Tunable moire spinons in magnetically encapsulated twisted van der Waals quantum spin-liquids

Guangze Chen and J. L. Lado

Department of Applied Physics, Aalto University, 02150 Espoo, Finland
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Quantum spin-liquid van der Waals magnets such as TaS$_2$, TaSe$_2$, and RuCl$_3$ provide a natural platform to explore new exotic phenomena associated with spinon physics, whose properties can be controlled by exchange proximity with ferromagnetic insulators such as CrBr$_3$. Here we put forward a twisted van der Waals heterostructure based on a quantum spin-liquid bilayer encapsulated between ferromagnetic insulators. We demonstrate the emergence of spinon flat bands and topological spinon states in such heterostructure, where the emergence of a topological gap is driven by the twist. We further show that the spinon bandstructure can be controlled via exchange proximity effect to the ferromagnetic leads. We finally show how by combining small magnetic fields with tunneling spectroscopy, magnetically encapsulated heterostructures provide a way of characterizing the nature of the quantum spin-liquid state. Our results put forward twisted quantum spin-liquid bilayers as potential platforms for exotic moire spinon phenomena, demonstrating the versatility of magnetic van der Waals heterostructures.

Magnetic van der Waals materials have risen as a highly versatile family of compounds in the two-dimensional realm[1–4]. These materials attracted much research interest as their two-dimensional nature provides a platform to electrically control magnetism[5–8], design magnetic tunnel junctions[9–12], topological superconductivity,[13, 14] and exploit magnetism in generic van der Waals heterostructures[15]. Remarkably, specific magnetic van der Waals materials such as TaS$_2$, TaSe$_2$ and RuCl$_3$ provide a realization of an exotic phase of matter, quantum spin-liquid (QSL) states[16–19].

In contrast with conventional magnets, QSL appear in magnetic systems featuring strong degrees of frustration, and are characterized by a quantum disordered ground state[20–24]. Interest in quantum-spin liquids has been fueled by their potential emergent Majorana physics[25] and their potential relation with high-temperature superconductivity[26, 27]. A variety of materials have been proposed as QSL candidates[28–40], yet it remains a remarkable challenge to unveil the nature of QSL and to experimentally identify them. Interestingly, magnetic van der Waals materials offer new directions for the engineering and detection of QSL, by exploiting the large flexibility of stacking and twisting of moire systems.

Stacking van der Waals heterostructures yields electronic structures sensitive to the relative twisting between different layers,[41, 42]. A paradigmatic example of these phenomena is twisted bilayer graphene, where the emergence of flat bands has lead to a variety of unconventional many-body states[43–47]. Interestingly, twist engineering generically provides a platform for correlated phases with electrical tunability[48–50] and topologically nontrivial electronic structures[51–56]. The versatility offered by stacked van der Waals heterostructures motivates the search for analogous phenomena in the realm of van der Waals magnets[57, 58] that can ultimately lead to novel spinon phenomena in moire quantum spin-liquids.

In this Letter, we put forward twist engineering in QSL van der Waals heterostructures as a powerful knob to control spinon physics. We show that twist engineering creates spinon flat bands at a specific twisting angle, with a topological gap opening leading to in-gap spinon edge modes. We show that the spinon spectra can be tuned by...
means of encapsulation between van der Waals magnets, leading to dramatic changes in their low energy spectra. Finally, we discuss how this exchange bias tuning provides a spectroscopic electrical method to characterize QSL states. Our results put forward magnetic van der Waals heterostructures formed by ferromagnets and QSLs as a tunable platform to explore and probe spinon phenomena in moire systems.

