]). Finally, the moiré-defined quantum dots preserve the three-fold rotational (C₃) symmetry, which implies that e.g. the interlayer excitons should inherit valley-contrastting properties [169]. These systems are currently under intense study to realize arrays of entangled quantum light emitters and realizing new exotic excitonic many-body phases (e.g. topological exciton insulator) [173, 174].

E. Generating flat bands from artificial gauge fields

A paradigmatic case of localized modes in a van der Waals material is non-uniform strained graphene (Fig. 6a). The appearance of flat bands in this system stems from the emergence of an artificial gauge field [57, 62, 178]. The effect of strain is a create a term in the system Hamiltonian that mimics a magnetic field (“pseudo-magnetic field”). However, this differs from a real magnetic field as the artificial gauge field manifests as a positive magnetic field for electrons in valley $K$ and a negative electric field for electrons in valley $K'$. That overall, the system does not break time-reversal symmetry.

The simplest instance of this is periodically rippled graphene monolayers[179–183] (Fig. 6b). The emergence of the gauge field can be easily rationalized from the graphene Hamiltonian[62]. For unstrained graphene, the low energy Hamiltonian in a single valley takes the form [184] $H = p_x \sigma_x + p_y \sigma_y$. In the presence of a global uniform strain, the Dirac point get displaced from the $K$ and $K'$ points, leading to Hamiltonian of the form $H = (p_x + A_x) \sigma_x + (p_y + A_y) \sigma_y$. Now, in a non-uniformly strained sample, we can take that there is a local strain that changes in real space, turning $A_x$ and $A_y$ spatially dependent. Noting that $\nabla$ enters in the Dirac Hamiltonian as a canonical momentum, we can then identify a strain-induced artificial magnetic field as $\tilde{B} = \nabla \times \tilde{A}$.

Twisted graphene bilayers represent another case in which spatial modulations give rise to an artificial gauge field[57, 60]. In twisted graphene bilayers, the stacking in space changes between AA, AB and BA. The modulation
FIG. 6. Sketch of the buckling of a graphene monolayer (a) and spatial profile of the induced gauge field (b) [175]. Change in the local structure of a graphene bilayer leading to a non-abelian gauge field (c) [57], and profile of the stacking in the moire unit cell (d). Panels (e,f) shows the topography (e) and dI/dV (f) of buckled graphene monolayer, showing the emergence of pseudo Landau levels [176]. Panel (g,h) shows the real-space STS (f) and dI/dV (g) of a twisted graphene bilayer at 1.8°, showing the emergence of van Hove singularities associated to non-abelian Landau levels [177].

The interlayer hopping due to the stacking brings up localized modes stemming from gauge fields, that can be rationalized as follows. A local Hamiltonian for a bilayer can be represented by a $4 \times 4$ Hamiltonian, in which the off-diagonal blocks contain the coupling between the two layers. Due to the modulated stacking (Fig. 6cd) the $2 \times 2$ interlayer coupling is modulated in space. Given the Dirac nature of the monolayer dispersion, that modulated hopping can be rationalized as an off-diagonal $2 \times 2$ gauge field, namely a non-abelian $SU(2)$ gauge field [57]. This non-abelian gauge field will thus give rise to associated pseudo Landau levels, the lowest one of them being the magic-angle flat bands at $1^\circ$ [55, 56, 60].

F. Experiments flat bands from gauge fields

The earliest experiments showing the emergence of pseudo-Landau levels appeared in non-uniform strained graphene, which naturally appear in graphene nanobubbles [185]. In these setups van Hove singularities in the density of states were shown to appear, in contrast with the semimetallic spectra of unstrained samples. The emergence of those resonances is associated to the emergent gauge field, which was shown to correspond to an effective field of 300 T [185]. These nanobubbles can also be realized with graphene deposited on a weakly interacting substrate, and in that case, the STM tip could be used to tune the strain and hence, the pseudo-magnetic field [186, 187]. Finally, suspended graphene drumheads have also been used to investigate the effects of pseudomagnetic fields and how they can confine the charge carriers in graphene [188, 189].