For the sake of concreteness, in the following, we focus on a specific gapless quantum spin liquid state on the triangular lattice, structurally analogous to the one proposed for TaS$_2$. For this sake, let us first briefly review the physics of a single-layer QSL. We start with a Heisenberg model on a triangular lattice of the form $H_0 = \sum_{i,j} J_{ij}^\mu \sigma_i^\mu \sigma_j^\mu$ where $J_{ij}^\mu$ are exchange coupling between spin components $\mu$, $\nu$ on sites $i$, $j$. The previous model is known to have a rich phase diagram and in particular it supports QSL states such as those realized in TaS$_2$[16, 59, 60], TaSe$_2$[18] and NaYbO$_2$[39, 40]. We now focus on the regime of the model yielding a QSL state with a linear density of states (DOS), and in particular the $U(1)$ Dirac spin liquid $\pi$-flux model[61, 62]. This state can be captured by performing a parrot transformation of the form $S = \frac{1}{2} f_{a}^\dagger \sigma_{a\beta} f_{\beta}$, with $f_{a}^\dagger$ and $f_{a}$ fermionic spinon operators and $\sigma_{a\beta}$ the spin Pauli matrices. With the previous replacement, the Heisenberg model can be solved at the spinon mean-field level, yielding a single-particle spinon Hamiltonian of the form $H_0 = t \sum_{i,j} \chi_{ij} f_{i}^\dagger f_{j}$, where $\chi_{ij}$ and $t$ are mean-field parameters. The $\pi$-flux model is defined by taking the mean-field solution $\chi_{ij}$ hosting an associated staggered $0$ and $\pi$ fluxes in neighboring triangles. We apply the gauge with real hoppings $\chi_{ij} = \pm 1$ such that the system has time-reversal symmetry. Under this gauge, the model has two Dirac cones in the first Brillouin zone located at time-reversal invariant momenta[63].

We move on to consider a twisted bilayer QSL as sketched in Fig.1(a). We start from the parent Heisenberg Hamiltonian for the twisted bilayer, that takes the form

$$H = \sum_{l,i,j} J_{||,ij}^\mu \sigma_{i,l}^\mu \sigma_{j,l}^\mu + \sum_{i,j} J_{\perp,ij}^\mu \sigma_{i,j,1}^\mu \sigma_{i,j,2}^\mu,$$

where $l$ labels the two layers, and $J_{||,ij}^\mu$ and $J_{\perp,ij}^\mu$ denote intra- and inter-layer spin exchange, respectively. In the regime $J_{||,ij}^\mu \gg J_{\perp,ij}^\mu$, the ground state of the system will consist on two coupled $U(1)$ QSL states. Therefore, we take as the mean-field solution for each layer the spinon $\pi$-flux model, with an effective inter-layer spinon coupling from inter-layer spin exchange:

$$H = t \sum_{l,i,j} \chi_{ij} f_{i,l}^\dagger f_{j,l} + \sum_{i,j} t_{\perp,ij}(f_{i,j,1}^\dagger f_{j,2} + h.c.),$$

where $t$, $\chi_{ij}$ and $t_{\perp,ij}$ are mean-field parameters that can be derived analogously from a mean-field replacement in Eq. (1). The mean-field parameter $t_{\perp,ij}$ will depend on the relative distance between sites $i$ and $j$, inherited from the parent Heisenberg coupling $J_{\perp,ij}^\mu$ in Eq. (1). We take a functional form for the interlayer coupling as[64] $t_{\perp,ij} = t_{\perp,0} e^{-\lambda(r_{ij}-d)}$, where $d$ is the inter-layer distance, $r_{ij}$ is the distance between sites $i$ and $j$, $\lambda$ is the parameter that controls the decay of the interlayer coupling, and $t_{\perp,0}$ is the largest possible inter-layer coupling realized at $r_{ij} = d$. In the following we take $t_{\perp,0} = 0.36t$, $\lambda = 10/a$, and $d = a$, where $a$ is the lattice constant of the triangular lattice. From the computational point of view, we will use the twist scaling relation for computational convenience[65, 66], and we compute the valley expectation $\langle \mathcal{V}_z \rangle = \pm 1$ by means of the valley operator[49, 67–70].

The $\pi$-flux hoppings $\chi_{ij}$ are subject to a $U(1)$ degree of freedom for each layer, respectively. However, the gauge difference between the two layers determines the relative position of Dirac cones of the two layers in reciprocal space[71]. As a result, the momentum difference between Dirac cones of the two layers, $\Delta k$, can be either large or small. When $|\Delta k| \gg 1/|R|$, where $R$ is the periodicity of $t_{\perp,ij}$ in real space, the Dirac cones are almost decoupled. In such case, the impact of $t_{\perp,ij}$ is small on low energy physics, keeping the two layers effectively
decoupled. In contrast, when $|\Delta k| \ll |1/R|$, $t_{\perp,ij}$ leads to significant coupling between the Dirac cones of different layers. From the energetic point of view, this gauge configuration couples the two layers and therefore will lower the many-body energy through spinon hybridization. With this gauge choice, the Moire Brillouin zone is shown in Fig.1(b), with two Dirac cones at time-reversal invariant momenta $M$ and $M'$. The spinon dispersion in the decoupled limit for twisting angle $\theta \approx 22^\circ$ is shown in Fig.1(c), where the two spinon Dirac cones of the two decoupled layers are observed.

Let us now move on to the case in which the two QSL are coupled through the interlayer exchange coupling. In this situation, we find that the interlayer coupling leads to a gap opening in the Dirac cones of the bilayer QSL upon twisting. This gap opening stemming from the twist is similar to the case of twisted double bilayer graphene[51] (Fig.2(a-c)), and stems from the broken $C_2T$ symmetry[72, 73] of the effective model. Besides the gap opening, we also observe the emergence of spinon flat bands at a specific fine tuned twisting angle $\theta/(t_{\perp}/t) \approx 2.6^\circ$, which for $t_{\perp} = 0.36t$ appears at $\theta = 0.93^\circ$ (Fig.2(d)), similar to other twisted Dirac materials[41, 42].

Interestingly, the emergence of a gap opening driven by the twist has been shown to give rise to topological states in van der Waals heterostructures based on graphene[74–76]. In particular, we find that the gap opening in the bilayer QSL has an associated valley Chern number of 2, giving rise to two counterpropagating channels at each edge with opposite valley polarization[77–81]. The previous phenomenology can be explicitly demonstrated by computing the spinon band structure of a twisted QSL nanoribbon at twisting angle $\theta = 3.5^\circ$ (Fig.2(c)). In particular, it is clearly seen the emergence of in-gap valley-polarized topological edge modes, associated with the topological valley Hall quantum spin-liquid state. We note that the topological edge modes are protected by the approximate valley charge conservation, and therefore perturbations giving rise to strong intervalley scattering can lead to intervalley mixing between topological spinon edge states[69, 70, 80, 82, 83].

After considering the emergent spinon spectra driven by the twist between the two QSL layers, we now move on to show how such emergent moiré bands can be controlled by a magnetic encapsulation. From the material science point of view, in the following we will consider that the QSL bilayer is sandwiched between two ferromagnetic insulators, for which both CrBr$_3[9, 84]$ and CrCl$_3$ would be suitable candidates. The top and bottom ferromagnets are expected to be antiferromagnetically aligned through a superexchange mechanism[85], as shown in Fig. 3(a).

To study the impact on the QSL state, we now integrate out the degrees of freedom of the ferromagnet, and consider their impact on the QSL Hamiltonian. The magnetic encapsulation yields an exchange proximity term in the Hamiltonian of the QSL bilayer, analogous to the exchange terms proposed for other van der Waals materials proximized to ferromagnets[49, 86–89]

$$\mathcal{H}' = \mathcal{H} + \sum_{i,\mu} \mathcal{J}_\perp S_{i,1}^\mu M_1^\mu + \sum_{i,\mu} \mathcal{J}_\perp S_{i,2}^\mu M_2^\mu, \quad (3)$$

where $\mathcal{J}_\perp$ denotes the exchange interaction between spin in the QSL $S_{i,1}^\mu$ and the magnetic moment of the ferromagnets $M_l^\mu$, with $l = 1, 2$ labelling the two different magnets and QSL layers. We consider sufficiently small $\mathcal{J}_\perp$ that does not lead to many-body reconstruction. In such case, the mean-field solution of Eq. (2) remains, and the effect of the ferromagnets can be projected onto the spinon mean-field Hamiltonian as

$$H' = H + \frac{1}{2} \sum_{i,\mu,l,x,s'} \mathcal{J}_\perp \sigma_{x,s'}^\mu M_l^\mu f_{i,x,l}^s f_{i,s',l}^s. \quad (4)$$