The buckling of graphene monolayers can also be created by choosing an appropriate substrate. In particular, recent experiments of graphene on top of NbSe$_2$ showed that graphene gets a spontaneous buckling on this structure. Associated to the buckling, a periodic non-uniform strain appears in the graphene monolayer, which gives rise to an elastic gauge field spontaneously [180, 182, 190]. Signatures of pseudo-Landau levels in this spontaneously buckled structure have been recently observed with STM [176].

The second example of pseudo-Landau levels corresponds to twisted bilayer graphene. Signatures of the lowest pseudo-Landau level, usually known as magic angle flat bands we observed early on, including some signature of symmetry breaking [177, 191, 192]. In particular, these pseudo-Landau levels show a strong localization at the AA stacking regions of the twisted bilayer [177]. Interestingly, higher index Landau levels can also lead to correlated states [193], show different localization in the moire unit cell, and in particular the next van Hove singularity shows a higher extension around the AA regions [177, 191, 192]. Subsequent experiments have further explored the nature of the lowest flat band, in particular, observing spontaneous rotational symmetry breaking due to electronic interactions [194].
V. OUTLOOK

The possibility of artificial engineering states of matter with van der Waals materials has demonstrated a huge potential in the last few years. Beyond the instances of topological insulators and superconductors, quantum spin-liquids and flat band physics, their tunability opens prospects potentially opening radical new directions in quantum matter.

Starting with topological superconductors, a challenge for future experiments will be to braid the emergent Majorana modes, in an analogous way as it has been proposed for semiconductor nanowires[17, 18]. The possibility of switching on and off topological superconductivity with local gates provides a direction for extending these schemes to two-dimensional materials. Furthermore, artificial engineering can also allow engineering higher-order topological superconductors, in which the braiding of corner modes[195, 196] can open up a potential new direction for topological quantum computing with van der Waals materials.

Quantum spin-liquids open up exciting new experimental possibilities well beyond their experimental confirmation. First, the emergence of fractional spinon excitations in these systems opens possibilities to controllable spinon transport, and ultimately its interface with current spintronic devices[197, 198]. Secondly, the emergence of anyonic excitations in certain quantum spin liquids[199, 200] motivate potential future application of these systems for topological quantum computing[200].

Flat band systems further offer novel possibilities for emergent quantum matter, going beyond the well-known possibilities for high-temperature superconductivity and symmetry broken states. In particular, the emergence of topologically non-trivial flat bands in twisted van der Waals materials provides an ideal starting point for fractional quantum Hall states in the absence of magnetic field, known as fractional Chern insulators[66, 201–204]. Analogous phenomenology for flat bands hosting spin-textured bands would further provide playgrounds for fractional quantum spin Hall physics, a state not found in nature yet. Ultimately, the combination of potential fractional quantum Hall physics and superconductivity in twisted multilayers provides an ideal starting point for engineering novel parafermion states[205, 206]. These states have resisted experimental realization so far due to the difficulty of having simultaneously fractional quantum Hall physics and superconductivity due to the large magnetic fields required. Such limitation would, however, not exist for intrinsic fractional quantum Hall states in graphene multilayers, providing an ideal solid state platform for parafermion physics.

Advances in the last few years have drastically proved the versatility of artificial engineering in van der Waals materials, revealing a variety of exotic phenomena previously only observed in rare compounds. While many of those proposals require further materials engineering and to further understand the physics of the underlying materials, the steady development of the field suggest that some of those goals may be achieved in the near future.

ACKNOWLEDGMENTS

We thank our group members - past and present - and colleagues for inspiration and insightful discussions. We acknowledge support from the European Research Council (ERC-2017-AdG no. 788185 “Artificial Designer Materials”) and Academy of Finland (Academy professor funding no. 318995 and Academy research fellow no. 331342).

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