In the case of the antiferromagnetic alignment as depicted in Fig. 3(a), the magnetic encapsulation creates an effective spin-dependent inter-layer spinon bias $J = \mathcal{J}_\perp (M_1^\mu - M_2^\mu)$ on the twisted Dirac QSL[49, 68, 86].
To reveal the impact of the magnetic encapsulation, we now consider the change of the signal with respect to an in-plane magnetic field $B$, that is used to control the direction of magnetism in the magnets. The magnetic field will tune the angle between magnetization of the two magnets from $\pi$ to $\pi - \alpha(B)$, and modifies spinon DOS in the QSL bilayer due to proximity effect. For twisting angle $\theta = 0.93^\circ$, and effective exchange bias $J = 0.1t$, the spinon DOS under different $\alpha(B)$ is shown in Fig. 4(b). The modified spinon DOS exhibits peaks at different frequencies than the original one. Taking the specific case of antiferromagnetic alignment $\alpha = 0$, it is seen that the spin structure factor has peak structures, inherited from the peaks in the spinon DOS (Fig. 4(c)). Due to the modification of the spinon DOS with the field, an analogous effect is expected in the $dI/dV^2$. For this sake, we now compute the change in the $dI/dV^2$ as a function of the magnetic field, defined as $\Delta S(\omega, \alpha) = S(\omega, \alpha) - S(\omega, 0)$, and shown in Fig. 4(d). In particular, we see in Fig. 4(d) the existence of deeps and peaks in the differential $dI/dV^2$, manifesting from the dramatic change of spinon DOS with the magnetic field of Fig. 4(b).

Finally, we comment on specific quantitative aspects of our proposal relevant for experiments. First, in our manuscript, we have considered exchange couplings between the two monolayer QSL on the order of $J_1 \approx 0.3t_1$. The specific prediction of such exchange coupling should be performed via first principle methods for the specific materials considered[95, 96], and could ultimately be controlled with pressure[97, 98]. For our phenomenology, a change in the exchange coupling just drifts the physics towards bigger or smaller angles, yet without qualitatively changing the overall behavior[99]. Second, when exchange proximity is considered, the exchange proximity must be smaller than the intralayer exchange, yet without perturb the QSL ground state. Third, in order to tilt the direction of the magnets by an external magnetic field, yet without breaking the QSL ground state, a soft magnetic axis is preferred. As a reference, taking CrBr$_3$ as the ferromagnet, the anisotropy energy can be overcome with a magnetic field of around 1 T[100], whose Zeeman energy scale of 0.04 meV is not expected to perturb QSL with exchange constants on the order of 5 meV. The application of a magnetic field is also expected to affect the magnons of the ferromagnetic encapsulation, yet those states will contribute with a uniform background to the $dI/dV^2$ signal that can be subtracted[9, 10]. Finally, although our analysis has focused on a Dirac QSL, analogous calculations can be performed with other QSL ground states, such as the gaped QSL of RuCl$_3$[25].

To summarize, we have shown that a van der Waals heterostructure based on a magnetically encapsulated bilayer QSL allows designing controllable spinon physics, and ultimately detecting moire spinons. We showed that
a twisted bilayer QSL gives rise to a topological gap opening purely driven by the twist, and in fine-tuned regimes spinon flat bands. Furthermore, we showed that encapsulating the twisted QSL bilayer with ferromagnets produces exchange bias via proximity effect, which allows to magnetically control the spinon bandstructure. Based on such magnetic tunability, we proposed an experimental identification of QSL phases, utilizing field-controlled magnetism in magnetic van der Waals materials as well as inelastic spectroscopy. Our results put forward twist engineering as a means of exploiting exotic spinon phenomena in quantum spin-liquids, highlighting the versatility of magnetic van der Waals heterostructures to explore emergent spinon phenomena.

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